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Persistent Organic Chemicals in the Irish Waste Stream

Authors: Stuart Harrad, Martin Sharkey, Daniel Drage, William Stubbings, Marie Coggins and Harald Berresheim



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- 5. Office of Communications and Corporate Services

The EPA is assisted by advisory committees who meet regularly to discuss issues of concern and provide advice to the Board.



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Identifying pressures

The EU is transitioning to a circular economy, in which resources are kept in use for as long as possible, for example by maximising recycling. A potential obstacle is the presence of regulated brominated persistent organic pollutants (POPs) in waste plastics, as articles containing recycled material may be contaminated with such chemicals. To minimise contamination, there are limits on the maximum permissible concentrations of POPs in waste, such that material exceeding these limits may not be recycled. Effective implementation of such limits presents enormous technical and economic challenges, as conventional methods for measuring POPs are technically demanding and expensive. This project evaluated the feasibility of using hand-held X-ray fluorescence (XRF) spectrometers to measure bromine in waste articles to check compliance with limit values. It also generated an extensive database on concentrations of POPs and related chemicals in waste plastic articles in Ireland. This permits assessment of whether recent bans on the use of some POPs has reduced their presence in Irish waste and establishes a baseline against which the success of possible future restrictions on other chemicals may be evaluated.

Informing policy

This research identifies ways to improve the effectiveness of hand-held XRF spectrometers to provide a faster, less expensive way of checking whether waste articles comply with limits on brominated POPs. Moreover, it provides the first data on per- and polyfluoroalkyl substances (PFAS) and chlorinated organophosphate esters (CI-OPEs) in the Irish waste stream. These data reveal very few exceedances of permissible limit concentrations of PFAS. In contrast, this research also shows that, if a limit on waste of 1000 mg/kg were to be introduced for CI-OPEs, a substantial proportion of articles tested would exceed this limit. Analysis of waste childcare articles like car seats reveals that some exceed existing and potential limits on brominated POPs and CI-OPEs. These data can help inform choices on materials used in childcare articles. Concentrations and limit value exceedances for brominated POPs in Irish waste have either declined or remained similar since 2015–2016. The impact of new, lower limits on concentrations of brominated POPs was evaluated. Although lowering limits prevents more brominated POPs from entering the recycling stream, it increases the mass of unrecyclable waste.

Developing solutions

Based on this research, the following recommendations are made.

The duration of XRF measurement of bromine to screen waste articles for compliance with limits on concentrations of brominated POPs should be conducted over a single 5-second period. This would substantially increase the rate at which waste articles may be checked, with minimal reduction in the accuracy with which XRF correctly identifies articles exceeding the limit.

Alongside measurements of total organic fluorine, future studies should measure a wider range of PFAS to ensure that articles containing elevated concentrations of PFAS are not overlooked.

A limit of 1000 mg/kg should be placed on concentrations of Cl-OPEs in waste articles above which such articles cannot be recycled. This will substantially reduce the quantity of these chemicals entering the recycling stream.

Further monitoring of concentrations of brominated POPs, CI-OPEs and PFAS in waste (including childcare articles) should be conducted to fully evaluate the impact of legislation designed to eliminate these chemicals from the waste stream.

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This report is based on research carried out between February 2019 and June 2022. More recent data may have become available since the research was completed.

The EPA Research Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.

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Executive Summary

Given their persistence and ability to bioaccumulate and cause adverse health effects, use of some halogenated flame retardants (HFRs) and per- and polyfluoroalkyl substances (PFAS) has been restricted. Selected HFRs and PFAS were measured in samples (n=743) collected from five broad categories of waste: construction and demolition extruded and expanded polystyrene foam (EPS/XPS) (n=25); endof-life vehicle (ELV) fabrics and foams (n = 111); soft furnishings (n = 124); waste electrical and electronic equipment (WEEE) (n=210); and childcare articles (n=273; child car seats, cot mattresses, changing mats, pushchairs, prams, etc.). No item exceeded the EU Persistent Organic Pollutants (POPs) Regulation limit of 50 mg/kg for perfluorooctane sulfonate. However, in 19/308 samples (3.7%), concentrations of at least one PFAS exceeded 10 mg/kg, and in 3/308 (1%), concentrations exceeded the new EU POPs Regulation limit of 1 mg/kg for perfluorooctanoic acid. No article exceeded the new EU POPs Regulation limit of 1 mg/kg for perfluorohexane sulfonate.

Compared with a previous study conducted in Ireland in 2015–16, concentrations and exceedances of limits for polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBP-A) were either similar or had declined in most waste categories. To illustrate, fewer samples exceeded limits for PBDEs and HBCDD in 2019–20 (7.8%) than in 2015–16 (8.7%).

Of the waste childcare article foam and fabric samples, 15/187 (8.0%) exceeded the limit of 1000 mg/kg for PBDEs, 15 (8.0%) exceeded the limit for HBCDD and 6 (3.2%) exceeded the limit for both. An even greater proportion contained concentrations > 1000 mg/kg of tris(1-chloro-2-propyl) phosphate (TCIPP) (75/273, 27%) and tris(1,3-dichloro-2-propyl) phosphate (TDCIPP) (58/273, 21%), with concentrations also > 1000 mg/kg for tris(2-chloroethyl) phosphate (TCEP) (14/273, 5.1%), 2-ethylhexyl tetrabromobenzoate (7/187, 3.7%), decabromodiphenyl ethane and bis(2-ethylhexyl)tetrabromophthalate (both 5/187, 2.7%). An estimated 10,200 kg of PBDEs, HBCDD and TBBP-A entered the Irish waste stream in nonchildcare waste articles in 2019, alongside an estimated 74,000 kg of TDCIPP, 70,000 kg TCIPP and 3000 kg TCEP. The 1000 mg/kg limit for PBDEs, HBCDD and TBBP-A renders unrecyclable approximately 2800 t waste (3.1% of the ≈90,000 t waste generated). By comparison, ≈7900 kg of PBDEs, HBCDD and TBBP-A (78% of that generated annually) are prevented from entering the recycling stream. Lowering the limit to 500, 200 and 100 mg/kg would prevent an estimated 82%, 84% and 85% of PBDEs, HBCDD and TBBP-A entering the recycling stream but increase the annual mass of unrecyclable waste to 4.0%, 4.9% and 5.6% of the total generated. Introducing a 1000 mg/kg limit on TDCIPP, TCIPP and TCEP would render unrecyclable 7200 t/year (24%) of waste building insulation foam and ELV and furniture foams and fabrics but prevent 144,000 kg (98% of the total) of these HFRs entering the recycling stream.

We explored ways of increasing the rate of sample testing using portable X-ray fluorescence (XRF) measurement of bromine without impairing its ability to correctly identify waste exceeding limit values for PBDEs and HBCDD. In 2015-16, 6.3% of non-childcare waste articles were "false exceedances", i.e. XRF incorrectly indicated that the limit was exceeded. In 2019-20, the incidence of false exceedances increased to 10%. While there were no "false negatives" in 2015-16 - i.e. XRF incorrectly indicating that the limit was not exceeded nine (1.9%) samples were false negatives in 2019-20. Decreasing the duration of XRF measurement of bromine to a single 5-s period increased estimated sample throughput to ≈180,000 articles/year, with minimal reduction in the accuracy with which XRF correctly identifies articles exceeding the limit.

Based on our findings, we recommend:

 Future studies should measure a wider range of PFAS alongside total organic fluorine, to identify articles containing elevated concentrations of PFAS.

- The impact of lowering the limit for PBDEs and HBCDD should be considered. While lowering the limit prevents more brominated flame retardants entering the recycling stream, it increases the mass of unrecyclable waste.
- Placing a limit of 1000 mg/kg on TDCIPP, TCIPP and TCEP in waste articles above which such articles cannot be recycled.
- Further monitoring of concentrations of HFRs and PFAS in waste to fully evaluate the impact of legislation designed to eliminate these chemicals from the waste stream.
- Ongoing monitoring of HFR and PFAS concentrations in waste childcare articles.

1 Introduction

1.1 Sources and Applications of HFRs and PFAS

A number of halogenated chemicals have found extensive use worldwide as halogenated flame retardants (HFRs) in a wide variety of commercial, domestic and industrial applications. HFRs are either brominated or chlorinated. With respect to brominated flame retardants (BFRs), the most widely used are polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and tetrabromobisphenol-A (TBBP-A) (BSEF, 2003). Applications of PBDEs include electrical and electronic equipment (EEE - e.g. televisions, personal computers, small domestic appliances (SDAs)) and soft furnishings (e.g. sofas, mattresses, curtains, pillows). In the former case, PBDEs are added both to the polymer casing for electronics (e.g. high-impact polystyrene or acrylonitrile butadiene styrene) and to internal circuit boards. In the latter case, they are added both to the foam fillings and to the fabric covers of soft furnishings, such as sofas and chairs in domestic, office or vehicular environments. With respect to HBCDD, the most important application (i.e. 96% of its use in the EU) is its widespread use as a flame retardant (FR) in expanded polystyrene (EPS) and extruded polystyrene (XPS), which are used in building insulation foam in the construction industry (ECHA, 2009), with some minor application in plastic housing for electronics (Weil and Levchik, 2008) and in furniture fabrics (Drage et al., 2018). Applications of TBBP-A are predominantly in the plastic casings of EEE (Abdallah et al., 2008). As of 2001 (the last reliable figures publicly available), Europe accounted for 2%, 16%, 14%, 57% and 9.7% of the annual global demand for penta-bromodiphenyl ether (BDE), octa-BDE, deca-BDE, HBCDD and TBBP-A, respectively (BSEF, 2003). As a result of legislative restrictions on the manufacture and new use of PBDEs and HBCDD (see section 1.3), use of so-called "novel" brominated flame retardants (NBFRs) as substitutes is likely to have increased. Examples of NBFRs include 2-ethylhexyl tetrabromobenzoate (EH-TBB), decabromodiphenyl ethane (DBDPE) and bis(2-ethylhexyl)tetrabromophthalate (BEH-TEBP). Of particular relevance to this project, concentrations

of DBDPE in indoor air and dust from Ireland exceed those reported elsewhere, suggesting its widespread use as a "drop-in" replacement for deca-BDE (Wemken *et al.*, 2019). With respect to EH-TBB and BEH-TEBP, these NBFRs have been used widely as FRs in polyurethane furniture foam and, although Irish data do not exist, both have been detected in >90% of samples of UK indoor air and dust (Tao *et al.*, 2016).

In addition to BFRs, chlorinated organophosphate esters (CI-OPEs), specifically tris(2-chloroethyl) phosphate (TCEP), tris(1-chloro-2-propyl) phosphate (TCIPP) and tris(1,3-dichloro-2-propyl) phosphate (TDCIPP), have been used extensively as FR additives in applications such as rigid foams used in construction blocks and panels used for building insulation purposes, and in flexible polyurethane foam (PUF) for sofas, chairs, vehicle seating and mattresses (Cooper *et al.*, 2016; European Union, 2008a; Marklund *et al.*, 2003; Stubbings *et al.*, 2016). Within Europe, total annual usage of phosphorous FRs (including CI-OPEs) is an estimated 89,640t (PINFA, 2017).

Per- and polyfluoroalkyl substances (PFAS) are a class of thousands of chemical substances. These chemicals have been produced since the 1950s and still find use in a wide variety of industrial applications and consumer products. The perfluoroalkyl moiety is both chemically and thermally stable and has hydrophobic and lipophobic properties. This makes this chemical class very useful and enduring when incorporated as surfactants in firefighting foams and coatings and as polymers used in textiles and food packaging materials (Buck *et al.*, 2011; Glüge *et al.*, 2020).

1.2 Toxicity of HFRs and PFAS

Human exposure to some BFRs has been associated with many adverse effects, such as endocrine disruption, liver microsomal enzyme induction, immunotoxicity, neurotoxicity and carcinogenicity (Darnerud, 2008; Vonderheide *et al.*, 2008). Animal studies have further shown neurodevelopmental and behavioural outcomes of exposure to PBDEs, such as hepatic abnormality, endocrine disruption and possibly cancer (Birnbaum and Staskal, 2004; Darnerud, 2008; Hakk, 2010; Wikoff and Birnbaum, 2011). In animals, HBCDD was found to induce hepatic cytochrome P450 enzymes and alter the normal uptake of neurotransmitters, while in humans HBCDD has been reported to trigger cancer through non-mutagenic mechanisms and disruption of the thyroid hormone system (Covaci et al., 2006; Darnerud, 2008; Law et al., 2005). As a consequence of the health concerns about PBDEs and HBCDD, a variety of jurisdictions around the world have evaluated the risk to human health, leading in some instances to health-based limit values. The European Food Safety Authority (EFSA, 2011a,b) has delivered scientific opinions that recommend benchmark doses for a number of PBDEs and HBCDD and concluded that, in the EU, dietary exposure to HBCDD and BDE-47, BDE-153 and BDE-209 was not of concern. However, the lower benchmark dose for BDE-99 (12µg/kg body weight (bw)/day) raised a potential health concern. Outside the EU, the United States Environmental Protection Agency (USEPA) has promulgated reference doses for some PBDEs. Specifically, these values are 100 ng/kg bw/day for both BDE-47 and BDE-99 (USEPA, 2008a,b), with a higher value (7000 ng/kg bw/day) for decabromodiphenyl ether (BDE-209) (USEPA, 2008c).

With respect to TBBP-A, studies on laboratory animals indicate that it may elicit adverse health effects, including immunotoxicity, neurotoxicity and disruption of the endocrine system (Zhou *et al.*, 2014).

Turning to NBFRs, the USEPA's 2014 assessment of alternatives to deca-BDE rates DBDPE as a similarly high hazard to deca-BDE with respect to developmental toxicity (USEPA, 2014). This is noteworthy, as the endpoint that drives USEPA's reference dose for deca-BDE is neurodevelopmental toxicity. Moreover, animal studies suggest other health risks associated with exposure to NBFRs, including endocrine disruption (DBDPE, EH-TBB, BEH-TEBP), neurodevelopmental and behavioural effects (EH-TBB, BEH-TEBP), hepatotoxicity (DBDPE), impaired reproductive physiology (DBDPE) and DNA damage (EH-TBB, BEH-TEBP) (Bearr et al., 2010; Egloff et al., 2011; Ezechiáš et al., 2012; Johnson et al., 2013; Larsson et al., 2006; Mankidy et al., 2014; McGregor et al., 1991; Nakari and Huhtala, 2010;

Noyes *et al.*, 2015; Park *et al.*, 2011; Pradhan *et al.*, 2013; Saunders *et al.*, 2013; Wang *et al.*, 2010).

Regarding CI-OPEs, human epidemiological studies suggest that exposure to TCIPP and TDCIPP adversely impact human hormone levels and semen quality parameters (Meeker and Stapleton, 2010). Related to this, in 2018 the European Chemicals Agency (ECHA) identified a risk to children from exposure to TCEP, TCIPP and TDCIPP (ECHA, 2018), proposing a ban on the use of these CI-OPEs in childcare articles and residential upholstered furniture. The proposal is now suspended and awaiting evaluation of the carcinogenicity of TCIPP by the US National Toxicology Program (ECHA, 2022a).

A recent report by EFSA highlighted a number of possible adverse human health effects of PFAS. The liver plays a part in the reabsorption of PFAS. Thus, there have been studies reporting adverse effects of PFAS on liver function (EFSA CONTAM Panel, 2020). Moreover, the EFSA CONTAM Panel found that "there may well be a causal association between perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) and birth weight" (EFSA CONTAM Panel, 2020). Another significant health effect of PFAS in humans is reduced antibody response to vaccination. Studies on birth cohorts from the Faroe Islands found negative correlations between vaccine antibodies against diphtheria and tetanus at ages 5 and 7 years and concentrations of PFAS in both maternal pregnancy and children's serum at 5 years (Grandjean et al., 2012, 2017). Similarly, Abraham et al. (2020) conducted a study on samples from a German cohort collected in the late 1990s and found significant associations between concentrations of PFOA in serum and adjusted levels of vaccine antibodies against tetanus, diphtheria and Haemophilus influenzae type b. Studies on highly exposed cohorts have also linked PFAS exposure and endocrine disruption (e.g. thyroid toxicity and some cancers) (Barry et al., 2013; Winquist and Steenland, 2014). A different study on a large cohort found positive associations between serum PFOA concentrations and kidney and testicular cancers (Barry et al., 2013). Nevertheless, as noted recently, both these adverse effects are detected at only high levels of exposure, and there is insufficient evidence to suggest an association between PFAS exposure at lower levels and cancers or thyroid disease (EFSA CONTAM Panel, 2020). Based on these findings, in

September 2020, EFSA proposed a group tolerable weekly intake of 4.4 ng/kg bw/day for the sum of PFOA, PFOS, perfluorononanoic acid (PFNA) and perfluorohexane sulfonate (PFHxS). EFSA also noted various reports suggesting that PFAS could have a wider impact on human health, but further studies are needed to confirm these findings (EFSA CONTAM Panel, 2020).

1.3 International Action to Limit Environmental Health Impacts

Over the last decade, the widespread use of PBDEs and HBCDDs has been a subject of concern, owing to their documented presence in the environment, including human tissues, coupled with evidence of their toxicity, as outlined in section 1.2. At a global level, this concern is exemplified by the listing of HBCDD and the penta-, octa- and deca-BDE commercial formulations under the United Nations Environment Programme Stockholm Convention on Persistent Organic Pollutants (POPs). Within the EU, manufacture and new use of penta- and octa-BDE has been banned since the mid-2000s, with that of deca-BDE restricted severely since 2008. Since the 2013 listing of HBCDD under the Stockholm Convention, use of this BFR has also been restricted, although there was a time-limited derogation that permitted its use within the EU in EPS and XPS for building insulation up to the end of 2018.

Likewise, the strong carbon-fluorine bond means that PFAS are resistant to thermal, chemical and biological degradation (Kissa, 2001) and are capable of bioaccumulation and long-range environmental transport, exemplified by their detection in the Arctic (Chaemfa et al., 2010). As a result, at the time of writing, the following PFAS are listed under the Stockholm Convention on POPs. Specifically, PFOS and its salts and perfluorooctane sulfonyl fluoride are listed in Annex B (Restriction), while PFOA, its salts and PFOA-related compounds (including precursor compounds) are listed in Annex A (Elimination). Moreover, following the recommendation of the POPs Review Committee (POPRC), the 10th meeting of the Conference of the Parties to the Stockholm Convention in June 2022 listed PFHxS, its salts and PFHxS-related compounds in Annex A to the convention, setting them for elimination, with no exemptions. POPRC is also currently considering

listing the following under the Stockholm Convention: the FR chemical dechlorane plus (DP), as well as long-chain perfluorocarboxylic acids (PFCAs), their salts and related compounds (specifically the C_g-C_{21} analogues of PFOA) (POPRC, 2022). In addition, perfluorobutane sulfonate (PFBS), PFHxS, PFOA, PFNA and perfluorodecanoic acid (PFDA) are listed under REACH (Regulation on Registration, Evaluation, Authorisation and Restriction of Chemicals) as substances of very high concern recommended for restriction (ECHA, 2022b). Furthermore, it is anticipated that ECHA will receive a proposal to place a "group restriction" on PFAS from several EU Member States in 2023.

1.4 Environmental Hazard Presented by Waste Containing HFRs and PFAS

Despite these recent and proposed restrictions, many items/products containing HFRs and PFAS remain in use. Moreover, in view of the turnover times of such articles, it is further apparent that there is a growing inventory of materials containing restricted chemicals that have entered or will shortly be entering the waste stream. To illustrate, one study estimated that 4200, 2651, 40,428 and 20,949t of EPS/XPS building insulation foam, end-of-life vehicle (ELV) fabrics/ foams, waste electrical and electronic equipment (WEEE), and soft furnishing fabrics and foams, respectively, were generated annually in Ireland (Drage et al., 2018). The massive scale on which such waste, which may contain HFRs and PFAS, has already entered and is expected to enter the waste stream over the next 5 years and beyond requires detailed research studies to generate the knowledge base needed to allow regulatory bodies to formulate effective policies to address this important issue.

In "Manifesto for a Resource-Efficient Europe" (EREP, 2012), the EU, in keeping with other jurisdictions, recognised that it has no choice but to transition to a resource-efficient and ultimately regenerative circular economy. An alternative to a traditional linear economy, a circular economy is one in which resources are kept in use for as long as possible. Maximum value is extracted from resources while in use, with products and materials recovered and regenerated at the end of each service life. While a highly desirable objective, implementation of a circular

economy is not entirely straightforward. One important consideration is that the presence of HFRs and related contaminants, such as PFAS, in plastic components of waste consumer products, such as electronics and building insulation, as well as furniture fabrics and foam, presents a potential (yet surmountable) barrier to the ongoing use, re-use and recycling of such waste products. Indeed, evidence exists that uncontrolled recycling of waste polymers containing HFRs leads to the unintentional presence of such chemicals in articles in which their presence is not required, such as food contact materials, children's toys and polystyrene packaging (Abdallah et al., 2018; Alghamdi et al., 2022; Guzzonato et al., 2017; Kuang et al., 2018; Leslie et al., 2016; Puype et al., 2015; Turner, 2018). In recognition of this, the EU has implemented low persistent organic pollutant concentration limit (LPCL) values for PBDEs and HBCDD, which forbid recycling of waste polymers containing such chemicals at concentrations exceeding 1000 mg/kg (European Union, 2019). Moreover, the EU classifies TBBP-A as both an H400 ("very toxic to aquatic life") and an H410 ("very toxic to aquatic life with long lasting effects") substance. Consequently, waste that contains TBBP-A above 1000 mg/kg is classified as "HP 14 - Ecotoxic", treated as hazardous waste and cannot be recycled (ECHA, 2022c; European Union, 2008b). With respect to PFAS, the EU specifies an LPCL value for PFOS of 50 mg/kg (European Union, 2019), with LPCL values for other PFAS in waste not specified. Moreover, although CI-OPEs are not under consideration for listing as POPs, given evidence that they are also entering new articles as unintentional contaminants (Alghamdi et al., 2022), combined with ECHA's restriction proposal (ECHA, 2018), it is not unrealistic to anticipate that similar limits will be placed on concentrations of TCEP, TCIPP and TDCIPP in waste to prevent their recycling. In a similar vein, the EU has agreed provisionally to lower the LPCL values for PFOA, HBCDD and PBDEs, and introduce an LPCL for PFHxS over the next few years (European Council, 2022).

The WAFER project reported concentrations of PBDEs, HBCDD and TBBP-A in ≈550 samples of waste polymeric materials collected from various sites in Ireland between 2015 and 2016 (Drage *et al.*, 2018; Harrad, 2018; Harrad *et al.*, 2019a). Importantly, we found that 8.7% of articles analysed in the WAFER project exceeded the LPCL value of 1000 mg/kg for

PBDEs and HBCDD. As the chromatographic-mass spectrometric techniques required to measure PBDEs, HBCDD and other HFRs are expensive (typically €500/sample), time-consuming and require expert operators, the WAFER project also evaluated whether portable X-ray fluorescence (XRF) measurements of bromine were a viable technique for screening waste for compliance with LPCL values for PBDEs and HBCDD. One of the key findings of the WAFER project was that, for a proportion of samples, XRF recorded a "false exceedance" of the LPCL. Specifically, while the XRF measurement of bromine indicated that the LPCL was exceeded, this was not supported by the corresponding measurements of PBDEs and HBCDD. While a substantial proportion of the false exceedances could be attributed to concentrations >1000 mg/kg of TBBP-A, the WAFER project concluded that many of the other false exceedances were probably caused by concentrations > 1000 mg/kg of NBFRs, which were not measured in the study. The WAFER project hypothesised that, as a consequence of the restrictions on PBDEs and HBCDD, while genuine exceedances of the LPCL for these BFRs would decline over time without concomitant relaxations in fire safety regulations, increased use of NBFRs such as DBDPE was likely. As a result, unless such NBFRs are also regulated via LPCLs, such a trend would lead to an increased incidence of false exceedances.

While such false exceedances could be viewed as an acceptable compromise, as they would prevent recycling of items containing TBBP-A and NBFRs, reducing the frequency of false exceedances is desirable. In this regard, antimony trioxide is known to be used with BFRs as a synergist during the manufacturing/treatment process. It has been suggested that antimony trioxide use is largely confined to PBDEs and thus the use of XRF measurements of elemental antimony has been proposed as a means of differentiating between genuine and false LPCL exceedances (Guzzonato et al., 2017). A previous study suggested that detection of bromine alongside antimony at ratios between ≈2:1 and 4:1 would confirm that concentrations of bromine measured by XRF that exceeded the LPCL in hard plastic samples were attributable to the presence of PBDEs, while this would not be observed if the bromine detected was due to other BFRs.

A further issue of relevance is that, although now suspended pending publication of the outcome of an evaluation of the carcinogenicity of TCIPP by the United States Toxicology Program (ECHA, 2022a), in 2018 ECHA proposed a ban on the use of the following CI-OPEs in childcare articles: TCEP, TCIPP and TDCIPP (ECHA, 2018). While, to our knowledge, concentrations of these CI-OPEs and other HFRs have not previously been measured in childcare articles in the EU, evidence from the USA suggests their widespread presence in cot mattresses and child car seats (Cooper *et al.*, 2016; Stapleton *et al.*, 2011).

1.5 Objectives

Against the preceding backdrop, the research objectives of this project were:

 to quantify the mass of target HFRs and PFAS in various components of the Irish waste stream (including childcare articles), thereby providing regulators with sound scientific information on the scale and precise nature of the issue (e.g. which HFRs and PFAS are prevalent in which waste material categories);

- to evaluate the efficacy of recent restrictions on PBDEs and HBCDD in reducing the presence of these contaminants in the Irish waste stream;
- to test the hypothesis that concentrations of NBFRs in Irish waste have increased since 2016, both in absolute terms and relative to those of PBDEs and HBCDD, and that, as a consequence, the incidence of false exceedances of LPCL values for PBDEs and HBCDD indicated by XRF measurements of bromine will have increased concomitantly;
- to test the hypothesis that knowledge of antimony:bromine ratios in hard plastic samples containing bromine at concentrations that indicate the LPCL for PBDEs is exceeded will assist evaluation of whether or not the exceedance is genuine.

2 Methods

In this chapter, we describe the sampling strategy and technical methods employed to obtain the data reported here.

2.1 Sampling

The first phase involved collection of samples of individual articles of waste goods and materials from waste streams considered to potentially contain HFRs and PFAS. As one purpose of this project was to provide samples that would facilitate comparison with PBDE, HBCDD and TBBP-A concentrations detected in the earlier WAFER project, our sampling strategy was designed to match the strategy followed in the WAFER project. We also highlight that samples collected were those of convenience, and this should be borne in mind when drawing conclusions from our data.

Samples were collected over 2019 and 2020. Table 2.1 summarises the numbers of different sample types collected as part of this project that were subjected to analysis for concentrations of HFRs, PFAS, bromine, chlorine and phosphorus. Further details regarding the collection of samples from each waste stream are provided below. Table 2.1 also includes the numbers of samples from each category collected in 2015–16 for which concentrations of PBDEs, HBCDD and TBBP-A were reported as part of the WAFER project. A smaller number of archived samples from the WAFER project were analysed for concentrations of NBFRs, CI-OPEs and PFAS as part of the current project.

2.1.1 Sample collection

2.1.1.1 Non-childcare articles

Samples were collected from several waste-handling facilities located in Ireland between 2019 and 2020. The sampling campaign addressed waste streams considered most likely to contain products treated with BFRs. To facilitate elucidation of temporal trends in concentrations of BFRs, the waste stream categories examined and the number of samples collected from each category were matched as closely as possible with the WAFER project conducted in 2015–16. A

total of 470 samples were collected from four broad categories of waste: construction and demolition (C&D) XPS and EPS foam (n=25), ELV fabrics and foams (n=111), soft furnishings (n=124) and WEEE (n=210). These four categories were further divided, as detailed in Table 2.1. Figure 2.1 shows (along with examples of childcare articles collected – see section 2.1.1.2) an ELV seat with both a fabric and a foam sample removed. Also shown in Figure 2.1 are fabric samples stored in sealed plastic bags.

During the WAFER project, samples were collected between 2015 and 2016 from a broad range of sites nationwide to assess any regional variation in BFR concentrations in the waste categories examined. No such variances were observed, and therefore all samples in this project were collected from waste collection or waste transfer sites within the County Galway region. These sites included three ELV scrapyards, two major recycling/waste transfer sites that process a wide range of household, commercial and C&D wastes, and a selection of construction/ demolition sites where some EPS/XPS samples were collected. Hard plastic samples were taken from various WEEE items representative of the major plastic component (i.e. largest surface area) of the item. The WEEE items included information technology (IT) and telecommunications equipment, large household appliances (LHAs), SDAs, display items and cooling equipment (i.e. fridges and freezers). Fabrics/upholstery and directly underlying PUFs, along with any other cushioning materials (wool, additional fabrics, etc.), were taken from ELV, furniture and mattress samples.

Although concentrations of PBDEs, HBCDD and TBBP-A are already available for samples from the WAFER project (Drage *et al.*, 2018; Harrad, 2018), we also measured concentrations of NBFRs in archived samples collected as part of the WAFER project for which sufficient material remained available for analysis. For NBFRs, concentrations were measured in the following archived samples from the WAFER project: carpets (n=29), curtains (n=11), ELV fabrics and foams (n=25) and WEEE (n=12).

		Number of	samples anal	ysed for
Category	Sub-category	BFRs	CI-OPEs	PFAS
Construction and demolition	EPS	12 (40)	12 (0)	0 (0)
	XPS	13 (20)	13 (0)	0 (0)
ELV upholstery foams and fabrics		111 (119)	111 (70)	111 (25)
Soft furnishings	Carpets	20 (31)	20 (29)	20 (29)
	Curtains	25 (15)	25 (14)	25 (11)
	Furniture fabrics	16 (22)	16 (15)	15 (0)
	Furniture foam filling	16 (20)	16 (12)	17 (0)
	Mattress foam filling	27 (17)	40 (10)	27 (0)
	Mattress fabric covering	20 (17)	20 (6)	20 (0)
WEEE	Large household appliances	21 (57)	0 (0)	0 (0)
	Cooling appliances	30 (30)	0 (0)	0 (0)
	Display	47 (43)	0 (0)	0 (0)
	Small domestic appliances	60 (29)	0 (0)	0 (0)
	Information technology and telecommunications	52 (78)	0 (0)	0 (0)
Childcare	Child car seat foam	35 (0)	62 (0)	26 (0)
	Child car seat fabrics	53 (0)	88 (0)	48 (0)
	Pushchair foam	2 (0)	2 (0)	0 (0)
	Pushchair fabric	32 (0)	44 (0)	0 (0)
	Pram foam	4 (0)	5 (0)	0 (0)
	Pram fabrics	18 (0)	19 (0)	0 (0)
	Cot mattress foam	6 (0)	6 (0)	0 (0)
	Cot mattress fabric	14 (0)	15 (0)	0 (0)
	Change mat foam	7 (0)	7 (0)	0 (0)
	Change mat fabrics	16 (0)	16 (0)	0 (0)
	Miscellaneous childcare article foam	0 (0)	1 (0)	0 (0)
	Miscellaneous childcare article fabrics	0 (0)	9 (0)	0 (0)

Table 2.1. Number of waste items collected in 2019–20 (2015–16 figures in parentheses) and analysed for HFRs and PFAS

2.1.1.2 Childcare articles

Samples of a variety of childcare articles (examples illustrated in Figure 2.1) were collected from several waste-handling facilities, as well as from individual donors of unwanted articles located in Ireland between 2019 and 2020. A total of 273 samples were collected from five broad categories of waste childcare articles: car seats, pushchairs, prams, cot mattresses and change mats. These categories were further divided (see Table 3.1) based on the materials collected, which included overlaying fabrics (polyurethane and polyvinylchloride) and filling foams (PUFs, wool and EPS).

For most samples, overlaying fabric samples were collected along with underlying cushioning foams directly beneath the fabric samples. Multiple foam samples were taken if various filling materials were present. For pushchairs, little cushioning materials were present; thus, samples of fabrics and (if available) foam cushioning were collected using the same methods. In the case of car seats, additional layers of EPS foam were present between a rigid plastic frame as an additional safety feature, and samples of these foams were also collected for analysis.



Figure 2.1. Examples of items analysed in this study.

2.2 Methods for Determination of Concentrations of HFRs and PFAS

The HFRs targeted in this study were PBDE-28, -47, -99, -100, -153, -154, -183, and -209, α -, β - and γ -HBCDD, TCEP, TCIPP, TDCIPP, DBDPE, tetrabromoethylcyclohexane (TBECH), 1,2-bis(2,4,6-tribromophenoxy)ethane, 2,4,6-tris(2,4,6tribromophenoxy)-1,3,5-triazine (TTBP-TAZ), pentabromobenzene, hexabromobenzene, pentabromotoluene and 2,3-dibromopropyl-2,4,6tribromophenyl ether, as well as the chlorinated FRs *anti*-DP and *syn*-DP.

Target PFAS in this study were PFBS, perfluoropentane sulfonate (PFPeS), PFHxS, perfluoroheptane sulfonate (PFHpS), PFOS, perfluorononane sulfonate (PFNS), perfluorodecane sulfonate (PFDS), perfluoroundecane sulfonate (PFuDS), perfluorododecane sulfonate, perfluorotridecane sulfonate (PFTrDS), perfluorobutanoic acid (PFBA), perfluoropentanoic acid, perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, PFNA, PFDA, perfluoroundecanoic acid (PFuDA), perfluorododecanoic acid (PFDoA), perfluorotridecanoic acid (PFTrDA), perfluorooctane sulfonamide (FOSA) and its methyl and ethyl derivatives (i.e. methyl perfluorooctane sulfonamide (MeFOSA) and ethyl perfluorooctane sulfonamide (EtFOSA)), as well as methyl perfluorooctane sulfonamidoethanol (MeFOSE) and ethyl perfluorooctane sulfonamido ethanol (EtFOSE). These PFAS were targeted on the basis that the C₄–C₁₃ PFCAs and perfluorosulfonic acids are specified under the EU Drinking Water Directive, with others included because of their previous detection in indoor air and dust in Ireland (Harrad et al., 2019b). Our list of target PFAS expanded through the lifetime of the project, accounting to some extent for new information and related legislation (e.g. the Drinking Water Directive). As stated in our recommendations (Chapter 5), emerging knowledge about PFAS suggests that future studies should include a range of additional PFAS.

Full details of the methods used are provided in the peer-reviewed publications emerging from this project (see Appendix 1). In summary, however, known masses/volumes of samples were treated with isotopically labelled internal standards before solvent extraction. Following extraction, sample extracts were purified via column chromatography before being subject to gas chromatography–mass spectrometry (applied to most HFRs) and liquid chromatography– mass spectrometry (applied to HBCDDs and PFAS).

To ensure the robustness of our data on concentrations of HFRs and PFAS, a number of quality checks were performed. Full details of these are provided in the peer-reviewed publications (see Appendix 1), but a summary is provided here.

For CI-OPEs, a reagent blank consisting of 100 mg of anhydrous sodium sulfate was analysed with every batch of 11 samples. Low masses of CI-OPEs were detected in blank samples. Where the concentration in the blank associated with a given batch was 5-25% of the sample concentration, the latter was corrected by subtracting the blank concentration. Where the concentration in the blank was >25% of the sample concentration, then the sample was reported as <LOQ (limits of quantification). The LOQs for CI-OPEs were reported as the average blank concentration (0.02 mg/kg for TCEP and TCIPP and 0.25 mg/kg for TDCIPP). In addition, we evaluated the accuracy of our method via analysis of matrix spikes. Matrix spikes (of pre-extracted PUF) were performed at 50 mg/kg (n=5) and 1000 mg/kg (n=5). All measured values were found to be within 80-120% of the spiked concentrations, with a relative standard deviation (RSD) of <15%. Matrix spikes of native target analytes were also performed with every other batch of samples analysed. For a batch to be accepted, the measured concentration for each compound was required to be within 80-120% of the spiked concentration.

For other HFRs, a reagent blank consisting of 100 mg of anhydrous sodium sulfate was analysed with every batch of 11 samples. "Negative control" samples were created using plastics and textiles that had previously been found to contain no detectable HFRs and were analysed throughout the study. Three such control samples were assessed for each matrix. None of the target compounds was found above the limits of detection (LODs) in the blanks. Therefore, the results were not corrected for blank residues, and method LODs and LOQs were estimated based on a signal to noise ratio of 3:1 and 10:1, respectively. LOQs for target compounds ranged from 0.1 to 0.5 mg/kg for PBDEs, and were 0.01 mg/kg for α -,

β- and γ-HBCDD and TBBPA; 0.2 mg/kg for TBECH, pentabromotoluene, DPTE, hexabromobenzene, 1,2-bis(2,4,6-tribromophenoxy)ethane, EH-TBB, BEH-TEBP and *anti*-DP; 0.6 mg/kg for *syn*-DP; 1.0 mg/kg for DBDPE; and 6.0 mg/kg for TTBP-TAZ.

Method accuracy and precision for PBDEs was assessed via repeated analysis of certified reference materials ERM-EC591 (polypropylene) and ERM-EC590 (polyethylene), in addition to textiles (polyester fabrics), XPS and EPS that had been previously measured by this laboratory and another. All values were found to be close to certified or indicative levels, with an RSD of <15%. Full details of method precision and accuracy can be found in Abdallah *et al.* (2017). Matrix spikes of native target analytes were also performed with every other batch of samples analysed. For a batch to be accepted, the measured concentration for each compound was required to be within 80–120% of the spiked concentration.

For PFAS, a reagent blank consisting of 100 mg of anhydrous sodium sulfate was analysed with every batch of nine samples. "Control" samples were created using textiles previously identified as containing none of our target PFAS and were analysed throughout the study. None of the target compounds was found above the LODs in the blanks. Therefore, results were not corrected for blank residues, and method LODs and LOQs were estimated based on a signal to noise ratio of 3:1 and 10:1, respectively. LOQs for target PFAS ranged from 0.1 to 0.4 mg/kg. In the absence of appropriate certified reference material, as part of the initial method validation, six matrix samples were analysed. Each matrix spike sample comprised a control textile sample free of target PFAS. Of this, an accurately weighed (100-mg) aliquot was analysed. For the initial validation exercise, one tube was left unspiked as a control and the remaining five tubes were spiked with 100 ng of target compounds (i.e. 1 mg/kg). Samples were left at <4°C to fortify overnight before analysis. All measured concentrations were 80-120% of the spiked concentration levels, with an RSD of <15%. The consistently high recoveries of target analytes (average = 98%, range = 91–104%), along with the low RSD between repeated measurements (average = 3.3%, range = 1-6.8%), demonstrates that this is an accurate, precise and robust method. As an ongoing check on accuracy, a matrix spike was analysed every 20th sample and

was required to be within 80–120% of the spiked concentration.

2.3 Methods for Determination of Concentrations of Elemental Bromine, Chlorine, Phosphorus and Antimony

The instrument used for analysis was a Niton XL3t GOLDD+ XRF analyser used in its "desktop" mode, i.e. mounted into a dedicated test-stand, specially designed to avoid escape of harmful primary or secondary X-rays during analysis of low-density materials. Quantification of bromine and antimony in samples was conducted using the "plastics" operational mode, using the Ka1 and Ka2 lines for bromine (11.9242 keV and 11.8776 keV) and antimony (2.6111 keV and 2.6359 keV). For elemental chlorine, the instrument was used in its plastics operational mode to optimise quantification in fixed-thickness low-density samples. For elemental phosphorus, the only operational mode available was designated for mining and mineral applications, i.e. much denser samples than investigated here. It is known, therefore, that quantified concentrations of phosphorus in these samples are likely to be subject to a substantial offset. A more accurate estimation of actual phosphorus content in the samples of each material type was determined using the observed ratios of antimony between the two operational modes (quantified in both plastics and minerals operational modes). For each material type, the fraction of the plastics modedetermined antimony was divided by the minerals mode determination and multiplied by the observed concentration of phosphorus from the minerals mode. This was applied to all sample types, with the exception of EPS and XPS, where elemental phosphorus could not be determined.

The instrument underwent annual calibration by Niton UK, using proprietary standards containing varying concentrations of relevant inorganic compounds in a polymer matrix. This calibration uses ranges of analytes of various concentrations in a sample of "infinite thickness", i.e. a sample in which virtually all the primary X-rays are attenuated, scattered and/or

absorbed by the material and its component elements. For materials of finite thickness, this introduces a source of error, as the instrument assumes a sample of infinite thickness unless a specific calibration factor is used to compensate for the disparity. The revised screening method outlined here made use of a dedicated test-stand to eliminate the issue of background interference, i.e. target elements being detected in underlying materials and/or materials of various density interfering with the instrument's estimations. For example, the instrument typically uses Compton scattering to determine the density of analyte materials, which is adversely affected by the presence of air within the samples - as is the case for PUF, EPS and XPS foams - or primary X-rays passing through thin and low-density samples - as is the case for fabrics and EEE plastic casings.

Our previous work on XRF screening established a conservative screening threshold of approximately 710 mg/kg bromine attributable to 1000 mg/kg penta-BDE congeners, the lowest brominated congener by molecular weight of the BFRs covered by the LPCLs (Sharkey et al., 2018). However, measurements in this study reveal BDE-209 to be dominant in each sample group (with the exception of EPS and XPS foams, in which HBCDD dominates). In the light of this, in this study we employed a revised screening threshold of 833 mg/kg bromine (attributable to BDE-209) for EEE, upholstery and foam samples, while a threshold of 743 mg/kg (attributable to HBCDD) was used for EPS/ XPS foam. In a similar vein, thresholds of 325 mg/kg of chlorine attributed to TCIPP and 72 mg/kg of phosphorus attributed to TDCIPP were applied.

All samples were analysed for 60 s, in triplicate, using the above parameters, with the resulting average concentrations and standard deviations used for comparison against mass spectrometric analysis results. Samples of WEEE hard plastics containing concentrations of BFRs exceeding 100 mg/kg were also screened using the XRF for shorter time intervals (10 and 5 s), in triplicate. XRF data obtained for the same samples using different analysis times were then compared to assess the feasibility of using decreased analysis times while maintaining acceptable accuracy.

3 Results

3.1 Concentrations of PFAS in Samples of Waste Non-childcare Articles

As is evident from Table 3.1, the concentrations of our target PFAS were low (i.e. <5 mg/kg of any individual target PFAS) in all of the non-childcare samples. As we were not aware of PFAS applications in building insulation foam or EEE, PFAS analysis was restricted to ELV and soft furnishing foams and fabrics, with a total of 235 such samples collected in 2019–20 analysed (see Table 2.1). The most frequently detected of our target PFAS were MeFOSE (detection frequency=3.4%) and PFDS (detection frequency=2.6%). The maximum concentration detected in these non-childcare samples was 4.7 mg/kg PFOA in a carpet sample. This sample and two others (a mattress foam and ELV fabric containing 2.7 mg/kg and 1.3 mg/kg of PFOA, respectively) were the only samples to exceed the new EU POPs limit of 1 mg/kg for PFOA. No sample contained PFOS above the LPCL value of 50 mg/kg, with the maximum concentration of PFOS detected being 0.14 mg/kg. Moreover, PFHxS was not detected in any sample, translating into zero exceedances of the new EU POPs limit for this compound.

PFAS concentrations were also measured in the following archived samples collected in 2015–16: 29 carpets, 11 curtains and 25 ELV fabrics and foams. As observed for samples collected in 2019–20, concentrations of PFAS were low. The maximum concentration detected of any PFAS in these 2015–16 samples was 17 mg/kg of PFHxA. The most

PFAS	Detection frequency (%)	Minimum	Median	Average	Maximum
PFBA	0	<0.4	<0.4	<0.4	<0.4
PFPeA	0	<0.4	<0.4	<0.4	< 0.4
PFHxA	2.1	<0.2	<0.2	<0.2	0.42
PFHpA	0.43	<0.1	<0.1	<0.1	1.5
PFOA	1.3	<0.1	<0.1	<0.1	4.7
PFNA	0	<0.1	<0.1	<0.1	<0.1
PFDA	0	<0.1	<0.1	<0.1	<0.1
PFUdA	0	<0.2	<0.2	<0.2	< 0.2
PFDoA	0	<0.2	<0.2	<0.2	<0.2
PFTrDA	0.43	<0.2	<0.2	<0.2	1.4
PFBS	0.85	<0.1	<0.1	<0.1	0.42
PFPeS	0	<0.1	<0.1	<0.1	<0.1
PFHxS	0	<0.1	<0.1	<0.1	<0.1
PFHpS	0	<0.1	<0.1	<0.1	<0.1
PFOS	0.43	<0.1	<0.1	<0.1	0.14
PFNS	0	<0.1	<0.1	<0.1	<0.1
PFDS	2.6	<0.1	<0.1	<0.1	<0.1
PFUdS	0	<0.1	<0.1	<0.1	<0.1
PFDoS	1.3	<0.1	<0.1	<0.1	0.12
PFTrDS	0.43	<0.1	<0.1	<0.1	0.15
FOSA	0.43	<0.1	<0.1	<0.1	0.20
MeFOSA	0	< 0.2	<0.2	<0.2	< 0.2
EtFOSA	0	<0.2	<0.2	<0.2	< 0.2
MeFOSE	3.4	< 0.2	<0.2	<0.2	1.1
EtFOSE	0	<0.2	<0.2	<0.2	<0.2

Table 3.1. Summary of concentrations (mg/kg) of target PFAS in non-childcare waste (n=235)

frequently detected of our target PFAS were PFHxA and PFOS (detection frequency of both = 15%). In addition, the following PFAS were detected at frequencies of >5%: FOSA (14%) and PFDoA (9.2%), as well as PFHpA and PFHpS (both 7.7%). No sample contained PFOS above the LPCL value of 50 mg/kg, nor PFOA or PFHxS above the new EU POPs limit of 1 mg/kg.

3.2 Concentrations of PFAS in Samples of Waste Childcare Articles

The concentrations of PFAS were determined in 74 samples of foam fillings and fabric coverings from child car seats collected in 2019–20 (see Table 2.1). Results are summarised in Table 3.2. While concentrations were generally still low, there were 19 instances where the concentration of one or more PFAS was > 10 mg/kg. The following PFAS were detected with a concentration > 10 mg/kg in at least one sample: PFNA (n=5), MeFOSE (n=4), PFBA (n=2), PFTrDA (n=2), PFDA (n=1), PFHpS (n=1), PFNS (n=1), EtFOSA (n=1), EtFOSE (n=1) and MeFOSA (n=1). The maximum concentration detected was 2500 mg/kg of MeFOSE. The most frequently detected of our target PFAS were PFNA, PFDA and FOSA (detection frequency of each=12%) and MeFOSE (detection frequency=11%). In keeping with the non-childcare samples, no samples contained PFOS above the LPCL value of 50 mg/kg. No sample exceeded the new EU POPs limits of 1 mg/kg for either PFOA or PFHxS.

3.3 Concentrations of HFRs in Samples of Waste Non-childcare Articles

Table 3.3 summarises the concentrations of Σ PBDEs (i.e. BDE-28, -47, -99, -100, -153, -154, -183, and -209), TBBP-A and Σ HBCDDs (α -, β - and γ -HBCDD) detected in samples from various categories of

PFAS	Detection frequency (%)	Minimum	Median	Average	Maximum
PFBA	2.7	<0.4	<0.4	2.5	130
PFPeA	0	< 0.4	<0.4	< 0.4	< 0.4
PFHxA	3	<0.1	<0.1	<0.1	2.9
PFHpA	4.1	<0.1	<0.1	0.11	1.9
PFOA	2.7	<0.1	<0.1	<0.1	0.55
PFNA	12	<0.1	<0.1	4.1	85
PFDA	12	<0.1	<0.1	0.49	13
PFUdA	3	< 0.2	<0.2	<0.2	<0.2
PFDoA	1.4	< 0.2	<0.2	<0.2	<0.2
PFTrDA	8	< 0.2	<0.2	2.9	120
PFBS	1.4	<0.1	<0.1	<0.1	<0.1
PFPeS	0	<0.1	<0.1	<0.1	<0.1
PFHxS	0	<0.1	<0.1	<0.1	<0.1
PFHpS	5.4	<0.1	<0.1	1.8	130
PFOS	0	<0.1	<0.1	<0.1	<0.1
PFNS	8	<0.1	<0.1	0.32	18
PFDS	0	<0.1	<0.1	<0.1	<0.1
PFUdS	0	<0.1	<0.1	<0.1	<0.1
PFDoS	0	<0.1	<0.1	<0.1	<0.1
PFTrDS	0	<0.1	<0.1	<0.1	<0.1
FOSA	12	<0.1	<0.1	<0.1	0.51
MeFOSA	4.1	< 0.2	<0.2	0.72	35
EtFOSA	1.4	< 0.2	<0.2	<0.2	13
MeFOSE	11	< 0.2	<0.2	35	2500
EtFOSE	2.7	<0.2	<0.2	5.0	360

Table 3.2. Summary of concentrations (mg/kg) of target PFAS in waste childcare articles (n=73)

Table 3.3. Summary of concentrations (mg/kg) of PBDEs, HBCDD and TBBP-A, and percentage of samples exceeding the 1000 mg/kg limit value in waste non-childcare articles collected in 2019–20 (this study) and 2015–16 (WAFER study)

			ΣHBCDD		ZPBDEs		TBBP-A		ΣPBDEs+HBCDD	HBCDD
Waste category Sub-category	Sub-category	Statistical parameter 2015–16	2015-16	2019–20	2015-16	2019–20	2015-16	2019–20	2015-16	2019–20
C&D	EPS	Average	2100	390	<loq< th=""><th><loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Median	100	<loq< th=""><th><loq< th=""><th><loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq-10,000< th=""><th>< LOQ-1600</th><th><loq <<="" th=""><th><pre>> </pre></th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq></th></loq-10,000<>	< LOQ-1600	<loq <<="" th=""><th><pre>> </pre></th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq>	<pre>> </pre>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		% > limit value	35	25	0	0	0	0	35	25
	XPS	Average	27	660	<loq< th=""><th><pre>> </pre></th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	<pre>> </pre>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Median	19	0.82	<loq< th=""><th><loq <<="" th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq></th></loq<>	<loq <<="" th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq-94< th=""><th>< LOQ-6000</th><th><loq< th=""><th><loq< th=""><th><loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<></th></loq<></th></loq<></th></loq-94<>	< LOQ-6000	<loq< th=""><th><loq< th=""><th><loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<></th></loq<></th></loq<>	<loq< th=""><th><loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<></th></loq<>	<loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<>	<loq< th=""><th></th><th></th></loq<>		
		% > limit value	0	15	0	0	0	0	0	15
ELVs	Foams and	Average	45	13	960	300	<loq< th=""><th>0.60</th><th></th><th></th></loq<>	0.60		
	upholstery	Median	<loq<< th=""><th>0.31</th><th>3.0</th><th>7.0</th><th><loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<></th></loq<<>	0.31	3.0	7.0	<loq<< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq-3300< th=""><th>< LOQ-550</th><th>< LOQ-31,000</th><th>0.80-8400</th><th><loq< th=""><th><loq-58< th=""><th></th><th></th></loq-58<></th></loq<></th></loq-3300<>	< LOQ-550	< LOQ-31,000	0.80-8400	<loq< th=""><th><loq-58< th=""><th></th><th></th></loq-58<></th></loq<>	<loq-58< th=""><th></th><th></th></loq-58<>		
		% > limit value	1.7	0	4.2	5.4	0	0	5.9	5.4
Soft furnishings	Carpets	Average	1.1	<loq< th=""><th>240</th><th>2.9</th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	240	2.9	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Median	<loq< th=""><th><loq< th=""><th>1.0</th><th><pre>> </pre></th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq<>	<loq< th=""><th>1.0</th><th><pre>> </pre></th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	1.0	<pre>> </pre>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<l0q–26< th=""><th><loq< th=""><th>< LOQ-7000</th><th><l0q-12< th=""><th><loq< th=""><th><loq-0.05< th=""><th></th><th></th></loq-0.05<></th></loq<></th></l0q-12<></th></loq<></th></l0q–26<>	<loq< th=""><th>< LOQ-7000</th><th><l0q-12< th=""><th><loq< th=""><th><loq-0.05< th=""><th></th><th></th></loq-0.05<></th></loq<></th></l0q-12<></th></loq<>	< LOQ-7000	<l0q-12< th=""><th><loq< th=""><th><loq-0.05< th=""><th></th><th></th></loq-0.05<></th></loq<></th></l0q-12<>	<loq< th=""><th><loq-0.05< th=""><th></th><th></th></loq-0.05<></th></loq<>	<loq-0.05< th=""><th></th><th></th></loq-0.05<>		
		% > limit value	0	0	3.2	0	0	0	3.2	0
	Curtains	Average	3.8	1.7	3.8	3.8	<loq< th=""><th>0.23</th><th></th><th></th></loq<>	0.23		
		Median	<loq< th=""><th>0.62</th><th><loq <<="" th=""><th><pre>> </pre></th><th><loq< th=""><th>0.20</th><th></th><th></th></loq<></th></loq></th></loq<>	0.62	<loq <<="" th=""><th><pre>> </pre></th><th><loq< th=""><th>0.20</th><th></th><th></th></loq<></th></loq>	<pre>> </pre>	<loq< th=""><th>0.20</th><th></th><th></th></loq<>	0.20		
		Range	<loq-56< th=""><th>< LOQ-8.7</th><th>< LOQ-53</th><th><loq-53< th=""><th><loq< th=""><th><loq-0.82< th=""><th></th><th></th></loq-0.82<></th></loq<></th></loq-53<></th></loq-56<>	< LOQ-8.7	< LOQ-53	<loq-53< th=""><th><loq< th=""><th><loq-0.82< th=""><th></th><th></th></loq-0.82<></th></loq<></th></loq-53<>	<loq< th=""><th><loq-0.82< th=""><th></th><th></th></loq-0.82<></th></loq<>	<loq-0.82< th=""><th></th><th></th></loq-0.82<>		
		% > limit value	0	0	0	0	0	0	0	0
	Furniture fabrics	Average	9200	640	6800	600	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Median	1.1	2.0	12	13	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq-51,000< th=""><th>0.47-4100</th><th>< LOQ-73,000</th><th>0.85–2100</th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq-51,000<>	0.47-4100	< LOQ-73,000	0.85–2100	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		% > limit value	27	19	27	31	0	0	40	50
	Furniture foam	Average	1100	20	660	470	25	<loq< th=""><th></th><th></th></loq<>		
	filling	Median	0.31	0.02	16	7.3	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq-8000< th=""><th>< LOQ-250</th><th>< LOQ-7800</th><th>0.70–1900</th><th><loq-250< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq-250<></th></loq-8000<>	< LOQ-250	< LOQ-7800	0.70–1900	<loq-250< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq-250<>	<loq< th=""><th></th><th></th></loq<>		
		% > limit value	25	0	15	19	0	0	35	19

			ΣHBCDD		ZPBDEs		TBBP-A		ΣPBDEs + HBCDD	HBCDD
	Waste category Sub-category	Statistical parameter	2015-16	2019–20	2015-16	2019-20	2015-16	2019–20	2015-16	2019–20
-	Mattress foam filling	Average	<loq< th=""><th>7.7</th><th>89</th><th>6.5</th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	7.7	89	6.5	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Median	<loq< th=""><th>0.30</th><th>8.4</th><th>1.6</th><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	0.30	8.4	1.6	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq< th=""><th>< LOQ-190</th><th>< LOQ-870</th><th><loq-92< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq-92<></th></loq<>	< LOQ-190	< LOQ-870	<loq-92< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq-92<>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		% > limit value	0	0	0	0	0	0	0	0
	Mattress fabric	Average	2.2	1.4	10	1	0.02	<loq< th=""><th></th><th></th></loq<>		
-	covering	Median	0.06	0.58	6.7	2.1	<loq< td=""><td><loq< td=""><td></td><td></td></loq<></td></loq<>	<loq< td=""><td></td><td></td></loq<>		
		Range	<l0q–12< th=""><th><l0q-6.2< th=""><th>0.17–49</th><th><loq-100< th=""><th><l0q-0.11< th=""><th><loq< th=""><th></th><th></th></loq<></th></l0q-0.11<></th></loq-100<></th></l0q-6.2<></th></l0q–12<>	<l0q-6.2< th=""><th>0.17–49</th><th><loq-100< th=""><th><l0q-0.11< th=""><th><loq< th=""><th></th><th></th></loq<></th></l0q-0.11<></th></loq-100<></th></l0q-6.2<>	0.17–49	<loq-100< th=""><th><l0q-0.11< th=""><th><loq< th=""><th></th><th></th></loq<></th></l0q-0.11<></th></loq-100<>	<l0q-0.11< th=""><th><loq< th=""><th></th><th></th></loq<></th></l0q-0.11<>	<loq< th=""><th></th><th></th></loq<>		
		% >limit value	0	0	0	0	0	0	0	0
	LHAS	Average	<loq< th=""><th>0.21</th><th>19</th><th>¢L0Q</th><th>36</th><th>4.1</th><th></th><th></th></loq<>	0.21	19	¢L0Q	36	4.1		
		Median	<loq< td=""><td><loq< td=""><td>0.039</td><td><loq< td=""><td><loq< td=""><td>0.41</td><td></td><td></td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>0.039</td><td><loq< td=""><td><loq< td=""><td>0.41</td><td></td><td></td></loq<></td></loq<></td></loq<>	0.039	<loq< td=""><td><loq< td=""><td>0.41</td><td></td><td></td></loq<></td></loq<>	<loq< td=""><td>0.41</td><td></td><td></td></loq<>	0.41		
		Range	<loq< th=""><th><l0q-2.1< th=""><th>< LOQ-190</th><th><loq< th=""><th><loq-2000< th=""><th><l0q-16< th=""><th></th><th></th></l0q-16<></th></loq-2000<></th></loq<></th></l0q-2.1<></th></loq<>	<l0q-2.1< th=""><th>< LOQ-190</th><th><loq< th=""><th><loq-2000< th=""><th><l0q-16< th=""><th></th><th></th></l0q-16<></th></loq-2000<></th></loq<></th></l0q-2.1<>	< LOQ-190	<loq< th=""><th><loq-2000< th=""><th><l0q-16< th=""><th></th><th></th></l0q-16<></th></loq-2000<></th></loq<>	<loq-2000< th=""><th><l0q-16< th=""><th></th><th></th></l0q-16<></th></loq-2000<>	<l0q-16< th=""><th></th><th></th></l0q-16<>		
		% > limit value	0	0	0	0	1.8	0	0	0
	Cooling appliances	Average	<loq< th=""><th>0.65</th><th>0.48</th><th>1.5</th><th><loq< th=""><th>4.1</th><th></th><th></th></loq<></th></loq<>	0.65	0.48	1.5	<loq< th=""><th>4.1</th><th></th><th></th></loq<>	4.1		
		Median	<loq< td=""><td><loq <<="" td=""><td><loq <<="" td=""><td><loq< td=""><td><loq< td=""><td>0.35</td><td></td><td></td></loq<></td></loq<></td></loq></td></loq></td></loq<>	<loq <<="" td=""><td><loq <<="" td=""><td><loq< td=""><td><loq< td=""><td>0.35</td><td></td><td></td></loq<></td></loq<></td></loq></td></loq>	<loq <<="" td=""><td><loq< td=""><td><loq< td=""><td>0.35</td><td></td><td></td></loq<></td></loq<></td></loq>	<loq< td=""><td><loq< td=""><td>0.35</td><td></td><td></td></loq<></td></loq<>	<loq< td=""><td>0.35</td><td></td><td></td></loq<>	0.35		
		Range	<loq< th=""><th>< LOQ-16</th><th>< LOQ-3.6</th><th><loq-16< th=""><th><loq< th=""><th><loq-59< th=""><th></th><th></th></loq-59<></th></loq<></th></loq-16<></th></loq<>	< LOQ-16	< LOQ-3.6	<loq-16< th=""><th><loq< th=""><th><loq-59< th=""><th></th><th></th></loq-59<></th></loq<></th></loq-16<>	<loq< th=""><th><loq-59< th=""><th></th><th></th></loq-59<></th></loq<>	<loq-59< th=""><th></th><th></th></loq-59<>		
		% > limit value	0	0	0	0	0	0	0	0
	Display	Average	46	5.8	1900	730	3000	1700		
		Median	0.014	0.07	<loq <<="" th=""><th>0.80</th><th>4.8</th><th>037</th><th></th><th></th></loq>	0.80	4.8	037		
		Range	<loq< th=""><th>< LOQ-140</th><th>< LOQ-62,000</th><th><loq-32,000< th=""><th><loq-274,000< th=""><th><loq-38,000< th=""><th></th><th></th></loq-38,000<></th></loq-274,000<></th></loq-32,000<></th></loq<>	< LOQ-140	< LOQ-62,000	<loq-32,000< th=""><th><loq-274,000< th=""><th><loq-38,000< th=""><th></th><th></th></loq-38,000<></th></loq-274,000<></th></loq-32,000<>	<loq-274,000< th=""><th><loq-38,000< th=""><th></th><th></th></loq-38,000<></th></loq-274,000<>	<loq-38,000< th=""><th></th><th></th></loq-38,000<>		
		% >limit value	0	0	4.7	4.3	19	6.4	4.7	4.3
	SDAs	Average	<loq< th=""><th>1.8</th><th>170</th><th>5.5</th><th>350</th><th>1.8</th><th></th><th></th></loq<>	1.8	170	5.5	350	1.8		
		Median	<loq< th=""><th><loq <<="" th=""><th>0.019</th><th><loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq></th></loq<>	<loq <<="" th=""><th>0.019</th><th><loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<></th></loq>	0.019	<loq< th=""><th><loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<></th></loq<>	<loq< th=""><th><loq< th=""><th></th><th></th></loq<></th></loq<>	<loq< th=""><th></th><th></th></loq<>		
		Range	<loq< th=""><th>< L0Q-42</th><th>< LOQ-1600</th><th><loq-140< th=""><th><loq-8600< th=""><th><l0q-42< th=""><th></th><th></th></l0q-42<></th></loq-8600<></th></loq-140<></th></loq<>	< L0Q-42	< LOQ-1600	<loq-140< th=""><th><loq-8600< th=""><th><l0q-42< th=""><th></th><th></th></l0q-42<></th></loq-8600<></th></loq-140<>	<loq-8600< th=""><th><l0q-42< th=""><th></th><th></th></l0q-42<></th></loq-8600<>	<l0q-42< th=""><th></th><th></th></l0q-42<>		
		% >limit value	0	0	6.9	0	3.4	0	6.9	0
_	IT and	Average	20	1.1	260	48	1700	74		
	telecommunications	Median	<loq< th=""><th><loq <<="" th=""><th>0.21</th><th><loq< th=""><th><loq< th=""><th>1.2</th><th></th><th></th></loq<></th></loq<></th></loq></th></loq<>	<loq <<="" th=""><th>0.21</th><th><loq< th=""><th><loq< th=""><th>1.2</th><th></th><th></th></loq<></th></loq<></th></loq>	0.21	<loq< th=""><th><loq< th=""><th>1.2</th><th></th><th></th></loq<></th></loq<>	<loq< th=""><th>1.2</th><th></th><th></th></loq<>	1.2		
		Range	<loq-1600< th=""><th>< LOQ-56</th><th>< LOQ-7600</th><th><loq-2200< th=""><th><loq-110,000< th=""><th><loq-900< th=""><th></th><th></th></loq-900<></th></loq-110,000<></th></loq-2200<></th></loq-1600<>	< LOQ-56	< LOQ-7600	<loq-2200< th=""><th><loq-110,000< th=""><th><loq-900< th=""><th></th><th></th></loq-900<></th></loq-110,000<></th></loq-2200<>	<loq-110,000< th=""><th><loq-900< th=""><th></th><th></th></loq-900<></th></loq-110,000<>	<loq-900< th=""><th></th><th></th></loq-900<>		
		% >limit value	1.3	0	6.4	1.9	5.1	0	7.7	1.9

Table 3.3. Continued

non-childcare waste articles collected in 2019–20. Table 3.1 also provides the percentage of samples that exceeded the current LPCL for PBDEs and HBCDD of 1000 mg/kg and the related limit value for TBBP-A. The equivalent data reported previously for samples from the same waste categories obtained in 2015–16 as part of the WAFER project are provided for comparison.

Table 3.4 summarises the concentrations of TCEP, TCIPP and TDCIPP detected in non-childcare samples collected in 2019–20. Table 3.4 also provides the percentage of samples for which concentrations of any individual CI-OPE exceeded 1000 mg/kg. Where available, the same information for archived samples obtained in 2015–16 in the WAFER project is provided. Table 3.4 also gives the *p*-value obtained for an independent *t*-test comparison of concentrations in samples from each waste category acquired in 2019–20 with those in samples acquired in 2015–16.

3.4 Concentrations of HFRs in Samples of Waste Childcare Articles

Table 3.5 summarises the concentrations of Σ PBDEs, Σ HBCDDs, CI-OPEs and NBFRs present at > 1000 mg/kg in at least one sample, in fabrics and foams from waste childcare fabrics and foams collected in 2019–20. Table 3.5 also provides the percentage of samples that exceeded 1000 mg/kg for each target HFR.

non-childcare ar	non-childcare articles collected in 2019–20 (this study) and 2015–16 (WAFER study))–20 (this study) and 2	2015–16 (WAFE	ER study)				
			TCEP		TCIPP		TDCIPP	
Waste category	Sub-category	Statistical parameter	2015-16	2019–20	2015–16	2019–20	2015–16	2019–20
C&D	EPS	Average		<loq< td=""><td></td><td>1.0</td><td></td><td>750</td></loq<>		1.0		750
		Median		<loq< td=""><td></td><td>0.32</td><td></td><td>75</td></loq<>		0.32		75
		Range		<loq< td=""><td></td><td>< LOQ-3.8</td><td></td><td>5.2-6100</td></loq<>		< LOQ-3.8		5.2-6100
		% > limit value		0		0		17
		<i>p</i> -value ^a						
	XPS	Average		0.04		7000		51
		Median		<loq< td=""><td></td><td>7500</td><td></td><td>4.0</td></loq<>		7500		4.0
		Range		<loq-0.43< td=""><td></td><td>< LOQ-22,000</td><td></td><td>< LOQ-150</td></loq-0.43<>		< LOQ-22,000		< LOQ-150
		% > limit value		0		62		0
		<i>p</i> -value						
ELVs	Foams	Average	220	370	170	5900	1000	30,000
		Median	26	14	130	630	25	250
		Range	1.2–2400	2.0-8400	3.6–570	61-100,000	6.7-15,000	< LOQ-340,000
		% > limit value	8.6	6.1	0	37	14	
		<i>p</i> -value	0.51		0.03		0.004	43
	Fabrics	Average	110	10	60	66	880	2300
		Median	7.2	<loq< td=""><td>60</td><td>27</td><td>62</td><td><loq< td=""></loq<></td></loq<>	60	27	62	<loq< td=""></loq<>
		Range	1.0–1200	<loq-220< td=""><td>5.6-150</td><td>< LOQ-1200</td><td>7.5-6300</td><td>< LOQ-17,000</td></loq-220<>	5.6-150	< LOQ-1200	7.5-6300	< LOQ-17,000
		% >limit value	2.9	0	0	1.6	26	26
		<i>p</i> -value	0.03		0.12		0.04	
Soft furnishings	Carnets	Averade	1		۵ ۲	0	ر ۵	32
0	- - -	Median	0.58	<loq< td=""><td>6.3</td><td><loq< td=""><td>8.3</td><td>< LOQ</td></loq<></td></loq<>	6.3	<loq< td=""><td>8.3</td><td>< LOQ</td></loq<>	8.3	< LOQ
		Range	< LOQ-10	<loq< td=""><td>0.39–48</td><td>< LOQ-1600</td><td><loq-140< td=""><td>< LOQ-350</td></loq-140<></td></loq<>	0.39–48	< LOQ-1600	<loq-140< td=""><td>< LOQ-350</td></loq-140<>	< LOQ-350
		% > limit value	0	0	0	5.0	0	0
		<i>p</i> -value	0.01		0.32		0.36	

Table 3.4. Summary of concentrations (mg/kg) of TCEP, TCIPP and TDCIPP, and percentage of samples exceeding the 1000 mg/kg limit value in waste

			TCEP		TCIPP		TDCIPP	
Waste category	Sub-category	Statistical parameter	2015-16	2019–20	2015-16	2019–20	2015-16	2019–20
	Curtains	Average	2.9	1.1	35	60	1.0	210
		Median	0.80	<loq< th=""><th>13</th><th><loq< th=""><th>0.45</th><th>36</th></loq<></th></loq<>	13	<loq< th=""><th>0.45</th><th>36</th></loq<>	0.45	36
		Range	< LOQ-10	<l0q-3.1< th=""><th>0.39–150</th><th>< LOQ-1500</th><th><loq-3.1< th=""><th>< LOQ-2700</th></loq-3.1<></th></l0q-3.1<>	0.39–150	< LOQ-1500	<loq-3.1< th=""><th>< LOQ-2700</th></loq-3.1<>	< LOQ-2700
		% > limit value	0	0	0	4.0	0	4.0
		<i>p</i> -value	0.25		0.69		0.08	
	Furniture fabrics	Average	61	53	130		37	340
		Median	19	0.39	130		8.3	110
		Range	2.9–320	<loq-700< th=""><th>12–330</th><th>< LOQ-3400</th><th>6.5-320</th><th>< LOQ-1600</th></loq-700<>	12–330	< LOQ-3400	6.5-320	< LOQ-1600
		% > limit value	0	0	0	6.3	0	13
		<i>p</i> -value	0.97		0.09		0.04	
	Furniture foam filling	Average	1300	940	11,000	5400	1100	5700
		Median		4.7	9500	220	13	24
		Range	-9000	<loq-14,000< th=""><th>0.35–25,000</th><th>1.8-40,000</th><th>1.1–13,000</th><th>4.8-44,000</th></loq-14,000<>	0.35–25,000	1.8-40,000	1.1–13,000	4.8-44,000
		% >limit value		6.3	83	38	8.3	19
		<i>p</i> -value	0.76		0.16		0.23	
	Mattress foam filling	Average	5.5	2.3	1700	3900	14	50
		Median	4.7	<loq< th=""><th>180</th><th>67</th><th>4.4</th><th><loq< th=""></loq<></th></loq<>	180	67	4.4	<loq< th=""></loq<>
		Range	<loq-13< th=""><th><loq-54< th=""><th>29–15,000</th><th>< LOQ-41,000</th><th>2.1–88</th><th><loq-1100< th=""></loq-1100<></th></loq-54<></th></loq-13<>	<loq-54< th=""><th>29–15,000</th><th>< LOQ-41,000</th><th>2.1–88</th><th><loq-1100< th=""></loq-1100<></th></loq-54<>	29–15,000	< LOQ-41,000	2.1–88	<loq-1100< th=""></loq-1100<>
		% >limit value	0	0	10	18	0	2.5
		<i>p</i> -value	0.12		0.31		0.24	
	Mattress fabric covering	Average	36	1.2	40	330	2.0	360
		Median	8.1	<loq< th=""><th>24</th><th>300</th><th>0.51</th><th>140</th></loq<>	24	300	0.51	140
		Range	1.9–110	<loq-8.4< th=""><th>2.9–130</th><th>< LOQ-1200</th><th><loq-10< th=""><th>< LOQ-2200</th></loq-10<></th></loq-8.4<>	2.9–130	< LOQ-1200	<loq-10< th=""><th>< LOQ-2200</th></loq-10<>	< LOQ-2200
		% >limit value	0	0	0	5.0	0	10
		<i>p</i> -value	0.14		0.001		0.14	

^ap-value derived from independent f-test comparing concentrations in samples from 2015–16 with samples from 2019–20.

Table 3.4. Continued

Table 3.5. Summary of concentrations (mg/kg) of selected HFRs, and percentage of samples exceedingthe 1000 mg/kg limit value in waste childcare article foam and fabric samples collected in 2019–20

Waste category	Statistical parameter	ТСЕР	TCIPP	TDCIPP	ΣPBDEs	DBDPE	ЕН-ТВВ	BEH- TEBP	ΣHBCDD
Child car	Median	6.7	120	15	4.8	<lod< td=""><td><lod< td=""><td><lod< td=""><td>1.3</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>1.3</td></lod<></td></lod<>	<lod< td=""><td>1.3</td></lod<>	1.3
seats	Average	1400	3600	25,000	1900	6.2	2800	1100	500
	Maximum	66,000	51,000	390,000	23,000	220	100,000	39,000	7400
	% > 1000 mg/kg	9.3	25	33	7.3	0	4.7	3.3	7.3
Pushchairs	Median	1.4	40	<lod< td=""><td>6.9</td><td>0.0</td><td><lod< td=""><td><lod< td=""><td>1.3</td></lod<></td></lod<></td></lod<>	6.9	0.0	<lod< td=""><td><lod< td=""><td>1.3</td></lod<></td></lod<>	<lod< td=""><td>1.3</td></lod<>	1.3
	Average	9.0	510	2300	630	80	0.15	<lod< td=""><td>5.1</td></lod<>	5.1
	Maximum	270	11,000	80,000	11,200	1200	5.2	<lod< td=""><td>74</td></lod<>	74
	% >1000 mg/kg	0	6.1	4.1	4.1	2.0	0	0	0
Prams	Median	0.49	580	72	11	<lod< td=""><td><lod< td=""><td><lod< td=""><td>2.5</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>2.5</td></lod<></td></lod<>	<lod< td=""><td>2.5</td></lod<>	2.5
	Average	15	5600	13,000	870	1200	0.82	<lod< td=""><td>7700</td></lod<>	7700
	Maximum	270	52,000	170,000	11,203	9600	18	<lod< td=""><td>140,000</td></lod<>	140,000
	% >1000 mg/kg	0	33	21	8.3	17	0	0	17
Cot	Median	1.1	310	<lod< td=""><td>0.59</td><td>0.06</td><td><lod< td=""><td><lod< td=""><td>0.33</td></lod<></td></lod<></td></lod<>	0.59	0.06	<lod< td=""><td><lod< td=""><td>0.33</td></lod<></td></lod<>	<lod< td=""><td>0.33</td></lod<>	0.33
mattresses	Average	2.0	16,000	0.3	37	0.12	0.05	<lod< td=""><td>0.35</td></lod<>	0.35
	Maximum	11.0	170,000	2.9	590	0.46	0.54	<lod< td=""><td>1.3</td></lod<>	1.3
	% >1000 mg/kg	0	38	0	0	0	0	0	0
Change mats	Median	1.8	1100	<lod< td=""><td>5.2</td><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>1.0</td></lod<></td></lod<></td></lod<></td></lod<>	5.2	<lod< td=""><td><lod< td=""><td><lod< td=""><td>1.0</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>1.0</td></lod<></td></lod<>	<lod< td=""><td>1.0</td></lod<>	1.0
	Average	7.0	13,000	3900	4.6	<lod< td=""><td>0.96</td><td><lod< td=""><td>1.0</td></lod<></td></lod<>	0.96	<lod< td=""><td>1.0</td></lod<>	1.0
	Maximum	48	84,000	60,000	11	0	12	<lod< td=""><td>1.5</td></lod<>	1.5
	% >1000 mg/kg	0	52	8.7	0	0	0	0	0
Miscellaneous	Median	0.51	640	4.6					
childcare	Average	0.55	5000	110					
articles	Maximum	0.86	44,000	570					
	% > 1000 mg/kg	0	50	0					
Childcare	Median	2.4	140	3.2	6.4	<lod< td=""><td><lod< td=""><td><lod< td=""><td>1.2</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>1.2</td></lod<></td></lod<>	<lod< td=""><td>1.2</td></lod<>	1.2
articles overall	Average	800	5700	16,000	1100	160	1400	540	1200
	Maximum	66,000	170,000	390,000	23,000	9600	100,000	39,000	140,000
	% > 1000 mg/kg	5.0	27	21	7.7	2.6	3.6	2.6	7.7

4 Discussion

4.1 PFAS in Waste Foam and Fabric Articles

Overall, concentrations of the PFAS targeted in this study were low, with no article containing PFOS at a concentration exceeding its LPCL of 50 mg/kg, low detection frequencies and only 19 articles (all foams and fabrics from child car seats) containing a PFAS >10 mg/kg. Of note is that in child car seat samples the highest concentration (2500 mg/kg) of any PFAS was that of MeFOSE, and that the most frequently detected PFAS was PFNA (detection frequency = 12%). This is not inconsistent with a report from Ireland that concentrations of both of these PFAS were significantly higher in air inside cars containing a child car seat than in cars without a child car seat (Harrad et al., 2019b). While we measured concentrations of 25 PFAS, we consider it likely that other non-targeted PFAS are present in waste foams and fabrics currently entering the waste stream in Ireland.

4.2 PBDEs and HBCDD and Exceedances of the LPCL in Irish Non-childcare Samples Collected in 2019–20 Compared with 2015–16 Samples

4.2.1 Construction and demolition EPS/XPS waste

As in the 2015–16 study (Harrad, 2018), no PBDEs were detected in any of the C&D samples collected in 2019-20. In contrast, of 25 C&D EPS/XPS samples, five (20%) contained HBCDDs above the LPCL of 1000 mg/kg. This is slightly lower than the 23% LPCL exceedance for HBCDD observed in 2015–16. Interestingly, while in the 2015–16 samples all LPCL exceedances for HBCDD were observed in EPS samples (35%), in the 2019-20 samples there were exceedances in both EPS (n=3/12, 25%) and XPS (n=2/13, 15%). As a result, concentrations of HBCDD in XPS were significantly higher (p < 0.05) in 2019–20 than in 2015–16. No such significant difference was observed for EPS, nor for EPS and XPS combined. This may suggest that the apparent increase in HBCDD concentrations in C&D XPS is an artefact of the small sample numbers analysed, while

also highlighting the probably long lag time between the introduction of restrictions on HBCDD use and its removal from the waste stream. For example, the service life of EPS and XPS building insulation materials has been estimated at 35–50 years for EPS and equal to the building lifetime for XPS (Kono *et al.*, 2016).

4.2.2 End-of-life vehicle waste fabrics and foams

Of the 111 ELV fabric and foam samples collected in 2019–20, concentrations of Σ PBDEs exceeded the LPCL in six samples (5.4%), while no LPCL exceedances were detected for HBCDD. The incidence of LPCL exceedances in the current project is broadly similar to that in 2015–16, where, of the 119 ELV samples studied, five (4.2%) and two (1.7%) exceeded the LPCL for PBDEs and HBCDD, respectively (Harrad *et al.*, 2019a). This similarity in LPCL exceedance was reflected in the absence of any significant temporal change in concentrations of both PBDEs and HBCDD.

4.2.3 Soft furnishings

As indicated in Table 2.1, the soft furnishing samples collected consisted of a mix of carpets and curtains, as well as fabric coverings and PUF fillings for mattresses, sofas and chairs. Consistent with what we reported for samples collected in 2015-16, concentrations of PBDEs and HBCDD in samples of carpets, curtains and mattresses collected in 2019-20 were low, with no LPCL exceedances in such samples. In contrast, in 2019-20, 8 of 16 (50%) samples of furniture fabrics exceeded the LPCL (five and three samples for PBDEs and HBCDD, respectively). This proportion of exceedances is consistent with that observed for samples collected in 2015-16, for which nine (41%) samples of furniture fabrics exceeded the LPCL (six for PBDEs only, six for HBCDD only, and three for both PBDEs and HBCDD in the same sample) (Harrad, 2018). With respect to furniture foam filling samples, both concentrations and the incidence of LPCL exceedances were lower than for furniture fabrics. Specifically, in 2015-16, seven

(35%) furniture foam samples exceeded the LPCL (two for PBDEs only, four for HBCDD only, and one for both PBDEs and HBCDD in the same sample). For the 2019–20 samples, there were fewer LPCL exceedances than in 2015–16, i.e. none for HBCDD and three (19%) for PBDEs. Despite this, there was no significant difference in concentrations of either PBDEs or HBCDD between 2015–16 and 2019–20.

4.2.4 Waste electrical and electronic equipment

As observed for such waste items collected in 2015–16 (Harrad, 2018), concentrations of both PBDEs and HBCDD in LHAs and cooling appliances collected in 2019–20 were low, with none exceeding the LPCL. Similarly, low concentrations and zero LPCL exceedances were also seen in SDAs collected in 2019–20. However, this contrasts with what was observed for SDAs in 2015–16, for which there were two (6.9%) LPCL exceedances for PBDEs only, and this was reflected in concentrations of **SPBDEs** declining significantly (*p*<0.05) between 2015–16 and 2019–20. For display items, the incidence of LPCL exceedances showed little change: two (4.3%) exceedances for PBDEs only in 2019–20, compared with two (4.7%) exceedances for PBDEs only in 2015-16. Finally, for IT and telecommunications items, while there was no significant difference between the two studies with respect to concentrations of PBDEs and HBCDD, LPCL exceedances were proportionally lower in 2019-20 than in 2015–16. Explicitly, while in 2015–16, 5 of 78 (6.4%) and 1 of 78 (1.3%) IT samples exceeded the LPCL for PBDEs and HBCDD, respectively, in 2019-20 the LPCL was exceeded for PBDEs in only a single sample (1.9%) out of 52 samples.

Figure 4.1 summarises the temporal trend in the percentage of waste articles containing > 1000 mg/kg of PBDEs and HBCDD for the different waste categories examined.

4.3 TBBP-A in Irish Waste Nonchildcare Samples Collected in 2019–20 Compared with 2015–16 Samples

In summary, and consistent with observations for samples collected in 2015–16 (Harrad, 2018), concentrations of TBBP-A in C&D wastes, ELVs,

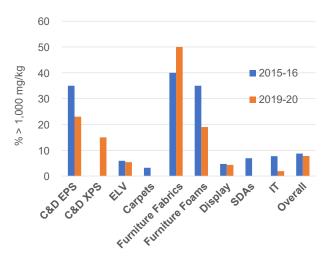


Figure 4.1. Temporal trend in percentage of waste articles containing concentrations >1000 mg/kg of PBDEs and HBCDD (zero exceedances unless shown).

soft furnishings and cooling appliances collected in 2019-20 were well below the limit value of 1000 mg/kg. We also detected only very low concentrations (<1000 mg/kg) of TBBP-A in LHAs collected in 2019-20. Although in 2015-16 there was one LHA sample that exceeded 1000 mg/kg TBBP-A, concentrations in LHAs did not differ significantly (p>0.05) between the two studies. In contrast, there were proportionally fewer display, SDA and IT samples in 2019–20 that contained > 1000 mg/kg TBBP-A than in 2015–16. Specifically, while in 2015–16, 19%, 3.4% and 5.1% of display, SDA and IT samples contained >1000 mg/kg TBBP-A, the corresponding figures in 2019-20 were 6.4%, 0% and 0%. While the decline in the proportion of IT samples containing > 1000 mg/kg TBBP-A between the two studies was not reflected by a significant difference in concentrations, the concentrations of TBBP-A in both SDA and display samples declined significantly (p < 0.05) between 2015-16 and 2019-20.

Figure 4.2 illustrates the temporal trend in the percentage of waste articles containing > 1000 mg/kg of TBBP-A for the different waste categories examined.

4.4 NBFRs in Irish Waste Nonchildcare Samples Collected in 2019–20 Compared with 2015–16 Samples

Our target NBFRs were rarely detected in samples collected in this study in 2019–20. Indeed, *anti*-DP

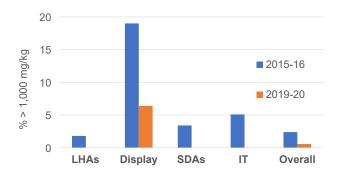


Figure 4.2. Temporal trend in percentage of waste articles containing concentrations >1000 mg/kg of TBBP-A (zero exceedances unless shown).

and DBDPE were the only NBFRs targeted that were detected in at least one waste category at a detection frequency > 20%, at maximum concentrations of 96 and 1100 mg/kg, respectively. As in the 2019–20 samples, the presence of our target NBFRs in samples (mainly soft furnishings and ELV fabrics and foams, as well as a small number of WEEE samples) collected in 2015-16 was negligible. As in 2019-20, only anti-DP and DBDPE were detected in >20% of samples from at least one waste category, at maximum concentrations of 33 and 8.8 mg/kg, respectively. However, despite these generally very low concentrations of most of our target NBFRs in most samples, it is important to note that in the 2019–20 study TTBP-TAZ was detected at concentrations of 22,000 mg/kg in one IT sample (an internet router) and at 14,000 and 32,000 mg/kg in two display samples (both TVs). Moreover, another display sample (a TV) contained 1100 mg/kg of DBDPE. While, to our knowledge, no data yet exist of concentrations of TTBP-TAZ in Irish indoor air and dust, the concentrations of DBDPE in indoor air and dust collected in Ireland in 2016-18 were the highest reported globally to date (Wemken et al., 2019). This suggests that, while concentrations of NBFRs in the Irish waste stream remain well below those of PBDEs, HBCDD and TBBP-A, the high concentrations observed in a very small number of items now entering the waste stream indicate that NBFRs such as TTBP-TAZ and DBDPE may become more prevalent in plastic items reaching the waste stream over the next decade.

4.5 Cl-OPEs in Irish Waste Nonchildcare Samples Collected in 2019–20 Compared with 2015–16 Samples

While no previous data exist on concentrations of CI-OPEs in Irish waste articles, our data are broadly consistent with those from a preliminary study of FRs in waste office furniture in the UK, in which seven out of nine furniture foam samples contained TCIPP at an average concentration of 19,000 mg/kg, with a further foam sample containing both TDCIPP and TCEP at concentrations of 11,000 and 5000 mg/kg, respectively (Stubbings *et al.*, 2016). Data reported in this study are also within the range of those reported for soft furnishing samples, such as sofas, chairs, mattresses, etc., collected in the USA, in which, for example, 25% of sofas/love seats contained TDCIPP and 4.6% contained TCIPP at a concentration > 1000 mg/kg (Cooper *et al.*, 2016).

4.5.1 Construction and demolition EPS/XPS waste

No archived C&D EPS or XPS building insulation foam samples were available for analysis. However, in the samples collected in 2019–20, while concentrations were below or only just above LOQs for TCEP in both EPS and XPS, for TCIPP in EPS and for TDCIPP in XPS, eight (62%) XPS samples and two (17%) EPS samples contained > 1000 mg/kg of TCIPP and TDCIPP, respectively. Maximum concentrations of TCIPP and TDCIPP were 22,000 and 6100 mg/kg in XPS and EPS, respectively. This is consistent with the reported application of these CI-OPEs in building insulation foam (European Commission, 2008).

4.5.2 End-of-life waste fabrics and foams

Of the ELV fabric (n=62) and foam (n=49) samples collected in 2019–20, concentrations of TCEP exceeded 1000 mg/kg in three (6.1%) foam samples, with no such exceedances observed in any of the ELV fabric samples. The proportion of samples containing > 1000 mg/kg of TCEP was greater in the samples collected in 2015–16, at three (8.6%) foam and one (2.9%) fabric sample. While this did not translate into a significant difference between the two studies for ELV foams, TCEP concentrations were significantly lower in ELV fabrics collected in 2019–20. In contrast, for both TCIPP and TDCIPP, the proportion of samples with concentrations > 1000 mg/kg were significantly higher in foam samples collected in 2019–20, for which maximum concentrations were 340,000 and 100,000 mg/kg for TDCIPP and TCIPP, respectively. The situation is less clear for ELV fabrics, as there was no significant difference between TCIPP concentrations in samples collected in different years, and while concentrations of TDCIPP were higher in ELV fabric samples collected in 2019–20, the proportion > 1000 mg/kg is the same, whichever year samples were collected.

4.5.3 Waste soft furnishings

As shown in Table 2.1, soft furnishing samples collected in 2019-20 were a mix of fabric coverings and PUF fillings for chairs, mattresses and sofas, as well as carpets and curtains. Of all the waste categories examined in this study, average concentrations and the proportion of samples containing concentrations > 1000 mg/kg were highest for all three target CI-OPEs in furniture foam, with at least one sample in each sub-category containing concentrations > 1000 mg/kg for one or more CI-OPE. Most strikingly, six (37%), three (19%) and one (6.3%) furniture foam sample contained > 1000 mg/kg of TCIPP, TDCIPP and TCEP, respectively, with maximum concentrations of 44,000 mg/kg of TCIPP, 25,000 mg/kg of TDCIPP and 9000 mg/kg of TCEP. Average concentrations of all CI-OPEs were lowest for curtains and carpets.

With respect to possible temporal trends, while average concentrations and the proportion of samples with a concentration > 1000 mg/kg were greater for all three CI-OPEs in furniture foam collected in 2015-16. notably 10 (83%) of such samples contained TCIPP concentrations > 1000 mg/kg; however, no significant differences were found in concentrations detected in the 2019–20 samples. Indeed, while *t*-test comparison of data for 2015–16 and 2019–20 samples revealed concentrations of TCEP to be higher in carpets in 2015–16, concentrations of TDCIPP to be higher in furniture fabrics in 2019-20 and concentrations of TCIPP to be higher in mattress fabric coverings in 2019–20, there were very few of any such samples that contained > 1000 mg/kg of the CI-OPEs in guestion, and the temporal trends observed are unlikely to reflect a meaningful trend in the application of CI-OPEs.

4.6 PBDEs and HBCDD and Exceedances of the LPCL in Irish Waste Childcare Article Samples Collected in 2019–20

Of the 187 samples analysed for BFRs overall, 7.7% (n = 15) exceed the LPCL for Σ PBDEs, with all exceedances due to BDE-209, which was by far the dominant PBDE congener detected. The same proportion of samples overall exceeded the LPCL for ΣHBCDD. For BDE-209, all samples containing >1000 mg/kg were fabric coverings. By contrast, while nine samples containing HBCDD at > 1000 mg/kg were fabrics, five were EPS components and one (which contained the maximum **SHBCDD** concentration in this study) was underlay material from a pram. There were no exceedances for either PBDEs or HBCDD for change mats and cot mattresses, with zero exceedances observed for HBCDD for pushchairs. Instead, exceedances were observed for car seats and prams (both PBDEs and HBCDD), as well as for pushchairs (PBDEs). The maximum concentrations detected were 23,000 mg/kg (2.3%) and 140,000 mg/kg (14%) for **SPBDEs** and **SHBCDD**, respectively, indicating intentional use of these BFRs in such articles. Data from a previous study on 101 samples of foam taken from similar childcare articles acquired in the USA in 2010 reports concentrations of those congeners associated with the penta-BDE formulation (e.g. BDE-47, -99, -100, -153, -154) to be on average 32,000 mg/kg, with a range of 17,000-52,000 mg/kg (Stapleton et al., 2011). In contrast, a more recent study of 10 foam samples from childcare articles (changing pads, sleep positioners and bath products) collected in the USA in 2015 did not detect either PBDEs or HBCDD above the detection limit of 1 mg/kg (Gloekler et al., 2021). Consistent with measurements of PBDEs in indoor dust from Ireland conducted in 2016-17 (Wemken et al., 2019), the PBDE profile in childcare products in Ireland is predominantly BDE-209 (predominant in the deca-BDE formulation), with lower brominated congeners associated with the penta-BDE formulation rarely detected.

4.7 NBFRs in Irish Waste Childcare Article Samples

Of the NBFRs targeted in this study, only three (DBDPE, EH-TBB and BEH-TEBP) were detected at concentrations > 1000 mg/kg in at least one sample.

Concentrations of these HFRs exceeded 1000 mg/kg in eight (3.7%), nine (4.2%) and seven (3.2%) of the articles tested, with maximum concentrations of 9600, 100,000 and 39,000 mg/kg for DBDPE, EH-TBB and BEH-TEBP, respectively. With respect to the latter two HFRs, we calculated the fraction of EH-TBB ($f_{\rm EH-TBB}$) in the nine articles in which the concentration of the former was > 1000 mg/kg. In line with a previous report (Ma *et al.*, 2012), $f_{\rm EH-TBB}$ is calculated as:

$$f_{\text{EH}_{\text{TBB}}} = \text{EH}_{\text{TBB}} / (\text{EH}_{\text{TBB}} + \text{BEH}_{\text{TEBP}})$$
 (4.1)

Values of $f_{\rm EH-TBB}$ ranged between 0.70 and 0.80, with a median of 0.72 and an arithmetic mean of 0.74. These data are in very close agreement with the value of 0.77 reported elsewhere for the FR commercial formulation known as Firemaster-550 (FM-550) (Ma *et al.*, 2012). Therefore, we suspect strongly that the articles in question had been treated with either FM-550 or Firemaster-BZ54, which also contains EH-TBB and BEH-TEBP at a similar ratio to FM-550 (Ma *et al.*, 2012).

The detection of DBDPE in a small number of childcare articles at concentrations up to 9600 mg/kg is unsurprising given that we recently reported elevated concentrations of DBDPE in indoor air and dust from Ireland (Wemken et al., 2019). We did not measure either EH-TBB or BEH-TEBP in that study; however, both have been detected frequently in indoor air and dust from the UK (Tao et al., 2016), but at concentrations that are an order of magnitude below those of HBCDD and BDE-209 and not suggestive of widespread use. While we do not have information about the provenance of the articles containing elevated EH-TBB and BEH-TEBP, we believe that it is plausible that they were imported directly from the USA where FM-550 and Firemaster-BZ54 were used widely (Ma et al., 2012).

4.8 Cl-OPEs in Irish Waste Childcare Article Samples

In summary, of the 274 samples analysed for CI-OPEs, concentrations exceeded 1000 mg/kg in 27% (n=75), 21% (n=58) and 5.0% (n=14) of samples for TCIPP, TDCIPP and TCEP, respectively. Samples for which concentrations exceeded 1000 mg/kg for TCIPP were observed in each of the different sample categories studied, reflected by average TCIPP concentrations of 3600 mg/kg, 510 mg/kg, 5600 mg/kg, 16,000 mg/kg,

13,000 mg/kg and 5000 mg/kg in car seats, pushchairs, prams, cot mattresses, change mats and miscellaneous childcare articles, respectively. At least one sample contained TDCIPP at a concentration > 1000 mg/kg in car seats, pushchairs, prams and change mats, with average concentrations in these items being 25,000 mg/kg, 2300 mg/kg, 13,000 mg/kg and 3900 mg/kg, respectively. By comparison, samples displaying TCEP concentrations > 1000 mg/kg were observed in car seats only. These findings point to widespread intentional use of CI-OPEs in childcare articles.

A particularly notable feature of our data is that, for a small number of samples, we recorded some very high concentrations of TDCIPP and to a lesser extent TCIPP – up to 390,000 mg/kg (39%) of TDCIPP in one instance. In total, there were 12 (10%) samples for which concentrations of TDCIPP or TCIPP exceeded 100,000 mg/kg, and these samples comprised nine car seats, two prams and one cot mattress.

Interestingly, while most samples containing at least one CI-OPE at a concentration > 1000 mg/kg were PUF fillings, we also detected high TCIPP and TDCIPP concentrations in nine PVC coverings of change mats. In five instances, the concentrations exceeded 29,000 mg/kg, with TDCIPP present at 60,000 mg/kg in one case. Moreover, concentrations of TCIPP, TDCIPP and TCEP exceeded 1000 mg/kg in 25, 25 and 5 fabric covering samples, respectively. In every such instance, the concentration of the CI-OPE in question was > 1000 mg/kg in a PUF sample from the same childcare item, and this suggests that CI-OPEs added intentionally to foam filling material transfer to overlying fabric coverings.

We compared our data for Ireland with those reported for foam samples obtained from childcare articles in previous studies from the USA. The largest such study reported concentrations of CI-OPEs in 101 samples collected in 2010 (Stapleton *et al.*, 2011). The products tested comprised car seats (n=21), changing table pads (n=16), infant sleep positioners (n=15), portable crib mattresses (n=13) and nursing pillows (n=11), plus a few miscellaneous others. Average concentrations (range in parentheses) of TCEP, TCIPP and TDCIPP were 5900 mg/kg (1100–5900 mg/kg), 5500 mg/kg (1100–14,000 mg/kg) and 39,000 mg/kg (2400–120,000 mg/kg), respectively. A later study measured CI-OPEs in child car seats (n=98), child mattresses (*n* = 36) and other childcare products (*n*=49) collected in 2014–16 in the USA (Cooper *et al.*, 2016). The authors reported whether or not an article contained a FR using a definition of a concentration > 10,000 mg/kg. Based on this definition, 50%, 14% and 29% of car seats, child mattresses and other childcare articles, respectively, contained TDCIPP, with corresponding figures for TCIPP being 27%, 8.3% and 22%. A more recent study measured TCIPP and TDCIPP in 10 foam samples from childcare articles on the US market (Gloekler *et al.*, 2021). TCIPP was detected in all samples tested (range 149–38,400 mg/kg) and TDCIPP was detected in a single sample (at 75 mg/kg).

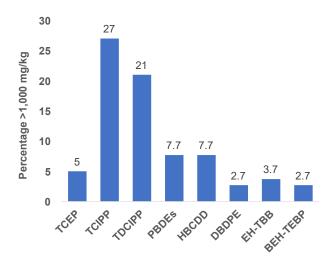


Figure 4.3. Percentage of childcare articles exceeding >1000 mg/kg for various HFRs.

Overall, our data for childcare foam and fabric samples collected in Ireland in 2019–20 are broadly consistent with those reported in the USA, although TDCIPP appears essentially absent from US articles purchased after 2011, likely because of its addition to Proposition 65 in California in 2011 (Cooper *et al.*, 2016). Such restrictions are not in place in Ireland and this may explain why TDCIPP is the major HFR detected in this study.

Figure 4.3 shows the percentage of all waste childcare articles containing > 1000 mg/kg of the various HFRs measured in this study. Figure 4.4 depicts the percentage of waste childcare articles containing > 1000 mg/kg for different HFRs according to article type.

4.9 Preliminary Estimation of Mass of Products Exceeding Limit Values and Mass of PBDEs, HBCDD, TBBP-A and Cl-OPEs Annually Entering the Nonchildcare Waste Streams Studied in Ireland

Estimates of the mass of our target waste materials generated in Ireland in 2019 were derived as described elsewhere (Drage *et al.*, 2022). We are not aware of data on the mass of waste childcare items generated annually in Ireland and thus have not considered them here. Combining these data

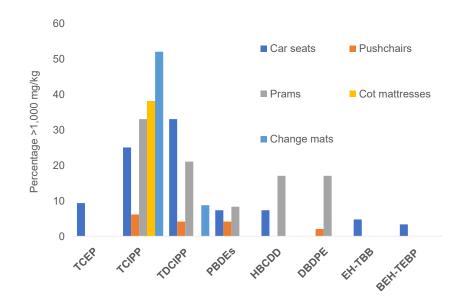


Figure 4.4. Percentage of waste childcare articles containing concentrations >1000 mg/kg of various HFRs, according to article type.

with our concentration data generated preliminary estimates of the mass of PBDEs, HBCDD, TBBP-A and CI-OPEs annually entering the waste streams studied in Ireland (Table 4.1). The uncertainties inherent in these estimates are acknowledged, and their preliminary nature underlined. Specifically, estimates of the mass of waste materials generated annually involve a substantial degree of uncertainty; for example, direct estimates of waste furniture foam for Ireland are not available, and we have therefore extrapolated on a population basis from estimated UK arisings of waste furniture (237,516t in 2011) and applied our own judgement to estimate that 15% of this is foam (Drage et al., 2022). Coupled with this, while this study and the earlier WAFER study are, to our knowledge, among the largest of their kind to date anywhere, the extent to which the samples analysed are representative of all such articles in Ireland is unknown. Notwithstanding these caveats, we believe that our estimates provide an informative overview of HFR contamination in the Irish waste stream.

Of note is that of the ≈10,200 kg ΣBFRs (comprising the sum of PBDEs, HBCDD and TBBP-A) estimated to be entering the Irish waste stream in 2019, 28.6% were associated with C&D insulation foam, with ELV foams/fabrics, display items, furniture foams and furniture fabrics contributing a further 23.8%, 21.6%, 12.5% and 10.6%, respectively. In contrast, only 2.9% were found in waste carpets, curtains, mattresses, cooling appliances, large and small household appliances, and IT/telecommunications articles combined. Such information may help focus monitoring resources on those waste categories most contaminated with PBDEs, HBCDD and TBBP-A.

Particularly notable is that the mass of CI-OPEs (principally TCIPP and TDCIPP) entering the Irish waste stream in 2019 was – at ≈147,000 kg – an order of magnitude greater than the ≈10,200 kg of PBDEs, HBCDD and TBBP-A estimated to be entering the Irish waste stream in 2019. Of the ≈74,000 kg of TDCIPP, 74% was associated with ELV foams/fabrics, with most of the remainder (20%) present in furniture foams. TCIPP contamination is more evenly distributed. Thirty-four per cent of the 70,000 kg of TCIPP entering the Irish waste stream in 2019 was found in mattress foam, with 29%, 20% and 14% associated with building insulation foam, furniture foam, and ELV foams and fabrics, respectively. Meanwhile, of the ≈3100 kg of TCEP entering the Irish waste stream in 2019, most (77%) was associated with furniture foam, with the bulk of the remainder (20%) found in ELV foams and fabrics. These data regarding the distribution of CI-OPEs across different waste categories may assist in directing monitoring resources towards those waste categories most contaminated with CI-OPEs.

4.10 Implications of Enforcement of Limit Values on Mass of HFRs Removed from the Waste Stream and Mass of Waste Rendered Unrecyclable

In October 2021, the European Parliament published a proposal for the upcoming revision of the POPs Regulation (EU 2019/1021) to include revised LPCLs for PBDEs and HBCDD (among other chemicals). Specifically, the Parliament cites revised LPCLs for both HBCDD and PBDEs of 500 mg/kg, reduced to 200 mg/kg 5 years after the entry into force of the POPs Regulation (European Union, 2022). A follow-up report from the European Parliament in February 2022 further proposed more stringent LPCLs for these POPs, reducing 200 mg/kg to 100 mg/kg after 5 years (Hojsik, 2022). Such revised LPCLs were since agreed in summer 2022 but are yet to be entered into legislation. Against this shifting legislative backdrop, we calculated what proportion of PBDEs, HBCDD and TBBP-A would need to be removed from the waste stream were these revised limit values in place in 2019.

Table 4.2 shows that enforcement of the current limit values for PBDEs, HBCDD and TBBP-A of 1000 mg/kg will result in 3.1% of the estimated ≈91,000 t/year of the waste materials studied, generated in Ireland in 2019, exceeding these limit values (≈2800 t). Balanced against this, the material exceeding limit values contains ≈7900 kg of ΣBFRs or 78% of the total mass of these BFRs associated with the waste materials studied. As noted above, proposals exist to lower the current limit values for PBDEs and HBCDD progressively to 500, 200 and 100 mg/kg. Therefore, we examined the potential impact of such changes to the limit values (which also assume similar changes in the limit value for TBBP-A to 500, 200 and 100 mg/kg) on the mass of unrecyclable waste generated, compared with changes in the extent to which the limit values remove BFRs from the waste stream. A

and CI-OPEs associated with such waste	ed with su	ch waste								
Category	t waste/y	t waste/y PBDEs (kg/y) HBCDD (kg/y)		POP-BFR (kg/y) ^a TBBP-A (kg/y) ZBFRs (kg/y) ^a TCEP (kg/y) TCIPP (kg/y) TDCIPP (kg/y) ZCI-OPEs (kg/y) ^a	TBBP-A (kg/y)	ΣBFRs (kg/y) ^g	TCEP (kg/y)	TCIPP (kg/y)	TDCIPP (kg/y)	ΣCI-OPEs (kg/y) ^g
C&D	5500	5.1	2900	2900	0.20	2900	0.15	20,000	2100	22,000
ELVs	3800	2300	110	2400	0.50	2400	630	10,000	55,000	66,000
Carpets	7600	22	91	110	0.08	110	0.08	690	240	930
Curtains	740	2.9	1.3	4.2	0.17	4.4	0.8	44	150	200
Furniture foam	2600	1200	51	1300	0.01	1300	2400	14,000	15,000	31,000
Furniture fabrics	880	530	560	1100	0.004	1100	47	470	300	810
Mattress foam	6100	40	47	87	0.61	88	14	24,000	310	24,000
Mattress fabrics	2500	27	3.5	30	0.04	30	3.0	830	910	1700
LHA⁵	37,000	0.09	0.02	7.7	0.15	7.9				
Cooling	10,000	1.5	0.65	2.2	4.1	6.3				
Display ^d	5000	660	5.2	660	1500	2200				
SDA€	8100	0.33	0.001	0.34	0.11	0.44				
IT and telecommunication articles ^f	1900	16	0.37	17	25	42				
Total ^g	92,000	4800	3800	8600	1600	10,000	3100	70,000	74,000	147,000
*Rounded to two significant figures	it finures									

Table 4.1. Estimated annual masses^a of different categories of non-childcare waste generated in Ireland in 2019, and mass^a of PBDEs, HBCDD, TBBP-A

^aRounded to two significant figures.

^bAssuming that 0.29% w/w of LHAs is bromine-containing plastic (WRc, 2012).

^cAssuming that 10% w/w of waste cooling appliances is bromine-containing plastic (WRc, 2012).

^dAssuming that 18% w/w of display waste is bromine-containing plastic (WRc, 2012).

Assuming that 0.75% w/w of SDAs is bromine-containing plastic (WRc, 2012).

Assuming that 18% w/w of IT equipment is bromine-containing plastic (WRc, 2012).

⁹Totals may differ because of rounding.

w/w, weight for weight.

Table 4.2. Estimated annual mass (t/y^a) of material in each non-childcare waste category studied that exceeds the limit value of 1000 mg/kg, and annual mass of PBDEs, HBCDD, TBBP-A and CI-OPEs (kg/y^a) associated with such material

		BFRs					CI-OPEs				
Category	PBDE assoc with 1 t waste >1000 >1000 mg/kg/y (kg/y)	PBDEs associated with material > 1000 mg/kg (kg/y)	HBCDD associated with material > 1000 mg/kg (kg/y)	POP-BFRs associated with material > 1000 mg/kg (kg/y)	TBBP-A associated with material > 1000 mg/kg (kg/y)	ΣBFRs associated with material >1000mg/kg (kg/y)	TCEP assoc with r t waste >1000 mg/kg/y (kg/y)	TCEP associated with material > 1000 mg/kg (kg/y)	TCIPP associated with material > 1000 mg/kg (kg/y)	TDCIPP associated with material >1000 mg/kg (kg/y)	ΣCI-OPEs associated with material > 1000 mg/kg (kg/y)
C&D	1100	0.78	2700	2700	0.01	2700	2200	0.12	20,000		22,000
ELVs	210	1100	4.2	1100	0.01	1100	1600	580	9600	55,000	65,000
Carpets	0	0	0	0	0	0	380	0.01		0	610
Curtains	0	0	0	0	0	0	59	0.79	44		120
Furniture foam	490	910	5.5	920	0.002	920	1300	2300	14,000	15,000	31,000
Furniture fabrics	440	520	560	1100	0.002	1100	170	0.02	280	200	480
Mattress foam	0	0	0	0	0	0	1100	8.0	24,000	220	24,000
Mattress fabrics	0	0	0	0	0	0	380	0.92	270	450	720
LHA ^b	0	0	0	0	0	0					
Cooling	0	0	0	0	0	0					
Display ^d	530	650	0.49	650	1500	2200					
SDA [€]	0	0	0	0	0	0					
IT and telecommunication articles ^f	36	14	0	4	0.10	14					
Total ^g	2800	3200	3300	6400	1500	7900	7200	2900	68,000	72,000	144,000
% captured by limit implementation		65	86	74	97	78		93	98	98	98
% > 1000 mg/kg	3.1						24				

^aRounded to two significant figures.

^bAssuming that 0.29% w/w of LHAs is bromine-containing plastic (WRc, 2012).

⁶Assuming that 10% w/w of waste cooling appliances is bromine-containing plastic (WRc, 2012).

^dAssuming that 18% w/w of display waste is bromine-containing plastic (WRc, 2012).

Assuming that 0.75% w/w of SDAs is bromine-containing plastic (WRc, 2012).

'Assuming that 18% w/w of IT equipment is bromine-containing plastic (WRc, 2012).

⁹Totals may differ because of rounding.

w/w, weight by weight.

summary of this evaluation is provided in Table 4.3. While lowering the limit value progressively to 500, 200 and 100 mg/kg removes, respectively, 82%, 84% and 85% of ΣBFRs (PBDEs, HBCDD and TBBP-A) from the waste stream, compared with the 78% removed under the existing 1000 mg/kg limit, the mass of waste rendered unrecyclable increases by an estimated 4.0%, 4.9% and 5.6%, compared with the 3.1% unrecyclable under the current limits. Notwithstanding the clear environmental benefits in minimising the mass of regulated BFRs able to enter the recycling stream, policymakers should take into consideration the associated implications for the circular economy of promulgating stricter limit values in reducing recycling of plastic waste materials. For example, while stricter limits on BFR concentrations in waste eligible for recycling will remove more BFRs, such limits will also increase the mass of plastic waste that cannot be recycled as a result of its POP content.

Moreover, while there is presently no legislation preventing waste from being recycled because of its CI-OPE content, ECHA's currently suspended restriction proposal on CI-OPE use (ECHA, 2018, 2022a) led us to consider the impact should a 1000-mg/kg limit be introduced on concentrations of TCEP, TCIPP and TDCIPP in future.

Table 4.2 also shows that enforcement of a limit value of 1000 mg/kg for each of TCEP, TCIPP and TDCIPP as individual contaminants will result in \approx 24% (\approx 7200t) of the estimated \approx 30,000 t/year of the waste materials studied (i.e. those generated in Ireland in 2019) exceeding these limit values. Balanced against this, this material, containing one or more of our target CI-OPEs with concentrations > 1000 mg/kg, contains \approx 144,000 kg or 98% of the total mass of these CI-OPEs associated with the waste materials studied. Clearly, notwithstanding the

Table 4.3. Summary of estimated annual mass (t/y^a) of material exceeding various limit values and associated ΣBFR removal percentages

Limit (mg/kg)	Annual mass waste >limit (t/y)	% removal ΣBFR
1000	2804	78
500	3600	82
200	4500	84
100	5200	85

^aTo two significant figures.

reduction in the quantity of waste recycled and the technical, logistical and economic issues associated with implementing a 1-000 mg/kg limit on CI-OPEs in waste, its implementation is likely to be very effective in removing CI-OPEs from the recycling stream.

4.11 Temporal Trends in Exceedances and "False Exceedances" of the LPCL for PBDEs and HBCDD

Of the 470 waste non-childcare articles in which PBDEs and HBCDD were measured, 25 (5.3%) were found to exceed the LPCL of 1000 mg/kg for at least one of these BFRs. These figures were lower than those observed in the WAFER project, in which 47 of the 538 (8.7%) articles tested exceeded the LPCL (Harrod et al., 2019a). However, the WAFER project analysed proportionally more samples from waste categories in which the frequency of LPCL exceedances are greater (e.g. C&D EPS). When corrected for the different mix of samples from different waste categories in the two studies, proportionally slightly fewer samples exceeded limit values for PBDEs and HBCDD in 2019-20 (7.8%) than in 2015-16 (8.7%). However, portable XRF measurements of bromine incorrectly identified 47 articles as exceeding the LPCL. This equates to a false exceedance frequency of 10%. By comparison, in the WAFER project, conducted in 2015-16, the false exceedance frequency was 6.3% (34/538) (Harrod et al., 2019a). Of the 47 false exceedances in the current project, 10 can be at least partially attributed to the presence of BFRs not covered by LPCLs, such as TBBP-A (n=5), TTBP-TAZ (n=3) and DBDPE (n=2). However, in the majority of cases (37/47), the false exceedance was not attributable to any of the BFRs we targeted. The cause(s) of the false exceedance in these 37 samples is unclear but will be a combination of (a) the presence of a brominated compound not targeted in this study, (b) inaccurate XRF determination of bromine (see discussion below) and (c) inhomogeneous distribution of PBDEs/HBCDD in the sample, such that XRF testing was conducted on a portion of the sample containing a low concentration of PBDEs or HBCDD.

In the WAFER project, no "false negatives", i.e. samples that exceeded the LPCL, but were not indicated by the corresponding portable XRF measurement of bromine, were recorded. In contrast, we identified nine (1.9%) samples collected in 2019-20 that were "false negatives". Seven of these "false negatives" occurred in furniture and ELV foam samples, with the other two observed in furniture upholstery fabric samples. Several of these "false negatives" are likely to be due to inhomogeneous distribution of BFRs in the samples, while others may be due to inaccurate XRF quantification of bromine, exacerbated by the relatively low-density nature of the samples concerned. For thin and low-density samples, such as foams and fabrics, the accuracy of measurements can be improved by folding/stacking thin materials (i.e. fabrics) or compressing low-density materials, as reported previously (Sharkey et al., 2018). A potential drawback, therefore, of using the test-stand is the inability to compress expanded foam samples or stack thin samples without interference: using the XRF in portable mode and pressing stacked materials into a surface will hold otherwise irregularly shaped fabrics in place. Finally, the two upholstery fabric samples identified as "false negatives" were composed of a linoleum-like material and were unique among the other upholstery samples collected and analysed (in terms of both material type and excess BFR presence).

4.12 The Potential Utility of Bromine: Antimony Ratios to Reduce False Exceedances of the LPCL for PBDEs in WEEE Plastics

Of the 31 hard plastic samples for which XRF analysis indicated an exceedance of the LPCL, three were genuine exceedances attributable to PBDEs with measured bromine: antimony ratios between 5.1 and 6.8, i.e. outside the range of 2:1-4:1 hypothesised by Guzzonato et al. (2017) as indicating the presence of PBDEs. Moreover, the apparent LPCL exceedance was not attributable to PBDEs in any of the 11 samples displaying bromine: antimony ratios between 2:1 and 4:1. Specifically, in 4 of these 11 samples, the bromine content was attributable to TBBP-A, with the bromine content in the other 7 samples due to either another brominated compound or an XRF measurement artefact. Bromine: antimony ratios in the remaining 17 samples were either not calculable because of the absence of detectable antimony or were between 4.4:1 and 14:1. Overall, our results strongly indicate that bromine: antimony ratios do not provide a means of differentiating between genuine and false exceedances of LPCL values for PBDEs.

4.13 Can Reducing XRF Measurement Time Increase Sample Throughput without Adversely Impacting Accuracy?

All XRF data referred to in the sections above are based on triplicate measurements, each conducted for 60s using the XRF in its "desktop" mode (i.e. in the dedicated XRF test-stand). However, we evaluated whether or not this length of measurement time was required for the purposes of screening samples for LPCL compliance. WEEE plastic samples that contained bromine above 100 mg/kg (45 of the total 210 WEEE samples) were measured using the same methodology as previously described (i.e. triplicate measurements of each sample), but using reduced measurement times (10s and 5s). One-way repeated measures analysis of variance conducted on these measurements showed no statistically significant influence of measurement time on the average bromine concentration (p=0.89). However, as illustrated in Figure 4.5, significant differences (p < 0.01) were observed between the RSDs of the measurements conducted at each measurement time, and this indicates that the reproducibility of XRF measurements of bromine decreases significantly as the measurement time decreases.

Assuming that all bromine in a given sample is due to BDE-209 gives a bromine-equivalent LPCL of 833 mg/kg. Taking into account the different RSD values for each measurement time, this gives a bromine-equivalent LPCL of 833 ma/kg plus the maximum RSD for each measurement time (35, 38 and 70 mg/kg for 60-, 10- and 5-s measurement times, respectively). Using a 60-s measurement time, 28 of the 210 WEEE samples screened gave false exceedances. Decreasing the measurement time to 10s did not cause any additional false exceedances, while further reduction to 5s resulted in just one additional false exceedance. Using these 210 data points, approximately 86.7% of WEEE was successfully screened for LPCL compliance using a 60-s screening time, while it is slightly reduced to 86.2% for 5-s measurements. These data indicate that reducing the measurement time from 60s to 5s has minimal impact on the accuracy with which XRF can correctly identify hard plastic articles exceeding the LPCL for PBDEs. Thus, we explored the extent to which using a 5-s measurement time would improve

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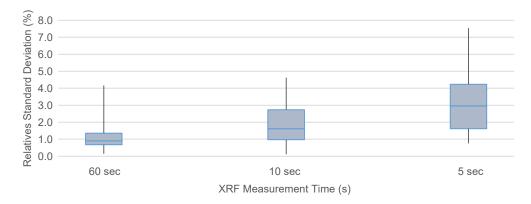


Figure 4.5. Box plot showing RSDs (%) for 60-, 10- and 5-s measurement times on 45 WEEE samples containing bromine concentrations >100 mg/kg from initial screening (60-s measurements in triplicate). Median, first and third quartiles, and maximum/minimum RSDs are shown.

sample throughput when using XRF to check WEEE plastics for LPCL compliance.

In Ireland, approximately 52,600t of WEEE (not including lamps) was collected and prepared for reuse or recycling in 2019 (EPA, 2021), which equates to roughly 22.3 million WEEE units (WEEE Ireland, 2020). The WAFER project calculated that a single portable XRF instrument conducting triplicate measurements each of 60-s duration (with a 35-s downtime between each triplicate measurement) could process 100 screened units over an 8-h working day and a 220-day working year, amounting to 66,000 items over a period of 3 years (Harrad *et al.*, 2019a). However, our data suggest that there is no need for triplicate measurements, as the RSD

for individual measurements does not affect the ability of the XRF to correctly identify whether an article complies with the LPCL and, therefore, a single measurement per item is sufficient. Assuming a single 60-s XRF screening (with a 30-s downtime between measurements) per item, some 70,400 units of WEEE per annum would be successfully screened for compliance with LPCL values by a single portable XRF. By comparison, a 5-s measurement time, with a 30-s downtime between measurements, would allow approximately 180,000 items per annum to be screened for LPCL compliance. Based on these calculations, around 125 portable XRF instruments would be needed to screen the current number of WEEE articles generated annually in Ireland.

5 Conclusions and Recommendations

This project measured a wide range of HFRs and PFAS in a large number of waste articles, covering EEE and polystyrene building insulation foam, as well as foams and fabrics from soft furnishings and childcare items. In general, the concentrations of the 25 PFAS targeted were low, with no items exceeding the low POP concentration limit value of 50 mg/kg for PFOS. However, there was a small proportion of instances (n=19) where concentrations of a PFAS exceeded 10 mg/kg. Only three samples (a carpet, mattress foam and ELV fabric containing 4.7 mg/kg, 2.7 mg/kg and 1.3 mg/kg of PFOA, respectively) exceeded the new EU POPs limit of 1 mg/kg for PFOA. No sample contained PFOS above the LPCL value of 50 mg/kg, with the maximum concentration of PFOS detected being 0.14 mg/kg. Moreover, PFHxS was not detected in any sample, translating into zero exceedances of the new EU POPs limit of 1 mg/kg for this compound. Notwithstanding the observed low concentrations of the targeted PFAS, it is considered likely that other PFAS not measured in this study are present in such articles.

Comparing these data with data for Ireland in 2015-16 (Harrad et al., 2019a) revealed that, for most waste categories examined, concentrations and exceedances of limits for PBDEs. HBCDD and TBBP-A were similar or had declined. For ELV fabrics and foams, HBCDD and SPBDE concentrations had declined significantly (p < 0.05) since 2015–16. Moreover, **SPBDE** concentrations in waste SDAs were significantly lower (p < 0.05) in 2019–20, with a similarly significant decline (p<0.05) for TBBP-A in waste IT and telecommunications articles. In contrast, HBCDD concentrations in waste XPS increased significantly (p < 0.05) between 2015–16 and 2019-20. For other waste categories studied, no statistically significant temporal trends are evident (p>0.05). The NBFRs targeted in this study were detected infrequently and predominantly at very low concentrations. However, TTBP-TAZ was detected in three display/IT product samples at 14,000-32,000 mg/kg, indicating that elevated concentrations of FRs used as alternatives to PBDEs and HBCDD are likely to increase in future.

HFRs and PFAS were measured for the first time in foams and fabrics from waste childcare articles collected in Ireland. Of such articles, 7.7% exceeded the LPCL of 1000 mg/kg for PBDEs (all due to BDE-209), an additional 7.7% exceeded the limit for HBCDD, and 3.2% of articles exceeded the limit for both PBDEs and HBCDD. An even greater proportion of articles contained concentrations exceeding 1000 mg/kg for TCIPP (27%) and TDCIPP (21%), with concentrations greater than 1000 mg/kg also observed for TCEP (5.0% articles), EH-TBB (3.7%), DBDPE (2.7%) and BEH-TEBP (2.7%). Overall, 115 samples contained at least one HFR at a concentration exceeding 1000 mg/kg. In addition to the waste management implications of our findings, our data raise concerns about child exposure to HFRs during the use phase of these everyday items.

Based on concentrations of HFRs in non-childcare waste articles, an estimated 10,200 kg of ΣBFRs (i.e. the sum of PBDEs, HBCDD and TBBP-A) entered the Irish waste stream in 2019. Of this, 28.6% was associated with polystyrene building insulation foam, with ELV foams and fabrics, display articles, furniture foam and furniture fabrics containing 23.8%, 21.6%, 12.5% and 10.6%, respectively. By comparison, an estimated 74,000 kg of TDCIPP, 70,000 kg of TCIPP and 3000 kg of TCEP were associated with waste foams and fabrics in 2019. For TDCIPP, 74% was present in ELV foams and fabrics, with 20% found in furniture foams. By comparison, the burden of TCIPP was more equally distributed across mattress foam (34%), building insulation foam (29%), furniture foam (20%) and ELV foams and fabrics (14%).

Enforcing the current limit value of 1000 mg/kg for PBDEs, HBCDD and TBBP-A renders ≈2800t of waste (3.1% of the ≈90,000t of waste generated) unrecyclable. Balanced against this, the limit prevents ≈7900 kg of PBDEs, HBCDD and TBBP-A (78% of the 10,200 kg generated annually) from entering the recycling stream. Anticipated stepwise lowering of the limit value to 500 mg/kg, 200 mg/kg and 100 mg/kg would prevent an estimated 82%, 84% and 85% of PBDEs, HBCDD and TBBP-A entering the recycling stream; however, it would increase the annual mass of unrecyclable waste to 4.0%, 4.9% and 5.6% of the total mass generated. While no limit value currently exists for the CI-OPEs, one may be introduced in the near future. In anticipation of this, we calculated that enforcing a 1000 mg/kg limit on each of TDCIPP, TCIPP and TCEP would render unrecyclable 7200t (24%) of the ≈30,000 t of waste building insulation foam, ELV and furniture foams and fabrics generated in Ireland in 2019 but would prevent 144,000 kg (98% of the total generated) of these CI-OPEs from entering the recycling stream.

When corrected for the different proportions of samples from different waste categories between the two years studied, proportionally slightly fewer samples exceeded LPCL values for PBDEs and HBCDD in 2019–20 (7.8%) than in 2015–16 (8.7%).

The project examined the effectiveness of using portable XRF to screen waste articles for compliance with LPCL values for PBDEs and HBCDD. Given the volume/mass of waste generated that requires checking for compliance with LPCLs, combined with the cost (typically €500/sample) and time (typically 1 day) involved in using the gas chromatographymass spectrometry and liquid chromatography-mass spectrometry methods used here to measure concentrations of PBDEs and HBCDD, portable XRF represents a potentially effective, rapid and more cost-effective alternative. A similar exercise in 2015–16 found that 6.3% of non-childcare waste articles examined were "false exceedances" of the LPCL, i.e. where XRF measurements of bromine incorrectly indicated that the LPCL had been exceeded. In the current study, the incidence of false exceedances in waste non-childcare articles increased to 10% (47/473 articles examined). Of these 47 false exceedances, only 10 could be attributed to the presence of a known BFR other than a PBDE or HBCDD. The remainder are attributable to one or more of the following factors: (a) presence of an unknown brominated compound, (b) a measurement artefact of the XRF instrument or (c) inhomogeneous distribution of the PBDE or HBCDD in the sample, such that XRF measurement is undertaken on an area of the sample surface that contains a concentration of PBDE or HBCDD that exceeds that present in the sample overall. Interestingly, while in 2015-16 no incidences of "false negatives" were recorded, i.e. where XRF measurements of bromine incorrectly indicate that the LPCL has not been

exceeded, nine (1.9%) samples were false negatives in the current study. The exact cause of these false negatives could not be ascertained, but they are likely to be attributable to one or more of the following: (a) inhomogeneous distribution of the PBDE or HBCDD in the sample, such that the XRF measurement is undertaken on an area of the sample surface that contains a concentration of PBDE or HBCDD that is well below that present in the sample overall or (b) a measurement artefact of the XRF instrument. As a previous study (Guzzonato et al., 2017) had highlighted that antimony trioxide was used in hard plastics as a synergist alongside PBDEs, but to a much lesser extent with other BFRs, this project explored whether XRF measurements of antimony and bromine would provide a means of identifying false exceedances. However, data on antimony and bromine concentrations in samples shown to be false exceedances suggested that the presence of antimony was not exclusively associated with PBDEs, and thus measurement of antimony and bromine was not a viable means of identifying false exceedances. In an earlier assessment of the potential use of portable XRF measurements of bromine to screen waste articles for compliance with LPCLs for PBDEs and HBCDD, bromine was measured using a 60-s screen time in triplicate. This project showed that decreasing this to a single 5-s measurement would increase estimated sample throughput from 22,000 articles per year to ≈180,000 articles annually, with minimal reduction in the ability of the XRF to correctly identify articles exceeding the LPCL.

Based on our findings, the following recommendations are made:

 Future studies should widen the scope of PFAS measured in waste articles. A – not exhaustive – list of additional PFAS that should be targeted in future studies is as follows: C₂-C₃ PFCAs and perfluorosulfonic acids; 6:2 and 8:2 polyfluoroalkyl phosphoric acids mono esters and diesters; perfluoro-1-octanesulfonamidoacetic acid, and its methyl and ethyl derivatives; 4:2, 6:2 and 8:2 fluorotelomer sulfonic acids; major components of the F53-B commercial product (9-chlorohexadecafluoro-3-oxanonane-1-sulfonate and 11-chloroeicosafluoro-3-oxaundecane-1-sulfonate); 2-perfluorohexyl ethanol (6:2), 2-perfluorooctyl ethanol (8:2) and 2-perfluorodecyl ethanol (10:2); 2,3,3,3-tetrafluoro-2(1,1,2,2,3,3,3-heptafluoropropoxy)propanoic acid; dodecafluoro-3H-4,8-dioxanonanoate; and perfluoro-4-ethylcyclohexanesulfonate. Measurements of additional PFAS should be undertaken alongside measurements of total organic fluorine. Combined, this will help identify waste articles containing elevated concentrations of PFAS. In addition, where the concentration of total organic fluorine in a sample exceeds the sum of concentrations of all targeted PFAS, it will highlight the likely presence of other unidentified PFAS.

- To further enhance understanding of the presence of other potentially hazardous chemicals present in the waste stream, measurement of other emerging contaminants should be undertaken. A particular example is organic ultraviolet stabiliser compounds, such as benzophenones and benzotriazoles. Among other applications, these are added to plastic casings of electrical and electronic goods to minimise ultraviolet degradation of the plastic.
- Consideration should be given to the impact on the circular economy of lowering the LPCL value for PBDEs and HBCDD. While lowering the limit will prevent more of these BFRs from entering the recycling stream, it will also increase the mass of waste that cannot be recycled.
- Using portable XRF instruments to check whether waste articles comply with LPCL values for PBDEs and HBCDD is technically feasible, although the technique incorrectly identifies whether the LPCL is exceeded in around 10% of articles. The speed with which articles can be checked for LPCL compliance by portable XRF can be considerably increased by scanning the XRF once per article for 5s to allow a single instrument to screen ≈180,000 articles per year. While portable XRF can be considered a potential pragmatic solution to the need to check that waste complies

with LPCL values, the large quantities of waste requiring checking means that research is urgently needed into how screening approaches such as portable XRF can be incorporated into a compliance checking approach that the waste management industry is able to implement "on the ground". Low-cost technologies such as density separation of waste plastics that separate out the denser material that is enriched with bromine (a heavy element) have been applied successfully by some recyclers, and detailed investigation of how such approaches can be combined with, for example, portable XRF within a viable compliance checking strategy is recommended.

- A concentration limit value of 1000 mg/kg on the permissible concentrations of TDCIPP, TCIPP and TCEP in waste articles above which such articles cannot be recycled is recommended. While enforcing such a limit would render ≈2800 t/year of waste unrecyclable, it would prevent 144,000 kg per year of these chemicals from entering the recycling stream.
- While comparison between data obtained for waste articles collected in 2015–16 and 2019–20 provide a preliminary indication of changes in concentrations of HFRs, further monitoring is required if the impact of legislation designed to eliminate HFRs from the waste stream is to be fully evaluated. Specifically, measurements of HFRs in similar samples as analysed in this study, and in the WAFER study conducted in 2015–16, are required to evaluate future temporal trends in HFR concentrations in the Irish waste stream.
- Given the elevated concentrations of many HFRs detected in foams and fabrics from waste childcare articles, ongoing monitoring of HFR concentrations in such articles is recommended. It is also recommended that the human exposure implications of the presence of such HFRs in childcare articles be evaluated.

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Abbreviations

BDE	Bromodiphenyl ether
BDE-209	Decabromodiphenyl ether
BEH-TEBP	Bis(2-ethylhexyl)tetrabromophthalate
BFR	Brominated flame retardant
bw	Body weight
C&D	Construction and demolition
CI-OPEs	Chlorinated organophosphate esters
DBDPE	Decabromodiphenyl ethane
DP	Dechlorane plus
ECHA	European Chemicals Agency
EEE	Electrical and electronic equipment
EFSA	European Food Safety Authority
EH-TBB	2-Ethylhexyl tetrabromobenzoate
ELV	End-of-life vehicle
EPS	Expanded polystyrene
EtFOSA	Ethyl perfluorooctane sulfonamide
EtFOSE	Ethyl perfluorooctane sulfonamide ethanol
FM-550	Firemaster-550
FOSA	Perfluorooctane sulfonamide
FR	Flame retardant
HBCDD	Hexabromocyclododecane
HFR	Halogenated flame retardant
IT	Information technology
LHA	Large household appliance
LOD	Limit of detection
LOQ	Limit of quantification
	Low persistent organic pollutant concentration limit
MeFOSA	Methyl perfluorooctane sulfonamide
MeFOSE	Methyl perfluorooctane sulfonamidoethanol
NBFR	Novel brominated flame retardant
PBDE	Polybrominated diphenyl ether
PFAS	Per- and polyfluoroalkyl substances
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonate
PFCA	
PFDA	Perfluorocarboxylic acid Perfluorodecanoic acid
PFDoA	Perfluorododecanoic acid
PFDOA	Perfluorodecane sulfonate
PFHpA	Perfluoroheptanoic acid
PFHpS	
РГПРЗ РFHxA	Perfluoroheptane sulfonate Perfluorohexanoic acid
PFHXA	
PFNA	Perfluorohexane sulfonate/perfluorohexanesulfonic acid Perfluorononanoic acid
PFNA	Periluorononane sulfonate
_	
PFOA	Perfluorooctanoic acid

DEOG	Derfluereestene culterete
PFOS	Perfluorooctane sulfonate
PFPeS	Perfluoropentane sulfonate
PFTrDA	Perfluorotridecanoic acid
PFTrDS	Perfluorotridecane sulfonate
PFuDA	Perfluoroundecanoic acid
PFuDS	Perfluoroundecane sulfonate
POP	Persistent organic pollutant
PUF	Polyurethane foam
RSD	Relative standard deviation
SDA	Small domestic appliance
TBBP-A	Tetrabromobisphenol-A
TBECH	Tetrabromoethylcyclohexane
TCEP	Tris(2-chloroethyl) phosphate
TCIPP	Tris(1-chloro-2-propyl) phosphate
TDCIPP	Tris(1,3-dichloro-2-propyl) phosphate
TTBP-TAZ	2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5-triazine
USEPA	United States Environmental Protection Agency
WEEE	Waste electrical and electronic equipment
XPS	Extruded polystyrene
XRF	X-ray fluorescence

Appendix 1 Peer-reviewed Publications Emerging from the Research Undertaken in This Project

- Daniel Drage, Martin Sharkey, Layla Salih Al-Omran, William A. Stubbings, Harald Berresheim, Marie Coggins, André Henrique Rosa, Stuart Harrad. Halogenated flame retardants in Irish waste polymers: concentrations, legislative compliance, and preliminary assessment of temporal trends. *Environmental Pollution* 309: 119796 (2022).
- Stuart Harrad, Martin Sharkey, William A. Stubbings, Misbah Alghamdi, Harald Berresheim, Marie Coggins, André Henrique Rosa, Daniel Drage. Chlorinated organophosphate esters in Irish waste foams and fabrics: concentrations, preliminary assessment of temporal trends and evaluation of the impact of a concentration limit value. Science of the Total Environment 859: 160250 (2023).
- Stuart Harrad, Daniel Drage, Martin Sharkey, Will Stubbings, Misbah Alghamdi, Harald Berresheim, Marie Coggins, André Henrique Rosa. Waste childcare articles from Ireland contain elevated concentrations of halogenated flame retardants. *Environmental Pollution* 317: 120732 (2023).
- Martin Sharkey, Daniel Drage, Stuart Harrad, William Stubbings, André Henrique Rosa, Marie Coggins, Harald Berresheim. POP-BFRs in consumer products: evolution of the efficacy of XRF screening for legislative compliance over a 5-year interval and future trends. *Science of the Total Environment* 853: 158614 (2022).
- Daniel Drage, Martin Sharkey, Harald Berresheim, Marie Coggins, Stuart Harrad. Rapid determination of selected PFAS in textiles entering the waste stream. *Toxics* 11: 55 (2023).

An Ghníomhaireacht Um Chaomhnú Comhshaoil

Tá an GCC freagrach as an gcomhshaol a chosaint agus a fheabhsú, mar shócmhainn luachmhar do mhuintir na hÉireann. Táimid tiomanta do dhaoine agus don chomhshaol a chosaint ar thionchar díobhálach na radaíochta agus an truaillithe.

Is féidir obair na Gníomhaireachta a roinnt ina trí phríomhréimse:

Rialáil: Rialáil agus córais chomhlíonta comhshaoil éifeachtacha a chur i bhfeidhm, chun dea-thorthaí comhshaoil a bhaint amach agus díriú orthu siúd nach mbíonn ag cloí leo.

Eolas: Sonraí, eolas agus measúnú ardchaighdeáin, spriocdhírithe agus tráthúil a chur ar fáil i leith an chomhshaoil chun bonn eolais a chur faoin gcinnteoireacht.

Abhcóideacht: Ag obair le daoine eile ar son timpeallachta glaine, táirgiúla agus dea-chosanta agus ar son cleachtas inbhuanaithe i dtaobh an chomhshaoil.

I measc ár gcuid freagrachtaí tá:

Ceadúnú

- > Gníomhaíochtaí tionscail, dramhaíola agus stórála peitril ar scála mór;
- Sceitheadh fuíolluisce uirbigh;
- Úsáid shrianta agus scaoileadh rialaithe Orgánach Géinmhodhnaithe;
- Foinsí radaíochta ianúcháin;
- Astaíochtaí gás ceaptha teasa ó thionscal agus ón eitlíocht trí Scéim an AE um Thrádáil Astaíochtaí.

Forfheidhmiú Náisiúnta i leith Cúrsaí Comhshaoil

- > Iniúchadh agus cigireacht ar shaoráidí a bhfuil ceadúnas acu ón GCC;
- Cur i bhfeidhm an dea-chleachtais a stiúradh i ngníomhaíochtaí agus i saoráidí rialáilte;
- Maoirseacht a dhéanamh ar fhreagrachtaí an údaráis áitiúil as cosaint an chomhshaoil;
- > Caighdeán an uisce óil phoiblí a rialáil agus údaruithe um sceitheadh fuíolluisce uirbigh a fhorfheidhmiú
- Caighdeán an uisce óil phoiblí agus phríobháidigh a mheasúnú agus tuairisciú air;
- Comhordú a dhéanamh ar líonra d'eagraíochtaí seirbhíse poiblí chun tacú le gníomhú i gcoinne coireachta comhshaoil;
- > An dlí a chur orthu siúd a bhriseann dlí an chomhshaoil agus a dhéanann dochar don chomhshaol.

Bainistíocht Dramhaíola agus Ceimiceáin sa Chomhshaol

- > Rialacháin dramhaíola a chur i bhfeidhm agus a fhorfheidhmiú lena n-áirítear saincheisteanna forfheidhmithe náisiúnta;
- Staitisticí dramhaíola náisiúnta a ullmhú agus a fhoilsiú chomh maith leis an bPlean Náisiúnta um Bainistíocht Dramhaíola Guaisí;
- An Clár Náisiúnta um Chosc Dramhaíola a fhorbairt agus a chur i bhfeidhm;
- Reachtaíocht ar rialú ceimiceán sa timpeallacht a chur i bhfeidhm agus tuairisciú ar an reachtaíocht sin.

Bainistíocht Uisce

- Plé le struchtúir náisiúnta agus réigiúnacha rialachais agus oibriúcháin chun an Chreat-treoir Uisce a chur i bhfeidhm;
- > Monatóireacht, measúnú agus tuairisciú a dhéanamh ar chaighdeán aibhneacha, lochanna, uiscí idirchreasa agus cósta, uiscí snámha agus screamhuisce chomh maith le tomhas ar leibhéil uisce agus sreabhadh abhann.

Eolaíocht Aeráide & Athrú Aeráide

- Fardail agus réamh-mheastacháin a fhoilsiú um astaíochtaí gás ceaptha teasa na hÉireann;
- Rúnaíocht a chur ar fáil don Chomhairle Chomhairleach ar Athrú Aeráide agus tacaíocht a thabhairt don Idirphlé Náisiúnta ar Ghníomhú ar son na hAeráide;

 Tacú le gníomhaíochtaí forbartha Náisiúnta, AE agus NA um Eolaíocht agus Beartas Aeráide.

Monatóireacht & Measúnú ar an gComhshaol

- Córais náisiúnta um monatóireacht an chomhshaoil a cheapadh agus a chur i bhfeidhm: teicneolaíocht, bainistíocht sonraí, anailís agus réamhaisnéisiú;
- Tuairiscí ar Staid Thimpeallacht na hÉireann agus ar Tháscairí a chur ar fáil;
- Monatóireacht a dhéanamh ar chaighdeán an aeir agus Treoir an AE i leith Aeir Ghlain don Eoraip a chur i bhfeidhm chomh maith leis an gCoinbhinsiún ar Aerthruailliú Fadraoin Trasteorann, agus an Treoir i leith na Teorann Náisiúnta Astaíochtaí;
- Maoirseacht a dhéanamh ar chur i bhfeidhm na Treorach i leith Torainn Timpeallachta;
- Measúnú a dhéanamh ar thionchar pleananna agus clár beartaithe ar chomhshaol na hÉireann.

Taighde agus Forbairt Comhshaoil

- Comhordú a dhéanamh ar ghníomhaíochtaí taighde comhshaoil agus iad a mhaoiniú chun brú a aithint, bonn eolais a chur faoin mbeartas agus réitigh a chur ar fáil;
- Comhoibriú le gníomhaíocht náisiúnta agus AE um thaighde comhshaoil.

Cosaint Raideolaíoch

- Monatóireacht a dhéanamh ar leibhéil radaíochta agus nochtadh an phobail do radaíocht ianúcháin agus do réimsí leictreamaighnéadacha a mheas;
- Cabhrú le pleananna náisiúnta a fhorbairt le haghaidh éigeandálaí ag eascairt as taismí núicléacha;
- > Monatóireacht a dhéanamh ar fhorbairtí thar lear a bhaineann le saoráidí núicléacha agus leis an tsábháilteacht raideolaíochta;
- Sainseirbhísí um chosaint ar an radaíocht a sholáthar, nó maoirsiú a dhéanamh ar sholáthar na seirbhísí sin.

Treoir, Ardú Feasachta agus Faisnéis Inrochtana

- > Tuairisciú, comhairle agus treoir neamhspleách, fianaisebhunaithe a chur ar fáil don Rialtas, don tionscal agus don phobal ar ábhair maidir le cosaint comhshaoil agus raideolaíoch;
- > An nasc idir sláinte agus folláine, an geilleagar agus timpeallacht ghlan a chur chun cinn;
- Feasacht comhshaoil a chur chun cinn lena n-áirítear tacú le hiompraíocht um éifeachtúlacht acmhainní agus aistriú aeráide;
- > Tástáil radóin a chur chun cinn i dtithe agus in ionaid oibre agus feabhsúchán a mholadh áit is gá.

Comhpháirtíocht agus Líonrú

> Oibriú le gníomhaireachtaí idirnáisiúnta agus náisiúnta, údaráis réigiúnacha agus áitiúla, eagraíochtaí neamhrialtais, comhlachtaí ionadaíocha agus ranna rialtais chun cosaint chomhshaoil agus raideolaíoch a chur ar fáil, chomh maith le taighde, comhordú agus cinnteoireacht bunaithe ar an eolaíocht.

Bainistíocht agus struchtúr na Gníomhaireachta um Chaomhnú Comhshaoil

Tá an GCC á bainistiú ag Bord lánaimseartha, ar a bhfuil Ard-Stiúrthóir agus cúigear Stiúrthóir. Déantar an obair ar fud cúig cinn d'Oifigí:

- 1. An Oifig um Inbhunaitheacht i leith Cúrsaí Comhshaoil
- 2. An Oifig Forfheidhmithe i leith Cúrsaí Comhshaoil
- 3. An Oifig um Fhianaise agus Measúnú
- 4. An Oifig um Chosaint ar Radaíocht agus Monatóireacht Comhshaoil
- 5. An Oifig Cumarsáide agus Seirbhísí Corparáideacha

Tugann coistí comhairleacha cabhair don Ghníomhaireacht agus tagann siad le chéile go rialta le plé a dhéanamh ar ábhair imní agus le comhairle a chur ar an mBord.



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