

TOXIC TOY OR TOXIC WASTE: OLD POPS IN NEW PRODUCTS



Jitka Strakova

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Jitka Strakova (Arnika – Toxics and Waste Programme)

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Abstract

This study brings forward evidence about recycling of plastic parts of electronic waste contaminated by POPs-PBDEs as well as by alternatives to PBDEs also raising concern into magic cube toys. The results raise concerns about Low POPs Content Level (LPCL) of PBDEs and E-waste management, including recycling materials containing flame retardants that result in further human and environmental exposure to substances listed in the Stockholm Convention for global elimination, particularly for E-waste workshop operators, recyclers and children.

Magic cubes made of black hard plastic purchased in Armenia, Czech Republic, Hungary, Serbia, and China, and the original Rubick's cubes® purchased in Hungary and Czech Republic, were screened for bromine using a handheld XRF analyzer and analyzed in the laboratory for polybromodiphenyl ethers (PBDEs) and new, alternative brominated flame retardants (nBFRs). Bromine was not detected at significant levels in samples from Armenia and Czech Republic. Laboratory analysis of 7 samples from Belarus, Hungary, Serbia and China found that six samples of magic cubes contained components of a commercial octaBDE mixture (hexabromodiphenyl and heptabromodiphenyl ethers), which is listed in the Stockholm Convention for global elimination, in concentrations ranging from 3 to 57 ppm and with a median value of 13 ppm. Content of substances resembling commercial octaBDE exceeded the provisional Low POPs Content Level of 50 ppm set for PCBs¹, in one of the samples. Four samples of magic cubes from Serbia and China exceeded the EU Low POPs Content limit for heptabromodiphenyl ether. Concentrations of the flame retardant decaBDE, which has been proposed for addition to the Convention, ranged from 36 to 153 ppm with the median value of 42 ppm. Only the original Rubick's cube® from Hungary did not contain significant levels of the PBDEs or nBFRs, indicating that only this sample was not very likely made of recycled E-waste plastics.

Acknowledgement

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¹ PBDEs levels are compared with LPCL for PCBs as PBDEs are effectively as hazardous as PCBs.

Introduction

Environmental impacts of brominated flame retardants

Toxic chemicals marketed as flame retardants are widely present in our homes, our bodies, and even in our wastes. These substances are being found throughout the global environment, wildlife, and even in areas far from where these substances are used or produced, like the Arctic. Due to increasing public health concerns, actions to regulate and eliminate them have occurred globally, nationally, and locally.

Penta- and Octabrominated diphenylethers (Penta- and OctaBDEs) global elimination

In May 2009, the 4th Conference of the Parties of the Stockholm Convention (COP4) listed certain congeners contained in commercial pentabromodiphenyl ether (PentaBDE) and octabromodiphenyl ether (OctaBDE) in Annex A for global elimination. PentaBDE was mainly used in polyurethane materials such as furniture foam and OctaBDE in the housings of electrical and electronic equipment, mainly in the plastic acrylonitrile butadiene styrene (ABS) of TV and computer casings.

Recycling exemptions for Penta and OctaBDEs

The decision included specific exemptions, which may last until 2030, allowing the recycling of materials containing these substances (such as plastics and foam) into new products. COP4 also requested the Persistent Organic Pollutants (POPs) Review Committee (POPRC) to evaluate this practice. Key recommendations from the POPRC included taking action to "...eliminate brominated diphenyl ethers from the recycling streams as swiftly as possible." The Committee noted that, "Failure to do so will inevitably result in wider human and environmental contamination and the dispersal of brominated diphenyl ethers into matrices from which recovery is not technically or economically feasible and in the loss of the long-term credibility of recycling."

Low POPs Content limit for Penta and OctaBDEs

The task of establishing Low POPs limits or the level of contamination above which a waste becomes a POPs waste was taken over by the Basel Convention in 2002. These limits apply to PBDEs that were newly listed in 2011, and define whether a waste, due to its POPs content, will have to be treated by the destruction or irreversible transformation methods defined in Annex V to the regulation. Nevertheless, those limits have been provisional until today. The first challenge is that, as many of the POPs "exhibit carcinogenic effects, there is a risk with any level of exposure" (Senes, 2002).

In August 2010 the European Union (EU) adopted Commission regulation 757/2010, which updated the EU POPs regulations with Low POPs Content limits for the POP PBDEs. The values included are 10 ppm for each of the Stockholm BDEs.

Based on the evaluation of major substance and waste flows, and given the expected trend for remaining stocks of PBDEs in used products, the ESWI/BiPro (2011) suggested a preferred option for LPCLs for new POPs. This option is 10 ppm for each of the Stockholm BDEs, as in the case of the EU regulation.

Contrary to the recommendation, drafts of Technical Guidelines for Environmentally Sound Management of PBDEs and the General Technical Guidelines stated that the provisional definition for PBDEs is as a sum of BDEs, and increased the value to 1000 ppm. This is also inconsistent with the provisional limit for PCBs (50 ppm), which are substances resembling PBDEs. In spite of the comparable toxicity, the proposed level for PBDEs is 20x weaker than the level for PCBs.

Decabrominated diphenylether (decaBDE)

The Stockholm Convention expert committee also agreed that decaBDE - a commonly-used chemical marketed as a flame retardant - warranted global action due to its harmful properties for humans, animals and the environment. It is used mainly in polystyrene, which is used in the television industry. This substance exhibits properties of POPs.

Nevertheless, industry distorted the risks to children by this substance, so it was not included into the Annex A of Stockholm Convention for global elimination.

Alternative halogenated flame retardants to PBDEs have toxic characteristics that raise concerns

In addition to polybrominated diphenylethers (PBDEs), various alternative halogenated flame retardants have been applied or were recently introduced by the industry to replace PBDEs. Overall, toxic chemicals marketed as flame retardants lack adequate toxicity information. However, the information that is available has raised concerns. For example, BTBPE, PBEB, PBT and HBB are persistent, bioaccumulative and travel long distances.

1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE)

BTBPE is one of the new flame retardants that replaced octaBDE. It is used in computers, TVs and cell phones. Its metabolite, 2, 4, 6-tribromophenol, is a thyroid-disrupting chemical that has been found in umbilical cord blood.

Oktabromtrimetylfenylindan (OBIND)

OBIND is another replacement of PBDEs that is used in electronic products. There is very little known about its toxicity.

Pentabromoethylbenzene (PBEB)

PBEB is a flame retardant that was used mainly in the 1970's and 1980's under the name FR-105. It was used in polymers and has poorly characterized toxicologically, but the substance is a brominated analogue of ethyl benzene, a carcinogen.

Pentabromtoluen (PBT)

PBT is used in polystyrene electronics, ABS plastics and other plastic polymers, and sold under the name FR-105 or Flammex. Studies confirmed histologic changes on laboratory rats; however, other than this fact, there is very little known about this substance. A significant property of this substance is an ability to be transported for long distances.

Hexabrombenzen (HBB)

HBB is a retardant mainly produced in Asia and applied to electronics. A significant property of this substance is an ability to travel long distances.

Decabromodiphenylethane (DBDPE)

DBDPE is a commercially important alternative to decaBDE used in the television industry. A significant property of this substance is an ability to be transported for long distances.

Survey Design and Methods

A principal recycling route of e-waste plastics containing brominated flame retardants (BFRs) is via toys. As this recycling practice has implications for consumer exposure, particularly exposure to children, we investigated whether PBDEs were present in magic cubes made of black plastic. The original Rubick 's cube® was purchased in both Czech Republic and Hungary, four samples of magic cubes were purchased in both China and Serbia, two magic cubes each were purchased in Czech Republic, Belarus, and Armenia, and one in Hungary. We tested the black parts of the cubes, as we anticipated that those contain BFRs, because manufacturers add black colour to the recycled plastics for aesthetic reasons.

To target articles likely to contain BFRs, samples were screened for bromine using an XRF device, and positive samples were analyzed for PBDEs and nBFRs at the Institute of Chemical Technology, an accredited laboratory in the Czech Republic.

The main components of commercial octaBDE mixtures listed in Annex A to the Stockholm Convention, meaning 2,2',4,4',5,5'-hexabromodiphenyl ether (BDE-153, CAS No: 68631-49-2), 2,2',4,4',5,6'-hexabromodiphenyl ether (BDE-154, CAS No: 207122-15-4), and 2,2',3,4,4',5',6'-heptabromodiphenyl ether (BDE-183, CAS No: 207122-16-5), were investigated.

Other components of commercial mixtures of penta-, okta- and dekaBDEs were also analysed. For purposes of calculation: components of commercial PentaBDE mixtures include the following congeners: BDE 28, 47, 49, 66, 85, 99, 100; components of commercial OctaBDE mixtures include the following congeners: BDE 153, 154, 183, 196, 197, 203, 206, 207; and the component of commercial decaBDE mixtures is BDE 209.

Additionally, new BFRs including BTBPE (1,2-bis(2,4,6-tribrom-fenoxy)etan), HBB (hexabrombenzen), PBEB (pentabrometylbenzen), PBT (pentabromtoluen), and OBIND (oktabromtrimetylfenylandan) were tested in all, and DBDPE (decabromodiphenylethane) in five samples.

Brominated flame retardants were extracted by n-hexan and the leach transferred into isooctane. Identification and quantification of flame retardants were accessed via gas chromatography/mass spectrometry in the mode of electron ionization (GC-MS/MS-EI). Limit of detection for PBDEs, PBT, PBEB, HBB, and BTBPE was 0.1 ppb and for OBIND and DBDPE was 20 ppb.

Results

Magic cubes from Armenia, Czech Republic, Hungary, Serbia, and China were screened for bromine using a handheld XRF analyser and analyzed in the laboratory for polybromodiphenyl ethers (PBDEs). Bromine was not detected at significant levels (above 1000 ppm) in samples from Armenia and Czech Republic.

Laboratory analysis of 7 samples from Belarus, Hungary, Serbia and China found that six samples contained hexabromodiphenyl and heptabromodiphenyl ether, which are listed in the Stockholm Convention for global elimination, in total concentrations ranging from 3 to 57 ppm (see Table No.1). Only the original Rubick's Cube did not contain any significant levels (above 1 ppm) of PBDEs listed in the Stockholm Convention. One cube from Serbia crossed the provisional Low POPs Content Level of 50 ppm listed for PCBs, substances resembling commercial octaBDE. Four samples exceeded the EU Low POPs Content limit of 10 ppm for heptabromodiphenyl ether.

Table No. 1: Concentration (ppm) of PBDEs listed in the Stockholm Convention in the investigated products

	Cube "Toys"	"Magic cube"	Cube "QJ"	Cube "Toys" small	"IQ Magic cube"	Cube "Toys" big	Rubick 's cube®
Country of purchase	Belarus	Belarus	China	Serbia	Serbia	Serbia	Hungary
hexaBDE*	0.676	0.897	1.966	7.935	2.220	8.726	<LOQ
heptaBDE*	2.545	4.432	11.359	41.831	10.917	48.319	0.003
Σhexa and heptaBDEs*	3.221	5.329	13.325	49.766	13.137	57.045	0.003
decaBDE	133.949	152.634	36.255	36.593	46.691	35.519	0.037

*PBDEs listed in Annex A of the Stockholm Convention

In addition to POP-PBDEs, new BFRs were identified. The most abundant representatives are hierarchically listed in Table No.2.

Table No. 2: Average and median values for most abundant BFRs (ppm)

	Average	Median
BTBPE	114.1	91.0
decaBDE	73.6	41.6
Commercial mixture of octaBDE⁺	43.0	27.2
Hexa and heptaBDEs*	23.6	13.2
DBDPE	11.9	13.3
OBIND	7.8	5.7
ΣPBDEs	116.9	123.2

⁺ Components of OctaBDE include the following congeners: BDE 153, 154, 183, 196, 197, 203, 206, and 207.

*Hexa and heptaBDEs, main components of commercial octaBDE listed in the Annex A of the Stockholm Convention

Discussion

Results of this brief survey indicate that toys made of hard recycled E-waste plastics are available at the market in Belarus, Serbia and China. All the BFR-positive samples come from cheap Chinese production except for one sample purchased in Belarus, where the country of manufacture was not indicated on the toy packaging. The original and more expensive Rubick's cube bought in Hungary was almost free of all tested BFRs.

BFR screening technique. The explanation for the focus on black plastic samples of thermo cups and kitchen utensils by Samsonek and Puype (2013) is that black is used for aesthetic and technological reasons. Firstly, recycled polymers from waste electrical and electronic equipment (WEEE) retain a brownish color, and secondly, adding black pigments hides the presence of BFR-contaminated WEEE. This explanation was confirmed in our study of toys articles. The combination of the focus on black-colored samples and application of XRF spectrometry for distinguishing bromine-positive samples seems to be an appropriate screening technique to indicate BFR content.

BFR concentration. The concentration of BFRs measured in magic cube toy products are summarized in Table No.1 and Appendixes. PBDEs were detected at significant levels in all cube toys except for the original Rubick's cube®. A median concentration of PBDE, expressed as a sum of the 16 congeners, was 123 ppm. The levels of PBDEs found in this study were substantially higher than those found by Chen et al. (2009) in a sample of hard toys in China, which had a median value of 53 ppm. Although our study involved two extra congeners in comparison to Chen's et al. study, those did not contribute to higher concentration, as BDE 85 was not detected in any sample and BDE 49 was either not detected or found in negligible amounts. In addition to our Chen et al (2009) study, a recent study by Arnika (Strakova and Krcmarova, 2014) revealed POP-BDEs in thermo cups and children's cutlery.

A non-PBDE BFR representative used as a commercial alternative to octaBDE, BTBPE occurred at the highest concentration of 281 ppm and 91 ppm of a median value. It was followed by decaBDE, with the highest concentration of 152 ppm and a median value of 41 ppm, hepta (median of 11 ppm) and hexa (median of 2 ppm) BDE. This indicates predominant contamination of BTBPE, deca and octa BDE as a result of E-waste plastics recycling. The same finding for decaBDE was detailed in a recent study by Samsonek and Puype (2013). This hypothesis is supported by occurrence of other POPs PBDE alternatives, such as DBDPE and OBIND. DecaBDE is likely to be a significant source for the current POP BDEs, because total production and use of penta and octa BDE commercial mixtures between 1970 and 2005 was much lower than the production of decaBDE (POPRC, 2010). It is very likely, therefore, that in the future the most important source of the current POP-BDEs will be the debromination of decaBDE as shown indicatively by Ross et al. (2009). Thus, whether or not it is finally listed as a POP itself cannot sensibly be ignored. The negligible (below 1 ppm or limit of detection) concentrations were observed for tri, tetra and pentaBDE, which are the main congeners of commercial pentaBDE products. This is expected, as the pentaBDE was used as a flame retardant in upholstering in homes and vehicles, and is being recycled into a foam product, such as carpet padding (DiGangi et al., 2011).

Congener assigning to commercial mixtures. Taking into consideration that commercial octaBDEs were always supplied in complex technical mixtures of hexa, hepta, octa and nona BDEs, the concentrations of the respective flame retardant corresponded not only exclusively to hexa and hepta BDEs officially assigned to commercial octa BDEs by the Stockholm Convention, but also to their more brominated counterparts (octa and nona). Therefore, concentrations of commercial octaBDE was in reality ranging higher than Table No.1 presents - from 9 to 91 ppm and a median value of 27 ppm. Consequently, concentration of octaBDE in one more case exceeded the EU limit (10 ppm), and in one more case the concentrations rose above the Basel provisional Low POPs Content Level for PCBs (50 ppm).

Recycling exemption for PBDEs. The data collected in this study demonstrate that main components of octaBDE listed in Annex A of the Stockholm Convention with the intention to be globally eliminated are being recycled into toys such as magic cubes. This finding is in accordance with the study of Chen et al. (2009) or analysis of POP-BDE

stream in the Netherland by Leslie et al. (2013), illustrating that 22% of the POP-BDE in WEEE is expected to end up in recycled plastics. Nevertheless, those discoveries contradict Article 6 of the Convention, which states that POPs in waste, including octaBDEs in E-waste, shall be “destroyed or irreversibly transformed so that they do not exhibit the characteristics of persistent organic pollutants.” Uncontrolled dissemination of POPs from E-waste into new products (including toys) via recycling processes can never fulfil those Convention provisions.

There are key concerns relating to octaBDE in recycled plastics, particularly those for sensitive uses including toys. According to Chen et al. (2009), the people most highly exposed were assessed to be children aged 3-18 months that mouthed toys. Chen therefore concluded that the exposures “are of special concern because of the relatively higher exposures observed for young children.” In terms of worker exposure, the POPRC (2010) summarizes that the actions of regrinding and moulding of polymers are among the worst PBDE waste treatment options.

Low POPs Content Level of PBDEs. The applied low POPs concentration limit value being establishing by the Basel Convention body highly influences the quantities of wastes to be managed as POP wastes. The levels currently proposed by the EU for Low POPs Content Level of PBDEs is 1000 ppm for the sum of the homologue groups (total BDEs). The correlation of low POPs concentration limit quantities of POPs waste in a study by ESWI/BiPro (2011) illustrates that for the LPCL of 1000 ppm, negligible proportion of waste-containing POP-PBDEs would be classified as POPs waste. As a consequence of the 1000 ppm provision, most of POPs E-waste would not require any destruction or irreversible transformation of the highly hazardous flame retardants. Taking into consideration that POP-PBDEs are effectively as hazardous as PCBs and are being recycled into children’s toys, having a LPCL of 1000 ppm undermines the intention of the Stockholm Convention and the effectiveness of the BAT-BEP Guidelines. Nevertheless, the EU-proposed threshold is being compared with an alternative limit of 200 ppm for penta, octa, and hexa BDE, and 1000 ppm for hepta BDE, meaning 1600 ppm for total BDEs. This is misleading, because those outdated levels were based on temporary and transitional arrangements by ESWI/BiPro and were to be revised. The updated proposal by ESWI/BiPro comes with the preferable threshold of 10 ppm for each of the homologue groups. This threshold is exceeded by four analysed samples, the threshold of 1000 ppm by none of them. It demonstrates that the threshold of 1000 is far too high to avoid dilution of PBDEs into the toys. After application of this limit, it will be hard to eliminate PBDEs from both product and waste streams. The stricter level would eliminate most of the PBDEs directly upon becoming waste. Taking into consideration that the provisional level for PCBs is 50 ppm, and one of our analysed samples exceeds this level with PCBs-resembling substances and that a second is very close to crossing the threshold, we may conclude that children play with toys that could be easily considered as hazardous waste.

Conclusions and recommendations

The study illustrates contamination of toys in Belarus, Serbia and China by POP-PBDEs as a result of E-waste recycling. It also demonstrates that the proposed Low POPs Content Level listed in the Technical Guidelines of the Basel Convention for POP-PBDEs is far too high to eliminate PBDEs from both product and waste streams. DecaBDE is to be a significant source for the current POPs BDEs. As noted, it is very likely, therefore, that in the future the most important source of the current POP-BDEs will be the debromination of decaBDE, and thus it cannot sensibly be ignored, whether or not it is finally listed as a POP itself.

The recycling of products containing POP-PBDEs certainly affects the range of products and wastes in which the PBDEs are used and generates new sources of wastes-containing POPs from the recycling processes. While the product types in which PBDEs have been used are known, details of the extent of current and likely future recycling flows are much less well quantified. For those reasons, inventories of PBDE flows in recycled materials needs to be undertaken to identify articles contaminated by PBDE and to enable comprehensive evaluation and review of brominated diphenyl ethers pursuant to paragraph 2 of parts IV and V of Annex A to the Stockholm Convention. At the next Conference of the Parties for the Stockholm Convention, results of the inventories shall be evaluated and actions taken to eliminate brominated diphenyl ethers from the recycling streams.

Having a Low POPs Content Level for PBDEs twenty times weaker than for PCBs, which are substances resembling PBDEs, may encourage the export of hazardous, POPs-contaminated wastes from developed to developing countries, and countries with economies in transition unable to deal with those wastes safely. IPEN believes that all the current provisional Low POPs Content limits are unreasonably high and are not consistent with the aims of the Convention. A more appropriate and protective standard for the Low POPs Content limit would be 10 ppm total concentration of the PBDE commercial mixture. Taking into account actions and negotiation, Low POPs Content Level for waste containing substances effectively as hazardous as PCBs should be established at least as low as for PCBs.

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Appendix 1: Concentration of nBFRs in the investigated products (ppb)

	Cube "Toys" Belarus	"Magic cube" Belarus	Cube "QJ" China	Cube "Toys" small Serbia	"IQ Magic Cube" Serbia	Cube "Toys" big Serbia	Rubick 's cube® Hungary
PBT	<0.1*	<0.1*	71.4	52.6	36	56.8	<0.1*
PBEB	<0.1*	<0.1*	2.62	1.63	1.2	1.8	<0.1*
HBB	86.4	441	288	226	122	217	<0.1*
BTBPE	12844	22140	119365	186317	62660	281291	41
OBIND	<20*	9594	15864	1124	1820	18332	<20*
DBDPE	N/A	N/A	7171	13942	13236	13401	<20*

*Limit of quantification (LOQ)

Appendix 2: BDE congeners and their potential distribution into commercial mixtures (ppb)

Analyt	Cube "Toys" Belarus	"Magic cube" Belarus	Cube "QJ" China	Cube "Toys" small Serbia	"IQ Magic Cube" Serbia	Cube "Toys" big Serbia	Rubick 's cube® Hungary
BDE 28	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*
BDE 47	14	18.2	<0.1*	258	66.8	<0.1*	<0.1*
BDE49	<0.1*	<0.1*	<0.1*	28.2	<0.1*	<0.1*	<0.1*
BDE 66	<0.1*	<0.1*	<0.1*	7.4	<0.1*	<0.1*	<0.1*
BDE 85	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*	<0.1*
BDE 99	<0.1*	<0.1*	<0.1*	335	89.8	55.7	<0.1*
BDE 100	<0.1*	<0.1*	22	135	58.2	107	1.6
comm. pentaBDE	14	18.2	22	763.6	214.8	162.7	1.6
BDE 153	620	847	1777	7182	1986	7949	<0.1*
BDE 154	55.90	50	189	753	234	777	5.26
BDE 183	2545	4432	11359	41831	10917	48319	3.45
BDE 196	561	791	1318	5918	1599	6341	<0.1*
BDE 197	616	1557	2984	11973	3175	12892	0.73
BDE 203	1277	3595	963	3664	1072	3989	<0.1*
BDE 206	1871	5979	3517	4826	6845	5537	<0.1*
BDE 207	1479	4265	2170	5356	4233	6111	<0.1*
comm. octaBDE	9024.9	21516	24277	81503	30061	91915	9.44
BDE 209 (decaBDE)	133949	152634	36255	36593	46691	35519	36.8
ΣPBDEs	142987.9	174168.2	60554	118859.6	76966.8	127596.7	47.8

*Limit of detection/quantification (LOQ)

Components of PentaBDE include the following congeners: BDE47, 49, 66, 85, 99, and 100.

Components of OctaBDE include the following congeners: BDE153, 154, 183, 196, 197, 203, 206, and 207.

DecaBDE includes BDE209.

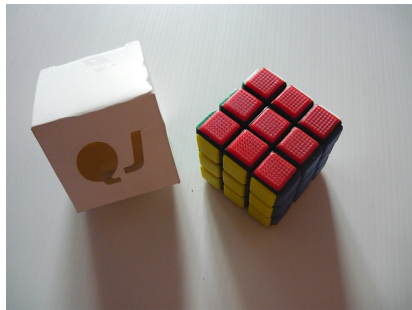
Photos



Picture 1: "Magic cube" sample, Belarus



Picture 2: "Toys" cube sample, Belarus



Picture 3: Cube "QJ" sample, China



Picture 4: Cube "Toys", small, Serbia



Picture 5: "IQ Magic Cube" sample, Serbia



Picture 6: Cube "Toys", big, Serbia



Picture 7: Rubick 's cube® sample, Hungary



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