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CONCENTRATIONS OF BROMINATED FLAME RETARDANTS IN PLASTICS OF ELECTRICAL AND ELECTRONIC EQUIPMENT, VEHICLES, CONSTRUCTION, TEXTILES AND NON-FOOD PACKAGING: A REVIEW OF OCCURRENCE AND MANAGEMENT

Pierre Hennebert *

INERIS - RISK/COSM, ARDEVIE La Villa Av Louis Philibert BP 33, Aix-en-Provence Cedex 4 13545, France

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ABSTRACT

A synthesis of 4000 published data from 37 references of brominated flame retardants (BFRs) concentrations, in plastics of electrical and electronic equipment, vehicles, construction products, textiles and non-food packaging is presented. For POP decabromodiphenylether, a median concentration of 50 mg/kg in plastics of electrical and electronic equipment (n=276), is reported, as well as 31 mg/kg in plastics of vehicles (n=80), 0 mg/kg in plastics of construction (n=81), and 0 mg/kg in plastics of textiles equipment and upholstery (n=75). The mean concentrations are 5200, 3100, 8700 and 6500 mg/kg, respectively. In non-food packaging (expanded or extruded polystyrene), hexabromocyclododecane is present in some samples. All these plastics always have at least some samples with one BFR with a mean concentration above the EU regulatory concentration limit for substances, products or hazardous waste. The distribution of all reported concentrations of PBDEs is skewed, with for instance, in plastics of vehicles, 84% of the data lower than 1000 mg decaBDE /kg, and some large values up to 150 000 mg/kg. The sorting and the up-to-date management technologies are for these categories of plastics (estimated to be 40% of the plastic use in the EU, the brominated fraction of them being a few percent) necessary to weed out banned substances in the circular economy.

1. INTRODUCTION

The EU is developing a strategy on plastics in a circular economy (EC 2017). Some plastic flows have a low rate of recycling and reuse of plastics due to, among other reasons, quality issues (presence of additives or mixing of different types of polymers). On the other hand, scientific progress in knowledge of substances results in the restriction or banning of elements or substances in products, and restricted management options when these substances are present in waste. This happens frequently as a legacy of previous practices. Research is active in this field. Human biomonitoring of global populations has identified an exposure to a range of plastic additives, detectable in some cases in most people (Galloway et al. 2019). A database of chemicals associated with (food) plastic packaging found 906 chemicals identified as likely, 3377 chemicals as possibly associated; the 148 most hazardous of them being classified in the CLP the United Nations' Globally Harmonized System (GHS). Some are classified as persistent, bioaccumulative and toxic (PBT) or very persistent and very bioaccumulative (vPvB), and endocrine disrupting chemicals (EDC) (Groh et al. 2019). A general overview of the plastic waste management options and the additives presents in plastics can be found in Hahladakis et al. (2018). The conclusion is that recycled plastics must be sorted in order to avoid the spreading of some unwanted additives in the new products.

The use of plastics in Europe is divided as follows: 39.7% for packaging (food- and non-food-), 19.8% for building and construction, 16.7% for medical equipment, plastic furniture and furniture equipment, technical parts used for mechanical engineering or machine-building, 10.1% for automotive, 6.2% for electrical and electronic equipment, 4.1% for household, leisure and sport, the rest being used in agriculture (3.4%) (PlasticsEurope 2018). For instance, hard plastics and foams now account for about 150 kg in modern cars, 16% of which could be brominated (this study). They are protected against fire with, among others, brominated flame retardants (BFR), some of them being now classified as hazardous or persistent organic pollut-

* Corresponding author:
Pierre Hennebert
email: pierre.hennebert@ineris.fr

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ants (POP). An important review on POP substances including BFRs in plastics is (EC 2011).

The compilation of PBDE production data prepared for the POPs Reviewing Committee of the Stockholm Convention estimated the total production of all PBDE from 1970 to 2005 as between 1.3 million and 1.5 million tons (UNEP 2017a). DecaBDE production is estimated at between 1.1 and 1.25 million tons. The three largest producers are Israel Chemical Limited - Industrial Products (ICL), Albemarle Corporation and Chemtura - Great lakes Solutions. They offer products or additives for construction (particularly insulation foam of polystyrene and polyurethane), electrical and electronic equipment (EEE), transport, electrical wires, coal flue gas cleaning, agriculture and oil refining, among others (Albemarle 2018, Chemtura 2018,, ICL 2012). A huge variety of BFR is produced. ICL has a specific product guide "Fire protection for automotive and transportation". The BFR are used in different polymers in the 1-15% range (Arias 2011, Alaee et al. 2003).

To assess the concentration of POP substances in the end-of-life plastics, published or reported data have been gathered and analyzed. The objective is to establish whether pre- or post-shredder sorting of plastics of different electrical and electronic equipment, end-of-life vehicle (ELV), textiles and construction waste is necessary to comply with the EU regulations. Particular attention is paid to the recently banned decaBDE and plastics of ELV.

The EU Waste Framework Directive (EU 2018) has set up a separate collection for plastics, and, by January 1, 2025, for textiles. It aims to promote selective demolition in order to enable the establishment of sorting systems for waste construction and demolition, and by extension, for plastic. Waste containing POP substances above concentration limits cannot be landfilled in the EU 2016. Energy recovery as solid recovered fuel must be done in installations fulfilling strict flue gas emission requirements. This paper offers to give some information on the contaminant concentration in plastics and on which type of sorting could be applied for these.

1.1 Abbreviations

ABS	Acrylonitrile Butadiene Styrene
ASR	Automotive shredder residue
BDE	Bromodiphenylether
BFR	Brominated flame retardant
CLP	Classification and Labelling of Preparations and Substances
c-pentaBDE	Commercial pentabromodiphenylether
DBDPE	Decabromodiphenylethane
DDC-CO	Dechlorane plus
decaBDE	Decabromodiphenylether
EDC	Endocrine disrupting chemicals
EEE	Electrical and electronic equipment
ELV	End-of-life vehicle
EPS	Expanded polystyrene
EU	European Union
GHS	United Nations' Globally Harmonized System
HBCDD	Hexabromocyclododecane
heptaBDE	Heptabromodiphenylether
hexaBDE	Hexabromodiphenylether
Hnnn	Hazard Statement Code of substances, H followed by 3-digits number
HP 1 to HP 15	Hazard Property of waste, 1 to 15
ICL	Israel Chemical Limited
PBB	Polybromobiphenyls
PBDE	Polybromodiphenylether
PBT	Persistent, bioaccumulative and toxic
PCB	Polychlorobiphenyls
pentaBDE	Pentabromodiphenylether
PFHxS	Perfluorohexanoic acid
PFOA	Pentadecafluorooctanoic acid
POP	Persistent Organic Polutant
PS	Polystyrene
PST	Post-shredder treatment
PUR	Polyurethane
REACh	Registration, Evaluation and Authorization of Chemicals
RoHS	Restriction of Hazardous Substances
SCCPs	Short chain chlorinated paraffins
TBBPA	Tetrabromobisphenol A
tetraBDE	Tetrabromodiphenylether
UNEP	United Nations Environmental Program
USGS	United States Geological Service
vPvB	Very persistent and very bioaccumulative
WEEE	Waste of electrical and electronic equipment
XPS	Extruded polystyrene

2. REGULATED SUBSTANCES IN PLASTIC MATERIAL (PRODUCTS AND WASTE)

2.1 Limitation of concentrations in products

In electrical and electronic equipment, the following elements and substances are regulated (Directive Restriction of Hazardous Substances (RoHS), EU 2011): cadmium: 100 mg / kg; chromium (VI), mercury, lead: 1 000 mg / kg; polybrominated diphenyl ethers (PBDEs): 1 000 mg / kg.

In products in general, persistent organic pollutants (POPs) are limited (EU 2016). These substances are defined in the Stockholm Convention as not biodegradable, bioaccumulative and dispersed in the natural environment. Some are prohibited, and actions of emissions reduction must be put in place and monitored (Annex III of EU 2016). The limits in the products are as follows: polychlorinated biphenyls (PCBs): banned from use (Annex III of EU 2016); hexabromocyclododecane (HBCDD): 100 mg / kg in products subject to review by the Commission by 22/03/2019; hexachlorobutadiene (HCBD): 100 mg / kg (EU 2017a) (not sought here); tetra- or penta- or hexa- or hepta-bromodiphenylethers (PBDE): 1 000 mg / kg when the products are recycled (EU 2016); decabromodiphenylether (decaBDE): 1 000 mg / kg (EU 2017a) after 02/03/2019, with a proposition of the European Parliament as unintentional contaminant of the sum of tetra-, penta-, hexa-, hepta- and decaBDE of 500 mg/kg, and decaBDE of 10 mg/kg (EP 2019; decided in 2019 to 1000 mg/kg); short chain chlorinated paraffins (SCCPs): 10 000 mg / kg (not sought here).

In addition, there are three substances and groups of substances that are currently POP "candidates" (UNEP 2017b): dicofol, pentadecafluorooctanoic acid (PFOA –

listed in the Stockholm Convention in 2019), its salts and related compounds, and perfluorohexanoic acid (PFHxS), its salts and related compounds. Dicofol is an insecticide (not used in plastics), while PFOA and PFHxS are water-repellent products used, among other things, in vehicle textiles. They were not sought in this study.

Other substances also are a matter of concern. Decabromodiphenylethane (DBDPE) has been reported in WEEE plastics (Wäger et al. 2012, Wäger et al. 2010). Dechlorane plus (abbreviation DDC-CO, CAS No. 13560-89-9), a flame retardant with 12 chlorine atoms, was added to the REACh list of substances that are extremely worrisome (RPA 2014). Dechlorane plus has two isomers (syn- and anti-). It has been found in plastic of vehicles (coarse and fine fractions), as well as in WEEE (Morin et al. 2017). Some data on these substances are reported in this paper.

2.2 Hazardous waste classification (by hazard properties and POP content)

For the classification as hazardous waste, the 15 EU hazard properties, HP 1 to HP 15, are defined by the presence or the concentration of substances with specific hazard statement codes (EU 2014a, EU 2017b, EC 2003a). The hazard properties of the relevant substances in plastic waste (with their hazard statement codes from the CLP) for hazard classification are: (i) HP 7 "Carcinogenic", if Sb_2O_3 (hazard statement code H351 1B)> 10 000 mg / kg, equivalent to Sb> 8 354 mg / kg, or if hexabromobiphenyl (H350 1B)> 1 000 mg / kg; (ii) HP 10 "Reprotoxic", if hexa- or hepta- or octaBDE (H360 1B)> 3 000 mg / kg, or if HBCDD (H361)> 30 000 mg / kg; (iii) HP 14 "Ecotoxic", if the sum of (tetraBDE, pentaBDE, HBCDD and TBBPA) (H410)> 2 500 mg / kg from July 2018 (EU 2014b, EU 2017b) (> 25 000 mg / kg before July 2018).

Regarding the POP substances, it should be noted that not all of them can classify a waste as hazardous.

A first group classifies waste as hazardous if the following substances (EU 2016, EU 2014b, EU 2017b) are present at a concentration exceeding a limit. About plastic waste, these substances are in practice (in order of increasing concentration): polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD / PCDF)> 15 µg / kg, which can be created unintentionally from RFBs by friction and localized heating of BFRs during waste treatment operations (not sought here); polychlorinated biphenyls (PCBs)> 50 mg / kg; hexabromobiphenyl (HBB)> 50 mg / kg.

A second group is the POP substances that do not make the waste hazardous because they are classified. However, those wastes must be managed specifically (section 2.4): the PBDEs and the HBCDD. They can trigger the classification as hazardous by their hazard statement codes and their concentrations for the hazard properties HP 4, 5, 10 and 14, but the classifying concentrations are higher than the ones for specific waste management by their content (s) in POPs (see below).

2.3 Waste management (by their hazard properties HP)

Waste can be landfilled if it meets maximum leachable content and total substance levels (EC 2003a, EC 2003b). In practice, non-hazardous and non-POP plastics can be landfilled in landfills for non-hazardous waste. For hazardous plastic waste, the loss on ignition (<10%) or the total organic carbon content (TOC <6%) are exceeded, and these wastes are not allowed to be landfilled in landfills for hazardous waste but must be incinerated. The ashes must meet the criteria for the disposal of hazardous waste, which is possibly the case after a stabilization with cement.

2.4 Waste management (by their content (s) in POPs)

The POP substances in waste exceeding the concentration limits of POP regulation for waste (being hazardous or not) must be irreversibly transformed so that the substances no longer have the characteristics of a persistent organic pollutant (EU 2016 Annex IV): polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD / PCDF)> 15 µg / kg; polychlorinated biphenyls (PCBs)> 50 mg / kg; hexabromobiphenyl (HBB)> 50 mg / kg; HBCDD< 1 000 mg / kg; sum of tetra-, penta-, hexa- and heptaBDE> 1 000 mg / kg; and under discussion: sum of tetra-, penta-, hexa-, hepta- and decaBDE> 1 000 mg / kg, and a possible lowering two years later to 500 mg / kg (EP 2019).

These wastes cannot be recycled and must be handled only by the following operations: D9 Physicochemical treatment; D10 Incineration on land and; R1 Main use as fuel or other means of producing energy, excluding wastes containing PCBs.

2.5 Sorting to separate parts with controlled substance(s) above concentration limits

For electrical and electronic equipment, polychlorinated biphenyls of capacitors and brominated flame retardants of plastics (among other substances) must be separated (EU 2016). A technical specification (CENELEC CLC/TS 50625-3-1) recommends the sorting of WEEE plastic with a concentration limit of 2000 mg / kg total bromine. In practice, the sorting is carried out either by density (by flotation or by in-line X-ray transmission), or by measurement of bromine (with a portable or in line X-ray fluorescence device).

For products in general, the POPs Regulation (EU 2016) specifies: 'During this elimination or recovery, any substance listed in Annex IV may be isolated from the waste, provided that it is subsequently eliminated…".

3. REPORTED CONCENTRATION IN PRODUCTS AND WASTE AND DISCUSSION

3.1 Sampling, analysis, reported data

The reported concentration of BFRs in plastic particles/ parts has a skewed distribution (see section 3.10), with many particles/ parts with nil or low concentration, and a small number of particles/ parts with scattered high concentrations. The principle of sampling is that a smaller (sub-)sample of a flow or a heap will have the same composition as a larger flow or heap if “enough” particles/ parts are taken. The sampling of waste is described in EN 14899 with five technical documents FD CEN/TR 15310-1 to -5. Considering the skewed distribution of the concentration of bromine per plastic scrap and the analytical variability of BFR analysis, it can be calculated EN 14899, EN 15002) that a representative sample should contain 100 000 particles (Hennebert 2019). For plastics of WEEE, a technical specification (CENELEC CLC/TS 50625-3-1) recommends the sampling per day of production of the shredder of 10 times 3 litres if the size of the largest particle is < 20 mm, 10 times 5 litres for a particle size of 20-50 mm, and 10 times 10 litres for particle size > 50 mm. The composite sample is coned, mixed and quartered two times (volume divided by 4) and 7.5, 12 or 25 litres are sent to the laboratory. The number of particles in these laboratory samples should be checked (Hennebert 2019). The treatment of those samples is described in (EN 15002). The laboratory will cryo-grind it in two or three steps ([10-5] mm and [1 - <0.1] mm) with an intermediate mixing and quartering steps. An aliquot of 0.1 g will be extracted thanks to a solvent, and some [0.1-0.01] ml of the purified extract will be injected in the analysis apparatus (EN 62321-6).

The extraction and analysis of BFR in plastics should be performed by certified industrial laboratories. A publication and a report on ELV plastics vehicles have performed an incomplete extraction of BFR (Table 1). In one case, the samples were crushed at 4 mm and extracted with an accelerated solvent extractor with toluene at 130°C three times (Morin et al. 2017). In the second case, the sample was crushed between 1 and 5 mm respectively and extracted by cold sonication with hexane for 15 min (SMTT 2012). The extraction yields were not controlled, and the laboratories were not accredited for theses analyses. The measured concentrations in automotive shredder residue are low (Table 1) and those 21 data have not been used in this study. Another case has been reported in car fluff (Norwegian Climate and Pollution Agency 2012).

Published data are heterogeneous. Some are single concentration data, and others are a range of concentration data, with the report of the range values number (n), the minimum (min), the mean (mean), and the maximum (max) of the range. A detailed analysis of these ranges of concentrations cannot be presented here, due to page limitations. The mean concentrations presented here are the mean of all the single concentrations and the min, mean and max concentrations of the ranges. Data reported as "< x mg/kg" (lower than the limit of detection or quantification of the laboratory) have been accounted as "x" mg/kg. Commercial bromodiphenylethers mixtures (c-BDE) were typically produced at three different degrees of bromination, in particular c-pentaBDE, and are reported as such, aside with pentaBDE. Some authors report the sum of BDE, that were classified POP at the time of their publication (tetra-, penta-, hexa- and hepta-BDE), under the name POP-BDE. PBDE is the sum of all the BDEs data for the same sample. Hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA) concentrations are significant and are presented.

TABLE 1: Cases of reported BFR concentrations in plastics of vehicles with presumed incomplete extraction during the laboratory analyses.

Mean concentration (mg/kg)	Morin et al. 2017	SMMT 2016
DecaBDE		3.10
PentaBDEs		0.11
HeptaBDEs		0.06
TetraBDEs		0.04
HexaBDEs		0.01
PBDEs	47.24	
HBCDD (Hexabromocyclododecane)		0.565
PBBs (Polybromobiphenyls)		0.05
Sum of PBBs, PBTs (Polybromoterphenyls), dechlorane plus anti and syn, hexabromobenzene, pentabromotoluene and pentabromoethylbenzene	0.36	

Polybrominated biphenyls (PBBs) and polychlorinated biphenyls (PCBs), banned for a long time, have no data for these plastics, and it is known that their concentrations are close to zero in WEEE and automotive plastics (Hennebert and Filella 2017, Hennebert 2018 a, b). Three authors have studied other BFRs. These data are numerous (1 440) and briefly presented in section 3.6. A total of 4 016 RFBs data and 264 Br and Sb data were used (total 4 280 data) (Table 2). The total number of data sorted by category and by bibliographic reference, for all reported substances and all elements, is presented in Table 3. The reference of the Norwegian Environment Agency - BIPRO (2016) includes of 17 citations other than the ones cited in the References, with less abundant data, and source not fully defined). A partial previous version of this work has been presented as an oral communication (Hennebert 2018b).

In this study, since the distribution of concentrations is never gaussian (normal), the minimum, the median, the mean and the maximum concentrations are presented, as well as the percentage of data exceeding a given (regulatory) concentration.

3.2 BFRs in electrical and electronic equipment / WEEE

A total of 962 data (including data of 40 ranges) have been gathered (Table 2). The equipment categories are small and large household (cool/heat, and non cool/heat) equipment, TV, screens, IT equipment, wires, white plastics, and so on, up to a total of 97 different categories. The summarized concentrations are presented in Table 4. No distributions are gaussian (normal): the median concentrations are a smaller fraction of the mean concentrations, and the maximum concentrations are larger multiples of the mean concentrations than in normal distributions (Hennebert 2019). These skewed distributions for the high concentrations are presented with more details in section 3.10. Concentrations > 1 000 mg/kg are highlighted (EC 2003a, EU 2011, EU 2012, EU 2017a).

Large household appliances and cooling and freezing apparatus (two references: Wäger et al. 2012 and Vojta et al. 2017) have a significantly lower concentration of decaBDE than the other equipment (Mann-Whitney test, $p < 0.0001$), but one reported concentration is equal to the concentration limit of 1 000 mg/kg (Table 5).

TABLE 2: Total number of data sorted by category from literature, by BFR and element (Br and Sb), and number of data presented in this study ("range data" is reported as a range of concentration, most of the time with the number of the range values, the minimum, the mean and the maximum of the range).

Parameter	Individual data and range data						Number of ranges					
Category	EEE/WEEE	Vehicles/ ELVV	Construction	Textiles	Packaging	Total	EEE/WEEE	Vehicles/ ELVV	Construction	Textiles	Packaging	Total
BFRs												
PBDE	781	215	716	437		2149	35	26	4	12		77
of which DecaBDE	276	80	81	75		512						
HBCDD	112	21	84	59	57	333	5	2	2	7		16
TBBPA	69	11	6		8	94						
Sub-total	962	247	806	496	65	2576	40	28	6	19		93
Non-regulated BFRS	257	22	771	374	16	1440						
Total BFRs	1219	269	1577	870	81	4016						
Elements												
Br	92	36	9	70	6	213	38	12	3	23	2	78
Sb	32	11	4	4		51						
Total elements	124	47	13	74	6	264	38	12	3	23	2	78
Grand total	1343	316	1590	944	87	4280						

3.3 BFRs in vehicles and end-of-life vehicles

A total of 247 data (including data of 28 ranges) have been gathered (Table 2). Reported concentrations of brominated flame retardants in cars, automotive shredder residue (ASR), and sorted post shredder treatment fractions (PST fractions) are presented in Table S1 (supplementary information). There are 117 data for Car, 46 data for ELV, 51 for ASR and 33 for PST (total 247 data). The results are indicative since these figures are unweighted means of single data, and min, mean and max of range data. The light fluff fractions are documented as "light ASR" or "mixed light plastic" of the ASR category, or the lighter fractions of the PST category. The PST fractions are sorted by flotation, the most brominated fraction being the densest. PST data treatment fractions are scarce. As a result, data from mixed waste (WEEE and ELV) are also presented for this category. A summary is presented in Table 4.

The discussion will focus on the concentration of decaBDE because this substance is the only one that allows direct comparison between categories (Table 6). Significant mean concentrations are observed in some plastic car parts (rounded mean 3 500 mg/kg, above the concentration limit of 1 000 mg/kg), including printed circuit boards, with the highest concentrations in seats, foam and upholstery (27 000 mg/kg). These high values raise the mean concentration, while the median concentration is only 50 mg/kg. The sampling of the parts can be biased: authors may have looked for (previously known) parts with high concentration. In some studies, it is a deliberate choice: a multitude of parts are first measured in the field for total bromine with a hand-held X-ray fluorimeter, and only the most concentrated parts are sent to the laboratory for BFRs analyses. But low concentrations are also reported. This peculiar distribution makes it difficult to understand easily this waste flow. Concentrations in ELV are roughly similar to concentrations reported in car parts.

Much lower concentrations are observed in ASR (rounded mean 400 mg/kg). The sampling of the ASR is probably not biased: sub-samples are taken at periodic interval from the scrap flow, without previous knowledge of concentration. The median of 44 mg/kg is comparable to the median of car parts. It is significantly lower than the mean: the distribution is here also skewed by some high values. Lower concentrations than in car parts (see details in Table S1) are probably because plastics and foam are only 20% of the mass of ASR.

The concentrations in sorted plastics (so-called post-shredder treatment of ASR plastics, here by density separation) are very low (mean 14 mg/kg). The effectiveness of sorting (here mainly by density difference) is proven. Details can be found in Leslie et al. (2016). A recent study (Swerea IVF 2018) confirms it for ELV sorted plastics (a French sample of PS/ABS contained 140 mg/kg decaBDE, and a UK sample of ABS contained 5 mg/kg decaBDE, 15 mg/kg TBBPA, and 14 mg/kg decabromodiphenylethane - DBDPE).

TABLE 3: Total number of data sorted by category from literature, for all reported substances and all elements.

Publication	E	V	C	T	P	Total
Abdallah et al. 2018			1		50	51
Allen et al. 2008	50			30		80
ARN 2015		1				1
Arp et al. 2015	67	25				92
Ballesteros-Gomez et al. 2013	4					4
Chen et al. 2010	17	5				22
COWI 2013	2	2				4
Drage et al. 2018	60	24	24	60		168
EMPA 2010	21					21
Federal office of the Environment (FOEN) 2017	33					33
Gallen et al. 2014	8	3		1		12
Guzzonato et al. 2016	93					93
IVM, IVAM 2013	43	28				71
Japanese Ministry of the Environment (MOE) 2011		6				6
Jinhui et al. 2017	5	2	2	2		11
Kajiwara et al. 2013	1			35		36
Leslie et al. 2016	45	88		7		140
MEPEX 2012		6				6
Morf et al. 2005	1					1
Niinipuu 2013		8				8
Norwegian Climate and Pollution Agency - Mepex 2012		232				232
Norwegian Environment Agency - BIPRO 2016 (17 citations)	47	37	4	11		99
Petreas et al. 2009	12	3				15
Puype et al. 2015	187					187
Rani et al. 2014			24		32	56
RPA 2014	6	4	4	4		18
Sindiku et al. 2015	4					4
Sinkkonen et al. 2004	3	3				6
Stubbings et al. 2014				2		2
Taurino et al. 2010	20					20
Turner et al. 2017 Antimony	4	4	4	4		16
Turner et al. 2017 Bromine	9		9	9		27
Vojta et al. 2017	552	48	1656	816		3072
Wäger et al. 2011	2					2
Wäger et al. 2012	1032					1032
WRc 2012	136					136
WRc addendum 2012	2	18				20
Grand total	**2466**	**547**	**1728**	**981**	**82**	**5804**

The concentrations are lower than what is reported and observed in the corresponding fractions of WEEE. In France, in unsorted shredded plastics of small household appliances, cathode ray tubes and flat screens, the concentrations were 1 950 mg/kg decaBDE in 2014 and 1 395 mg/kg in 2015 (n = 10 - in triplicates) (Hennebert and Filella 2017). For sorted plastics (fraction < 2 000 mg/kg Br), the mean concentrations were 148 mg/kg decaBDE in 2014 and 522 mg/kg decaBDE in 2015 (n = 4).

For elemental concentrations, the most concentrated element is bromine (mean concentration in car parts and ASR = 8 564 mg/kg, n = 27). The concentration used in WEEE for sorting (2 000 mg/kg, (CENELEC CLC/TS 50625-3-1) is trespassed. The second most concentrated element

TABLE 4: Summary of concentration of some brominated flame retardants in plastics of EEE/WEEE, vehicles/ELV, construction, textiles and non-food packaging from literature data, and percentage of data lower and larger than the concentration limit of 1000 mg/kg (**hazardous waste** [bold], concentration allowed in products exceeded [underlined], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]), in yellow: to be sorted for recycling) (in green: lowest % data per category).

Category	BFR	Concentration data (mg/kg)					% data < 1 000 mg/kg	% data < 500 mg/kg
		n	Min	Median	Mean	Max		
EEE/WEEE	PBDE	781	0	7	*2663*	*154000*	90%	87%
	of which DecaBDE	276	0	50	*5216*	*150000*	84%	80%
	HBCDD	112	0	200	137	1600	99%	99%
	TBBPA	69	20	20	*3155*	*63000*	93%	91%
Vehicles/ELV	PBDE	215	0	6	*1623*	*85000*	92%	88%
	of which DecaBDE	80	0	31	3102	85000	88%	78%
	HBCDD	21	0	10	386	***4400***	90%	90%
	TBBPA	11	20	20	27	87	100%	100%
Construction	PBDE	716	0	0	1713	*300000*	99%	99%
	of which DecaBDE	81	0	0	*8662*	*300000*	95%	94%
	HBCDD	84	0	0	317	***10000***	92%	89%
	TBBPA	6	0	0	0	0	100%	100%
Textiles and upholstery	PBDE	437	0	0	*2080*	*130000*	95%	94%
	of which DecaBDE	75	0	0	*6511*	*120000*	84%	81%
	HBCDD	59	0	0	***3465***	***51000***	86%	86%
	TBBPA	-	-	-	-	-	-	-
Non-food Packaging	PBDE	-	-	-	-	-	-	-
	of which DecaBDE	-	-	-	-	-	-	-
	HBCDD	57	0	11	232	***5897***	91%	88%
	TBBPA	8	0	-	0	1	-	-

TABLE 5: Concentration of decaBDE in large household appliances and other categories of EEE/WEEE from literature data (concentration allowed in products exceeded [underlined]).

DecaBDE (mg/kg)	n	Minimum	Median	Mean	Maximum	Standard deviation
Large household appliances, cooling and freezing appliances	36	0	50	84	1000	162
Other categories	240	0	50	5 985	150 000	22 935

is lead (mean concentration in car parts = 6 499 mg/kg, n = 13) (result not shown, Hennebert 2018a). The concentration of the Restriction of Hazardous Substances (RoHS) (EU 2011) applicable to EEE is trespassed (1 000 mg/kg). According to some authors, it is due to printed circuit boards soldering.

TABLE 6: Concentrations of decaBDE in car parts, automotive shredder residue (ASR) and post shredder treatment fractions – unweighted mean concentrations and median from literature data (concentration allowed in products exceeded [underlined], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]).

Concentration (mg/kg)	DecaBDE		
Category	Mean	Median	n
Car - parts	*3 469*	50	35
ASR	386	44	21
PST – sorted plastics	(101)	(6)	(11)
Of automotive shredder residue	14	3	6
(Of mixed automotive and WEEE shredder residue)	(205)	(29)	5

3.4 BFRs in construction plastics

In total, 806 data (including data from 6 ranges) were collected (Table 2). The various samples are presented in Table S2 by group of BFR present or not (last column) and by decreasing mean concentration. The distinction between the PBDE - decaBDE groups on the one hand and HBCDD on the other hand is very clear, with more foams and polystyrenes for the latter group. However, HBCDD concentrations are too low to be flame retardant: the maximum is 0.54%, while 0.8 to 4% is recommended (Arias 2001, Alaee et al. 2003). This may be the index of products derived from recycling a mixture of brominated and non-brominated plastics . The RFB-free group includes paint, wood panels, paper-based insulation, etc. A summary is presented in Table 4. The samples from continental Europe (69 out of

85 samples) have low to very low concentrations (results not shown).

3.5 BFRs in textile equipment

A total of 496 data (including data from 19 ranges) were collected (Table 2). Plastics also cover a wide range (0-300 000 mg/kg-Table S3) presented by group of BFR present or not (last column) and by decreasing average concentration. A first PBDE - decaBDE group without HBCDD can be identified, mostly with non - flame retardant concentrations (at least 5% according to Arias 2001, Alaee et al. 2003). A second and smaller group contains HBCDD with or without PBDEs. The group without BFR is made up of articles that do not seem different from the first two groups.

A summary of concentrations for all samples is presented in Table 43. The average concentration of decaBDE is of 6 500 mg / kg (n = 75), which is above the limit for the products. This result is consistent with the high level observed in automotive seats and foams (Hennebert 2018a, this paper). The average HBCDD concentration is hazardous and above the limit for the products. The plastics of textile equipment should therefore be sorted. Here also, the samples from continental Europe (36 out of 55) have low to very low concentrations (results not shown).

3.6 BFRs in non-food packaging

In total, 65 data (without data ranges) were collected (Table 2). These plastics are exclusively expanded polystyrene (EPS) and extruded polystyrene (XPS) with HBCDD as RFB. The concentrations are presented in Table 4. Concentrations are low, but 31 samples have a HBCDD concentration> 10 mg / kg, the highest being polystyrene packaging laboratory equipment, appliances and printers (Abdallah et al. 2018, FOEN 2017). The maximum HBCDD content exceeds the concentration limit for hazardous waste, and the average is greater than what is allowed for the products. These plastics should therefore be sorted.

3.7 BFRs in food-contact packaging

This point is not the focus of this study but is however presented since, according to some authors, there is evidence of recycling of brominated plastics (maybe from construction insulation foams) in food-contact articles. According to two authors (Rani et al. 2014, Abdallah et al. 2018), six data out of 66 of disposable plate and cup, menu box, takeaway food container, PS cold box, vegetable packaging, and similar, all in expanded polystyrene or extruded polystyrene, have a concentration of HBCDD > 10 mg/kg (1516, 50, 29, 15, 14 and 10 mg/kg). This indicates the usefulness of sorting EPS and XPS (particularly from construction plastics) for bromine and BFRs.

3.8 Other BFRs

Three authors (Rani et al. 2014, Abdallah et al. 2018, FOEN 2017) measured 12 unregulated RFBs nowadays (Table 7) on a total of 144 samples. Only the maximum concentrations are presented. The concentrations are low or close to zero. For e-waste, the highest concentrations are reported by (FOEN 2017). Decabromodiphenylethane (DBDPE) has been a substitute for decaBDE for two decades in plastics of EEE (Wäger et al. 2012). For construction plastics, a rubber sample and a recycled plastic material sample have a dechlorane concentration greater than 64 mg/kg and 17 mg/kg, respectively (Vojta et al. 2017).

TABLE 7: Maximum concentrations in non-regulated RFBs.

Maximum concentration (mg/kg)			Category					
Abbreviation	**Name**	**CAS No**	**EEE/WEEE**	**Vehicles/ ELV**	**Construction**	**Textiles**	**Non-food packaging**	**n**
BTBPE	1,2-Bis(2,4,6-tribromophenoxy) ethane	37853-59-1	150	0	1	0	0	144
DBDPE (DBDPER)	Decabromodiphenylethane or 1,2-bis(pentabromodiphenyl) ethane	84852-53-9	340		0		0	15
DBE-DBCH (TBECH)	Tetrabromoethylcyclohexane (sum of alpha- and beta-)	3322-93-8	0	0	0	0		128
DBHCTD (HCDBCO)	Hexachlorocyclopentenyl-dibromocyclooctane	51936-55-1	0	0	0	1		128
DDC-CO	Dechlorane Plus (sum of anti- and syn-)	13560-89-9	33	0	64	1		129
HBB	Hexabromobenzene	87-82-1	0	0	1	1		128
PBEB	Pentabromoethylbenzene	85-22-3	0	0	0	0		128
PBT	Pentabromotoluene	87-83-2	0	0	0	0		128
TBCO	1,2,5,6-Tetrabromocyclooctane (sum of alpha- and beta-)	3194-57-8	0	0	0	0		128
TBP-BAE (BATE)	2-Bromoallyl-2,4,6-tribromophenyl ether	3728-89-5	0	0	0	0		128
TBP-DBPE (DPTE)	2,3-Dibromopropyl-2,4,6-tribromophenyl ether	35109-60-5	0	0	0	0		128
TBX (pTBX)	2,3,5,6-Tetrabromo-p-xylene	23488-38-2	0	0	0	0		128
Total			**340**	**0**	**64**	**1**	**0**	**1440**

3.9 Total Br and Sb in plastics

A total of 265 data (including data of 78 ranges) have been gathered (Table 8). All these plastics are brominated to some extent, the highest being the EEE/WEEE plastics, followed by vehicles/ELV and textiles plastics, and then with lower concentrations construction plastics and non-food packaging plastics. The WEEE technical specification (CENELEC CLC/TS 50625-3-1) recommends the sorting of plastics batches or flows with mean concentration of total Br > 2 000 mg/kg (measured on a representative laboratory sample). If applied to other plastics, the first four categories of Table 8 should be sorted.

These plastics also contain antimony. Two plastic samples are classified as hazardous based on their concentration of antimony (in bold): "construction - plumbing" at 13 000 mg Sb / kg and "dressing - padding" at 9 922 mg Sb / kg.

3.10 Synthesis of regulated BFRs concentrations and distribution of concentration

The 2 576 data from the literature summarized in Table 4 indicate that the five categories of plastics studied always have at least one BFR at medium or maximum concentration classified hazardous, not allowing the recycling of the products, or having a total bromine content> 2 000 mg / kg, and therefore not recyclable without sorting. A large majority of the data is lower than 1 000 mg/kg or 500 mg/kg. Some distributions are illustrated in Figure 1. The interval of concentration in the histograms is 1 000 mg/kg. The first interval of concentration [0-1 000] mg/kg has been set to the concentration limit for unintentional (re)use in products. The distributions of data are skewed by large values, as illustrated by the significant differences between the median and the mean (Table 4). The same distribution applies to individual scraps (Hennebert 2019). If the last percentiles of scraps are sorted out, the mean concentration falls drastically, and these plastics can therefore be recycled.

4. MANAGEMENT OPTIONS

For plastic recycling, the so-called "brominated fraction" should be separated, either by sorting for total bromine by X-ray transmission on line, or by flotation in a bath with a different density, as practiced today for WEEE (Hennebert and Filella 2017, Leslie et al. 2016, Swerea IVF 2018).

The POP fractions above the concentration limits should be destroyed in incinerators or irreversibly transformed (EU 2016). The incinerators must fulfil the emissions requirements of (chloro- and bromo-) dioxins and furans in the flue gas, and of leachable antimony (Sb_2O_3 is used as a synergist of BFRs in plastics) in the ashes and in the air pollution control residue. Corrosion of the installation by bromine is intense (Buekens and Zhou 2014). Bromine is present partly as HBr and partly as elemental bromine Br_2 (g). HBr is easily neutralized by solid sodium bicarbonate or captured by wet scrubbing with sodium hydroxide, but not Br_2. If not destroyed, POP substances will continue their cycle (Norwegian Climate and Pollution Agency 2012, Leslie et al. 2016). POP substances are found in landfill leachate (Morin et al. 2017). Traces of BFRs have been found in food-contact articles (Puype et al. 2015), together with rare earth elements from electronic goods. Large reviews point out the ubiquity of these substances (Norwegian Environment Agency 2015, 2016, 2017). These legacy substances can hamper the achievement of the compulsory objective of recycling in the EU. This point has been well identified by the European Commission (EC 2017). Finland has suggested that the EU recycling targets for ELV (85% of recycling including scraps and 10% energy recovery) be calculated from the material left after removal of hazardous substances and components (specified in Annex VII of the WEEE Directive 2012/19/EU) (Häkkinen 2016).

The "brominated plastics" are today mainly incinerated in incinerators for hazardous waste, in cement kiln, or in non-ferrous smelters refineries, with special management for corrosion by bromine, for the emission of elemental bromine and brominated dioxins / furans in the fumes, and for the presence of leachable antimony in the ashes, slags and air pollution control residues (Norwegian Environment Agency 2017). A non-ferrous smelter, Umicore, claims 70% recovery of Sb, a critical raw material, according to the EU and the USGS. If the POP concentration limit is not exceeded, they could theoretically be reused as flame-retarded plastics. But in practice, they are refused by manufacturers who have developed precise formulated compounds. They are mainly used as solid recovered fuel (emissions in the fumes should be secured) or landfilled in landfills for non-hazardous waste. For instance, in France, 40% of the ELV plastics are burned in different installations and 29% are landfilled (Deloitte and ADEME 2017).

Chemical recycling by cracking of plastics or solvolysis is up till now hampered by the incomplete separation of brominated compounds from the useful un-brominated

TABLE 8: Concentration of total Br and Sb in plastics of EEE/WEEE, vehicles/ELV, construction, textiles and non-food packaging from literature data (**hazardous waste** [bold], concentration of technical specification for sorting of WEEE plastics *Br> 2000 mg/kg* exceeded [italics]).

Plastics	Br					Sb				
	n	Min	Median	Mean	Max	n	Min	Median	Mean	Max
EEE/WEEE	92	0	145	*13374*	*171000*	32	0	1000	3381	**58900**
Vehicles/ELV	36	0	314	*8197*	*106800*	11	34	975	1521	6020
Construction	9	0	45	*2122*	*9410*	4	103	984	3768	**13000**
Textiles and upholstery	70	0	99	*7175*	*128300*	4	90	944	2975	**9922**
Packaging	6	0	10	1153	*5600*	-	-	-	-	-

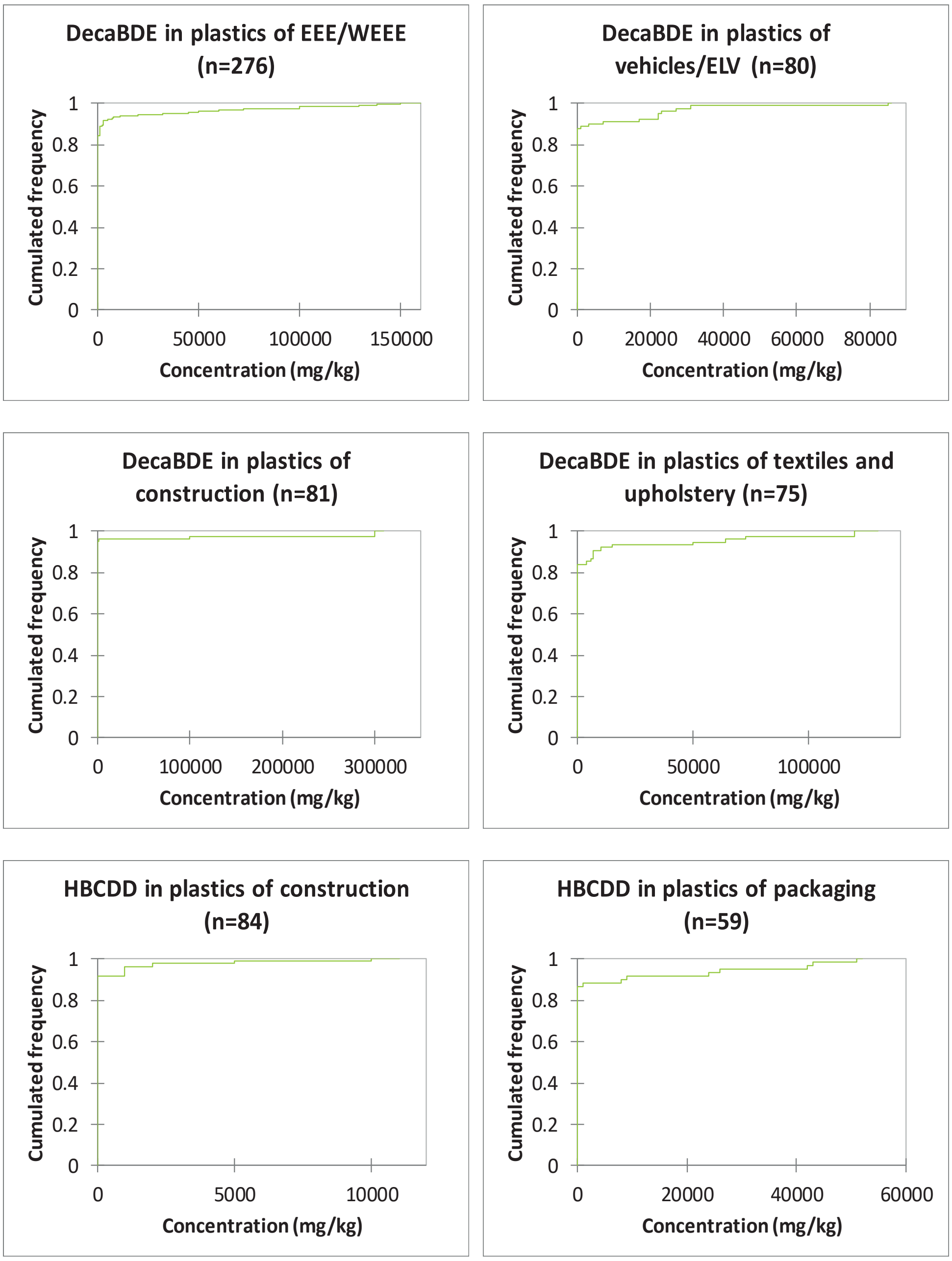

FIGURE 1: Distribution of decaBDE concentrations in plastics of EEE, vehicles, construction and textiles, and of HBCDD in expanded polystyrene foam of non-food packaging from literature data (frequency of data per concentration class of 1000 mg/kg).

feedstock chemicals. The production of syngas or liquid fuel by pyrolysis (heating without oxygen) faces the same obstacle. Exportation is in practice restricted since China decided in January 2018 to stop the importation of unsorted or contaminated plastics. Following others, a pyrolysis technology with recovery of heat, bromine and antimony is claimed at pilot scale in 2018 (Hense et al. 2018). A short synthesis of the advantages and drawbacks of the different management options is presented in Table 9.

5. CONCLUSION

The EU plastics strategy in a circular economy stipulates that plastics containing regulated BFRs must be sorted and managed separately from the non-brominated fraction. Sorting is essential to avoid the uncontrolled dispersion of controlled substances in recycled raw materials.

According to the EU, regulations are in force for each category. These unsorted plastics are hazardous (average or maximum for a BFR), they cannot be reincorporated as such into products, and they exceed 2000 mg Br/kg (operational sorting concentration for WEEE plastics). This study shows that approximately 16%, 12% 8%, 16% and 9% of published concentrations of plastics from EEE/WEEE, vehicles/ELV, building materials, textile equipment and non-food packaging could be greater than 1 000 mg/kg of regulated RFBs. Overall, the results indicate that the sorting

TABLE 9: Management options of brominated and antimoniated plastics.

POP waste? / Technology	Restriction of use (POP, RoHS)?	Hazardous waste?	Recycling as flame-retarded plastics	Chemical destruction	Co-Incineration H waste	Co-Incineration NH waste	Solid Recovered Fuel with flue gas treatment	Landfill H waste	Landfill NH waste	Co-Fuel for cement kiln	Co-Raw material for non-ferrous smelters-refineries (flue gas treatment)	Pyrolysis and post-combustion (flue gas treatment)	Export
Classification													
Yes	-	-		X	X					?	?		
No	Yes	Yes	X			X	X	X?		X	X		
		No	X			X	X		X	X	X		
	No	Yes	X			X	X		X	X	X		
		No	X			X	X		X	X	X		X
Technology													
Recovery	Polymer												
	Heat				X	X	X			X	X		
	Oil											X	
	Gas											X	
	Br										HBr?		
	Sb										70%		
	Metals of non-plastics										X	X	
Drawbacks	C content							X					
	Br2gas				X	X	X			X	X	X	
	Leach. Sb				X	X	X			?	30%?	X	
	Leach. POP							X	X				
	Corrosion				X	X	X			X	X	X	
Technology Maturity level	Routine				X	X	X		X	X	X		X
	R&D											X	
	Laboratory		X	X								X	
	Not practiced		X	X				X				X	

of bromine content for these categories of plastics (except for food packaging) is necessary.

Literature also shows that sorting is effective: data are scarce but sorted ELV plastics have a mean decaBDE concentration of 14 mg/kg. The efficiency of sorting was confirmed in France with WEEE plastics in 2014 and 2015 (Hennebert and Filella 2017). As the plastics of construction and demolition waste must be sorted in the EU and that separate collection of textiles is foreseen at the latest in 2025 (EU 2018), all these plastics (excepted food-packaging) will have to be sorted to weed out regulated substances (PBDE, HBCDD) in a circular economy. The amount to be sorted could reach 40% of the amount of plastics used in the EU, representing 20 million of tons per year (calculated from PlasticsEurope 2018). The amount that could not be recycled is a few percent of that mass. It must be emphasized that the samples of construction plastics and textile and furniture plastics from continental Europe have low to very low concentrations.

To conclude, it should be checked by analytical campaigns whether the different (sorted) plastics fractions have a POP waste status or not, and whether the major actual management practices of energy recovery and landfilling are in line with the EU regulations. Research in technology to recover energy, bromine and antimony is active and promising on a pilot scale.

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REFERENCES

Abdallah, M. A., M. Sharkey, et al. (2018). "Hexabromocyclododecane in polystyrene packaging: A downside of recycling?" Chemosphere 199: 612-616.

Alaee, M., Arias, P., Sjodin, A., Bergman, A. (2003). An overview of commercially used brominated flame retardants, their applications, their use patterns in different countries/regions and possible modes of release. Environ. Int. 29, 683-689.

Albemarle 2018. Albemarle 2018. Fire Safety Solutions - Product Selector Guide. www.albemarle.com . Consulted Jan 2018.

Arias P. 2001. Brominated flame retardants-an overview. The Second International Workshop on Brominated Flame Retardants. Stockholm: AB Firmatryck, 2001. p. 17- 9.

Arp, H.P.H., Morin, N.A.O., Hale, S.E., Okkenhaug, G., Breivik, K., Sparrevik, M., 2017. The mass flow and proposed management of bisphenol A in selected Norwegian waste streams. Waste Manag. 60, 775–785. https://doi.org/10.1016/j.wasman.2017.01.002

Auto Recycling Netherlands (ARN Recycling BV) (2015). Turning waste into raw materials - ELV automotive plastics recycling

Ballesteros-Gómez, A., Brandsma, S.H., De Boer, J., Leonards, P.E.G., 2014. Direct probe atmospheric pressure photoionization/atmospheric pressure chemical ionization high-resolution mass spectrometry for fast screening of flame retardants and plasticizers in products and waste. Anal. Bioanal. Chem. 406, 2503–2512. https://doi.org/10.1007/s00216-014-7636-8

Buekens A, Zhou X (2014). Recycling plastics from automotive shredder residues: a review. J Mater Cycles Waste Manag (2014) 16:398-414.

CEN FD CEN/TR 15310-1 to -5:2007. Characterization of waste - Sampling of waste material. CEN, Brussels, Belgium

CENELEC CLC/TS 50625-3-1:201.5 Requirements for collection, logistics and processing for Waste Electrical and Electronic Equipment (WEEE) - Part 3-1: Specifications for depollution. CENELEC, Brussels, Belgium.

Chemtura Great lakes Solution (2018). Great Lakes Solution 2017. LANXESS Bromine Solutions. Flame retardants product guide. Consulted Jan 2018.

Chen S.J., Ma Y.J., Wang J., Tian M., Luo X.J., Chen D., Mai B.X. (2010). Measurement and human exposure assessment of brominated flame retardants in household products from South China. Journal of Hazardous Materials, 176

COWI (consulting company) (2014). End-of-Life vehicles and environmental pollutants in material flows at shredder plants - An overview. Trondheim, Norway, COWI.

Deloitte and ADEME (2017). Deloitte Développement Durable (Salès K, Lornet L, Benhallam R.) and ADEME (Lecointre E). Octobre 2017. Rapport Annuel de l'Observatoire des Véhicules Hors d'Usage - Données 2015. 118 pages.

Drage, D.S., Sharkey, M., Abdallah, M.A.E., Berresheim, H., Harrad, S., 2018. Brominated flame retardants in Irish waste polymers: Concentrations, legislative compliance, and treatment options. Sci. Total Environ. 625, 1535–1543. https://doi.org/10.1016/j.scitotenv.2018.01.076

EC 2003a. Directive 2003/11/EC of the European Parliament and of the Council of 6 February 2003 amending for the 24th time Council Directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (pentabromodiphenyl ether, octabromodiphenyl ether). OJEU 15.2.2003 L 42/45

EC 2003b. Council Decision of 19 December 2002 establishing criteria and procedures for the acceptance of waste at landfills pursuant to Article 16

EC 2011. European Commission. Service request under the framework contract No ENV.G.4/FRA/2007/0066. Final report "Study on waste related issues of newly listed POPs and candidate POPs" 25 March 2011 (Update 13 April 2011). Consortium ESWI Expert Team to Support Waste Implementation. by Umweltbundesamt, Bipro, Enviroplan. 841 p. http://ec.europa.eu/environment/waste/studies/pdf/POP_Waste_2011.pdf

EC 2017. Strategy on Plastics in a Circular Economy. 26/01/2017. 4 p.

EN 14899:2015. Characterization of Waste - Sampling of waste materials - Framework for the preparation and application of a Sampling Plan. CEN, Brussels, Belgium

EN 15002:2015. Characterization of waste - Preparation of test portions from the laboratory sample. CEN, Brussels, Belgium

EN 62321-6: 2015. Determination of certain substances in electrotechnical products - Part 6: polybrominated biphenyls and polybrominated diphenyl ethers in polymers by gas chromatography-mass spectrometry (GC-MS). CEN, Brussels, Belgium

EP 2019. European Parliament legislative resolution of 18 April 2019 on the proposal for a regulation of the European Parliament and of the Council on persistent organic pollutants (recast) (COM(2018)0144 - C8-0124/2018 - 2018/0070(COD)). http://www.europarl.europa.eu/doceo/document/TA-8-2019-0436_EN.html

EU 2011. Directive 2011/65 of the European Parliament and of the Council of 8 June July 2011 on the restriction of the use of certain hazardous substances (RoHS) in electrical and electronic equipment (recast). Official Journal of the European Union, L174, 1.7.2011, p 88-110.

EU 2012. Directive 2012/19/EU of the European Parliament and of the Council of 4 July 2012 on waste electrical and electronic equipment (WEEE) (recast). Official Journal of the European Union 24.7.2012. L 197/38.

EU 2014a. Commission Decision 2014/955/EU of 18 December 2014 amending Decision 2000/532/EC on the list of waste pursuant to Directive 2008/98/EC of the European Parliament and of the Council. http://eurlex.europa.eu/legalcontent/EN/TXT/PDF/?uri=CELEX:32014D0955&rid=1

EU 2014b. Commission Regulation (EU) No 1357/2014 of 18 December 2014 replacing Annex III to Directive 2008/98/EC of the European Parliament and of the Council on waste and repealing certain Directives. Official Journal of the European Union. 19.12.2014. L 365/89.

EU 2016. Regulation (EC) No 850/2004 of the European parliament and of the Council of 29 April 2004 on persistent organic pollutants(POP) and amending Directive 79/117/EEC. Official Journal of the European Union, L 158, p. 7, 30.4.2004, last amended Commission Regulation (EU) 2016/460 of 30 March 2016, Official Journal of the European Union, L 80, p. 17, 31.3.2016.

EU 2017a. Commission Regulation (EU) 2017/227 of 9 February 2017 amending Annex XVII to Regulation (EC) No 1907/2006 of the European Parliament and of the Council concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) as regards bis(pentabromophenyl)ether. Official Journal of the European Union. L35/6. 10.2.2017

EU 2017b. Council Regulation (EU) 2017/997 of 8 June 2017 amending Annex III to Directive 2008/98/EC of the European Parliament and of the Council as regards the hazardous property HP 14 'Ecotoxic'. Official Journal of the European Union. 14.6.2017. L 150/1.

EU 2018. Directive (EU) 2108/851 of the European Parliament and of the Council of 30 May 2018 amending Directive 2008/98/EC on waste. Official Journal of the European Union. 14.6.2018. L 150/109. 32 p.

Federal Office for the Environment FOEN (2017). Substance flows in Swiss e-waste - Metals, non-metals, flame retardants and polychlorinated biphenyls in electrical and electronic devices. Bern, 2017. 8 p. Summary of the publication "Stoffflüsse im Schweizer Elektronikschrott" www.bafu.admin.ch/uz-1717-d

Galloway T S, Lee B P, Buric I, Steele A M, BPA Schools Study Consortium, Kocur A L, Pandeth A G, Harries L W. 2019. Plastics Additives and Human Health: A Case Study of Bisphenol A (BPA). Issues in Environmental Science and Technology 2019(47):131-155

Groh K J, Backhaus T, Carney-Almroth B, Geueke B, Inostroza P A, Lennquist A, Leslie H A, Maffini M, Slungef D, Trasande L, Warhurst A M, Muncke J. 2019. Overview of known plastic packaging-associated chemicals and. their hazards. Science of the Total Environment 651 (2019) 3253-3268

Hahladakis J N, Velis C A, Weber R, Iacovidou E, Purnell P. 2018. An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling. Journal of Hazardous Materials 344 (2018) 179-199

Häkkinen E. (2016). Finnish Guidance on POPs waste and pre-treatment of ELVs. Finnish Environment Institute. 22 November 2016, ppt 12 slides. Project on development of the environmental protection of transport, storage and pre-treatment operations of ELVs (Project report published in January 2016), Guidance on management of POPs waste (Published in September 2016).

Hennebert P, Filella M. (2017). WEEE plastic sorting for bromine essential to enforce EU regulation. Waste Management, 71, January 2018, 390-399.

Hennebert P. (2018a). Concentrations of brominated flame retardants in plastics of electrical and electronic equipment, vehicle, textiles and construction: a short review of occurrence and management. Proceedings of the 6th International Conference on Industrial and Hazardous Waste Management. Chania (Greece). 04-07/09/2018. 13 p.

Hennebert P. (2018b). Revue bibliographique des concentrations en substances réglementées dans les plastiques de véhicules hors d'usage. Rapport INERIS DRC-18-173977-02943B. 18/12/2018. 56 p.

Hennebert P. (2019). Sorting of waste for circular economy: sampling when (very) few particles have (very) high concentrations of contaminant or valuable element (with bi- or multi-modal distribution). 17th International Waste Management and Landfill Symposium (Sardinia 2019), 30/09 - 04/10/2019, Cagliari, Italy

Hense P, Aigner J, Franke M, Hornung A. (2018). Enabling the recycling of high-tech metals -Joined treatment of halogenated plastics with metal containing fractions. 17th International Electronics Recycling Congress - IERC 2018. January 18th, 2018, Salzburg.

ICL Industrial products (2012). Fire protection for automotive and transportation. 6 p. http://icl-ip.com/wp-content/uploads/2012/10/FR-Transportation-2012.pdf ; http://icl-ip.com/segment_category/automotive/ . Consulted Jan 2018.

Institute for Environmental Studies (IVM) and University of Amsterdam (IVAM) (2016). POP-BDE waste streams in the Netherlands: analysis and inventory. Report R13-16

Japanese Ministry of the Environment (MOE) (2011). Survey to identify the characteristics of automotive shredder residue (summary)

Jinhui, L., Yuan, C., Wenjing, X., 2017. Polybrominated diphenyl ethers in articles: a review of its applications and legislation. Environ. Sci. Pollut. Res. 24, 4312–4321. https://doi.org/10.1007/s11356-015-4515-6

Kajiwara, N., Takigami, H., 2013. Emission behavior of hexabromocyclododecanes and polybrominated diphenyl ethers from flame-retardant-treated textiles. Environ. Sci. Process. Impacts 15, 1957–1963. https://doi.org/10.1039/c3em00359k

Leslie H A, Leonard P E G, Brandsma S H, de Boer J, Jonkers N. (2016). Propelling plastics into the circular economy - weeding out the toxics ?rst. Environment International 94 (2016) 230-234.

Morf, L.S., Tremp, J., Gloor, R., Huber, Y., Stengele, M., Zennegg, M., 2005. Brominated flame retardants in waste electrical and electronic equipment: Substance flows in a recycling plant. Environ. Sci. Technol. 39, 8691–8699. https://doi.org/10.1021/es051170k

Morin N.A.O, Andersson P L, Hale S E, Arp H P. (2017). The presence and partitioning behavior of flame retardants in waste, leachate, and air particles from Norwegian waste-handling facilities. J of Environmental Sciences, 62 (2017), 115-132.

Niinipuu M. (2013). A comparative evaluation of brominated compounds in end-of-life vehicles. Polybrominated diphenyl ethers and polybrominated dibenzo-pdioxins and dibenzofurans in car seats. Master´s thesis at the Department of Chemistry, Umeå University, Sweden

Norwegian Climate and Pollution Agency (2012). Assessment of the need for new requirements for the environmentally sound treatment of end-of-life vehicles. March 2012. Mepex, Frode Syversen. 74 p.

Norwegian Climate and Pollution Agency, Mepex, 2012. Assessment of the need for new requirements for the environmentally sound treatment of end-of-life vehicles.

Norwegian Environment Agency (2015). Literature Study - DecaBDE in waste streams. Final Report. Reference number: 2015/10094. 11 December 2015. BiPRO GmbH, Alexander Potrykus. 160 p. (Meta reference presenting results of 17 citations other than the ones cited in this reference list, with less abundant data, and source not fully defined).

Norwegian Environment Agency (2016). Consultancy service on collecting, summarising and analysing information on c-decaBDE in waste - Analysis of the information received by the Basel Convention related to c-decaBDE as called for in decision BC-12/3. Reference number: 2016/4072. Final Report. BIPRO. Potrykus A. 25 p.

Norwegian Environment Agency (2017). Consultancy service on updating the Basel Convention Technical Guidelines to include information on decaBDE. Case number: 2017/5815. Supplementary Information on Destruction Technologies. Alexander Potrykus, BiPRO GmbH. 30th November 2017. 19 p.

Petreas M., Oros D. (2009). Polybrominated diphenyl ethers in California wastestreams. Chemosphere, 74

PlasticsEurope 2018. Plastics - the Facts 2018 - An analysis of European plastics production, demand and waste data. 60 p. https://issuu.com/plasticseuropeebook/docs/plastics_the_facts_2018-_afweb

Puype F, Samsonek J, Knoop J, Egelkraut-Holthus M, Ortlieb M. (2015). Evidence of waste electrical and electronic equipment (WEEE) relevant substances in polymeric food-contact articles sold on the European market. Food Addit. Contam. Part A 32, 410-426.

Rani, M., W. J. Shim, et al. (2014). "Hexabromocyclododecane in polystyrene-based consumer products: An evidence of unregulated use." Chemosphere 110: 111-119.

Risk and Policy Analysis (consulting company) (Georgalas B, Sanchez A, Zarogiannis P) (2014). European Chemical Agency 2014. Support to an annex XV dossier on bis-(pentabromophenyl) ether (decaBDE). J832/ECHA DecaBDE Final report

Sindiku, O., Babayemi, J.O., Tysklind, M., Osibanjo, O., Weber, R., Watson, A., Schlummer, M., Lundstedt, S., 2015. Polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) in e-waste plastic in Nigeria. Environ. Sci. Pollut. Res. 22, 14515–14529. https://doi.org/10.1007/s11356-015-5260-6

Sinkkonen S., Paasivirta J., Lahtiperä M., Vattulainen A. (2004). Screening of halogenated aromatic compounds in some raw material lots for an aluminum recycling plant. Environment International, 30

SMMT - Society of Motor Manufacturers and Traders (2016). Analysis of Automotive Shredder Residue from the Composition, Recycling and Recovery Trial for End of Life Vehicles in the United Kingdom. Arrichiello J and Davison P. Mayer Environmental. April 2016. 14 p.

Stubbings, W.A., Harrad, S., 2014. Extent and mechanisms of brominated flame retardant emissions from waste soft furnishings and fabrics: A critical review. Environ. Int. 71, 164–175. https://doi.org/10.1016/j.envint.2014.06.007

Swerea IVF (2018). Strååt M, Nilsson C. Decabromodiphenyl ether and other flame retardants in plastic waste destined for recycling. Project Report 5170721 M-973|2018. Contract number: 16128142. 2018.02.23. 29 p.

Swiss Agency for the Environment, Forests and Landscape (2003). Selected polybrominated flame retardants PBDEs and TBBPA, Berne.

Taurino, R., Pozzi, P., Zanasi, T., 2010. Facile characterization of polymer fractions from waste electrical and electronic equipment (WEEE) for mechanical recycling. Waste Manag. 30, 2601–2607. https://doi.org/10.1016/j.wasman.2010.07.014

Turner, A., Filella, M., 2017a. Field-portable-XRF reveals the ubiquity of antimony in plastic consumer products. Sci. Total Environ. 584–585, 982–989. https://doi.org/10.1016/j.scitotenv.2017.01.149

Turner, A., Filella, M., 2017b. Bromine in plastic consumer products – Evidence for the widespread recycling of electronic waste. Sci. Total Environ. 601–602, 374–379. https://doi.org/10.1016/j.scitotenv.2017.05.173

UNEP (2017a). United Nations Environmental Programme and Stockholm Convention. 2017. UNEP/POPS/COP.8/32. Stockholm Convention on Persistent Organic Pollutants. Conference of the Parties to the Stockholm Convention on Persistent Organic Pollutants. Eighth meeting. Geneva, 24 April-5 May 2017. Report of the Conference of the Parties to the Stockholm Convention on Persistent Organic Pollutants on the work of its eighth meeting. SC-8/10: Listing of decabromodiphenyl ether (commercial mixture, c-decaBDE). P 64/114. 13 July 2017. http://chm.pops.int/TheConvention/ConferenceoftheParties/Meetings/COP8/tabid/5309/Default.aspx

UNEP 2017b. Proposal to list perfluorohexane sulfonic acid (CAS No: 355-46-4, PFHxS), its salts and PFHxS-related compounds in Annexes A, B and/or C to the Stockholm Convention on Persistent Organic Pollutants. UNEP/POPS/POPRC.13/4*. Stockholm Convention on Persistent Organic Pollutants. 5 June 2017

Vojta S, Betanova J, Melymuk L, Komprdova K, Kohoutek J, Kukucka P, Klanova J (2017). Screening for halogenated flame retardants in European consumer products, building materials and wastes. Chemosphere 168: 457-466.

Wäger P, Schluep M, Müller E, Gloor R. (2012). RoHS regulated Substances in Mixed Plastics from Waste Electrical and Electronic Equipment. Environ. Sci. Technol. 2012, 46, 628?635.

Wäger P, Schluep M, Müller E. (2010). RoHS Substances in Mixed Plastics from Waste Electrical and Electronic Equipment. Final Report, September 17, 2010. Empa - Swiss Federal Laboratories for Materials Science and Technology (EMPA). 113 p.

WRc (consulting company) (2012). Peacock J., Turrell J., Lewin K., Glennie E. 2012. Analysis of Poly-Brominated Biphenyl Ethers (PBDEs) in Selected UK Waste Streams: PBDEs in waste electrical and electronic equipment (WEEE) and end of life vehicles (ELV). Final report for Defra, Report No.: UC8720.05

WRc (consulting company) Addendum (2012). Peacock J., Turrell J., Lewin K., Glennie E. 2012. Analysis of Poly-Brominated Diphenyl Ethers (PBDEs) in UK Waste Streams: PBDEs in end of life vehicles (ELV) - Addendum to WRc Report UC8720.04 (January 2012).

SUPPLEMENTARY INFORMATION

TABLE S1: Concentrations of some brominated flame retardants in car parts, end-of-life vehicles, automotive shredder residue (ASR) and post shredder treatment fractions – unweighted mean concentrations from literature data.

Mean Concentration (mg/kg)	PBDE	of which DecaBDE	HBCCD	TBBPA	All
Car (117 data)	3212	5432	431	27	2651
1990-1994 - Printed circuit board	77	200	10	26	53
1995-1999 #1 - Printed circuit board	4	10	10	20	8
1995-1999 #2 - Printed circuit board	10	10	10	87	26
2000-2004 - Printed circuit board	27	50	10	20	22
Airbag	27	50	50	20	30
Car	128	128			128
Car interior	8	14			8
Car seat cover	256	256			256
Car seats	66	66			66
European ELV parts	3469	5751			3469
Interior 1	5677	17000	50	20	3420
Interior material 1	9				9
Interior material 2	137				137
Interior Mazda 1998	52	52			52
Interior Pontiac 1997	18	18			18
Luggage compartment	27	50	50	20	30
Printed circuit boards sample 1	2				2
Printed circuit boards sample 3	2				2
Printed circuit boards sample 4	2				2
PUF from old car seats	1	1			1
PUF from old car seats, high contamination	900				900
PUF from old car seats, low contamination	1				1
PUF from US car seats	24	9			24
PUF Pontiac 1997	8676	522			8676
PUR foam for automotive applications	40000				40000
Radiator, outer	27	50	50	20	30
Rail vehicles	85000	85000			85000
Seat cover	9020	27000	50	20	5426
Seat cover material	51				51
Seat cover Mazda 1998	22700	22700			22700
Seat cover Pontiac 1997	8625	22500			8625
Soundproofing 1	27	50	4400	20	900
Soundproofing 2	2343	7000	50	20	1420
US/Asian ELV plastic parts	21	37			21
ELV (46 data)	925	3253	337		797
ELV foams	111	33	1		74
ELV upholstery	4053	8101	842		2983
Hyundai - foam	0	0	0		0
Škoda - foam	0	0	0		0
ASR (51 data)	179	386			179
ASR	254	429			254
Autoshredder Waste	44	44			44
Input Car shredder residue	6	17			6

Light ASR (foam and textile)	21				21
Mixed light plastic	3				3
Mixed SR	255	413			255
PST (33 data) (ELV and WEEE)	44	101			44
1,1 < density < 1,3	364	810			364
1,1<d<1,3 (black hard)	1	2			1
1,1<d<1,3 (black soft)	2	6			2
1,1<d<1,3 (coloured)	0	1			0
1,1<d<1,3 (white/grey)	5	3			5
d<1,1 (hard plastic)	0	0			0
density (0 - 1)	10	27			10
density < 1,1	2	6			2
Fiber fraction (foam)	43	113			43
Input for PST	10	29			10
Total (247 data)	**1623**	**3102**	**386**	**27**	**1447**

TABLE S2: Concentrations of some brominated flame retardants in plastics of construction - Unweighted average concentrations from bibliographic data.

Mean concentration (mg/kg)	PBDE	of which DecaBDE	HBCDD	TBBPA	Mean PBDE HBCDD TBBPA	n	Interpretation
epoxy adhesive	300000	300000			300000	1	with PBDE - decaBDE
electrical insulation	200000	200000			200000	2	
polyurethane foam (PUR)	130000				130000	4	
construction 2	4799				4799	1	
moisture-resistant membrane / film	1000	1000			1000	1	
pipe insulation	63	626	0		57	11	
recycled plastics	2	18	1		2	11	
phenolic foam insulation	0	4	0		0	11	
foam insulation	0	1	0		0	33	
blue sealing foam	0	0	5400		491	11	with HBCDD
expanded polystyrene (EPS)	0	0	1995	0	816	22	
pale mounting foam	0	0	832		76	11	
air conditioning - aluminum foil	1	6	545		50	11	
polystyrene	0	0	469		43	11	
polystyrene construction	0	0	469		43	11	
air conditioning - inner sheet	0	0	412		37	11	
fiber network	0	0	250		23	11	
polystyrene board	0	0	127		12	11	
extruded polystyrene (XPS)	0	0	28	0	10	14	
air conditioning - cellophane sheet	0	0	16		1	11	
drywall	0	0	9		1	11	
HARDSIL NT insulation	0	0	8		1	11	
air conditioning - fiberglass foam	0	0	8		1	11	
yellow mounting foam	0	0	3		0	11	
green mounting foam	0	0	2		0	11	
polyacrylate material	0	0	1		0	11	
laminate plastic flooring	0	0	1		0	11	
window corner cap	0	0	0		0	11	without BFR

heat exchanger	0	0	0		0	11	
insulation board	0	0	0		0	11	
unknown (construction site)			0		0	1	
insulation aluminum foil	0	0	0		0	11	
paper insulation	0	0	0		0	11	
exterior paint	0	0	0		0	11	
wood fiber insulation	0	0	0		0	33	
rubber	0	0	0		0	11	
plaster	0	0	0		0	11	
asphalt insulation	0	0	0		0	11	
formica	0	0	0		0	11	
oriented strand board (OSB)	0	0	0		0	55	
construction 1	0				0	1	
chipboard	0	0	0		0	55	
green sealing foam	0	0	0		0	11	
water resistant paint	0	0	0		0	11	
window finishing tip	0	0	0		0	22	
cotton insulation	0	0	0		0	11	
mastic	0	0	0		0	22	
paper insulation of recycled beverage cartons	0	0	0		0	11	
decorative polystyrene	0	0	0		0	11	
insulation hemp rope	0	0	0		0	11	
Drain pipe	0	0	0		0	11	
foam	0	0	0		0	11	
plank of wood	0	0	0		0	22	
brown chipboard	0	0	0		0	11	
elastic linoleum	0	0	0		0	22	
linoleum	0	0	0		0	22	
blown cellulose insulation	0	0	0		0	11	
plaster with fire retardant foam	0	0	0		0	11	
blow insulation made of paper	0	0	0		0	11	
building polystyrene panel	0	0	0		0	11	
drinking water pipe	0	0	0		0	11	
Total	**1713**	**8662**	**317**	**0**	**1555**	**806**	

TABLE S3: Concentrations of some brominated flame retardants in textile plastics - Unweighted average concentrations from literature.

Mean concentration (mg/kg)	PBDE	of which DecaBDE	HBCDD	TBBPA	Mean PBDE HBCDD TBBPA	n	Interpretation
commercial decaBDE treated polyester upholstery fabrics used in the manufacture of curtains	120000	120000			120000	2	with PBDE - DecaBDE
PUR foam for upholstered furniture	41040				41040	11	
various textiles	39850	39850			39850	11	
adhesive layer of reflective tapes	30000	30000			30000	11	
polyester	28131	120000	3		25318	11	
PUR foam for mattresses	25000				25000	2	
textile 2	20003				20003	12	
textile 1	11843				11843	11	
foam padding	7023	7023			7023	2	

window blinds	4799	4799			4799	11	
several carpets	907	1810	7		607	44	
several mattresses	115	230	3		78	12	
carpet	85	85			85	12	
velvet (70-80 g / m2)	27	27			27	22	
flat woven (30-80 g / m2)	21	21			21	22	
cotton (30-40 g / m2)	13	13			13	11	
tents	2	2			2	12	
insulation / carpet	1	2			1	11	
polyester	1	5	42500		4251	46	with HBCDD and with or without PBDE - decaBDE
furnishing	9999	19953	15050		11683	1	
curtain	0	0	8333		1087	2	
furniture foam filling	1060	2119	2275		1465	12	
curtains	7	14	15		10	4	
Persian rug	0	0	1		0	11	
treated textile	0				0	11	Sans RFB
Red carpet	0	0	0		0	10	
gray carpet	0	0	0		0	20	
blanket	0	0	0		0	2	
green carpet	0	0	0		0	5	
tablecloth	0	0	0		0	11	
foam	0	0	0		0	22	
textile material	0	0	0		0	1	
blue carpet	0	0	0		0	11	
brown carpet	0	0	0		0	1	
coconut fiber	0	0	0		0	1	
textile cover	0	0	0		0	11	
stuffing material	0	0	0		0	44	
pillow	0	0	0		0	1	
plush	0	0	0		0	1	
insulation of the textile bottle	0	0	0		0	33	
wardrobe	0	0	0		0	2	
bed-cover	0	0	0		0	2	
textile	0	0	0		0	1	
Total	**2080**	**6511**	**3465**		**2244**	**496**	