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Assessment of brominated flame retardants in a small mixed waste electronic and electrical equipment (WEEE) plastic recycling stream in the UK

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Abstract

Identifying the presence of brominated flame retardants (BFRs) within individual polymer types prior to extrusion has given us a unique perspective on which polymers may be problematic in meeting European Union (EU) low persistent organic pollutant (POP) content limits (LPCLs) and the potential for mixed engineering plastics (MEP) to be used as a viable recycled product. Our findings suggest that careful management of the polymer types within the feed chips prior to extrusion could deliver extruded polymer pellets that meet the EU LPCL values for POP-BFRs (i.e. <1000 mg/kg). Within this study, three fractions of extruded polymer pellets ("light", "medium", and "heavy" MEP) were created using density separation. Each fraction was characterised for 28 legacy and novel BFRs with brominated diphenyl ether-209 (BDE-209) (68–37,000 mg/kg) and tetrabromobisphenol-A (TBBP-A) (17–120,000 mg/kg) both predominant and ubiquitous. Portable X-ray fluorescence (XRF) was utilised to measure Br in 120 individual MEP chips of various polymer types. Those chips that XRF

flagged as having high Br concentrations (>2500 mg/kg) were subjected to further evaluation for BFR content *via* mass spectrometry analysis and the results compared with the XRF Br data. This revealed that in 22% of the 120 chips studied, XRF incorrectly identified the LPCL to be exceeded. Our data also identifies the presence of the novel BFRs decabromodiphenyl ethane (DBDPE) and 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE) in plastics derived from waste electronic and electrical equipment (WEEE). While the "light-MEP" samples contained POP-BFR concentrations below LPCLs, the "medium-MEP" and "heavy-MEP" fractions exceeded such limits. Management of the polymer chips by colour sorting resulted in significant reductions in concentrations of all BFRs in the clear polymers such that LPCL limits were not exceeded; however, concentration reductions in white polymers were insufficient to meet LPCLs.

1. Introduction

The move to a circular economy is vital for the development of industry and the protection of the environment (European Commission, 2018). Plastics are materials constantly in the spotlight and have received a substantial degree of public and political interest. Waste electronic and electrical equipment (WEEE) is one the fastest growing waste streams globally with an estimated 1.6 million tonnes generated in the UK, 12.3 million tonnes generated within the European Union (EU), and 44.7 million tonnes generated globally in 2016 (Baldé et al., 2017). Of this, the estimated proportion recycled in 2016 was 41.3% across the 28 member states of the EU and 49.8% in the UK (Eurostat, 2020). It has been estimated that the average plastics content of WEEE is ~30% by weight (Schlummer et al., 2007).

A significant volume of WEEE plastic is collected but not currently recycled and is destined for disposal via incineration. This material is referred to as mixed engineering plastic (MEP),

48 which comprises a wide range of polymers that can contain hazardous legacy additives of
49 potential concern when recycling the material (Drage et al., 2018). The current EU practice in
50 material recycling of WEEE plastics separates only a fraction of light polyolefin blend
51 polymers and all other MEP polymers are sent for disposal. However, at a small number of
52 the largest facilities, a minor quantity of other polymers can also be separated for recycling
53 using advanced techniques. In the UK, it is estimated that 134000 tonnes of "small mixed
54 WEEE" (SMW) was collected from business and consumer waste in 2020, of which ~40000
55 tonnes of SMW plastics are estimated to be collected (30% by weight), from which 27000
56 tonnes of MEP are estimated to be generated (Comply Direct, 2020; pers. comm. Axion). In
57 the EU, the SMW stream includes a broad range of WEEE items including: small household
58 appliances, information technology and communications equipment, powered tools, toys and
59 sports equipment, medical devices, control instruments, smoke detectors, and dispensers. It
60 does not include other larger WEEE items such as: cathode ray tube and flat screen
61 televisions and monitors, refrigeration equipment, large domestic appliances, and fluorescent
62 lamps (Environment Agency, 2014). SMW accounted for 26-31% of total WEEE collected in
63 the UK between 2015-2019 (Comply Direct, 2020).

64 A particular concern is that MEP contains brominated flame retardants (BFRs), of which
65 polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) are
66 classed as persistent organic pollutants (POPs) under the United Nations Environment
67 Programme (UNEP) Stockholm Convention. Typically, some polymers used in WEEE, most
68 commonly acrylonitrile butadiene styrene (ABS), and high impact polystyrene (HIPS) are
69 known to have been treated with BFRs at percent by weight levels in order to meet fire safety
70 regulations (UNEP 2006; UNEP 2010). While the principal BFRs detected in EEE polymers
71 are decabromodiphenyl ether (Deca-BDE), and tetrabromobisphenol-A (TBBP-A) (Drage et
72 al, 2018), evidence exists of application of HBCDD in a limited proportion of EEE plastics

(Harrad et al, 2009; Drage et al, 2018). Uncontrolled recycling of POP-BFR (i.e. PBDE and HBCDD) contaminated WEEE polymers has led to the presence of these banned BFRs in recycled plastic products (e.g. children's toys and kitchen utensils) with consequent health and exposure implications (Guzzonato et al., 2017; Kuang et al., 2018; Fatunsin et al., 2020). As a result, the presence of these and other chemical contaminants in polymers has led to the classification of WEEE as hazardous waste. Moreover, EU Regulation 850/2004 *inter alia* specifies in Annex IV "low POP content limits" (LPCLs) that apply to PBDEs as a sum of all representative PBDE congeners; including Penta-, Octa-, and/or DecaBDE formulations, and also separately to HBCDD as a sum of all isomers (European Commission, 2004). Waste materials in which PBDE or HBCDD concentrations exceed the LPCL (1000 mg/kg; each) cannot be recycled or re-used and must be treated in such a way as to ensure the POP-BFR content is destroyed or irreversibly transformed - most likely in a high temperature incineration process, losing all value in the material and negatively impacting the environment. However, these LPCLs are currently under review with the aim of adopting a legislative limit value lowered to 500 mg/kg as quickly as possible and no later than 16th July 2021 (European Commission, 2019). Furthermore, the EU has also introduced an unintentional trace contaminant (UTC) fixed threshold value for tetra-, penta-, hexa-, hepta- and decaBDE equal to or below 10 mg/kg for each homologue group (European Commission, 2018), with UTC limits for HBCDD equal to or below 100 mg/kg, to be introduced no later than July 2021 (European Commission, 2019). These UTC limits are designed to prevent intentional and meaningful use of these chemicals in products while allowing recycling and enabling control and enforcement by being high enough to be detected by current detection methods.

In order to verify compliance with LPCLs and UTCs, concentrations of PBDEs and HBCDD must be determined in waste polymers. However, this currently requires the use of gas or

liquid chromatography coupled with mass spectrometry and is thus an expensive, time consuming, and destructive process. Recently, several studies have suggested the use of portable X-ray fluorescence (XRF) measurements of elemental Br as a surrogate measure of POP-BFR concentrations in EEE plastics (Gallen et al., 2014; Aldrian et al., 2015; Guzzonato et al., 2016; Sharkey et al., 2018). However, use of portable XRF spectrometry to identify items exceeding LPCL values has been shown in some instances (~6 % of 538 waste EEE, foam, and fabric samples tested in the Republic of Ireland; Harrad et al., 2019) to result in false exceedances (defined as situations where the concentration of bromine but not PBDEs or HBCDD exceeds the LPCL). This was attributed to the presence of other currently non-regulated Br-containing compounds used in EEE plastics such as TBBP-A and potentially decabromodiphenyl ethane (DBDPE) (Harrad et al., 2019). The "Sink/Float" method is the most commonly used method for separating BFR containing fractions and is simply based on the higher density of plastics containing BFRs (Sofies, 2020).

Identification of potentially recyclable waste polymers that comply with LPCL values is important as there is increasing global demand to meet sustainability targets and increase the proportion of waste plastics recycled for all key applications. The EU has set ambitious targets of recycling greater than 50% of all plastic waste generated in Europe by 2030 (European Commission, 2018). Minimum recycling and reuse targets for different categories of WEEE are set out in Annex V to Directive 2012/19/EU on WEEE and range between 50% and 80% to be prepared for reuse and/or recycled, depending on the category (European Commission, 2012). Currently, large-scale, efficient separation of mixed plastics into individual polymers, or even specific grades so the materials can be reused and/or recycled, is at best a work in progress and at worst prohibitively expensive. For example, the use of near infra-red (NIR) technology widely used in plastic recycling facilities to sort polymers into different categories, is prevented by the presence of carbon black pigments and therefore

cannot be employed to sort MEP waste streams where a large proportion of polymers are black (WRAP, 2020). Current recovery activities at large scale WEEE recyclers in Europe only allow for separation of three of the >15 different polymer types (see Table S3; SI) with variable efficiencies. New polymer separation technologies such as Raman spectroscopy, mid-range infra-red spectroscopy (Mid-IR), and X-ray transmission (XRT) have high capital costs and are difficult to implement at larger scales. XRT for example may result in over sorting and larger volumes of plastic sent to incineration than if XRF were implemented. Moreover, the potential presence of PBDEs and HBCDD in WEEE creates a significant obstacle to achieving greater rates of recycling (European Commission, 2018). Due to the scale of the challenge, it is clear that innovative and creative solutions are required to transform the plastics industry into a full-circle recycling service. The manufacture of MEP material provides a potential route to utilise this otherwise unrecyclable mix of polymer material and recover some of its original value. Within a recycling context, MEP material is produced by sorting and combining heterogeneous mixtures of chipped or shredded polymers into predefined compositions to meet specific engineering characteristics. These are then passed through a melt extruder thus resulting in "homogenised" extruded pellets of MEP that can be utilised downstream to produce new recycled polymer products. Normally, a blend extrusion of different plastics would not be considered viable due to problems with engineering properties (such as incompatible melting or degradation points). However, due to high disposal costs via incineration, exploration of the possibility of creating a product that is composed of multiple polymer types, but with lowered engineering qualities (e.g. impact strength), has become a viable opportunity for specific product applications, providing they can also meet LPCLs and UTCs.

In order to effectively recycle this material, two developments are thus required: (1) a useable product from MEP that meets required engineering characteristics; and (2) enhanced

148 understanding of whether POP-BFRs are present and if so, which fractions of the overall
149 MEP feedstock contain PBDEs and HBCDD at concentrations that do not exceed the LPCL.
150 This knowledge can be used alongside an understanding of the engineering properties of such
151 fractions to produce a final recycled product that is both useable in an engineering context
152 while meeting the LPCLs.

153 Against this backdrop, this paper reports concentrations of both regulated and non-regulated
154 BFRs within different fractions of a waste polymer stream consisting of SMW plastics.
155 Additionally, we evaluate further the use of handheld XRF as a tool for screening for
156 compliance with LPCL values. Identifying the presence of BFRs within individual polymer
157 types prior to extrusion has given us a unique perspective on which polymers may be
158 problematic in meeting LPCLs and the potential for MEP to be used as a viable product.

159

160 **2. Materials and methods**

161 **2.1 Chemicals**

162 All solvents used for extraction and both gas chromatography–mass spectrometry (GC-MS)
163 and liquid chromatography–tandem mass spectrometry (LC–MS/MS) analyses were of HPLC
164 grade (Fisher Scientific, Loughborough, UK). Concentrated sulfuric acid (99.7%) was
165 purchased from Sigma–Aldrich (St. Louis, MA, USA).

166 Individual α-, β- and γ-HBCDD standards, $^{13}C_{12}$-α-, β- and γ-HBCDD, d_{18}-γ-HBCDD, as
167 well as individual standards of BDEs 28, 47, 77, 99, 100, 128, 153, 154, 183, 209, $^{13}C_{12}$-BDE
168 209, and $^{13}C_{12}$-TBBP-A were purchased from Wellington Laboratories (Guelph, ON,
169 Canada). A full list of all target analytes are presented in the Table S1 of the supporting
170 information.

171 Certified reference materials (CRMs) for low density polyethylene (LDPE) (ERM-EC590)
172 and polypropylene (PP) (ERM-EC591) were purchased from IRMM (Brussels, Belgium).

173 **2.2 Sampling**

174 Sample collection at the polymer recycling facility and extrusion of MEP pellets was
175 conducted by Axion Recycling Ltd. All samples of polymeric material were provided by
176 Axion Recycling Ltd. to the University of Birmingham. Axion Polymers are a major recycler
177 of plastics derived from waste electronic and electrical equipment (WEEE). They operate one
178 of the most advanced sorting and recycling facilities in Europe and already recycle over
179 150,000 tonnes of shredder residue per annum.

180

181 **2.3 Sample treatment**

182 Batches of MEP chips were separated into three, roughly equal by mass, fractions using
183 density separation: “Light-MEP”, “Medium-MEP”, and “Heavy-MEP”. The criteria for
184 density separation are Axion's proprietary information and thus cannot be included here.
185 Samples were generated from production by taking several grab samples of chipped (<15 mm
186 diameter) MEP over time (typically 1 – 2 kg, up to 5 times per day) from each of these three
187 fractions. Typically, the samples were then mixed well within their class before 2 kg of
188 plastic chips were selected and extruded using a single screw lab scale extruder to form the
189 “homogenised” MEP pellets, with subsamples (ca. 200 mg) of this extruded material taken
190 for analysis. In all instances PVC chips were removed prior to extrusion. Batches of ~500 g
191 of extruded pellets were prepared for each MEP fraction.

192 **2.3.1 FTIR analyses to characterise polymer type**

193 A representative random grab sample of the mixed polymer stream infeed was taken and the
194 material dried. This was taken as part of a routine test conducted on a weekly basis to identify
195 the constituent polymer of each chip. From this grab sample, 1 kg of chipped polymer (<15
196 mm diameter) was selected with visible contaminants (e.g. metal, wires, printed circuit
197 boards, wood, and rubber and other elastomers) removed. Polymer chips were selected and
198 cleaned of any dirt or debris before the polymer type was determined using Fourier-transform
199 infrared spectroscopy (FTIR) on site by Axion Recycling Ltd., Salford, UK. Using this
200 process, 217 individual polymer chips were grouped by polymer type and sent to University
201 of Birmingham for determination of concentrations of Br and BFRs (97 chips were analysed
202 by MS for BFRs + 120 were screened by XRF (of which 27 were then analysed by MS for
203 BFRs) = 217 total individual chips). The chips analysed in this study were obtained in
204 September 2019.

205

206 **2.3.2 Determination of concentrations of Br**

207 Concentrations of Br in polymer chips were determined using a handheld XRF device (Niton
208 XL3t 700 GOLDD+; Niton, UK). The sample to be analysed (minimum polymer sample
209 thickness 2 mm) was mounted onto the sampling stand with a lead lined housing and the
210 surface of the sample cleaned of any visible dust and dirt using a clean tissue. Measurement
211 of Br was then carried out by irradiating for no fewer than 30 seconds using the large (X-ray)
212 spot size, with the device locked into position and centred over the target area in the housing
213 throughout the measurement period. Full methodology and QA/QC procedures have been
214 reported previously (Sharkey et al., 2018). The limit of detection (LOD) of the Niton XRF
215 Analyzer varies according to the element targeted, the matrix and the (X-ray) spot size used.

Overall, it ranges between 1-10 mg/kg. For Br in plastics using the large spot size, LOD = 3-8 mg/kg. Using the small spot size, LOD = 5-20 mg/kg with an average of ~10 mg/kg.

2.3.3 Measurement of BFRs in MEP chips and extruded pellets

2.3.3.1 Polymer extraction and extract purification

The method used to extract BFRs from polymer samples and purify resultant extracts was adapted from a previously validated method (Abdallah et al., 2017), where dichloromethane (DCM) was used as extraction solvent and samples were not required to be pulverised prior to extraction. Extruded pellets were taken at random from a 500 g sample of MEP pellets, cut into small pieces (<2 mm^2) using metal shears, then mixed, and aliquots (ca. 200 mg) accurately weighed before being transferred into 15 mL glass centrifuge tubes. Each of the "white" and "clear" fraction extruded pellets were analysed in triplicate, whereby three (ca. 200 mg) subsample aliquots were taken from the extruded pellets of each colour fraction. When analysing individual polymer chips pre-identified using FTIR, in almost all instances the whole chip was used and cut into smaller pieces (< 2 mm^2) to enhance extraction.

Samples were extracted by adding approximately 3 mL of DCM before vortexing for 2 min. followed by ultrasonication for 5 min to effect dissolution of the polymer. The resultant extract was collected in a separate centrifuge tube and the process repeated twice, collecting all DCM extracts in the same centrifuge tube. The combined extracts were then evaporated to approx. 2 mL at 40 °C under a gentle stream of nitrogen. Two mL of hexane were then added to the sample to precipitate dissolved polymer.

The sample was then evaporated to < 1 mL and reconstituted in 2 mL in hexane (to ensure removal of DCM) and vortexed for 2 min. Approx. 2 mL of >98% concentrated sulfuric acid was added to the sample and vortexed for 30 s. Samples were left for at least 1 h followed by

centrifugation at 3000 × *g* for 5 min to separate the aqueous and organic layers. 100 µL of the clean supernatant hexane layer was collected in a glass tube and diluted to 10 mL in hexane. A further 100 µL of the diluted sample was taken and spiked with 30 ng of all internal (surrogate) standards (IS) ($^{13}C_{12}$ α-, β- and γ-HBCDD, BDEs 77 and 128, and $^{13}C_{12}$-TBBP-A), except for $^{13}C_{12}$-BDE-209, of which 120 ng was added to the sample extract before reconstitution in 100 µL toluene containing 0.2 ng/µL $^{13}C_{12}$-BDE-100 and 0.2 ng/µL d_{18}-γ-HBCDD as recovery determination standard. BDEs 77 and 128 were used to quantify concentrations for BDEs 28, 47, 99, 100, 153, 154, and 184, and also NBRs: PBBz, HBBz, and BTBPE. While $^{13}C_{12}$-BDE-209 was used to quantify concentrations of BDE-209 and DBDPE, $^{13}C_{12}$-TBBP-A was used to quantify concentrations of TBBP-A. After reconstitution, the sample extract was transferred to a glass inserted auto-sampler vial for quantitative analysis of PBDEs, novel brominated flame retardants (NBFRs) and TBBP-A by GC–MS and HBCDD by LC-MS/MS.

2.3.3.2 GC-MS and LC-MS/MS methods

Quantitative analysis of PBDEs, NBFRs, and TBBP-A was performed in a single injection on a ThermoFisher Trace 1310 gas chromatograph coupled to a ThermoFisher ISQ mass spectrometer. The mass spectrometer was operated in electron ionisation mode using selective ion monitoring (SIM). One µL of the purified extract was injected for analysis using a programmable temperature vaporiser (PTV) onto a Restek Rxi-5Sil MS column (15 m × 0.25 mm × 0.25 µm film thickness). Helium was used as the carrier gas at a flow rate of 1.5 mL/min with methane as the reagent gas. More details on instrument conditions and ions monitored are provided in Abdallah et al. (2017).

HBCDDs were measured using a Shimadzu LC–20AB Prominence binary pump liquid chromatograph (Shimadzu, Kyoto, Japan), equipped with a SIL-20A auto-sampler, a DGU-20A3 vacuum degasser coupled to an AB Sciex API 2000 triple quadrupole mass spectrometer (Applied Biosystems, Foster City, CA, USA). Chromatographic separation was achieved using an Agilent Pursuit XRS3 C18 column (150 mm × 2 mm id, 3 μm particle size) and a mobile phase of (i) 1:1 methanol/water with 2 mM ammonium acetate and (ii) methanol at a flow rate of 180 μL/min. Molecular ionisation was achieved using an ESI source operated in negative ion mode. MS/MS detection operated in the multiple reaction monitoring mode was used for quantitative determination of HBCDD isomers based on m/z 640.6→79, m/z 652.4→79 and m/z 657.7→ 79 for the native, $^{13}C_{12}$-labelled and d_{18}-labelled diastereomers, respectively.

2.4 Quality assurance / quality control

Two CRMs, low density polyethylene (LDPE - ERM-EC590), and polypropylene (PP - ERM-EC591), were used in determining method accuracy and precision in the analysis of PBDEs from plastic polymers. Five replicate (ca. 50 mg) analyses of each CRM were conducted using the extraction and clean-up method before commencement of analysis of polymer samples. The method precision and accuracy data obtained are considered satisfactory for these analyses (see Table S2; SI). Mean recoveries of PBDEs in the CRMs were generally very good (80-100% range) and are indicative that other BFRs (of similar physicochemical properties) would have similarly good recoveries.

2.5 Data analysis

286 Basic and descriptive statistics were calculated using IBM SPSS Statistics 26, and Microsoft
287 Excel 2016 software. Plots were generated using SigmaPlot 13 (Systat Software Inc.). The
288 distribution of concentrations for each compound within the data set were evaluated for
289 normality using Shapiro-Wilk test (due to the sample size being <50). The results, combined
290 with visual inspection of frequency diagrams, revealed that both the ΣPOP-BFR and ΣBFR
291 concentration data were log-normally distributed. Hence, Pearson correlation coefficients
292 were obtained for log-transformed data. Total Br concentrations from BFRs quantified *via*
293 GC-MS analysis (MS-Br values) were calculated to facilitate comparison with the XRF
294 measured total Br (XRF-Br) concentrations in these tests (see section S4, SI).

295

296 **2.6 Sample codes**

297 The naming convention used for samples is as follows: Density fraction-polymer type-sample
298 ID number (e.g M-ABS-1 = Medium-MEP, ABS, sample ID 1.) Acronyms for all polymer
299 types used in this paper can be found in Table S3 of the SI.

300

301 **3. Results and discussion**

302 **3.1 Characterisation of the MEP feed**

303 The SMW feed consists of a mixture of 15 predominant polymer types (Table S3).

304 **3.2 Characterisation of the extruded MEP**

305 Figure 1 provides a detailed overview of BFR concentrations within the three main extruded
306 MEP fractions. A statistical summary of BFR concentrations for all extruded MEP fractions
307 analysed is provided in Table S4, with concentrations of BFRs in four subgroupings within
308 the "Medium-MEP" fraction shown in Figure 2. Unsurprisingly, given the relatively low

309 application of HBCDD in the polymers comprising MEP feedstock (Drage et al, 2018),
310 concentrations of HBCDD were below LOQ in all samples analysed.

311 Within the "Light-MEP" fraction, BDE-209 dominates (arithmetic mean 410 mg/kg),
312 followed by the Tri-Octa BDEs (arithmetic mean 67 mg/kg), DBDPE (arithmetic mean 35
313 mg/kg), and TBBP-A (arithmetic mean 30 mg/kg). Notably, the arithmetic mean ΣPBDE and
314 HBCDD concentrations within this fraction are low enough to comply with both current and
315 prospective LPCLs (<1000 mg/kg and <500 mg/kg respectively). However, we do note that
316 in one Light-MEP sample the BDE-209 concentration was 650 mg/kg and in another 500
317 mg/kg. Additionally, we caveat that these results are based on only five replicate analyses and
318 further testing is required to confirm that they consistently pass LPCLs before this fraction
319 could be approved for recycling. We estimate that recycling of the "Light-MEP" fraction
320 would result in an additional 9000 tonnes of SMW recycled per annum (see section S5; SI).

321 BFR concentrations are considerably higher in the "Medium-MEP" fraction when compared
322 to the other MEP fractions examined. This is particularly true for the higher molecular weight
323 BFRs, i.e. TBBP-A, BDE-209, and DBDPE. TBBP-A is the dominant BFR in this density
324 range (arithmetic mean 15,000 mg/kg), followed by BDE-209 (arithmetic mean 6100 mg/kg),
325 the Tri-Octa BDEs (arithmetic mean 2700 mg/kg), and DBDPE (arithmetic mean 660
326 mg/kg).

327 Given these elevated concentrations in the overall "Medium-MEP" fraction, four additional
328 sub-fractions of "Medium-MEP" were examined; referred to here as "Medium-MEP" a-d.
329 These groupings were made to determine if proprietary refinements in the separation of the
330 feed chips could yield lower BFR content in the extruded MEP pellets. The results of these
331 analyses are given in Figure 2 and summarised in Table S4. One sample in the "Medium-
332 MEP-c" group had a TBBP-A concentration of ~12% by weight. Moreover, all sub-fractions

333 contained PBDEs (principally BDE-209) at concentrations that exceed the EU's LPCL of
334 1000 mg/kg.

335 The "Heavy-MEP" fraction contains considerably lower concentrations of BFRs than
336 "Medium-MEP", but these still exceed those found in "Light-MEP" with consequent
337 exceedances of the LPCL values. The greatest contributors to the BFR content of this fraction
338 are BDE-209 (arithmetic mean 1000 mg/kg), DBDPE (arithmetic mean 800 mg/kg), TBBP-A
339 (620 mg/kg), with a relatively low Tri-Octa-BDE contribution (arithmetic mean 99 mg/kg).

340 The variation in BFR concentrations observed within each density class is linked to the
341 heterogeneity of the feed material. Within one pellet may be many different chips melted
342 together and re-extruded. The exact blend is impossible to homogenise completely. SMW is a
343 very diverse feed of different equipment, each piece of SMW equipment can be different
344 depending on the manufacturer, country of origin, or year of production.

345

346 **3.3 Colour separated extruded MEP**

347 Within the "Medium-MEP" fraction, polymer feed chips were hand sorted into two further
348 subdivisions based on colour before extrusion: white (opaque) chips and clear (transparent
349 and translucent) coloured chips (see Fig. S3; SI). Polymer colour categories were measured
350 large scale as part of a routine test conducted on a weekly basis on site by Axion Recycling to
351 characterise the MEP polymer feed. Within the "Medium" fraction, "white" coloured
352 polymers represented 18.8% of the MEP feed, "clear" coloured polymers represented 7.1%,
353 while all other colours represented 74.1% (pers. comm. Axion Recycling Ltd., Salford, UK).
354 Table 1 provides an overview of the concentrations within the "white" and "clear" colour
355 extruded MEP, with the unsorted "mixed colour" Medium-MEP also given as reference.
356 Sorting of the polymer chips by colour prior to extrusion results in a marked reduction of

357 Σtri-octa-BDEs in both the white (arithmetic mean: 51 mg/kg) and clear (arithmetic mean:
358 1.5 mg/kg) MEP, when compared to the "mixed colour" (arithmetic mean: 890 mg/kg).
359 However, sorting by colour makes little difference in reducing the concentration of BDE-209
360 (arithmetic mean: 5000 mg/kg) and TBBP-A (arithmetic mean: 8300 mg/kg) in the "white"
361 MEP. The reduction of POP-BFRs within the white sorted polymers is not sufficient to pass
362 the EU LPCLs and therefore cannot be considered a viable means to do so. Conversely,
363 significant reductions in BFR concentrations are observed for all target compounds in the
364 "clear" sorted MEP resulting in the LPCLs being met. This is likely due to the composition of
365 the polymers included, with an increase in the proportion of polymers containing no or low
366 BFR concentrations (such as poly(methyl methacrylate); PMMA), resulting in a dilution of
367 BFR concentrations. We estimate that recycling of the "clear" Medium-MEP fraction would
368 result in an additional 639 tonnes of SMW recycled per annum.

369

370 **3.4 Concentrations of BFRs in individual chips of different polymer types**

371 Individual SMW polymer chips were separated, and their constituent polymer identified
372 using FTIR prior to the extrusion process. The major polymer types for the "Medium-MEP"
373 and "Heavy-MEP" fractions were identified in this fashion and five chips (ca. 200 mg each)
374 per polymer type were analysed for their BFR content. The "Light-MEP" fraction was not
375 examined here owing to the low concentrations of BFRs within this fraction resulting in
376 compliance with the LPCL. In total, 97 individual chips comprising ten different polymer
377 types were analysed for BFR content. Table 2 provides an overview of the concentrations of
378 BFRs determined in each polymer type within each density fraction.

379 In the "Medium-MEP" fraction; individual chips identified *via* FTIR as: acrylonitrile
380 butadiene styrene (ABS), polycarbonate-acrylonitrile butadiene styrene (PC-ABS),

polyamide (PA), polypropylene (PP), polystyrene-high impact polystyrene (PS-HIPS), PMMA, and polycarbonate (PC) were analysed for BFR content. As indicated by Table 2, PC-ABS and PA did not contain any of the targeted BFRs above detection limits. Moreover, consistent with its absence from extruded pellets, HBCDD was not detected in any individual polymer chips. Of the PC chips, while one sample contained 1100 mg/kg TBBP-A, no other BFRs were detected in the other PC chips. With respect to PP, one chip contained BDE-209 and DBDPE at 200 mg/kg and 250 mg/kg, respectively. All ABS, PMMA, and PS-HIPS chips analysed contained TBBP-A in the range 190–28000 mg/kg, 24–55 mg/kg, and 5.1–47000 mg/kg, respectively. 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE) was determined in 6 of 15 ABS chips mostly in the range of 0.90–40 mg/kg with one chip containing 27000 mg/kg (2.7% wt.). Moreover, BDE-209 was detected in one PMMA sample at 96 mg/kg, while two PS-HIPS chips contained DBDPE at 140 and 330 mg/kg.

Notably, two additional non-regulated BFRs, pentabromobenzene (PBBz) and hexabromobenzene (HBBz), were also present in some PS-HIPS chips at 2.0-13 mg/kg and 6.3-20 mg/kg, respectively. HBBz is a known FR additive used in polymers and EEE, and was detected in five chips, while PBBz was detected in four of the same five chips, suggesting a reductive debromination pathway of HBBz (Watanabe & Sakai, 2003). Tri-octa-BDE congeners were detected at relatively low levels in 9 of 15 “Medium-MEP” chips identified as ABS and in 6 of 13 chips identified as PS-HIPS.

One PS-HIPS chip was found to contain a mixture of PBBz (3.3 mg/kg), HBBz (16 mg/kg), BDE-153 (1.1 mg/kg), BDE183 (7.4 mg/kg), BDE-209 (25000 mg/kg), DBDPE (330 mg/kg), and BTBPE (51 mg/kg). While it is clear that the chip in question was predominantly treated with Deca-BDE, it is interesting that other FR compounds (e.g. HBBz, DBDPE, and BTBPE) are also present within the same chip albeit at much lower concentrations. Possible explanations include: that an unknown proprietary mixture was used to impart flame

406 retardancy, that there was some “carry over” of residues between batches during
407 manufacturing, and/or in the case of PBBz and the lower brominated PBDEs, that some
408 reductive debromination of compounds has taken place during the product lifecycle.
409 However, the most likely explanation is that the chip contained some recycled material,
410 supported by similar findings in a survey of Irish WEEE where quite a few items contained
411 more than one BFR at appreciable concentrations (Drage et al., 2018; Sharkey et al. 2018).

412 These data reveal that in general, ABS and PS-HIPS are the major contributors to the overall
413 BFR concentrations in the “Medium-MEP” fraction. TBBP-A is known to be extensively
414 used in as an additive in HIPS and ABS for EEE (Morf et al., 2003). This likely explains its
415 ubiquity in these polymers in this study at substantial concentrations, often in the percent
416 range and as high as 12% by weight.

417 In the “Heavy-MEP” fraction, individual chips of ABS, PC-ABS, PA, PP, polyvinyl chloride
418 (PVC), POM, and polyethylene terephthalate (PET) were analysed for BFR content, with the
419 results shown in Table 2.

420 PVC, PA, PP, POM did not contain any of the targeted BFRs above detection limits, with the
421 only exception being one PP chip which contained 55 mg/kg TBBP-A. ABS, PC-ABS and
422 PET were the only polymers containing BFRs in the “Heavy-MEP” fraction. Specifically, tri-
423 octa BDE congeners were detected at relatively low levels in 2 of 9 “Heavy-MEP” ABS
424 chips and BDE-209 in 3 of 9 samples. Meanwhile, seven ABS chips contained TBBP-A
425 between 9.0–25,000 mg/kg, one PC-ABS chip contained DBDPE at 350 mg/kg, all ten PET
426 chips contained TBBP-A in the range 13-1300 mg/kg, while one sample also contained
427 DBDPE at 520 mg/kg and another, BTBPE at 52 mg/kg.

428 Overall, PBDEs were only detected in 21 out of the 97 chips analysed. This highlights the
429 heterogeneity of BFR contamination of individual chips, because PBDEs and in particular

BDE-209 are ubiquitous within the extruded MEP mixtures. BFR analysis of a larger quantity of polymer identified individual chips is recommended for future research.

3.5 Handheld XRF screening

After the first round of analysis was completed (measuring BFR concentrations in individual polymers - detailed in the previous section) we then decided to screen the remaining 120 FTIR-identified individual SMW polymer chips by handheld XRF to identify chips with Br concentrations consistent with the elevated presence of BFRs. Of the 120 individual chips tested by handheld XRF, this screening identified a total of 27 high Br concentrations (>2000 mg/kg) within 14 of 20 ABS, 8 of 15 PS/HIPS, and 5 of 15 PET chips. The remaining 93 chips tested (6 ABS, 10 PA, 22 PC-ABS, 10 PET, 10 PMMA, 8 POM, 10 PP, 7 PS/HIPS, and 10 PVC) were identified as having low Br concentrations (<2000 mg/kg, typically 0-250 mg/kg) and were thus excluded from the next stage of analysis. The 27 high-Br content chips were then analysed for BFRs, with the paired Br and BFR concentrations for each of these chips given in Table 3.

The XRF Br overestimates both the BFR concentration and the POP-BFR concentration substantially in 26 of these 27 high BFR content samples with the exception of sample M-PS/HIPS-8. Whether the overestimation of BFR concentration is due to a systematic issue with XRF at these high concentrations (e.g. calibration of Br at higher concentrations) or the widespread presence of unknown BFRs is not possible to say. Pearson's correlation coefficient tests were performed, and a significant negative correlation was observed between the (log-transformed) XRF-Br and (log-transformed) ΣPOP-BFR MS-analysis Br concentration data (r = -0.500, p = 0.035; n = 19, 8 chips contained no PBDEs or HBCDD so were excluded from the analysis). Despite this, no correlation was observed between the (log-

transformed) XRF-Br and (log-transformed) ΣBFR MS-analysis Br concentration data (r = 0.052, p = 0.797; n = 27). The correlation seen for ΣPOP-BFR MS-analysis Br concentration data is owed to one sample (M-PS/HIPS-8 sample) skewing the trendline, with the correlation no longer significant when this sample is omitted from the analysis. For the Medium-MEP PS/HIPS-8 sample, XRF Br (10700 mg/kg) underestimates the POP-BFR concentration total MS-Br (20900 mg/kg), perhaps due to measurement inaccuracies of the portable XRF instrument and/or inhomogeneity of BFR distribution in the polymer chip sample analysed. For reference, Figure S1 (SI) provides the regression plot for the total Br concentrations derived from XRF-Br and MS-Br and Figure S2 the regression analysis between XRF-Br and MS-POP-BFR Br values. The inference of this overestimation of POP-BFR concentrations is that in 27 high Br samples tested, XRF incorrectly identified an individual chip as exceeding an LPCL of 1000 mg/kg. However, 93 "low-Br" (<2000 mg/kg) chips were not tested, of which 90 were shown to be <1000 mg/kg XRF-Br. For these 90 chips it is likely that MS-Br would also be <1000 mg/kg, thus the overall extent of false exceedances is expected to be much lower at 26 of 120 chips (~22%). This is still higher than previously reported in the Republic of Ireland where the rate of false exceedances for WEEE was ~11% (Harrad et al 2019, WAFER project report). These observed inaccuracies could be due to the limited sample size.

3.6 Exploration of the use of waste polymer management to reduce concentrations of PBDEs and HBCDD in recycled polymer products.

The findings of this study are consistent with the findings of the WAFER project which conducted a comprehensive survey of POP-BFRs within the Irish polymer waste stream (including WEEE plastics). In the WAFER project only 4.7% (n=2) of the 43 WEEE display

items examined contained PBDEs at concentrations >1000 mg/kg, yet the average concentration in such samples was 1900 mg/kg (Drage et al., 2018). Consistent with this, our data reveal that although PBDEs may only be present in a relatively small proportion of WEEE plastic chips in total, the relatively high PBDE concentrations (often in the percent range) of those few chips within the MEP feed mixture, results in PBDEs being present at concentrations approaching or exceeding the LPCL within homogenised extruded MEP pellets.

Our data reveals that while several polymer types (PA, PC, PC-ABS, PET, POM, and PVC) did not contain PBDEs, they were detected at substantial concentrations in ABS, PS-HIPS, and to a lesser extent PP and PMMA. This implies that effective sorting and separation of these polymer types from the SMW feed prior to extrusion could result in a recycled product that complies with LPCLs. Additionally, careful, effective management of the blend of polymer types within the feed chips prior to extrusion could allow extruded MEP pellets to meet current or proposed EU LPCLs, while also retaining desirable engineering properties (e.g. material strength and density) to deliver viable products. Furthermore, separations resulting in "Light-MEP" and colour separation ("clear" MEP) can also provide useful, simple, practical means of increasing the volume of WEEE plastic recycling.

The use of portable XRF analysis as a screening tool for determining compliance with POP-BFR LPCL values in the SMW polymer feed works to prevent POP-BFRs from entering the MEP recycling stream. However, our data indicates that its use as a screening tool for WEEE plastic results in a high proportion of false exceedances, where the measurement of XRF-Br overestimates exceedances of legislative limits on POP-BFR concentrations. This is mostly due to the high detection frequency of TBBP-A in these samples as well as other unidentified BFRs. Most of the Br content in the SMW polymer is attributed from the contribution of unknown brominated compounds not measured in this study. In turn, this can lead to an

503 overestimation of the volume of SMW polymer chips that are incorrectly identified as non-
504 recyclable. However, TBBP-A is subject to the H14 criteria of hazardous waste for which
505 waste can be classified as hazardous if TBBP-A is higher than 2500 ppm (Wagner and
506 Schlummer 2020).

507 Furthermore, plastics recycling from WEEE treats ~ 1 tonne per hour. This volume cannot be
508 pre-screened by handheld XRF. However, development of technologies such as XRT may
509 provide future solutions.

510

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637 **Table 1: Statistical summary of concentrations (mg/kg) of BFRs in colour separated extruded**
638 **MEP**

BFR		Colour sorted MEP		
		Mixed (n=20)	White (n=3)	Clear (n=3)
Σtri-octa BDE[1]	DF (%)	100	100	100
	Arithmetic mean	890	51	1.5
	Range	150–5800	46–61	1.4–1.7
BDE-209	DF (%)	100	100	100
	Arithmetic mean	6100	5000	60
	Range	2100–9300	4100–6100	32–80
DBDPE	DF (%)	100	100	100
	Arithmetic mean	660	59	23
	Range	180–1700	40–90	22–26
TBBP-A	DF (%)	100	100	100
	Arithmetic mean	150,000	8300	140
	Range	7800–27000	7600–9100	110–190
BTBPE	DF (%)	0	100	0
	Arithmetic mean	<0.01	370	<0.01
	Range	<0.01	320–460	<0.01
PBBz	DF (%)	0	100	0
	Arithmetic mean	<0.01	0.69	<0.01
	Range	<0.01	0.45–0.97	<0.01
HBBz	DF (%)	0	67	0
	Arithmetic mean	<0.01	0.71	<0.01
	Range	<0.01	<0.01–1.1	<0.01

639 [1] Σtri-octa-BDE is defined here as the sum of six primary congeners: BDE-47, BDE-99, BDE-100, BDE-153,
640 BDE-154, and BDE-183. DF = detection frequency; HBCDD was below limits of detection in all samples
641 (<0.05 mg/kg). When calculating arithmetic means, samples beneath limits of detection were assumed to be
642 equal to zero.
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645

Table 2: Statistical summary of concentrations (mg/kg) of BFRs in 97 SMW individual polymer chips

Polymer type	Σtri-octa BDE[1]			BDE-209			DBDPE			TBBP-A			BTBPE			PBBz			HBBz		
	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range	DF (*n*)	Arithmetic mean	Range
Medium-MEP ABS (n=15)	9	4.0	<0.01-18	1	43	<0.05-640	2	60	<0.2-480	15	13000	190-28000	6	1800	<0.01-27000	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PC-ABS (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PA (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PC (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	1	210	<0.5–1100	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PMMA (n=5)	0	<0.01	<0.01	1	19	<0.05–96	0	<0.2	<0.2	5	34	24–55	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PP (n=5)	0	<0.01	<0.01	1	40	<0.05–200	1	49	<0.2–250	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Medium-MEP PS/HIPS (n=13)	6	1.1	<0.01-13	1	1900	<0.05-25000	2	25	<0.2–330	13	4600	5.1–47000	1	3.9	<0.01-51	4	1.6	<0.01-13	5	5.3	<0.01-20
Heavy-MEP ABS (n=9)	2	0.56	<0.01-2.5	3	87	<0.05-330	0	<0.2	<0.2	7	7600	<0.5–25000	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP PC-ABS (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	1	71	<0.2–350	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP PA (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP PET (*n*=10)	0	<0.01	<0.01	0	<0.05	<0.05	1	52	<0.2–520	10	360	13–1300	1	5.2	<0.01-52	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP POM (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP PP (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	1	11	<0.5–55	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01
Heavy-MEP PVC (n=5)	0	<0.01	<0.01	0	<0.05	<0.05	0	<0.2	<0.2	0	<0.5	<0.5	0	<0.01	<0.01	0	<0.01	<0.01	0	<0.01	<0.01

[1] Σtri-octa-BDE is defined here as the sum of six congeners: BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183. DF = detection frequency; HBCDD was below limits of detection in all samples (<0.05 mg/kg). When calculating arithmetic means, samples beneath limits of detection were assumed to be equal to zero.

Table 3: Paired analysis concentrations (mg/kg) of XRF-Br and MS-Br in 27 individual SMW polymer type chips

Polymer type / Sample ID	XRF-Br	MS-Br for individual BFR compounds									ΣPOP-BFRs[1] MS-Br	ΣBFRs MS-Br
		BDE-99	BDE-153	BDE-183	BDE-209	DBDPE	TBBP-A	BTBPE	PBBz	HBBz		
M-ABS-1	103000±2070	<0.00708	3.79	1.11	<0.0417	346	14,000	0.80	<0.00845	<0.00869	4.91	14400
M-ABS-2	107000±2590	<0.00708	<0.00745	3.10	<0.0417	<0.165	466	<0.0069	<0.00845	<0.00869	3.10	469
M-ABS-3	78800±1450	<0.00708	2.52	2.85	<0.0417	<0.165	13,600	3.9	<0.00845	<0.00869	5.37	13600
M-ABS-4	38100±487	<0.00708	0.069	2.97	<0.0417	<0.165	434	<0.0069	<0.00845	<0.00869	3.02	437
M-ABS-5	111000±2530	<0.00708	3.55	4.28	<0.0417	<0.165	16,200	0.63	<0.00845	<0.00869	7.83	16200
M-ABS-6	106000±2210	<0.00708	1.70	9.78	531	<0.165	1120	8.6	<0.00845	<0.00869	542	1670
M-ABS-7	104000±2130	<0.00708	2.84	<0.00774	<0.0417	397	14,100	28	<0.00845	<0.00869	2.84	14500
M-ABS-8	68800±1120	<0.00708	2.00	3.62	<0.0417	<0.165	11,400	<0.0069	<0.00845	<0.00869	5.62	11400
M-ABS-9	113000±2560	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	365	18800	<0.00845	<0.00869	0.00	19200
M-ABS-10	94200±2130	<0.00708	1.06	<0.00774	<0.0417	<0.165	15,000	<0.0069	<0.00845	<0.00869	1.06	15000
M-PS/HIPS-1	129000±2870	2.81	<0.00745	<0.00774	<0.0417	<0.165	327	<0.0069	1.69	5.45	2.81	337
M-PS/HIPS-2	122000±2650	<0.00708	0.125	<0.00774	<0.0417	<0.165	197	<0.0069	<0.00845	<0.00869	0.125	197
M-PS/HIPS-3	106000±2120	<0.00708	<0.00745	<0.00774	160	<0.165	1020	<0.0069	2.70	14.4	160	1200
M-PS/HIPS-4	125,000±2880	<0.00708	1.18	<0.00774	<0.0417	<0.165	14,100	<0.0069	<0.00845	<0.00869	1.18	14100
M-PS/HIPS-5	88200±1700	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	481	<0.0069	<0.00845	<0.00869	0.00	481
M-PS/HIPS-6	118000±2600	<0.00708	0.281	<0.00774	<0.0417	<0.165	7640	<0.0069	10.6	17.3	0.281	7670
M-PS/HIPS-7	88300±1710	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	774	<0.0069	<0.00845	9.56	0.00	784
M-PS/HIPS-8	10700±130	<0.00708	0.827	5.75	20,900	272	712	36	2.78	13.6	20900	21900
H-ABS-1	79200±1490	<0.00708	<0.00745	<0.00774	176	<0.165	426	<0.0069	<0.00845	<0.00869	176	602
H-ABS-2	139000±3280	<0.00708	1.87	<0.00774	200	<0.165	14,600	<0.0069	<0.00845	<0.00869	202	14800
H-ABS-3	126000±2950	<0.00708	1.90	<0.00774	<0.0417	<0.165	14,900	<0.0069	<0.00845	<0.00869	1.90	14900
H-ABS-4	170000±4570	<0.00708	<0.00745	<0.00774	275	<0.165	534	<0.0069	<0.00845	<0.00869	275	809
H-PET-1	69900±1290	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	304	<0.0069	<0.00845	<0.00869	0.00	304
H-PET-2	79900±1620	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	778	<0.0069	<0.00845	<0.00869	0.00	777
H-PET-3	65000±1310	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	316	<0.0069	<0.00845	<0.00869	0.00	316
H-PET-4	58000±1110	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	163	<0.0069	<0.00845	<0.00869	0.00	163
H-PET-5	45700±780	<0.00708	<0.00745	<0.00774	<0.0417	<0.165	247	36	<0.00845	<0.00869	0.00	284

[1] ΣPOP BFRs is defined here as the sum of seven primary PBDE congeners (BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, and BDE-209) and HBCDD. BDE-47 and HBCDD were below limits of detection in all samples (<0.01 mg/kg and <0.05 mg/kg, respectively). M= Medium MEP; H=Heavy MEP

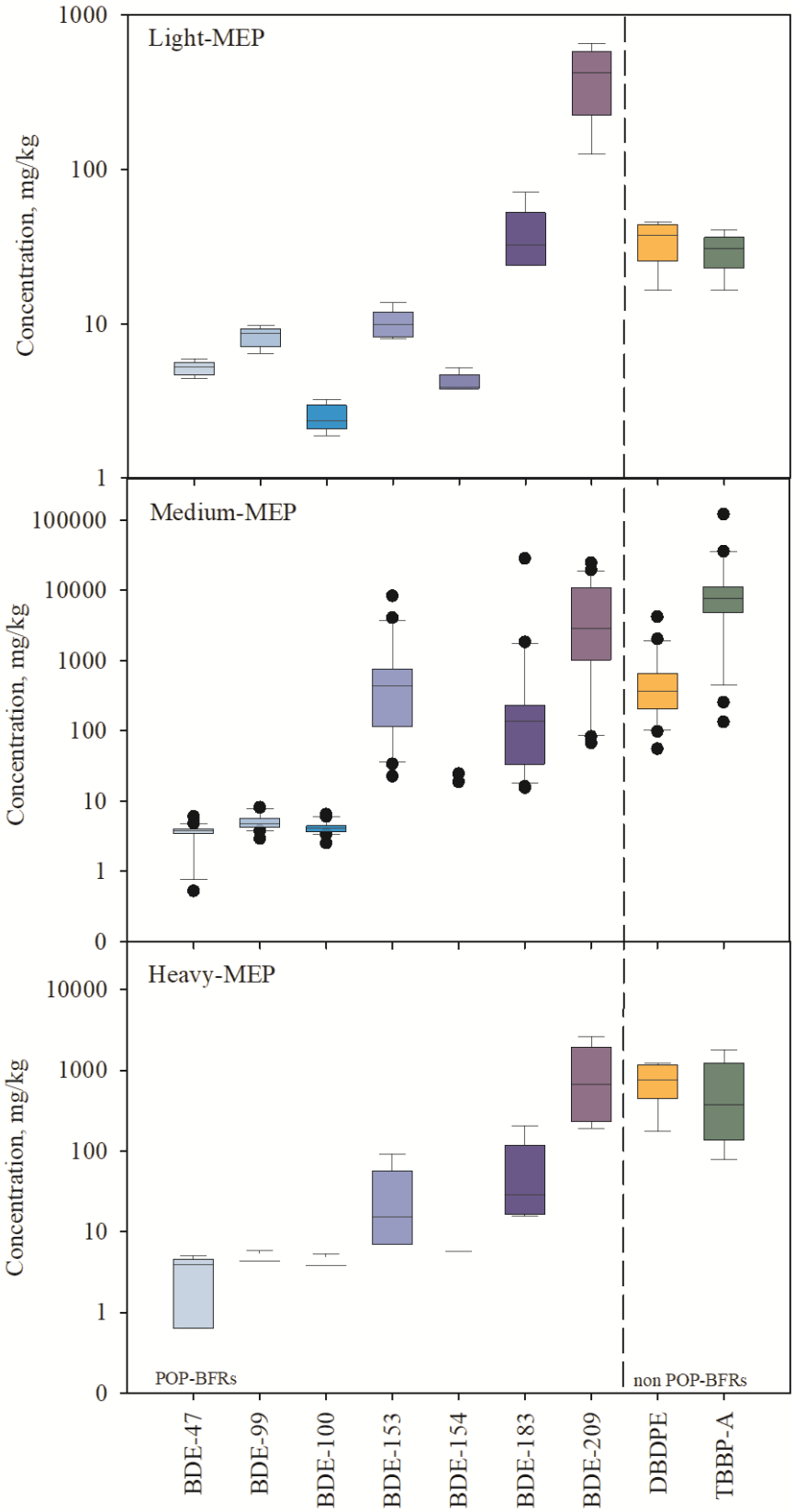

Figure 1: BFR concentrations (mg/kg) for each MEP grade ("Light-MEP", *n*=5; "Medium-MEP", *n*=20; "Heavy-MEP", *n*=5). The boxes indicate the 25th to 75th percentiles, the whiskers indicate the 10th and 90th percentiles, the black line is the median, and the black dots represent outliers. The y-axis is plotted using log scale.

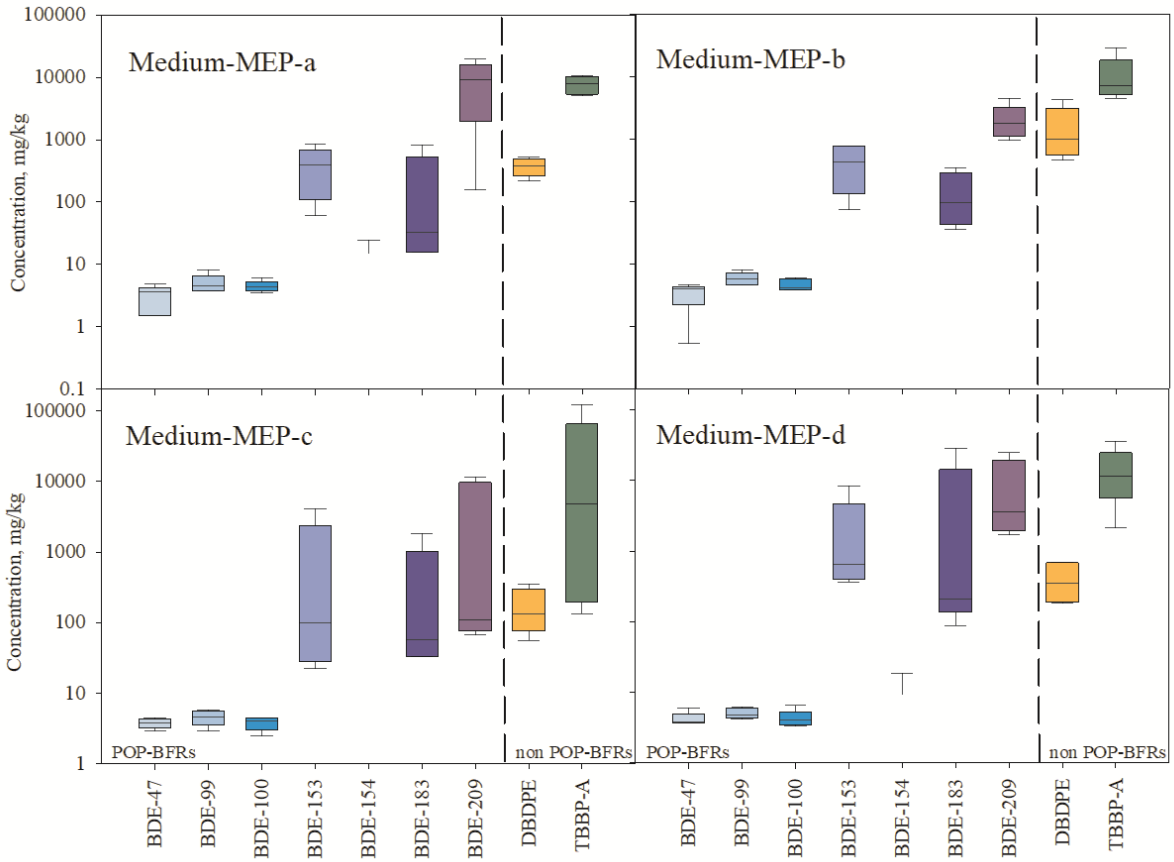

Figure 2: BFR concentrations (mg/kg) for each "Medium MEP" subgrouping a-d (*n*=5 each). The boxes indicate the 25th to 75th percentiles, the whiskers indicate the 10th and 90th percentiles, the black line is the median, and the black dots represent outliers. The y-axis is plotted using log scale.