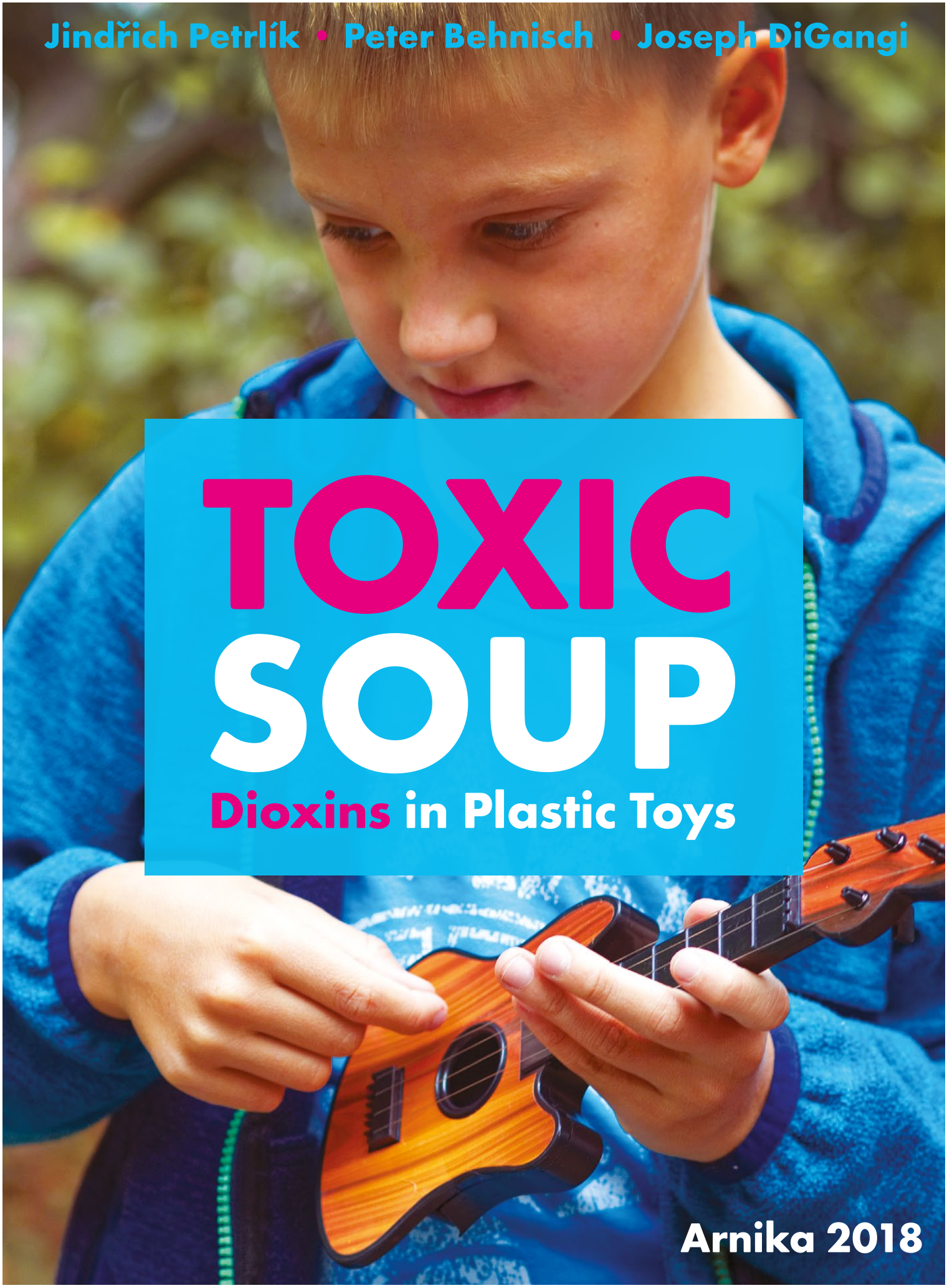
Jindřich Petrlík • Peter Behnisch • Joseph DiGangi



TOXIC SOUP

Dioxins in Plastic Toys

Arnika 2018

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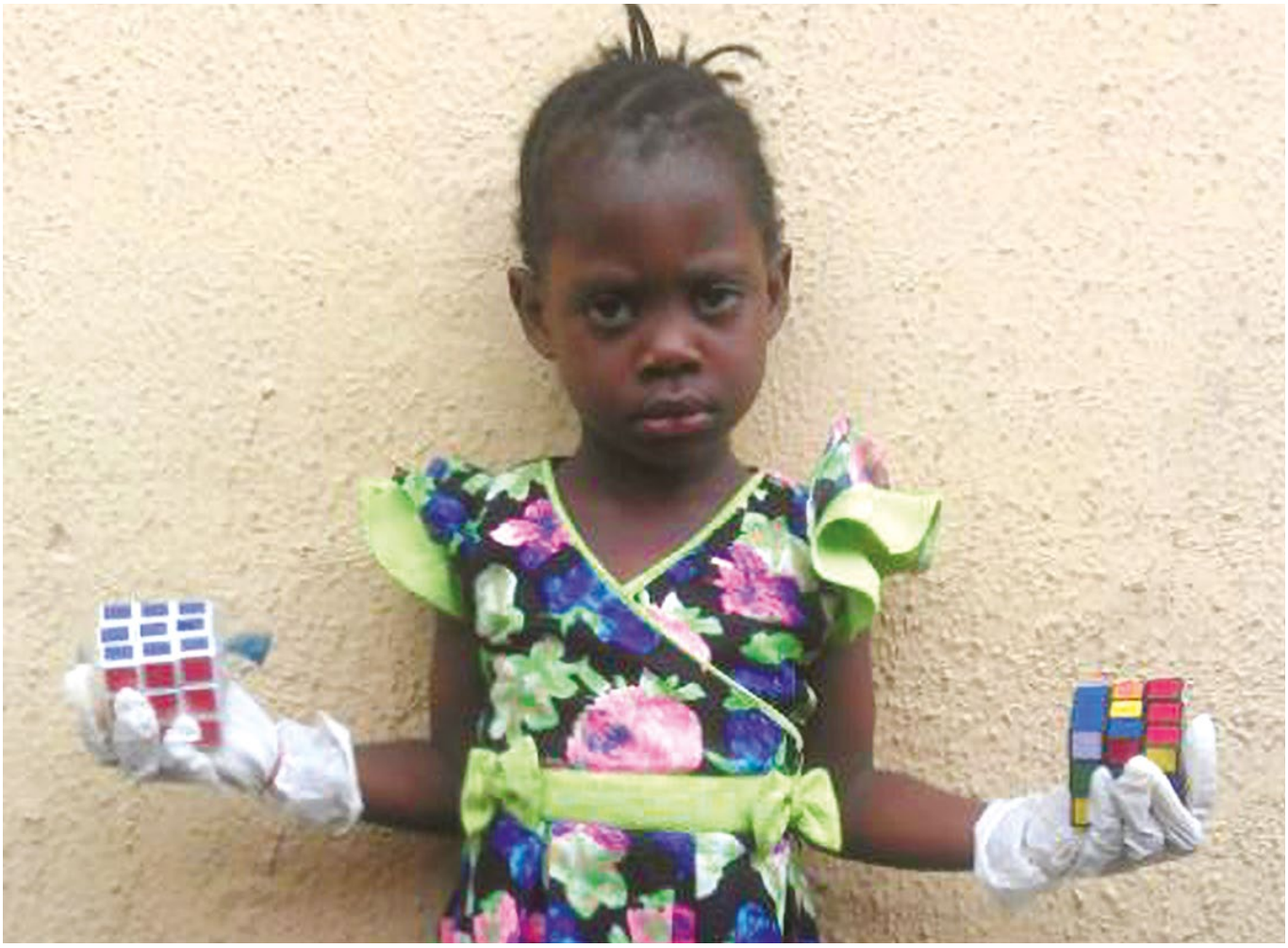
LIST OF ABBREVIATIONS

BDE – brominated diphenyl ether
BFRs – brominated flame retardants
GC-MS – gas chromatography – mass spectrometry
HBCD – hexabromocyclododecane
HpBDF – heptabromodibenzofuran
OBDF - octabromodibenzofuran
PBDD/Fs – polybrominated dibenzo-p-dioxins and dibenzofurans
PCBs – polychlorinated biphenyls
PCDD/Fs – polychlorinated dibenzo-p-dioxins and dibenzofurans
PBDEs – polybrominated diphenyl ethers
POPs – persistent organic pollutants
TBBPA – tetrabromobishpenol A (one of the brominated flame retardants)
TEF – toxic equivalency factor
TEQ – toxic equivalents
WEEE – waste electric and electronic equipment
WHO – World Health Organization

CONTENTS



EXECUTIVE SUMMARY

This study found significant levels of toxic brominated dioxins (PBDD/Fs) accompanying brominated flame retardants in nine samples of consumer products made from recycled plastics. PBDD/Fs are as toxic as the better-known chlorinated dioxins and furans (PCDD/Fs). In addition, PBDEs and PBDD/Fs are endocrine disrupting chemicals, which may impact the hormone levels in children through uptake from e.g. house dust and toys. They can, for example, affect brain development, damage the immune system and fetus, or induce carcinogenesis.

Analyzed consumer products were children's toys and a hair clip from 7 countries on 4 continents which had already been analyzed for PBDE content in previous studies. The significant contamination of children's products by PBDD/Fs ranged from 5,600 – 386,000 pg/g (56 – 3,800 pg WHO-TEQ/g). This is on the scale of PBDD/Fs found in a variety of hazardous wastes, including waste incineration bag filter ash, waste incineration bottom ash, residues of burned printed circuit boards, and in waste incineration ash after de novo synthesis. Dioxin-like activity measured by the DR CALUX method was found at similarly serious levels in these consumer products. This screening method allows monitoring of PBDD/Fs in a cost- and time-efficient manner.

A toy from Germany made of recycled plastic with levels of PBDEs that meet the current regulatory proposal (1,000 or 500[1] ppm PBDEs) contained 386,000 pg/g PBDD/Fs (3,800 pg WHO-TEQ/g). This signals that weak regulation of PBDEs can pose potential harms not only from PBDEs, but also from PBDD/Fs. In addition, the German regulation of PBDD/Fs does not address the most problematic PBDD/F congeners occurring as impurities in PBDEs.

TO PROVIDE MORE PROTECTIVE REGULATIONS, THE FOLLOWING POLICIES ARE NEEDED:

1. Not to allow the proposed 1,000 ppm limit for DecaBDE in recycled plastics, but rather establish a 10 ppm limit

2. A more stringent limit for the definition of POPs waste (Low POPs Content Level),[2] ideally to establish it at a level of 50 ppm for the sum of all regulated PBDEs

3. To withdraw the recycling exemptions for commercial PentaBDE and OctaBDE as they are currently established under the Stockholm Convention and registered in the EU and several other states.

4. Add PBDD/Fs to the Stockholm Convention for global reduction and elimination[3]

5. Improve the definition of electronic waste within the framework of the Basel Convention

PBDD/Fs in electronic-waste, electronic devices (such as TVs) and automotive plastics represent serious threats for recycling of plastics and consumer products made of it. The total amount of PBDD/Fs as an impurity in the produced volume of PBDEs has been recently estimated to be at least 1,000 t. This amount represents a very significant burden for human health and the environment considering the fact that PBDD/Fs are suggested to exhibit toxicity similar to PCDD/Fs.

There is also a need to stop the uncontrolled movement of electronic waste and BFR-contaminated plastics from automotive industry into developing and transition countries. Plastics from these waste flows were the likely origin of recycled plastics for the production of toys analyzed in this study.

1 This level was proposed by the European Parliament as an alternative to the current EU level of 1,000 ppm for Low POPs Content level.

2 This limit is called ”Low POPs Content“ level according to the definition in the Stockholm Convention, and its definition is established by the *technical* experts group under the Basel Convention in General Technical Guidelines for ESM of POPs waste. This level is defined in Annex IV to POPs Regulation No 850/2004 (POPs Regulation) in the EU.

3 Analysis guidelines for screening and chemical analysis of all toxic relevant PBDD/Fs, PBBs and PCDD/Fs in recycled plastic and products thereof could also help.

1. INTRODUCTION

Progress in scientific knowledge and efforts to protect consumers, as well as public pressure, are contributing to restrictions over toxic chemicals in consumer products. Mouthing toys for children, food contact materials, and kitchen utensils belong to products that are regulated in Europe. Nevertheless, there are huge legislative loopholes ignoring contamination of those critical items by a class of chemicals known as persistent organic pollutants (POPs). POPs are considered unmanageable due to their persistence, bioaccumulation, long-range transport and toxicity. The Stockholm Convention is a legally-binding global treaty that seeks to protect human health and the environment by reduction and elimination of POPs. The treaty's list of 28 substances includes brominated flame retardants (BFRs) commonly used in the plastic casings of computers, televisions, electronic office equipment, and many other electrical and electronic items.

Other representatives of the most toxic substances addressed by this treaty are polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs).[4] These substances are unintentional by-products of industrial processes and impurities accompanying PCBs, and other chemicals [2]. Brominated dioxins (PBDD/Fs), which are counterparts of PCDD/Fs, await global elimination (see the chapter 1.2).

The recycling of plastic casings of electric and electronic devices that contain POPs can contaminate new products with POPs and other toxic substances [3-10]. This study examines whether PBDD/Fs occur alongside PBDEs-contaminated toys and hair clips made of recycled plastics.

1.1 Previous studies on brominated flame retardants in plastic toys

Previous studies have found BFRs in electronic waste recycled into new plastic consumer products such as toys, hair accessories, kitchen utensils and many others. This includes substances prohibited by the Stockholm Convention

4 These two groups of chemicals are called "dioxins" in the brief.

[11-13], such as certain polybrominated diphenyl ethers (PBDEs)[4, 5, 9, 14-16] and hexabromocyclododecane (HBCD) [7, 8, 17].

"Toxic Loophole" study [5] confirmed that toys, hair accessories and kitchen utensils contaminated with high levels of PBDEs can be found on the market in 19 European countries. The study noted that regulatory loopholes permit the recycling of materials containing PentaBDE and OctaBDE, and that standards for toxic substances in recycled products are much weaker than for new products . The study also found a variety of unregulated toxic BFRs in consumer products.

Unintentionally-produced toxic substances are also associated with BFRs in electronic waste, but have not been previously investigated in consumer products on the market. These substances include brominated dioxins and furans (PBDD/Fs) and polybrominated biphenyls (PBBs). In this study, we investigated whether eight toys and a hair clip made of recycled plastic contain PBDDs/F. The samples were analyzed for their overall dioxin-like activity by cell-based reporter gene assay DR CALUX [18], and then content of PBDD/Fs by instrumental chemical GC-HRMS analysis.

1.2 Brief introduction of brominated dioxins

The term "brominated dioxins" includes two groups of chemicals called polybrominated dibenzo-p-dioxins (PBDDs) and polybrominated dibenzofurans (PBDFs). Both groups of chemicals have been known to be potential by-products of commercial PBDE mixtures since 1986 [19]. This is similar to the situation with chlorinated dioxins, which have been observed as impurities in PCBs. PBDFs have also found to be formed by sunlight exposure during normal use, as well as during disposal/recycling processes of flame-retarded consumer products [20].

Chlorinated dioxins (PCDD/Fs) are known to be extremely toxic. Numerous epidemiologic studies have revealed a variety of human health effects linked to chlorinated dioxin exposure including cardiovascular disease, diabetes, cancer, porphyria, endometriosis, early menopause, alteration of testosterone and thyroid hormones, and altered immune system response among others [21, 22]. Chlorinated dioxins became known to the public in the 1970s as a result of their contamination in Agent Orange, a defoliant pesticide mixture sprayed by the US during the Vietnam war.[5] The production of 2,4,5 T pesticide as basic ingredient for Agent Orange left one of the most seriously contaminated sites in Europe [24-26] and sick workers with many symptoms of exposure to most toxic of dioxin congeners 2,3,7,8-TCDD [27, 28]. PBDDs/F have been found to exhibit similar toxicity and health effects as their chlorinated analogues (PCDD/Fs) [1, 29-32]. They can for example affect brain development, damage the immune system and fetus or induce carcinogenesis [1].

"Both groups of compounds show similar effects, such as induction of aryl hydrocarbon hydroxylase (AHH)/EROD activity, and toxicity, such as induction of wasting syndrome, thymic atrophy, and liver toxicity." [31].

In general, brominated dioxins are less regulated than chlorinated dioxins. For example, PBDD/Fs are not currently listed under the Stockholm Convention [11], although there is clear evidence that they contain very similar properties to PCDD/Fs, which have been listed in Annex C of the Convention since its origin in 2001. In 2010, the Stockholm Convention POPs Review Committee recommended further assessment of PBDD/Fs including, *"releases from smelters and other thermal recovery technologies, including secondary metal industries, cement kilns and feedstock recycling technologies."* [33].

Because brominated dioxins tend to be less regulated, there is less data about their presence in the environment. There is also very little information about their presence in consumer products and food, where they can have direct impacts on human health, including in vulnerable groups such as children and women of childbearing age.

5 According to estimates provided by the Government of Vietnam, 400,000 people were killed or maimed by the pesticide; 500,000 children were born with birth defects ranging from retardation to spina bifida; and a further two million people have suffered cancers or other illnesses, which can be also related to dioxins as impurities in the Agent Orange mixture. It is estimated that in total, the equivalent of at least 366 kilograms of pure dioxin were dropped. 23. York, G. and H. Mick. *Last ghost' of the Vietnam War*. 2008 April 27, 2018 [cited 2018 19-11-2018]; Available from: https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all.

2. TESTING OF BROMINATED DIOXINS – SAMPLES AND METHODS

2.1 Selection of samples for the analyses

Eight toys and one hair clip made from black recycled plastic were selected for analysis. These samples had already been analyzed for PBDEs from previously researched sets of samples. We selected those which had a total content of PBDEs of more than 500 ppm, and above 250 ppm of DecaBDE. We selected two samples from Nigeria (NIG_06 and NIG_11), one sample from India (IND_11) and one sample from Argentina (ARG_04), among the toys collected in 2016 from Africa, Asia and Latin America and analyzed in 2017 [34]. We also selected two samples (JI_11 and SIX_02) from the collection analyzed in the Czech project and report released in 2017 [6, 17]. Three more samples were selected from those presented in the recent Toxic Loophole report [5] from France (FR-T-3), Germany (D-T07), and Portugal (PT-T-10a). The toys and the hair clip samples analyzed for this report are listed in Table 1.

2.2 Analytical methods

Selected samples were delivered either personally or via special post delivery services to the laboratories in Netherlands for DR CALUX assay (BioDetection Systems - BDS, Science Park, University of Amsterdam) and Germany for GC-HRMS[6] analysis of PBDD/Fs (MAS, Muenster). Analyses for brominated flame retardants were done previously in a laboratory at the University of Chemistry and Technology, Prague, Czechia, and are described in previous reports [5, 34].

2.2.1 Sample preparations

Firstly, the black plastic parts were separated from all other parts (such as the colored plastic or metal parts) of the samples and cut into small pieces. Next, between 5 and 30 g plastic material was put into a glass bottle (Schott) and 30 to 60 ml hexane were added so that all plastic was covered by hexane. The sample was then further broken down in small particles and homogenized with an Ultraturrax homogenization apparatus. Then the

6 Gas Chromatography – High Resolution Mass Spectrometry

dioxin-like compounds were extracted for 24 hours in hexane. After 24 hours the hexane solvents were collected, and two times more extracted with newly added 30 to 60 ml hexane. Then all hexane extraction fractions were collected together and further evaporated to ca. 1 ml hexane (Dionex SE500). This final hexane extract was then twice (at a minimum) cleaned-up by sulfuric acid silica gel clean-up columns (focusing on sulfuric acid oxidative stable dioxin-like POPs). Finally, part of the hexane solvent extracts (3 to 15 g in 25 to 100 µl hexane) were used for the chemical GC/MS analysis and the other part was evaporated and exchanged to DMSO (50 to 235 µl) for the DR CALUX bioanalysis. All DR CALUX analyses were performed on the basis of the ISO/IEC 17025:2005 accredited BDS method for the analysis of dioxins and dioxin-like compounds in solid materials (such as soil and fly-ash).

2.2.2 DR CALUX analyses

Briefly, H4IIE cells (70–95 % confluence) stably transfected with an aryl hydrocarbon receptor (AhR)-controlled luciferase reporter gene construct were cultured in α-minimum essential medium (MEM) supplemented with 10 % (v/v) fetal calf serum (FCS) under standard conditions (37 °C, 5 % carbon dioxide (CO2), 100 % humidity). Cells were exposed in triplicate on 96-well microtiter plates containing the standard 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) calibration range, a DMSO blank (99.9 %) and samples. Following a 24-h incubation period, cells were lysed. Finally, 100 µL of luciferin-containing solution was added, and the luminescence was measured using a luminometer (Centro XS3, Berthold Technologies, Germany). The 2,3,7,8-TCDD calibration curve, generated from DR CALUX readings, resulted from a serial dilution of 2,3,7,8-TCDD standards (0.3, 0.6, 1.0, 2.0 and 3.0 pM per well). The amount of 2,3,7,8-TCDD bioanalytical equivalents (TEQs) was quantified by comparison of the response in the test sample with the calibration curve for 2,3,7,8-TCDD [35]

2.2.3 GC-HRMS analysis for PBDD/Fs

For chemical PBDD/F analysis, the extract portions provided by BDS were taken up with fresh hexane and adjusted to a defined volume. Aliquots of these solutions were fortified with eleven $^{13}C_{12}$-labeled PBDD/F internal standards and further purified by several liquid chromatographic clean-up steps. Prior to the HRGC/HRMS analysis, additional $^{13}C_{12}$-labeled PBDD/F standards were added to the PBDD/F fractions as recovery standards (all standards from CIL, Round Rock, TX 78665, U.S.A.).

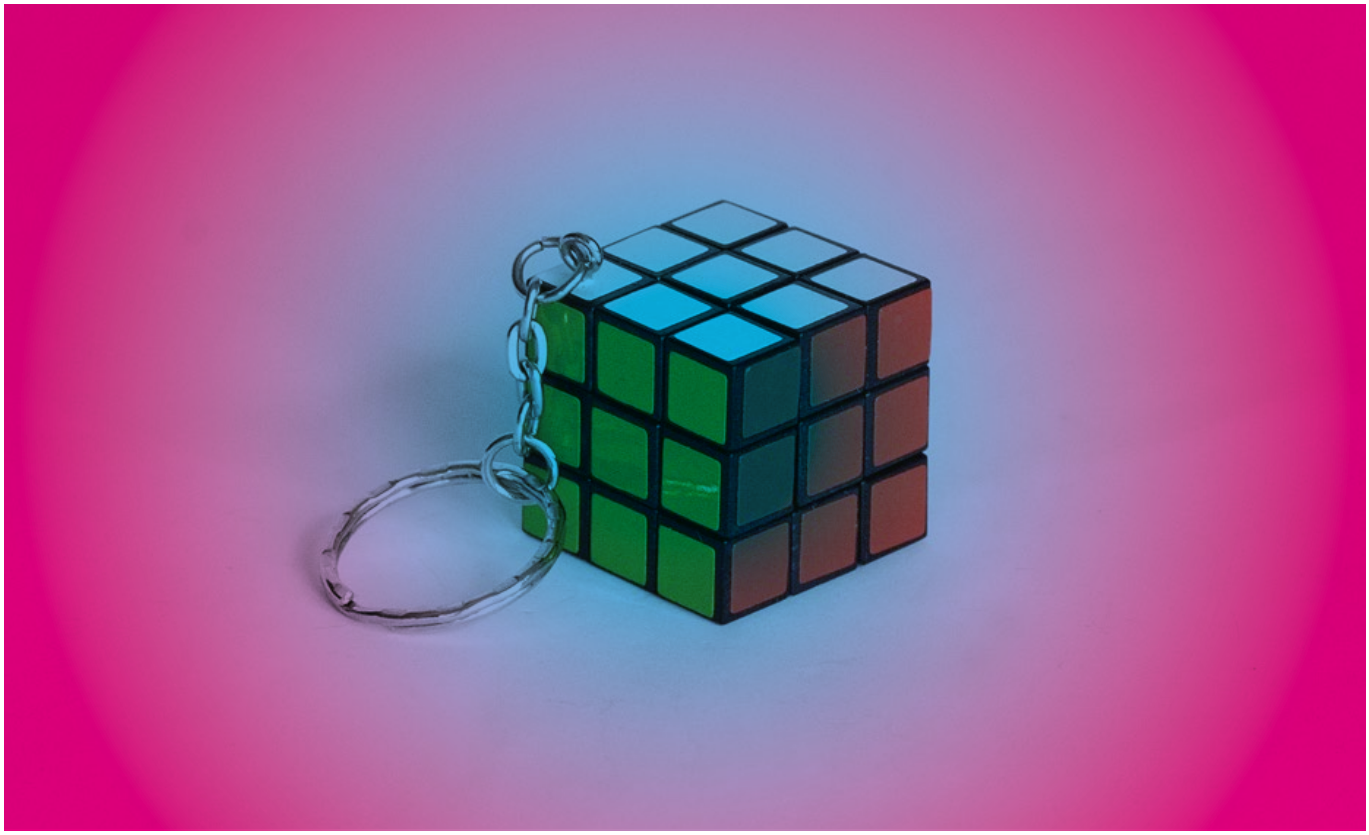
A capillary gas chromatograph (Thermo Scientific GC-Ultra), coupled with a high-resolution mass spectrometer (Thermo Scientific MAT 95XP HRMS), was used for instrumental PBDD/F analysis. The GC was equipped with a PTV injection port (Programmable Temperature Vaporizer) and a 30 m DB-5MS capillary column (Agilent J&W GC column, 0,25 mm inner diameter and 0,1 µm film thickness). The HRMS was operated in the SIM–Mode, monitoring selected masses of the molecular ion cluster. Furthermore, masses of the molecular ions of PBDEs were monitored to check for potential co-elution of PBDEs with PBDFs, which can lead to false positive results. Native PBDD/F congeners were quantified via the internal $^{13}C_{12}$-labeled PBDD/F standards (Isotope dilution and method of the internal standard).

All PBDD/F analyses were performed on the basis of the DIN EN ISO/IEC 17025:2005 accredited MAS test method MAS_PA002:2013-10 for the analysis of PCDD/Fs, PBDD/Fs and PCBs in solid matter.

Toxic Equivalency Factors (TEFs) for polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) were used for calculation of WHO-TEQ levels of PBDD/Fs in samples, as suggested by World Health Organization (WHO) experts [36, 37]

Table 1: Overview of the samples selected for the analyses in this report

Sample	Country of Purchase	Description	Year of sampling
ARG_04	Argentina	Rubik's-like cube	2016
JI_11	Czechia	Cube with picture of a mole	2017
SIX_02	Czechia	Hair clip	2017
FR-T-3	France	Toy revolver gun	2018
D-107	Germany	Key fob with magic cube	2018
IND_11	India	Rubik's-like cube	2016
NIG_06	Nigeria	Rubik's-like cube	2016
NIG_11	Nigeria	Rubik's-like cube	2016
PT-T-10a	Portugal	Small guitar	2018



3. RESULTS AND DISCUSSION

3.1 Results

Dioxin-like activity and PBDD/Fs were found in all nine consumer products (Table 2). Levels of DR CALUX dioxin-like activity ranged between 210 to 17,000 pg TEQ/g. Note that 1,000 pg TEQ/g can be considered as equivalent to the proposed hazardous waste limit[7] for chlorinated dioxins in the Stockholm Convention [38, 39]. Consumer products exceeding this proposed chlorinated dioxin hazardous waste limit included a toy cube from Czechia and Rubik's-like cubes from Argentina, India, and Nigeria. In addition, dioxin-like activity found in many of these consumer products is similar to levels found in waste incineration bag filter ash, which can contain 370 – 25,000 pg TEQ/g as measured by DR CALUX [40]. Toy samples with levels of dioxin-like activity similar to waste incineration bag filter ash included a cube toy from Czechia, toy revolver from France, key fob from Germany and Rubik's-like cubes from Argentina, India, and Nigeria.

Levels of 17 toxic PBDD/F congeners ranged from 5,600 – 386,000 pg/g and 56 – 3,800 pg WHO-TEQ/g[8] respectively. These levels are similar to and in some cases higher than PBDD/F levels measured in waste incineration bottom ash from Taiwan (1,600 – 31,000 pg/g

7 We mean the Low POPs Content Level, which defines whether waste is considered as POPs waste or not. POPs waste should be according to Article 6 of the Stockholm Convention: „Disposed of in such a way that the persistent organic pollutant content is destroyed or irreversibly transformed so that they do not exhibit the characteristics of persistent organic pollutants or otherwise disposed of in an environmentally sound manner when destruction or irreversible transformation does not represent the environmentally preferable option or the persistent organic pollutant content is low," and at the same time it is *„(iii) Not permitted to be subjected to disposal operations that may lead to recovery, recycling, reclamation, direct reuse or alternative uses of persistent organic pollutants; and (iv) Not transported across international boundaries without taking into account relevant international rules, standards and guidelines;"* 11. Stockholm Convention, *Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes.* 2010: Geneva. p. 64.

8 PBDD/Fs are measured in absolute weight values for its 17 toxic congeners and expressed then be recalculation according their dioxin-toxicity by using WHO toxic equivalent factors (TEFs). It was suggested to use the same TEF factors as for PCDD/Fs by WHO expert panel. We express here both absolute level of all 17 congeners as well as its equivalent in WHO-TEQ, as different studies use either one or another way of expresion of PBDD/Fs level in certain sample.

and 28 – 61 pg TEQ/g) [41]. In addition, levels observed in some toys exceeded those found in residues of pyrolyzed printed circuit boards (231–490 pg I-TEQ/g) [42] and in waste incineration ash (after de novo synthesis) (7,200 pg/g PBDD/Fs) [43].

Taken together, the data showed that the sampled children's products contained significant levels of PBDD/Fs. The measured PBDD/F levels were on the scale found in a variety of hazardous wastes, including waste incineration bag filter ash, waste incineration bottom ash, residues of burned printed circuit boards, and in waste incineration ash after de novo synthesis. Half of the products exceeded the proposed chlorinated dioxin hazardous waste limit.

3.2 Previous studies finding brominated dioxins in plastics

PBDD/Fs have been found in plastics treated with a variety of BFRs [44, 45]. It is well documented that PBDD/Fs are formed as unintentionally produced POPs during production of different flame retardants [46, 47]. They can also be formed during further reprocessing when plastics waste containing BFRs experience thermal stress; i.e. is shredded, melted and shaped into new products [48].

Hanari et al. [47] reported 30,000,000 - 50,000,000 pg/g PBDD/Fs in commercial DecaBDE and 10,000,000 - 19,000,000 pg/g in commercial OctaBDE, flame retarding chemicals applied to plastic casing of electric and electronic goods. These levels are higher than the later results from Ren et al. [46], where PBDD/F levels in commercial DecaBDE ranged between 3,400,000 – 13,600,000 pg/g with a mean concentration of 7,800,000 pg/g. The lowest PBDD/F levels were detected in the commercial PentaBDE at 260,000 pg/g [46]. PentaBDE was used as flame retardant in furniture and transport vehicles upholstery.

Sakai et al. [49] measured PBDD/F concentrations in discarded television sets and personal computers in Japan and found an average concentration of 280,000,000 pg/g in waste television cabinets and from 38,000,000 to 380,000,000 pg/g in printed circuit boards. Television casings contained PBDD/F concentrations of 23,000,000 pg/g, with a predominant proportion of OBDF (19,000,000 pg/g) in later research in Japan [20]. Takigami et al. [50] also showed that these high PBDD/F levels in TV casings lead to increased levels in house dust, which is a potential source of contamination for toddlers.

PBDD/Fs, consisting mainly of PBDFs, were detected in plastics at electronic waste sites in Nigeria, with a median (mean) concentration of 18,000,000 pg /g (41,000,000 pg/g) [44].

Levels of PBDD/Fs found in toys in this study are orders of magnitude lower than those in electronic waste from Nigeria. However, the use of contaminated plastic for toys and personal care products is a direct route of consumer exposure not only to PBDEs, but also to PBDD/Fs.

PBDD/Fs are also reported in flue gases and solid waste residues from waste incineration or pyrolysis of wastes containing BFRs [48, 51-53], and in waste electric and electronic equipment (WEEE) in particular [54]. That demonstrates the additional consequences of

Table 2: Results of analyses for PBDD/Fs, DR CALUX, PBDEs, HBCD and TBBPA

Country / Sample	Type	PBDD/Fs (pg TEQ/g)[1]	PBDD/Fs (pg/g)[1]	DR CALUX (pg TEQ/g)	PBDEs (ug/g)[2]	HBCD (ug/g)	TBBPA (ug/g)
Argentina ARG_04	Rubik's-like cube	727	170,754	1,200	708	1	na[3]
Czechia JI_11	Cube	2,159	224,097	17,000	2,614	91	na[3]
Czechia SIX_02	Hairclip	60	5,593	210	1,623	8	na[3]
France FR-T-3	Toy revolver	2,058	219,385	520	1,077	1	314
Germany D-TO7	Key fob	3,821	385,856	820	511	2	307
India IND_11	Rubik's-like cube	690	87,416	1,300	593	2	na[3]
Nigeria NIG_06	Rubik's-like cube	860	203,362	4,800	1,780	9	na[3]
Nigeria NIG_11	Rubik's-like cube	56	6,694	370	1,218	8	na[3]
Portugal PT-T-10a	Toy small guitar	1,137	98,745	270	3,318	2	37

1 Zero was used for PBDD/F congeners, declared in the analytical protocol by MAS as not detected above the limits of quantification.

2 Reflects the sum of congeners contained in commercial PentaBDE, OctaBDE and DecaBDE (BDE 28, 47, 49, 66, 85, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 209).

3 Not analyzed

plastics treated with BFRs and extends the exposure to PBDD/Fs to the next stage of recycled plastics' lifecycles when they become waste for final disposal.

3.2.1 Photodegradation of DecaBDE

Photodegradation of technical DecaBDE mixture incorporated into high-impact polystyrene and television casings under natural sunlight conditions yielded PBDFs [20, 55]. One study [20] has shown that exposure of high-impact polystyrene containing DecaBDE to sunlight increased concentrations of PBDFs by 40 - fold in 7 days. Similarly, exposure of television casings to sunlight for more than 224 days increased PBDFs concentrations by 20 - fold from the initial concentration of 23,000,000 pg/g. 52 OBDF was the predominant congener formed, accounting for 60 – 80% of the total PBDFs concentrations in television casings [20]. Photolysis occurs at a more rapid rate for PBDD/Fs than for PCDD/Fs [55-57]. That means that even the presence of plastics containing high levels of DecaBDE can increase exposure to PBDFs through their increased levels in the room.

3.3 Origin of the brominated dioxins in plastic toys

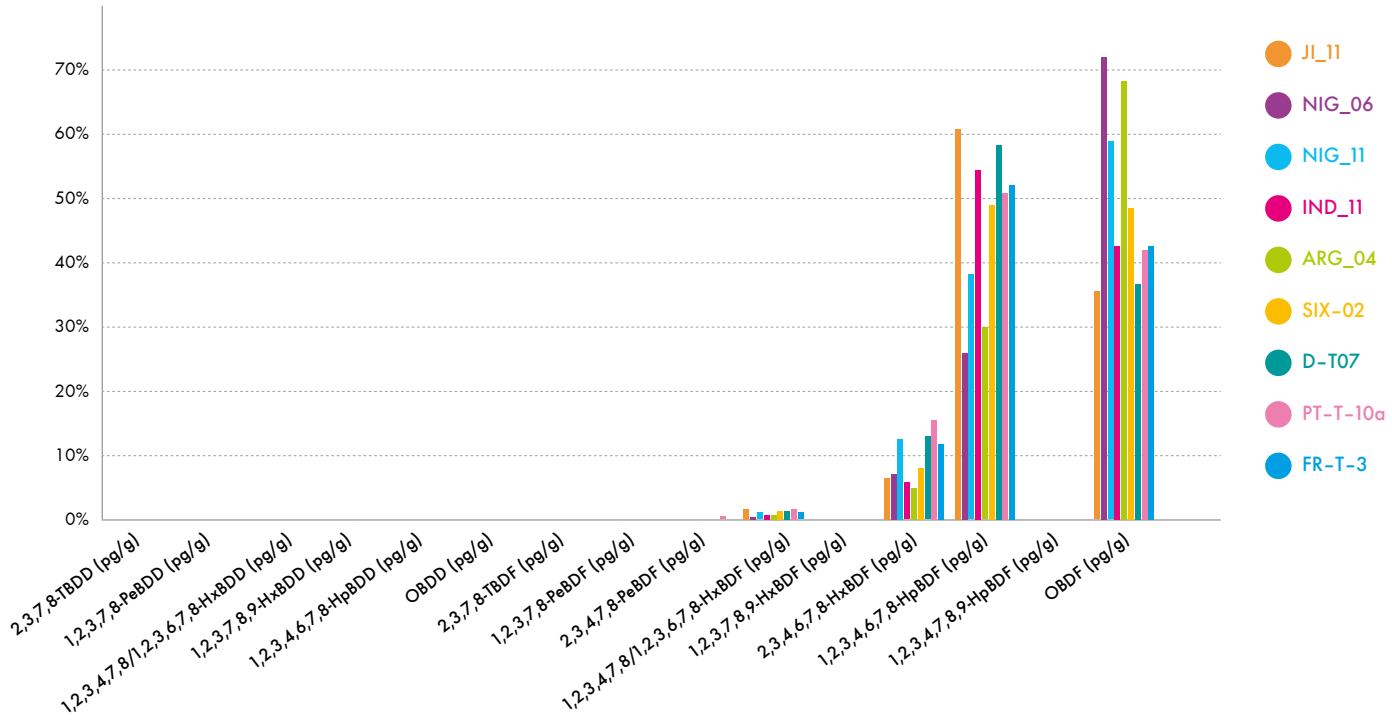
Similar to chlorinated dioxins (PCDD/Fs), PBDD/Fs have a specific congener pattern that can reveal their most likely source. The basic pattern of PBDD/Fs in the samples analyzed in this study is shown in Figure 1. Congeners 1,2,3,4,6,7,8 HpBDF and OBDF were most abundant in all samples. In individual samples, there is a difference in ratio between these two congeners and in the percentage of less brominated PBDF congeners.

The pattern of PBDD/Fs congeners in these samples resembles the pattern found as impurities in commercial DecaBDE mixtures (see Figure 2) [46].

Some less brominated PBDF congeners contributed significantly to the overall toxicity of the PBDD/F mixture because their toxicity factor is considered to be higher (see Figure 3). The toxic equivalency factors (TEFs) for PCDD/Fs are used for an evaluation of the toxicity of PBDD/Fs, as suggested by an international panel of WHO experts [36]. Figures 4 and 5 demonstrate slight differences between samples from Portugal (PT-T-10a) and Nigeria (NIG-06), although the basic balance between contribution of different PBDD/F congeners is somewhat similar. The patterns of these two samples represent the scale of differences within the analyzed collection of recycled plastics in this study.

It is likely that the PBDD/Fs observed in consumer products in this study are due to the impurities in the original e-waste plastic treated with DecaBDE. This indicates that allowing the recycling of plastics containing DecaBDE also allows significant amounts of PBDD/Fs to be recycled into new products as well [5, 9, 34]. Part of the PBDD/Fs could also occur as a result of thermal stress during the recycling process [48].

Figure 1: PBDD/F congeners in nine consumer products (percentage of total absolute levels of 17 toxic congeners).



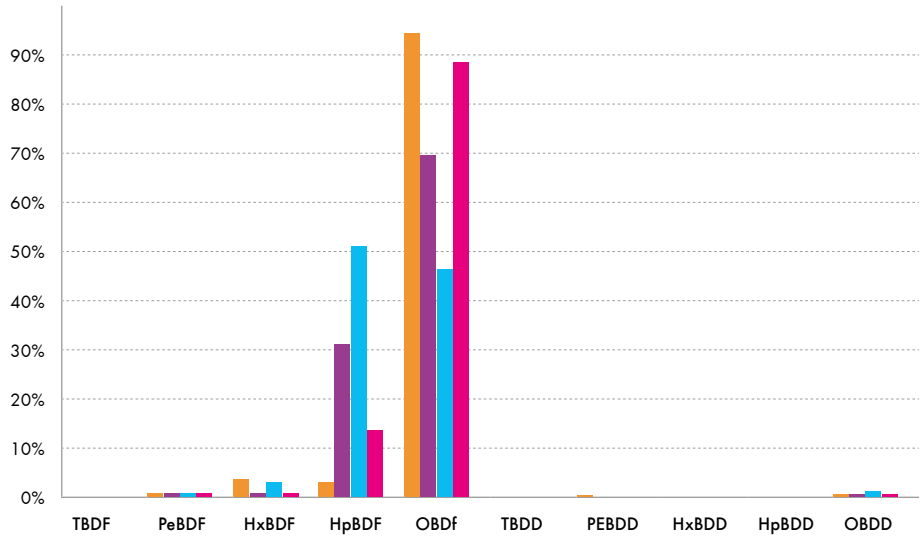
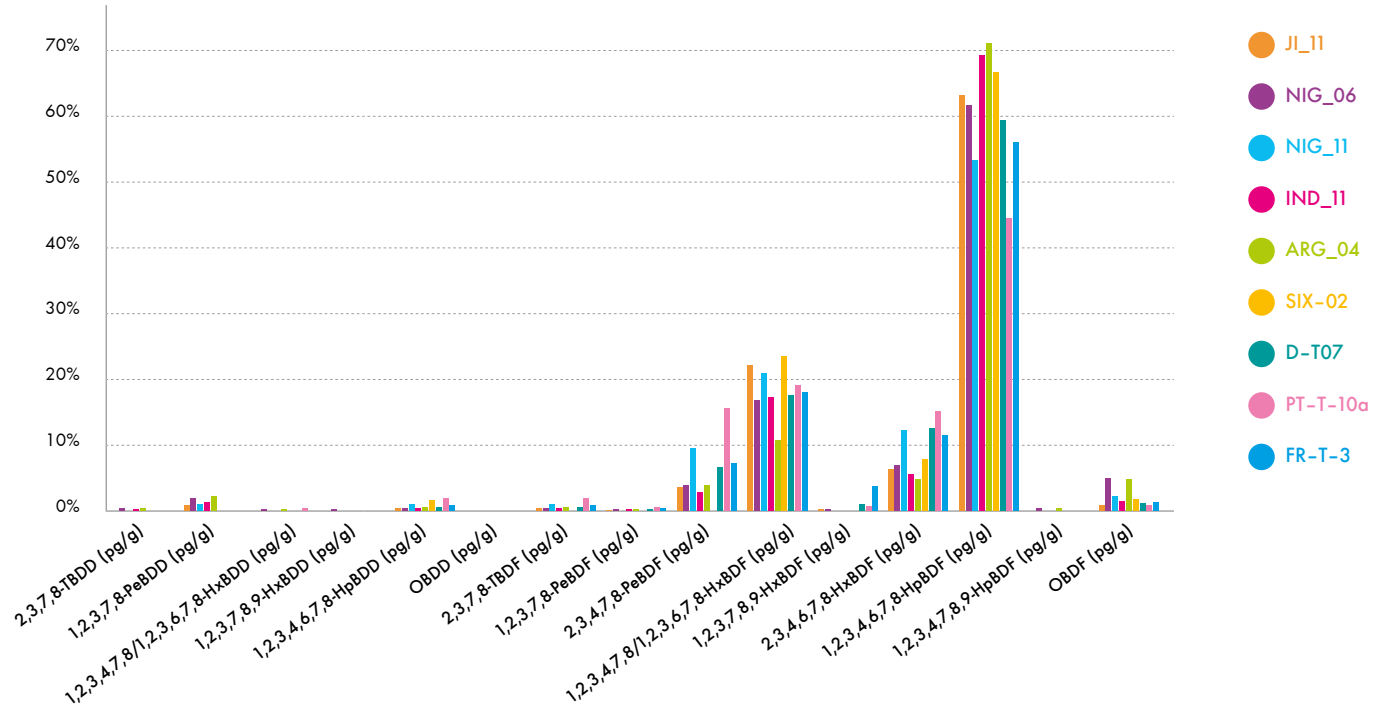
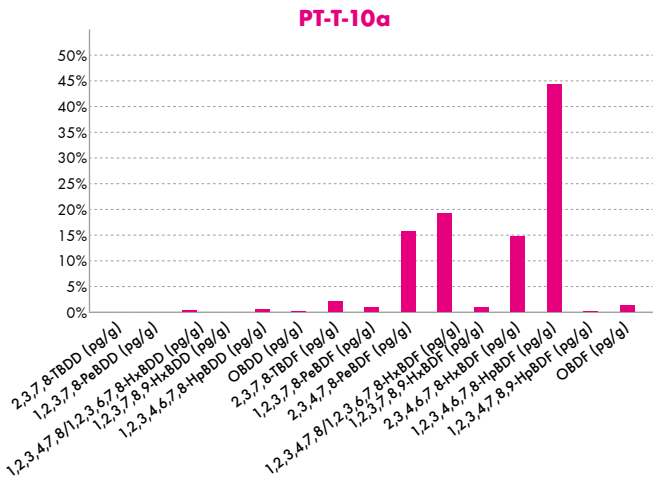


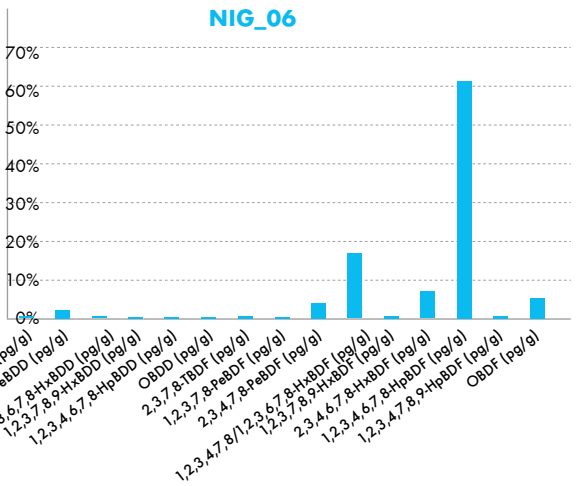
Figure 2: Relative proportions of individual PBDD/F homologues in commercial DecaBDE. Source: [46]

Figure 3: PBDD/F congener pattern in samples from consumer products in this study expressed as the congeners' contribution to levels of TEQ (percentage of individual congeners on WHO-TEQ level of samples).



Figures 4 and 5: PBDD/F congener pattern in samples from Portugal (PT-T-10a) and Nigeria (NIG_06) expressed as the congeners' contribution to WHO-TEQ levels.





3.4 Legislative tools to prevent exposure to brominated dioxins

Apparently, the only regulation prohibiting brominated dioxins in consumer products is a German law which limits certain PBDD/F congeners in substances and mixtures [58]. The regulation with such limits has been in force since 1996 [59]. At that time, only a few calibration standards for PBDD/Fs were available, and therefore only 8 from at least 17 highly toxic PBDD/Fs could be regulated. Therefore, only one of the nine samples, a toy bought in Germany, exceeded the limits set in that regulation. However, if also (like in the case of PCDD/Fs) all other toxic PBDD/Fs congeners would be added, most of the samples would exceed these limits. There is no other legislative measure to prevent or stop consumer exposure to significant levels of PBDD/Fs in products, children's toys, daily care products for women or kitchen utensils.

Some indirect regulation of PBDD/Fs in consumer products could be achieved by elimination of BFRs in products. Eliminating BFRs in products could also help eliminate the PBDD/F impurities that accompany them. However, the existing EU legislation does not prevent the occurrence of BFRs or PBDD/Fs in children's products, as demonstrated in Table 2.

The data in this study also reveal the inadequacy of a proposal to allow 1,000 ppm of DecaBDE in recycled plastics and maintain recycling exemptions for PentaBDE and OctaBDE, setting the same limit for recycled products at a level of 1,000 ppm. For example, a toy obtained in Germany would comply with these proposed limits because it had PBDEs at a level of 500 ppm; however, it contained 386,000 pg/g PBDD/Fs (3,800 pg WHO-TEQ/g) – a level higher than PBDD/Fs in hazardous waste.

Table 3: Comparison of PBDD/Fs in consumer products with existing legislative limit values in products (and waste)

Country / Sample or Limit	Type	PBDD/Fs (pg TEQ/g)	PBDD/Fs (pg/g)[3]	PBDEs (µg/g)	Sum of PBDD/Fs selected congeners (in pg/g)[2]
EU Proposed PBDE waste limit[1]				1,000	
German limit sum: 2378-TBDD, 2,378-TBDF, 12378-PeBDD, 23478-PeBDF (in pg/g)[2]					1,000
German limit sum: Sum: 2378-TBDD, 2,378-TBDF, 12378-PeBDD, 23478-PeBDF, 123478-HxBDD, 123678-HxBDD, 123789-HxBDD, 12378-PeBDF (in pg/g)[2]					5,000
Argentina ARG_04	Rubik's-like cube	727	170,754	708	160
Czechia JI_11	Cube	2,159	224,097	**2,614**	410
Czechia SIX_02	Hairclip	60	5,593	**1,623**	0
France FR-T-3	Toy revolver	2,058	219,385	**1,077**	711
Germany D-TO7	Key fob	3,821	385,856	511	**1090**
India IND_11	Rubik's-like cube	690	87,416	593	110
Nigeria NIG_06	Rubik's-like cube	860	203,362	**1,780**	190
Nigeria NIG_11	Rubik's-like cube	56	6,694	**1,218**	20
Portugal PT-T-10a	Toy small guitar	1,137	98,745	**3,318**	823

1 The EU proposal submitted as the option during debate in the European Parliament included a limit value of 1,000 ppm for all PBDE congeners listed under the Stockholm Convention as limit for the definition of POPs waste. Thus, the EU could also apply this limit to recycled plastic [60].

2 German Federal Regulation [58]

3 Zero was counted for PBDD/F congeners marked by the MAS laboratory as not detected at levels above the limits of quantification (LOQ) in the protocol from GC-HRMS analysis for PBDD/Fs.

A comparison of the measured levels of both PBDD/Fs and PBDEs in consumer products analyzed in this study with currently available (or proposed) limit values in different legislations is shown in Table 3. As noted above, one sample exceeded the German legislation limiting PBDD/Fs in consumer products. Note that it would be quite expensive and labor intensive to implement the German regulation for PBDD/Fs in consumer products due to its requirement for GC-HRMS analysis. This would make it prohibitively expensive in developing and transition countries. More importantly, the German regulation will not prevent contamination of consumer products by significant levels of less-but-still-toxic congeners of PBDFs, as demonstrated in this study (see Table 3). The HpBDF and OBDF congeners, which are not on the list of regulated polybrominated dibenzofurans in the German regulation, are the most abundant impurities in commercial DecaBDE. Introduction of stricter limits for PBDEs in products seems to be a more straight-forward solution, which could address toxic contamination by PBDD/Fs at the same time.

It is also urgently needed to work on world-wide acceptance of such low-cost screening methods for analysis of polyhalogenated dioxins and biphenyls such as DR CALUX analysis used in this study.[9]

The current weak limit for the definition of POPs waste (Low POPs Content Level)[10] at 1,000 ppm for PBDEs will result in new products made of recycled plastic contaminated with PBDEs and PBDD/Fs. IPEN, in agreement with other civil society organizations, suggested lowering the PBDE low POPs content limit to 50 ppm [38]. The results of this study support this suggestion, as levels of even a few hundred ppm of PBDEs are accompanied by toxic PBDD/Fs at significant levels.

Note that all samples met current limits for commercial PentaBDE and OctaBDE mixtures that are regulated in the EU POPs regulation and the Stockholm Convention as TetraBDE, PentaBDE, HexaBDE and HeptaBDE (data not shown).

3.5 Scope of the problem

This study examined only a small number of samples of consumer products made of recycled plastic containing PBDEs and other BFRs. An effort was made to select a representative sample from different countries, and within a certain range of content of PBDEs potentially accompanied by PBDD/Fs as impurities [46]. All samples displayed remarkable levels of dioxin-like toxicity and PBDD/Fs. Levels of PBDD/Fs exceeded 500 pg WHO-TEQ/g in seven out of the nine measured samples. This raises the question about the potential scope of the problem that could be encountered in a more comprehensive sampling of plastic consumer products.

The results of this study are consistent with the conclusion of Hanari et al. [47] that „*… the ongoing use of PBDE-containing products and the continued use of Deca-BDE together suggest that human and environmental exposures to PBDFs are likely to increase in the future.*“

No detailed estimates of the scale of PBDD/Fs content in consumer products has been made. However, *“It was estimated that the 237,000 t of CRT casings stockpiled in Nigeria contain between 2 and 8 t of PBDD/Fs. The total PBDD/F contamination in polymers arising from total historic PBDE production/use is estimated in the order of 1,000 t.”* [44]. The problem of PBDD/Fs as contaminants in plastic and their potential route into recycled plastics is potentially extremely large. This also raises concerns about the overall control of environmental contamination by PBDD/Fs. An important component of PBDD/Fs reduction and elimination would be their listing under Annex C to the Stockholm Convention, in addition to their regulation through currently listed POPs.

9 DR CALUX is accepted so far under: European Commission Regulation 2017; Dutch National Standard Specie 07, 2006; Japanese International Standard 463, 2009; US-EPA standard 4435, 2008; Chinese standard for Solid waste—Screening of PCDD/Fs—Chemical activated luciferase expression, 2018.

10 This limit is called the ”Low POPs Content“ level according to the definition in the Stockholm Convention, and its definition is established by the technical experts group under the Basel Convention in the General Technical Guidelines for ESM of POPs waste. This level is defined in Annex IV to POPs Regulation No 850/2004 (POPs Regulation) in the EU. 60. European Parliament and Council, *Regulation (EC) No 850/2004 of the European Parliament and of the Council of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC (OJ L 158 30.4.2004, p. 7. Amended by: Council Regulation (EC) No 1195/2006 of 18 July 2006; Council Regulation (EC) No 172/2007 of 16 February 2007; Commission Regulation (EC) No 323/2007 of 26 March 2007; Regulation (EC) No 219/2009 of the European Parliament and of the Council of 11 March 2009; Commission Regulation (EC) No 304/2009 of 14 April 2009; Commission Regulation (EU) No 756/2010 of 24 August 2010; Commission Regulation (EU) No 757/2010 of 24 August 2010;Commission Regulation (EU) No 519/2012 of 19 June 2012;Commission Regulation (EU) No 1342/2014 of 17 December 2014;Commission Regulation (EU) 2015/2030 of 13 November 2015; Commission Regulation (EU) 2016/293 of 1 March 2016; Commission Regulation (EU) 2016/460 of 30 March 2016, and Corrected by: Corrigendum, OJ L 229, 29.6.2004, p. 5 (850/2004).* European Parliament and Council, Editor. 2016: Brussels. p. 1-28, 61. Basel Convention, *General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants*, in *Technical Guidelines*. 2017: Geneva.

4. CONCLUSIONS AND RECOMMENDATIONS

The data showed that the sampled children's toys and hair clip contained significant levels of PBDD/Fs ranging from 5,600 – 386,000 pg/g and 56 – 3,800 pg WHO-TEQ/g. The measured PBDD/F levels were on the scale found in a variety of hazardous wastes, including waste incineration bag filter ash, waste incineration bottom ash, residues of burned printed circuit boards, and in waste incineration ash after de novo synthesis. Half of the products exceeded the proposed chlorinated dioxin hazardous waste limit.

Levels of PBDD/Fs found in children's products in this study significantly lower than those in electronic waste from Nigeria. However, the use of contaminated plastic for toys and personal care products is a direct route of consumer exposure in children and adults, not only to PBDEs, but also to PBDD/Fs.

A toy from Germany made of recycled plastic with levels of PBDEs that meet current regulatory proposals of 1,000 or 500[11] ppm PBDEs contained 386,000 pg/g PBDD/Fs (3,800 pg WHO-TEQ/g). This signals that weak regulation of PBDEs can pose potential harms not only from PBDEs, but also from PBDD/Fs. In addition, the German regulation of PBDD/Fs does not address the most problematic PBDD/F congeners[12] occurring as impurities in PBDEs, as demonstrated in Table 3.

The problem of PBDD/Fs as contaminants in plastic and their potential route into recycled plastics appears to be

11 This level was proposed as an alternative to level 1,000 ppm for Low POPs Content level.

12 The reason for that is explained in chapter 3.4 focused on legislation.

TO PROVIDE MORE PROTECTIVE REGULATIONS, THE FOLLOWING POLICIES ARE NEEDED:

1. Not to allow the proposed 1,000 ppm limit for DecaBDE in recycled plastics, but rather establish a 10 ppm limit

2. A more stringent limit for the definition of POPs waste (Low POPs Content Level),[13] ideally to establish it at a level of 50 ppm for the sum of all regulated PBDEs

3. To withdraw the recycling exemptions for commercial PentaBDE and OctaBDE as they are currently established under the Stockholm Convention and registered in the EU and several other states.

4. Add PBDD/Fs to the Stockholm Convention for global reduction and elimination[14]

5. Improve the definition of electronic waste within the framework of the Basel Convention

significant. The total amount of PBDD/Fs as an impurity in the produced volume of PBDEs is estimated to be 1,000 t [44]. This amount represents a significant burden for human health and environment, considering that PBDD/Fs exhibit toxicity similar to PCDD/Fs [36].

There is also a need to stop the uncontrolled movement of electronic waste and BFR-contaminated plastics from the automotive industry into developing and transition countries. Up to 80% of electronic waste produced globally was likely *"dumped, traded or recycled under inferior conditions"* [62]. Large volumes of PBDD/Fs have been found in electronic waste disposed in Nigeria [44, 63, 64]. Plastics from these types of waste flows were the likely origin of recycled plastics for the production of toys analyzed in this study.

13 This limit is called "Low POPs Content" level according to the definition in the Stockholm Convention, and its definition is established by the *technical* experts group under the Basel Convention in General Technical Guidelines for ESM of POPs waste. This level is defined in Annex IV to POPs Regulation No 850/2004 (POPs Regulation) in the EU.

14 Analysis guidelines for screening and chemical analysis of all toxic relevant PBDD/Fs, PBBs and PCDD/Fs in recycled plastic and products thereof could also help.

ANNEX 1: BROMINATED DIOXINS (PBDD/FS)

A.1.1 Introduction

Several summaries of PBDD/Fs characteristics and toxicity are available [1, 30, 36, 51, 65-69], although they were written in different years and can differ according to the current stage of knowledge at the time. The purpose of this Annex is not to gather all the information available about PBDD/Fs to date, but rather to highlight data related to the subject matter of this study. More data and information are available either in the text of the report itself or in the above-mentioned summaries.

A.1.2 Basic characteristics and occurrence

Polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) is the full and correct name for the two groups of chemicals called, in short, "brominated dioxins". They have been known to be potential by-products of commercial PBDE mixtures since 1986 [19], similar to chlorinated dioxins that have been observed as impurities in PCBs, and other chlorinated chemicals. Their properties are similar to the PCDD/Fs; however, they have been studied less extensively than their chlorinated analogues.

Polybrominated dibenzo- *p*-dioxins (PBDDs) and polybrominated dibenzofurans (PBDFs) are aromatic compounds. PBDD/Fs are a large class of substances with a potential existence of 75 PBDDs and 135 PBDFs. In addition, a large number of mixed halogenated congeners – 1550 brominated/chlorinated dibenzo-*p*-dioxins (PXDDs) and 3050 brominated/chlorinated dibenzofurans (PXDFs) - are theoretically possible. *"Because of the complexity of the analytical procedures and paucity of analytical reference standards, it has been possible to characterize and determine only a small number of these compounds. The most toxic congeners are those substituted at positions 2, 3, 7, and 8."* [65] Widespread experience with analyses of PBDD/Fs is lacking, and this is even more true for PXDD/Fs, although analytical capacities have developed since the evaluation by WHO in 1998.

In the 20-year old Environmental Health Criteria 205 [65] it states, *"They are not intentionally produced but are formed as undesired by-products in various processes. They can be formed by chemical, photochemical, or thermal reactions from precursors or by so-called de novo synthesis (from organic materials with bromine). … Thermolysis of brominated flame retardants, in particular PBDEs, has been implicated as an obvious source of PBDDs/PBDFs. Heating and burning of products containing such brominated compounds can cause emission of PBDDs/PBDFs. PBDDs/PBDFs have also been detected in traffic-related emissions.*

PBDDs/PBDFs have been found as contaminants in brominated organic chemicals (e.g. bromophenols) and, in particular, in flame retardants, such as polybrominated diphenyl ethers (PBDEs), decabromobiphenyl (decaBB or DBB), 1,2-bis(tribromophenoxy) ethane, tetrabromobisphenol A (TBBPA), and others. They have been detected in distillation residues of some bromophenols and bromoanilines and in wastes from chemical laboratories." This information is still valid, but more research needs to be performed on this important class of chemicals.

A.1.2.1 Natural formation of PBDD/Fs

It is known that some organobromine compounds are naturally produced. Tri - and tetra - bromodibenzo - *p* - dioxins produced by algae and/or cyanobacteria and assimilated in blue mussels in the Baltic Sea have been reported [70]. It has been hypothesized that some PBDDs are naturally produced by marine organisms in the littoral zone [71]. However, most PBDDs/F are produced unintentionally during production of brominated compounds or combustion processes involving bromine [72-74].

A.1.3 Toxicity, health effects, and exposure routes

A.1.3.1 Toxicity and health effects

PBDDs/F have been found to exhibit similar toxicity and health effects as their chlorinated analogues (PCDD/Fs) [1, 29-32], such as neuro- and developmental toxicity.

Haijima et al. [75] explored the neurotoxicity of TBDD in utero and lactational exposure at doses that did not affect the dams but altered the acquisition and retention of fear memory in mouse offspring. The results suggested disruption of the functions of memory and emotion in male mouse offspring even at low doses of TBDD. When compared with TCDD, no obvious difference was found between the developmental toxicities of these two compounds [75].

In the zebrafish model, brominated dioxins in feed were later found in female fish and transferred to their eggs. TBDD exposure reduced spawning success, altered the morphology of the ovaries, and induced aryl hydrocarbon genes [76].

PBDD/Fs can alter endocrine function [77], so they belong to the category of endocrine disrupting chemicals (EDCs). Thyroid hormones were affected in rats after single [78] and subchronic [79, 80] exposures to 2,3,7,8-TeBDD. Because PBDD/Fs can alter thyroid hormone levels, these compounds have the potential to affect early life stages of organisms by affecting brain development [1].

Studies have shown that PBDD/Fs bind and activate the aryl hydrocarbon receptor and exert their toxic effects through the same mechanism as their chlorinated analogs. Dioxins and dioxin

- like compounds bind to and thereby activate the aryl hydrocarbon receptor , which induces a series of severe pathophysiological consequences, such as developmental defects, immunosuppression, and carcinogenesis [32, 65, 81-84].

An early study by Birnbaum et al. [85] confirmed the teratogenicity of certain PBDF congeners.

It has also been shown that very low doses of PBDDs and PCDDs affect the immune system of the marmoset monkey [86-88].

Brief information about toxicity and health effects is also provided in chapter 1.2.

A.1.3.2 Dust as a potential exposure route for PBDD/Fs

Studies have shown that BFRs and PBDD/Fs are present in indoor dust samples collected in homes, offices, and electronic-waste facilities [50, 89, 90]. The possible sources are electronic appliances, such as televisions (TV), computers, and fabric products. Similar to TV cabinet samples, dust samples collected in electronic waste facilities contained high levels of PBDD/Fs [91, 92]. Total concentrations of tetra- to octa-BDD/Fs in electronic-waste floor dust samples from China (5 samples) and Vietnam (10 samples) were in the range of 7,700,000 – 143,000,000 pg/g dry wt [89, 91]. These studies confirmed that uncontrolled electronic-waste recycling processes are a major source of environmental contamination by PBDD/Fs.

Although the concentrations of PBDD/Fs in indoor house dust samples are lower than those measured in dust from electronic appliances and in dust samples from electronic-waste facilities, indoor dust can be a major exposure pathway of PBDD/Fs to the general human population.

A.1.3.3 PBDD/Fs in biota and food as exposure route

Studies on food chain transfer of PBDD/Fs are not available. However, based on the information available for PCDD/Fs, it can be inferred that PBDD/Fs can bioaccumulate and possibly biomagnify in the food chain. Miyake et al. [66] detected PBDD/Fs congeners in seafood collected from southeastern China and found that crab samples had the highest concentrations of PBDD/Fs at 892 pg/g lipid. A survey of UK shellfish found PBDD/Fs in oysters, scallops and cockles [93, 94]. PBDD/Fs have also been found in Baltic wild salmon [95] and fish in Japan [96].

Recent reports showed that PBDD/Fs were detectable in animal origin food, such as milk and eggs, which contained mean concentrations of PBDD/Fs of 0.2 pg TEQ/g lipid for milk samples and 0.3 pg TEQ/g lipid for egg samples [97]. High levels of PBDD/Fs measuring 27 and 16 pg TEQ/g lipid respectively were found in free range chicken egg samples from a neighborhood of a municipal waste incinerator in China and an electronic waste artisanal recycling workshop in Thailand, respectively [98-100].

A.1.3.4 Levels in human tissues

Ericson Jogsten et al. [101] reported that in the Swedish general population, dibromodibenzofurans (2,7/2,8 - BDF) were detected in one - third of adipose tissue samples at concentrations ranging from 0.19 to 0.30 pg/g lipid, and the tetra - substituted 2,3,7,8 - TBDF was detected in all samples at concentrations ranging from 0.27 to 2.24 pg/g lipid [101]. Choi et al. [102] analyzed adipose tissue samples among the Japanese general population and reported that 3 PBDD/Fs congeners (2,3,7,8 - TBDD, 2,3,7,8 - TBDF, and 2,3,4,7,8 – PeBDF) were found at median concentrations (ranges) of 5.1 (3.4 – 8.3) and 3.4 (1.9 – 5.3) pg/g lipid.

PBDD/Fs have also been found in the blood of workers involving in manufacturing resins containing PBDEs at levels ranging up to 112 pg/g 2,3,7,8-TBDF and 448 pg/g 2,3,7,8-TBDD [103]. Firefighters are another group of workers potentially exposed to PBDD/Fs, especially in fires involving flame-retarded materials containing PBDEs and other brominated substances. The first study to measure PBDD/Fs in firefighters found PBDD/Fs ranging from 0.2 – 734 pg/g lipid weight TEQ. In fact, the toxicity (expressed as TEQ) due to PBDD/Fs was 21 times higher than that due to PCDD/Fs [104].

Various studies have reported the presence of PBDD/Fs in breast milk in Belgium [105], Ireland [106], Japan [107], Sweden [108], and Vietnam [109] with levels up to 269 pg/g lipid weight in Japan and 300 pg/g wet weight PBDFs in Sweden. A 2005 study estimated that PBDD/Fs could contribute up to 12% of the dioxin-like toxicity in human milk [110].

REFERENCES

1. Kannan, K., C. Liao, and H.-B. Moon, *Polybrominated dibenzo-p-dioxins and dibenzofurans*, in *Dioxins and health Including Other Persistent Organic Pollutants and Endocrine Disruptors. Third Edition*, A. Schecter, Editor. 2012, Wiley: USA. p. 255-302.
2. UNEP and Stockholm Convention, *Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs under Article 5 of the Stockholm Convention*. 2013, United Nations Environment Programme & Stockholm Convention Secretariat: Geneva. p. 445.
3. POP RC, *Work programmes on new persistent organic pollutants as adopted by the Conference of the Parties. Annex: Technical Review of the Implications of Recycling Commercial Pentabromodiphenyl Ether. UNEP/POPS/POPRC.6/2/Rev.1*. 2010: Geneva. p. 45.
4. DiGangi, J., J. Strakova, and A. Watson, *A survey of PBDEs in recycled carpet padding.* Organohalog Compd, 2011. **73**: p. 2067-2070.
5. Straková, J., et al., *Toxic Loophole - Recycling Hazardous Waste into New Products*. 2018, Arnika, IPEN, HEAL: Brussels, Prague, Gothenburg. p. 36.
6. Straková, J., Petrlík, Jindřich, Pulkrabová, Jana, Gramblička, Tomáš, *Toxic Recycling, or how unsorted waste may contaminate consumer products in the Czech Republic*, in *Abstracts Book of the Dioxin 2018 : 38th International Symposium on Halogenated Persistent Organic Pollutants & 10th International PCB Wowkshop*. 2018: Kraków, Poland. p. 994-996.
7. Abdallah, M.A.-E., et al., *Hexabromocyclododecane in polystyrene packaging: A downside of recycling?* Chemosphere, 2018. **199**: p. 612-616.
8. Strakova, J., et al., *Hexabromocyclododecane (HBCD) found in e-waste is widely present in children's toys (available at http://www.dioxin2017.org/uploadfiles/2017/9997.pdf)*, in *Dioxin 2017*. 2017: Vancouver, Canada.
9. Kuang, J., M.A.-E. Abdallah, and S. Harrad, *Brominated flame retardants in black plastic kitchen utensils: Concentrations and human exposure implications.* Science of The Total Environment, 2018. **610-611**(Supplement C): p. 1138-1146.
10. Samsonek, J. and F. Puype, *Occurrence of brominated flame retardants in black thermo cups and selected kitchen utensils purchased on the European market.* Food Additives & Contaminants: Part A, 2013. **30**(11): p. 1976-1986.
11. Stockholm Convention, *Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes*. 2010: Geneva. p. 64.
12. Stockholm Convention, *An amendment to Annex A adopted by the Conference of the Parties to the Stockholm Convention on Persistent Organic Pollutants at its sixth meeting (Decision SC-6/13)*. 2013: Geneva.
13. Stockholm Convention, *Listing of decabromodiphenyl ether. Decision SC-8/10*. 2017: Geneva.
14. DiGangi, J. and J. Strakova, *Recycling of plastics containing brominated flame retardants leads to contamination of plastic childrens toys.* Organohalog Compd, 2016. **78**(2016): p. 9-11.
15. Drage, D.S., et al., *Brominated flame retardants in Irish waste polymers: Concentrations, legislative compliance, and treatment options.* Science of The Total Environment, 2018. **625**: p. 1535-1543.
16. Guzzonato, A., F. Puype, and S.J. Harrad, *Evidence of bad recycling practices: BFRs in children's toys and food-contact articles.* Environmental Science: Processes & Impacts, 2017. **19**(7): p. 956-963.
17. Strakova, J. and J. Petrlik, *Toy or Toxic Waste? An Analysis of 47 Plastic Toy and Beauty Products Made from Toxic Recycling*. 2017. p. 17.
18. European Commission, *Commission Regulation (EU) 2017/644 of 5 April 2017 laying down methods of sampling and analysis for the control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs and repealing Regulation (EU) No 589/2014 (Text with EEA relevance)*, European Commission, Editor. 2017: Official Journal of the European Union. p. 9-34.
19. Buser, H., *Polybrominated dibenzofurans and dibenzo-p-dioxins: thermal reaction products of polybrominated diphenyl ether flame retardants.* Environ Sci Technol, 1986. **20**(4): p. 404-408.
20. Kajiwara, N., Y. Noma, and H. Takigami, *Photolysis Studies of Technical Decabromodiphenyl Ether (DecaBDE) and Ethane (DeBDethane) in Plastics under Natural Sunlight.* Environmental Science & Technology, 2008. **42**(12): p. 4404-4409.
21. White, S.S. and L.S. Birnbaum, *An Overview of the Effects of Dioxins and Dioxin-Like Compounds on Vertebrates, as Documented in Human and Ecological Epidemiology.* Journal of Environmental Science and Health, Part C, 2009. **27**(4): p. 197-211.

22. Schecter, A., *Dioxins and health Including Other Persistent Organic Pollutants and Endocrine Disruptors. Third Edition*. 2012, USA: Wiley.

23. York, G. and H. Mick. *Last ghost' of the Vietnam War*. 2008 April 27, 2018 [cited 2018 19-11-2018]; Available from: https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all.

24. Kubal, M., et al., *Treatment of solid waste polluted by polychlorinated contaminants (pilot-scale demonstration)*, in *International Conference on Waste Management and the Environment No2*, S. WIT Press, ROYAUME-UNI (2004) (Monographie), Editor. 2004, WIT Press: Rhodes. p. 13-23.

25. Weber, R., et al., *Dioxin- and POP-contaminated sites—contemporary and future relevance and challenges. Overview on background, aims and scope of the series.* Environ Sci Pollut Res, 2008. **15**: p. 363-393.

26. Zemek, A. and A. Kocan, *2,3,7,8-Tetrachlorodibenzo-p-dioxin in soil samples from a trichlorophenol-producing plant.* Chemosphere, 1991. **23**(11-12): p. 1769-1776.

27. Pelclová, D., et al., *Adverse health effects in humans exposed to 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin (TCDD).* Reviews on environmental health, 2006. **21**(2): p. 119-138.

28. Bencko, V. and F.Y.L. Foong. *The History, Toxicity and Adverse Human Health and Environmental Effects Related to the Use of Agent Orange*. 2013. Dordrecht: Springer Netherlands.

29. Mason, G., et al., *Polybrominated and chlorinated dibenzo-p-dioxins: synthesis biologic and toxic effects and structure-activity relationships.* Chemosphere, 1987. **16**(8-9): p. 1729-1731.

30. Piskorska-Pliszczyńska, J. and S. Maszewski, *Brominated dioxins: little-known new health hazards-a review.* Bull Vet Inst Pulawy, 2014. **58**: p. 327-335.

31. Behnisch, P.A., K. Hosoe, and S.-i. Sakai, *Brominated dioxin-like compounds: in vitro assessment in comparison to classical dioxin-like compounds and other polyaromatic compounds.* Environment International, 2003. **29**(6): p. 861-877.

32. Birnbaum, L., D. Staskal, and J. Diliberto, *Health effects of polybrominated dibenzo-p-dioxins (PBDDs) and dibenzofurans (PBDFs).* Environ Int, 2003. **29**(6): p. 855-60.

33. POP RC, *Report of the Persistent Organic Pollutants Review Committee on the work of its sixth meeting.*, in *Persistent Organic Pollutants Review Committee Sixth meeting*. 2010: Geneva. p. 45.

34. DiGangi, J., J. Strakova, and L. Bell, *POPs Recycling Contaminates Children's Toys with Toxic Flame Retardants*. 2017, IPEN, Arnika. p. 20.

35. Vetter, W., et al., *Formation of polybrominated dibenzofurans (PBDFs) after heating of a salmon sample spiked with decabromodiphenyl ether (BDE-209).* Environ Sci Pollut Res Int, 2014.

36. van den Berg, M., et al., *Polybrominated Dibenzo-p-dioxins (PBDDs), Dibenzofurans (PBDFs) and Biphenyls (PBBs) - Inclusion in the Toxicity Equivalency Factor Concept for Dioxin-like Compounds.* Toxicological Sciences, 2013.

37. van den Berg, M., et al., *The 2005 World Health Organization reevaluation of human and Mammalian toxic equivalency factors for dioxins and dioxin-like compounds.* Toxicol Sci, 2006. **93**(2): p. 223-41.

38. IPEN, *Make Low POPs Content Levels Low Enough for Health and Environment Protection: Keep the Promise, Eliminate POPs! A Brief for Delegates*. 2017.

39. BiPRO, *Study to facilitate the implementation of certain waste related provisions of the Regulation on Persistent Organic Pollutants (POPs)*. 2005, European Commission: Brussels. p. 469.

40. Behnisch, P., et al., *Low-Temperature Thermal Decomposition of Dioxin-like Compounds in Fly Ash: Combination of Chemical Analysis with in Vitro Bioassays (EROD and DR-CALUX).* Environ. Sci. Technol., 2002. **36**(23): p. 5211-5217.

41. Tu, L.-K., et al., *Distribution of polybrominated dibenzo-p-dioxins and dibenzofurans and polybrominated diphenyl ethers in a coal-fired power plant and two municipal solid waste incinerators.* Aerosol and Air Quality Resarch, 2011. **11**(5): p. 596-615.

42. Lai, Y., et al., *Inhibition of polybrominated dibenzo-p-dioxin and dibenzofuran formation from the pyrolysis of printed circuit boards.* Environ Sci Technol, 2007. **41**(3): p. 957-62.

43. Kawamoto, K., *Potential formation of PCDD/Fs and related bromine-substituted compounds from heating processes for ashes.* Journal of Hazardous Materials, 2009. **168**(2–3): p. 641-648.

44. Sindiku, O., et al., *Polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) in e-waste plastic in Nigeria.* Environmental Science and Pollution Research, 2015. **22**(19): p. 14515-14529.

45. Schlummer, M., et al., *Polymers in waste electric and electronic equipment (WEEE) contain PBDD/F in the ppb-range.* Organohalogen Compounds, 2004(66): p. 859-863.

46. Ren, M., et al., *PBDD/F impurities in some commercial deca-BDE.* Environmental Pollution, 2011. **In Press, Corrected Proof**.

47. Hanari, N., et al., *Occurrence of Polybrominated Biphenyls, Polybrominated Dibenzo-p-dioxins, and Polybrominated Dibenzofurans as Impurities in Commercial Polybrominated Diphenyl Ether Mixtures* Environ. Sci. Technol., 2006. **ASAP Article 10.1021/ es060559k S0013-936X(06)00559-1**

48. Ebert, J. and M. Bahadir, *Formation of PBDD/F from flame-retarded plastic materials under thermal stress.* Environ Int, 2003. **29**(6): p. 711-6.

49. Sakai, S.-i., et al., *Combustion of brominated flame retardants and behavior of its byproducts.* Chemosphere, 2001. **42**(5-7): p. 519-531.

50. Takigami, H., et al., *Transfer of brominated flame retardants from components into dust inside television cabinets.* Chemosphere, 2008. **73**(2): p. 161-169.

51. Dumler, R., et al., *Thermal formation of polybrominated dibenzofurans and dioxins from decabromodiphenyl ether flame retardant: Influence of antimony(III) oxide and the polymer matrix.* Chemosphere, 1990. **20**(10-12): p. 1867-1873.

52. Dumler, R., et al., *PBDF and PBDD from the combustion of bromine containing flame retarded polymers: A survey.* Chemosphere, 1989. **19**(12): p. 2023-2031.

53. Sedlak, D., et al., *Polyhalogenated dibenzo-p-dioxins and dibenzofurans in the exhaust air during textile processings.* Chemosphere, 1998. **37**(9-12): p. 2071-2076.

54. Schlummer, M., et al., *Report: Recycling of flame-retarded plastics from waste electric and electronic equipment (WEEE).* Waste Management Research, 2006. **24**(6): p. 573-583.

55. Ritterbusch, J., W. Lorenz, and M. Bahadir, *Determination of polyhalogenated dibenzo-p-dioxins and dibenzofurans in analytical laboratory waste and their decomposition by UV-photolysis.* Chemosphere, 1994. **29**(9-11): p. 1829-1838.

56. Lenoir, D., et al., *Photochemical degradation of brominated dibenzo-p-dioxins and -furans in organic solvents.* Chemosphere, 1991. **22**(9): p. 821-834.

57. Ritterbusch, J., et al., *UV-photolysis of PXDD/F-contaminated bromophenols and wastes of chemical laboratories.* Chemosphere, 1994. **29**(3): p. 457-464.

58. German Federal Government, *Verordnung zur Neuregelung nationaler Vorschriften über das Inverkehrbringen und die Abgabe von Chemikalien. Vom 20. Januar 2017(Regulation on the revision of national regulations on the placing on the market and supply of chemicals. January 20, 2017 - in German)*, G.F. Government, Editor. 2017: Bundesgesetzblatt Jahrgang 2017 Teil I Nr. 4, ausgegeben zu Bonn am 26. Januar 2017. p. 94-105.

59. Schlummer, M., et al., *Analysis of flame retardant additives in polymer fractions of waste of electric and electronic equipment (WEEE) by means of HPLC–UV/ MS and GPC–HPLC–UV.* Journal of Chromatography A, 2005. **1064**(1): p. 39-51.

60. European Parliament and Council, *Regulation (EC) No 850/2004 of the European Parliament and of the Council of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC (OJ L 158 30.4.2004, p. 7. Amended by: Council Regulation (EC) No 1195/2006 of 18 July 2006; Council Regulation (EC) No 172/2007 of 16 February 2007; Commission Regulation (EC) No 323/2007 of 26 March 2007; Regulation (EC) No 219/2009 of the European Parliament and of the Council of 11 March 2009; Commission Regulation (EC) No 304/2009 of 14 April 2009; Commission Regulation (EU) No 756/2010 of 24 August 2010; Commission Regulation (EU) No 757/2010 of 24 August 2010;Commission Regulation (EU) No 519/2012 of 19 June 2012;Commission Regulation (EU) No 1342/2014 of 17 December 2014;Commission Regulation (EU) 2015/2030 of 13 November 2015; Commission Regulation (EU) 2016/293 of 1 March 2016; Commission Regulation (EU) 2016/460 of 30 March 2016, and Corrected by: Corrigendum, OJ L 229, 29.6.2004, p. 5 (850/2004).* European Parliament and Council, Editor. 2016: Brussels. p. 1-28.

61. Basel Convention, *General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants*, in *Technical Guidelines*. 2017: Geneva.

62. Ilankoon, I.M.S.K., et al., *E-waste in the international context – A review of trade flows, regulations, hazards, waste management strategies and technologies for value recovery.* Waste Management, 2018. **82**: p. 258-275.

63. Babayemi, J., et al., *Substance flow analysis of polybrominated diphenyl ethers in plastic from EEE/ WEEE in Nigeria in the frame of Stockholm Convention as a basis for policy advice.* Environmental Science and Pollution Research, 2015 a. **22**: p. 14502–14514.

64. Lundstedt, S., et al., *Brominated dioxins in plastics–Emissions during fires*, in *PIC2015 – the 14th International Congress on Combustion By-Products and Their Health Effects, 14-17 June 2015*. 2015: Umeå, Sweden.

65. WHO, *Polybrominated dibenzo-p-dioxins and dibenzofurans*, in *Environmental Health Criteria, 205. XXI+303P. ISBN 92-4-157205-1; 205 (0). 1998. i-xxi; 1-303.* 1998, World Health Organization: Geneva, Switzerland.

66. Miyake, Y., et al., *Preliminary health risk assessment for polybrominated diphenyl ethers and polybrominated dibenzo-p-dioxins/furans in seafood from Guangzhou and Zhoushan, China.* Marine Pollution Bulletin, 2008. **57**(6-12): p. 357-364.

67. Mason, G., et al., *Polybrominated dibenzo-p-dioxins and related compounds: Quantitative in vivo and in vitro structure-activity relationships.* Toxicology, 1987. **44**(3): p. 245-255.

68. Zhou, Y. and J. Liu, *Emissions, environmental levels, sources, formation pathways, and analysis of polybrominated dibenzo-p-dioxins and dibenzofurans: a review.* Environmental Science and Pollution Research, 2018.

69. Haglund, P. and I. Skogman, *Polybrominated Dioxins and Dibenzofurans: A global Concern.* Organohalogen Compounds,, 2009. **71**: p. 2347-2352.

70. Haglund, P., et al., *Temporal Variations of Polybrominated Dibenzo-p-Dioxin and Methoxylated Diphenyl Ether Concentrations in Fish Revealing Large Differences in Exposure and Metabolic Stability.* Environmental Science & Technology, 2010. **44**(7): p. 2466-2473.

71. Haglund, P., et al., *Brominated Dibenzo-p-Dioxins: A New Class of Marine Toxins?* Environmental Science & Technology, 2007. **41**(9): p. 3069-3074.

72. Li, H., et al., *Severe PCDD/F and PBDD/F pollution in air around an electronic waste dismantling area in China.* Environ Sci Technol, 2007. **41**(16): p. 5641-6.

73. Ortuño, N., et al., *De Novo Synthesis of Brominated Dioxins and Furans.* Environmental Science & Technology, 2014. **48**(14): p. 7959-7965.

74. Weber, R. and B. Kuch, *Relevance of BFRs and thermal conditions on the formation pathways of brominated and brominated-chlorinated dibenzodioxins and dibenzofurans.* Environ Int, 2003. **29**(6): p. 699-710.

75. Haijima, A., et al., *In utero and lactational exposure to low doses of chlorinated and brominated dioxins induces deficits in the fear memory of male mice.* NeuroToxicology, 2010. **31**(4): p. 385-390.

76. Norman Haldén, A., et al., *Retention and maternal transfer of brominated dioxins in zebrafish (Danio rerio) and effects on reproduction, aryl hydrocarbon receptor-regulated genes, and ethoxyresorufin-O-deethylase (EROD) activity.* Aquatic Toxicology, 2011. **102**(3): p. 150-161.

77. Ding, L., et al., *Effects of brominated flame retardants and brominated dioxins on steroidogenesis in H295R human adrenocortical carcinoma cell line.* Environmental Toxicology and Chemistry, 2007. **26**(4): p. 764-772.

78. Ivens, I., et al., *Toxicity of 2,3,7,8-tetrabromodibenzo-p-dioxin in rats after single oral administration.* Toxicology, 1992. **73**(1): p. 53-69.

79. Löser, E. and I. Ivens, *Preliminary results of a 3 month toxicity study on rats with 2,3,7,8,-Tetrabromodibenzo-p-dioxin (2,3,7,8-TBDD).* Chemosphere, 1989. **19**(1): p. 759-764.

80. Ivens, I., et al., *Subchronic toxicity of 2,3,7,8-tetrabromodibenzo-p-dioxin in rats.* Toxicology, 1993. **83**(1-3): p. 181-201.

81. Mennear, J.H. and C.C. Lee, *Polybrominated dibenzo-p-dioxins and dibenzofurans: literature review and health assessment.* Environ Health Perspect, 1994. **102 Suppl 1**: p. 265-74.

82. Weber, L.W. and H. Greim, *The toxicity of brominated and mixed-halogenated dibenzo-p-dioxins and dibenzofurans: an overview.* J Toxicol Environ Health, 1997. **50**(3): p. 195-215.

83. Bock, K.W. and C. Köhle, *Ah receptor: Dioxin-mediated toxic responses as hints to deregulated physiologic functions.* Biochemical Pharmacology, 2006. **72**(4): p. 393-404.

84. Birnbaum, L., *The mechanism of dioxin toxicity: relationship to risk assessment.* Environ Health Perspect, 1994. **102 Suppl 9**: p. 157-67.

85. Birnbaum, L.S., R.E. Morrissey, and M.W. Harris, *Teratogenic effects of 2,3,7,8-tetrabromodibenzo-p-dioxin and three polybrominated dibenzofurans in C57BL6N mice.* Toxicology and Applied Pharmacology, 1991. **107**(1): p. 141-152.

86. Neubert, D., *Peculiarities of the toxicity of polyhalogenated dibenzo-p-dioxins and dibenzofurans in animals and man.* Chemosphere, 1991. **23**(11): p. 1869-1893.

87. Neubert, R., et al., *Polyhalogenated dibenzo-p-dioxins and dibenzofurans and the immune system. 1. Effects on peripheral lymphocyte subpopulations of a non-human primate (Callithrix jacchus) after treatment with 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD).* Archives of Toxicology, 1990. **64**(5): p. 345-359.

88. Neubert, R., et al., *Effects of Small Doses of Dioxins on the Immune System of Marmosets and Rats.* Annals of the New York Academy of Sciences, 1993. **685**(1): p. 662-686.

89. Ma, J., et al., *Polybrominated Dibenzo-p-dioxins/ Dibenzofurans and Polybrominated Diphenyl Ethers in Soil, Vegetation, Workshop-Floor Dust, and Electronic Shredder Residue from an Electronic Waste Recycling Facility and in Soils from a Chemical Industrial Complex in Eastern China.* Environ Sci Technol, 2009. **43**(19): p. 7350-7356.

90. Suzuki, G., et al., *Dioxin-like Activity in Japanese Indoor Dusts Evaluated by Means of in Vitro Bioassay and Instrumental Analysis: Brominated Dibenzofurans Are an Important Contributor.* Environmental Science & Technology, 2010: p. null-null.

91. Tue, N.M., et al., *Evaluation of Dioxin-Like Activities in Settled House Dust from Vietnamese E-Waste Recycling Sites: Relevance of Polychlorinated/Brominated Dibenzo-p-Dioxin/Furans and Dioxin-Like PCBs.* Environmental Science & Technology, 2010. **44**(23): p. 9195-9200.

92. Takigami, H., et al., *Flame retardants in indoor dust and air of a hotel in Japan.* Environment International, 2009. **35**(4): p. 688-693.

93. Rose, M. and A. Fernandes. *Are BFRs responsible for brominated dioxins and furans (PBDD/Fs) in food?* in *BFR2010*. 2010.

94. Fernandes, A., et al., *Brominated dioxins (PBDD/Fs) and PBDEs in marine shellfish in the UK.* Food Additives & Contaminants: Part A, 2009. **26**(6): p. 918-927.

95. Zacs, D., J. Rjabova, and V. Bartkevics, *Occurrence of Brominated Persistent Organic Pollutants (PBDD/DFs, PXDD/DFs, and PBDEs) in Baltic Wild Salmon (Salmo salar) and Correlation with PCDD/DFs and PCBs.* Environmental Science & Technology, 2013. **47**(16): p. 9478-9486.

96. Ashizuka, Y., et al., *Determination of brominated flame retardants and brominated dioxins in fish collected from three regions of Japan.* Molecular Nutrition & Food Research, 2008. **52**(2): p. 273-283.

97. Fernandes, A.R., et al., *Polybrominated diphenylethers (PBDEs) and brominated dioxins (PBDD/Fs) in Irish food of animal origin.* Food Additives & Contaminants: Part B, 2009. **2**(1): p. 86-94.

98. Teebthaisong, A., et al., *POPs contamination at 'recycling' and metallurgical site in Thailand*, in *Abstracts Book of the Dioxin 2018 : 38th International Symposium on Halogenated Persistent Organic Pollutants & 10th International PCB Wowkshop*. 2018: Kraków, Poland. p. 1040-1043.

99. Petrlik, J., A. Teebthaisong, and A. Ritthichat, *Chicken Eggs as an Indicator of POPs Pollution in Thailand. Results of sampling conducted in 2015 – 2016*. 2017, Arnika - Toxics and Waste Programme, EARTH: Bangkok, Prague. p. 32.

100. Petrlik, J., *Persistent Organic Pollutants (POPs) in Chicken Eggs from Hot Spots in China (Updated version)*. 2016, Arnika - Toxics and Waste Programme, IPEN and Green Beagle: Beijing-Gothenburg-Prague. p. 25.

101. Ericson Jogsten, I., et al., *Analysis of POPs in human samples reveal a contribution of brominated dioxin of up to 15% of the total dioxin TEQ.* Chemosphere, 2010. **78**(2): p. 113-120.

102. Choi, J., et al., *Polybrominated dibenzo-p-dioxins, dibenzofurans, and diphenyl ethers in Japanese human adipose tissue.* Environ Sci Technol, 2003. **37**(5): p. 817-21.

103. Zober, M., et al., *Morbidity study of extruder personnel with potential exposure to brominated dioxins and furans. I. Results of blood monitoring and immunological tests.* Br J Ind Med, 1992. **49**(8): p. 532-44.

104. Shaw, S.D., et al., *Persistent organic pollutants including polychlorinated and polybrominated dibenzo-p-dioxins and dibenzofurans in firefighters from Northern California.* Chemosphere, 2013. **91**(10): p. 1386-1394.

105. Croes, K., et al., *Determination of PCDD/Fs, PBDD/Fs and dioxin-like PCBs in human milk from mothers residing in the rural areas in Flanders, using the CALUX bioassay and GC-HRMS.* Talanta, 2013. **113**: p. 99-105.

106. Pratt, I., et al., *Brominated and fluorinated organic pollutants in the breast milk of first-time Irish mothers: is there a relationship to levels in food?* Food Additives & Contaminants: Part A, 2013. **30**(10): p. 1788-1798.

107. Ohta, S., et al., *Levels of PBDEs, TBBPA, TBPs, PCDDs/DFs, PXDDs/DFs and PBDDs/DFs in human milk of nursing women and dairy milk products in Japan.* Organohalogen Compd, 2004. **66**: p. 2857-2862.

108. Brorstrom-Lunden, E., et al., *Results From the Swedish National Screening Programme 2008—Subreport 4: Screening of Unintentionally Produced Organic Contaminants*. 2010: ISER Institute: Goteborg, Sweden.

109. Tue, N.M., et al., *Dioxin-related compounds in breast milk of women from Vietnamese e-waste recycling sites: Levels, toxic equivalents and relevance of non-dietary exposure.* Ecotoxicology and Environmental Safety, 2014. **106**(0): p. 220-225.

110. Kotz, A., et al., *PBDE, PBDD/F and mixed chlorinated-brominated PXDD/F in pooled human milk samples from different countries.* Organohalogen Compounds - Volume 67 (2005), 2005. **67**.

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