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LITERATURE EVIDENCE ON THE BROMINATED FLAME RETARDANT CONTENT IN PLASTICS OF CONSTRUCTION, OF TEXTILES/FURNITURES AND OF NON-FOOD PACKAGING: SHOULD THEY BE SORTED BEFORE RECYCLING?

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ABSTRACT: The objective of this study is to bring information to check if sorting of plastics of construction, of textiles and of non-food packaging is necessary before reuse in circular loops. Data from literature on these plastics has been gathered from 8 publications and reports plus one report quoting 6 other reports or publications from 2013 to 2018. Published data are heterogeneous, and are presented by sample, with synthesis table (n, min, media, mean, max, centile for 1000 mg/kg and 500 mg/kg). Data of total bromine and antimony are also presented. The global literature data collected here show that approximately 8%, 16% and 9% of the reported concentrations of plastics from building materials, textile products and non-food packaging exceed the EU concentration limit of 1000 mg/kg of regulated BFRs. Samples from continental Europe (69 out of 85 for construction plastics and 36 out of 55 for plastics for textile and furniture equipment) have very low concentrations, excepted four samples from Switzerland, Norway and Sweden. These findings should be checked by characterization campaign to decide if sorting of these plastics is necessary before recycling.

Keywords: PBDE, DecaBDE, HBCDD, Br, Sb

1. INTRODUCTION

The presence of brominated flame retardants (BFR) in plastics of electrical and electronic equipment and vehicles, and waste thereof, is well established. However, despite availability of scientific papers and technical reports, the common awareness is not so clear for plastics of construction (plumber pipes and electric sheath, windows frame, profiles, insulation foam, floor and wall covering, connexion boxes, ventilation devices, joints, seals, air and water sealing films, glue...), for plastics and synthetic fabric from textile equipment, furniture and upholstery, and for non-food plastics of packaging. The use of plastics in Europe in these sectors is important: 19.8% of total use of plastics for building and construction, 16.7% for medical equipment, plastic furniture and furniture equipment, technical parts used for mechanical engineering or machine-building, and 4.1% for household, leisure and sport (PlasticsEurope 2018), totalling so about 40% of the plastics used in EU, as much as packaging (food and non-food) plastics. Are some of these plastics brominated with concentrations hampering their recycling?

2. MATERIAL AND METHODS

Data from literature on plastics of construction, of textiles and of non-food packaging has been gathered from 8 publications and reports plus one report quoting 6 other reports or publications dealing with the plastics studied here. In total 14 sources from 2003 to 2018 were used. Published data are heterogeneous. Some data are single concentration data, and other data are range of concentration, with the number (n), the minimum (min), the median, the mean (mean), and the maximum (max) of the range. The mean concentrations presented here are the mean of all the single concentrations and the minimum, mean and maximum 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. Data of textiles are sometimes reported as quantity per item or per square meter, hence not comparable with the other data of this study and were not used. Only data for the most frequently measured BFRs (PBDES, including decaBDE, and HBCDD) are presented here.

3. RESULTS AND DISCUSSION

The distributions of concentrations gathered here depends on the geographic origins of the samples (national regulations for fire protection) and their number per country, the potential choice of the samples by the authors (voluntary choosen because suspected to contain BFR or not), the individual weight of the plastic items... These data nevertheless seem usefull for documenting the question of the sorting of these plastics. The distributions of BFR are heterogeneous and it is useful to detail the results by category of plastics, by sample and by country (Table 1 and Table 2). The samples are sorted for decreasing polybromodiphenylether (PBDE), decabromodiphenylether (DecaBDE) and hexobromocyclododecane (HBCDD) concentrations. A synthesis is presented in Table 3, with the number of data, the median, mean and maximum concentrations, and the centile of data < 1000 mg/kg for BFR (a typical regulatory concentration limit), < 2000 mg/kg for bromine (the recommended concentration limit in the standard CENELEC CLC/TS 50625-3-1 for sorting plastics of small household appliances and screens), and < 8354 mg/kg for antimony (the concentration limit rendering the plastic carcinogenic according to HP7 criteria in the EU legislation). Details on regulatory concentrations in EU can be found in Hennebert and Filella (2018).

Table 1: Literature-reported concentrations of some brominated flame retardants in plastics of construction

Sample / Concentration (mg/kg)	PBDE	DecaBDE	HBCDD	Country	Category
Epoxy adhesive	300000	300000		Norway	
Electrical insulation	200000	200000		Sweden	
PUR foam 1	140000			USA	
PUR foam 2	120000			Not known	
construction 2	4799			Global literature	With PBDE
Damp-proof membrane/film	1000	1000		Switzerland	
pipe insulation	63	626	0	Czechia	
recycled plastic materials	2	18	1	Czechia	
blue sealant foam	0	0	5400	Czechia	
EPS D	0	0	3050	Ireland	
EPS 5			1609	Korea	
EPS 4			1519	Korea	
EPS 8			1335	Korea	
EPS 7			1098	Korea	
pale mounting foam	0	0	832	Czechia	
air conditioning - alu foil	1	6	545	Czechia	
polystyrene	0	0	469	Czechia	
building polystyrene	0	0	469	Czechia	
air conditioning - inside foil	0	0	412	Czechia	With HBCDD
fibre net	0	0	250	Czechia	1
EPS 6	1		194	Korea	1
polystyrene board	0	0	127	Czechia	1
XPS D	0	0	35	Ireland	1
air conditioning - cellophane foil	0	0	16	Czechia	1
drywall	0	0	9	Czechia	1
insulation HARDSIL NT	0	0	8	Czechia	
air conditioning - glass fibre foam	0	0	8	Czechia	
yellow mounting foam	0	0	3	Czechia	
green mounting foam	0	0	2	Czechia	
phenolic foam Insulation	0	4	0	Czechia	
foam insulation 2	0	2	0	Czechia	
foam insulation 3	0	1	0	Czechia	
wood fibre insulation 3	0	1	0	Czechia	
heat exchanger	0	0	0	Czechia	
paper insulation	0	0	0	Czechia	
rubber	0	0	0	Czechia	
construction 1	0			Global literature	
outdoor paint	0	0	0	Czechia	
insulation panel	0	0	0	Czechia	
window finishing bead 1	0	0	0	Czechia	
foam insulation 1	0	0	0	Czechia	
recycled beverage cartons paper insulation	0	0	0	Czechia	
insulation hemp rope	0	0	0	Czechia	
insulation aluminium foil	0	0	0	Czechia	1
waste pipe	0	0	0	Czechia	1
resilient linoleum 2	0	0	0	Czechia	1
linoleum 2	0	0	0	Czechia	1
water-resisting paint	0	0	0	Czechia	Without BFR
linoleum 1	0	0	0	Czechia	1
paper sound insulation	0	0	0	Czechia	1
polyacrylate material	0	0	1	Czechia	1
laminated plastic floor covering	0	0	1	Czechia	1
oriented strand board 2	0	0	0	Czechia	1
window corner bead	0	0	0	Czechia	1
plaster	1	0	0	Czechia	1
asphalt insulation	0	0			1
formica	0	0	0	Czechia	
	-	-	0 0	Czechia Czechia	
oriented strand board 1	0	0			
	0	0	0	Czechia	
oriented strand board 1	0 0 0	0 0 0	0 0	Czechia Czechia	
oriented strand board 1 chipboard 3	0 0 0 0	0 0 0 0	0 0 0	Czechia Czechia Czechia	
oriented strand board 1 chipboard 3 wood fibre insulation 1 green sealant foam	0 0 0 0 0 0	0 0 0 0 0 0	0 0 0 0	Czechia Czechia Czechia Czechia	
oriented strand board 1 chipboard 3 wood fibre insulation 1	0 0 0 0 0 0 0	0 0 0 0 0 0 0	0 0 0 0 0	Czechia Czechia Czechia Czechia Czechia	
oriented strand board 1 chipboard 3 wood fibre insulation 1 green sealant foam cotton insulation	0 0 0 0 0 0 0 0	0 0 0 0 0 0 0 0	0 0 0 0 0 0	Czechia Czechia Czechia Czechia Czechia Czechia Czechia	
oriented strand board 1 chipboard 3 wood fibre insulation 1 green sealant foam cotton insulation sealant 1	0 0 0 0 0 0 0 0 0	0 0 0 0 0 0 0 0 0	0 0 0 0 0 0 0	Czechia Czechia Czechia Czechia Czechia Czechia	

chipboard - brown	0	0	0	Czechia	
oriented strand board 3	0	0	0	Czechia	
chipboard 2	0	0	0	Czechia	
oriented strand board 4	0	0	0	Czechia	
chipboard 4	0	0	0	Czechia	
chipboard 1	0	0	0	Czechia	
window finishing bead 2	0	0	0	Czechia	
blown cellulose insulation	0	0	0	Czechia	
fireproof foam plaster	0	0	0	Czechia	
building polystyrene board	0	0	0	Czechia	
drinking water pipe	0	0	0	Czechia	
resilient linoleum 1	0	0	0	Czechia	
foam 2	0	0	0	Czechia	
decorative polystyrene	0	0	0	Czechia	
chipboard 5	0	0	0	Czechia	
wood fibre insulation 2	0	0	0	Czechia	
wooden board 1996	0	0	0	Czechia	
XPS 12			0	Korea	
Unknown (from construction site)			0	Ireland	

Table 2: Literature-reported concentrations of some brominated flame retardants in plastics of textiles and furniture

Sample / Concentration (mg/kg)	PBDE	DecaBDE	HBCDD	Country	Category
Polyester textile C	130000	120000	3	Japan	
c-decaBDE treated polyester upholstery textiles used in curtain				-	
manufacture	120000	120000		Japan	
PUR foam for upholstered furniture	41040			USA	
adhesive layer of reflective tapes	30000	30000		Asia to Norway	
PUR foam for mattresses	25000			USA	With PBDE
textile 2	20003			Global literature	
textile 1	11843			Global literature	
upholstery foam	7023	7023		India	
window blind	4799	4799		India	
carpet	85	85		Turkey	
polyester Textile A	0	1	43000	Japan	
polyester textile B	2	10	42000	Japan	
curtain textile A polyester blue			26000	Japan	
curtain textile B polyester black			24000	Japan	With HBCDD and
furniture fabrics	9999	19953	15050	Ireland	with or without
furniture foam filling	1060	2119	2275	Ireland	PBDE
curtain D	7	14	15	Ireland	
carpets	907	1810	7	Ireland	
mattresses	115	230	3	Ireland	
Insulation/carpet padding	1	2		The Netherlands	
Textile treated with DecaBDE	0			Japan	
persian carpet	0	0	1	Czechia	
curtain 2006	0	0	1	Czechia	
grey carpet 1	0	0	0	Czechia	
green carpet 1981	0	0	0	Czechia	
table cloth 1996	0	0	0	Czechia	
foam 1	0	0	0	Czechia	
red carpet	0	0	0	Czechia	
blanket	0	0	0	Czechia	
textile material 1	0	0	0	Czechia	
curtain 2005	0	0	0	Czechia	Without BFR
textile material 2	0	0	0	Czechia	WILLIOUL BFK
blue carpet	0	0	0	Czechia	
grey carpet 2	0	0	0	Czechia]
brown carpet	0	0	0	Czechia]
green carpet 2006	0	0	0	Czechia	
upholstery material 1	0	0	0	Czechia	
foam 2	0	0	0	Czechia]
coir (coconut fibre)	0	0	0	Czechia	1
cover textile	0	0	0	Czechia	
upholstery material 3	0	0	0	Czechia	1
pillow fill	0	0	0	Czechia	1
textile material 3	0	0	0	Czechia]

plush	0	0	0	Czechia
curtain 2008	0	0	0	Czechia
foam 1981	0	0	0	Czechia
textile bottle insulation	0	0	0	Czechia
foam	0	0	0	Czechia
textile material 4	0	0	0	Czechia
hanging	0	0	0	Czechia
bed cover	0	0	0	Czechia
curtain 2007	0	0	0	Czechia
upholstery material 2	0	0	0	Czechia
table cloth 1986	0	0	0	Czechia
textile 2010	0	0	0	Czechia

For non-food packaging, 65 data (without data ranges) were collected and are summarized in Table **3**. These plastics are exclusively EPS (expanded polystyrene) and XPS (extruded polystyrene) with HBCDD as BFR. Concentrations are low, but 31 samples have a HBCDD concentration > 10 mg/kg, the highest being polystyrene packaging laboratory equipments, appliances and printers (Rani et al (2014), Abdallah et al (2018)). The maximum HBCDD content exceeds the concentration limit for hazardous waste according to EU legislation.

For BFR, the distributions of concentration are totally skewed by some large concentrations, as shown in the tables and by the difference between the medians and the means (Table 3). The medians are all < 0.1 mg/kg or lower, excepted HBCDD in non-food packaging (11 mg/kg, n=57). The mean concentrations are the concentration that would be measured if all the samples (having all the same mass) were mixed and analysed. It is determined by the last few centiles of samples with very high concentrations, illustrated here by the maximum concentration, and by the centile of data with a concentration > 1000 mg/kg (8% for construction plastics, 16% for textile and furniture, and 9% for non-food packaging). For bromine and antimony, data are less numerous and indicates that 22%, 29% and 31% for the data exceed 2000 mg Br/kg for the three category of plastic, and one data from four data of construction and of textile and furniture exceeds the antimony concentration rendering this plastic hazardous.

Category	BFR, Br, Sb	Concentration (mg/kg)					data < 1 000 mg/kg	data < 500 mg/kg
		n	Min	Median	Mean	Max		
Construction	PBDE	716	0	0	9819	300000	99%	99%
	Of which DecaBDE	81	0	0	6792	300000	95%	94%
	HBCDD	84	0	0	223	5400	92%	89%
	ТВВРА	6	0	0	0	0	100%	100%
	Br	9	0	45	2122	9410	% < 2000 mg/kg : 78%	
	Sb	4	103	984	3768	13000	% < 8354 mg/kg : 75%	
Textile and	PBDE	437	0	0	7583	130000	95%	94%
furniture	Of which DecaBDE	75	0	0	6376	120000	84%	81%
	HBCDD	59	0	0	3463	43000	86%	86%
	ТВВРА	-	-	-	-	-	-	-
	Br	70	0	99	7175	128300	% < 2000 mg/kg : 71%	
	Sb	4	90	944	2975	9922	% < 8354 mg/kg : 75%	
Non-food	PBDE	_	-	-		-		
packaging	Of which DecaBDE	-	-	-	-	-	-	-
packaging	HBCDD	- 57	0	11	232	5897	91%	- 88%
	ТВВРА	8	0	0	0	1	91%	0070
		-	-	-	-	-	-	-
	Br	6	0	10	1153	5600	% < 2000 mg/kg : 69%	
	Sb	-	-	-	-	-	-	

Table 3: Summary of literature-reported concentrations of some brominated flame retardants, total bromine and antimony in plastics of construction, of textiles and furniture, and of non-food packaging

4. CONCLUSIONS

The European plastics regulation in the context of a circular economy stipulates that plastics containing regulated BFRs (i.e. BFR recognized as persistent organic pollutants by the Stockholm Convention) must be sorted and managed separately from the non-brominated fraction in order to avoid the uncontrolled dispersion of these substances in recycled raw materials.

The global literature data collected here show that approximately, in the selected publications, 8%, 16% and 9% of the reported concentrations of plastics from building materials, textile products and nonfood packaging exceed the EU concentration limit of 1000 mg/kg of regulated BFRs. Samples from continental Europe (69 out of 85 for construction plastics and 36 out of 55 for plastics for textile and furniture equipment) have very low concentrations, excepted seven samples from Ireland, Norway, Sweden and Switzerland,.

Major waste fractions including plastics from construction and demolition waste must be sorted in the EU to facilitate re-use and recycling (EU 2018). Mandatory separate collection of textiles is planned in the EU by 2025 at the latest. Characterization campaigns should be considered to confirm if their RFB content involve a specific sorting before recycling for some category of waste among them.

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