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REVIEW ARTICLE

Levels and distribution of polybrominated diphenyl ethers in humans and environmental compartments: a comprehensive review of the last five years of research

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Abstract

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants (BFRs), present in the environment, animals, and humans. Their levels, distribution, and human exposure have been studied extensively, and over the last decade, various legal measures have been taken to prohibit or minimize their production and use due to the increasing amount of evidence of their harmful effects on human and animal health.Our aim here was to make a comprehensive and up-to-date review of the levels and distribution of PBDEs in the aquatic environment, air, and soil, in indoor dust, and in humans. To fulfill this, we searched through *Web of Science* for literature data reported in the last five years (2015–2019) on levels of at least six key PBDE congeners in abovementioned matrices. According to our summarized data, significant PBDE mass concentrations/fractions are still being detected in various sample types across the world, which implies that PBDE contamination is an ongoing problem. Secondary sources of PBDEs like contaminated soils and landfills, especially those with electronic and electrical waste (e-waste), represent a particular risk to the future and therefore require a special attention of scientists.

Keywords Brominated flame retardants · PBDE · Persistent organic pollutants · POPs · Human exposure · Environmental matrices · Review

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants (BFRs) that came into use in the 1970s as additives to retard/reduce the combustibility of a variety of textile materials, furniture fillers (polyurethane foam), and electronic equipment (UNEP 2015).

There are 209 theoretically possible PBDE congeners divided into 10 congener groups depending on the number of bromine atoms, but they all share a common structure consisting of a brominated diphenyl ether molecule with two benzene rings connected with an oxygen atom (Fig. 1). For commercial use, they are mixed in various percentages (La Guardia et al. 2006) and marketed in three formulations named after the prevalent congener group in the mixture: (1) the "penta" formulation, (2) the "octa" formulation, and (3) the "deca" formulation. Respectively, they accounted for 11%, 6%, and 83% of the global production in 2001 (La Guardia et al. 2006; US EPA 2010).

In contrast to certain BFRs that are mixed with plastic before polymerization to form covalent bonds, PBDEs are added to polymers without forming chemical bonds with the materials and can therefore easily migrate into the surrounding air, dust, soil, and water during their lifetime (Zhang et al. 2011; UNEP 2015; Anh et al. 2017). They were first measured in the environment in the 1980s (Andersson and Blomkvist 1981), and have penetrated all environmental compartments ever since (US EPA 2010). In 2009, the United Nations Environment Programme's Stockholm Convention added them to the group of persistent organic pollutants (POPs) because they have an environmental half-life of several years, can travel long distances in the atmosphere, have a tendency to bioaccumulate and biomagnify in the food web, and are toxic to humans and animals (UNEP 2001; De Wit 2002). To reduce these risks, the European Union banned the use of the penta and octa commercial mixtures in 2004 and the use of the

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decaBDE mixture in electrical and electronic equipment in 2008. The USA followed suit by discontinuing production of the penta and octa mixtures in 2004 (Harrad et al. 2006; US EPA 2010) and of decaBDE in 2013 (US EPA 2014). In 2009, the Stockholm Convention banned the production, use, import, and export of the penta and octa commercial mixtures and in 2017 of decaBDE (Bramwell et al. 2017).

Despite these actions, massive reserves of products containing PBDEs are still in circulation and will continue to release them into the environment for a long time (Abbasi et al. 2015). Part of the problem is the rapidly growing e-waste (Ohajinwa et al. 2019) consisting of discarded electrical and electronic equipment that contains several toxic chemicals, including PBDEs (Robinson 2009; Ilankoon et al. 2018). It is the most prominent in Asian countries that import and recycle e-waste through legal and illegal channels. In these countries, soils at e-waste sites are an important secondary source of pollution (Leung et al. 2007; Tue et al. 2013).

The objective of this review is to summarize the data published in the last five years about the distribution and content of PBDEs in humans and various environmental compartments worldwide. It also addresses concerns about high PBDE levels detected in the areas where e-waste is collected or processed.

Environmental fate and human exposure

Semivolatile pollutants like PBDEs get into the atmosphere as a result of combustion from domestic and industrial sources, emissions from waste incineration or motor vehicles, and (il)legal e-waste landfills (Farrar et al. 2004; Pozo et al. 2016; Degrendele et al. 2018). Their levels and gas/particle partitioning in the atmosphere depend on the physicochemical properties of a particular PBDE congener, environmental conditions, and the abundance, composition, and size of suspended particles (Besis et al. 2017; Degrendele et al. 2018). Once PBDEs are sorbed onto airborne particles, they reach aquatic and terrestrial environments. Atmospheric transport can take them over long distances, which has been confirmed by the detection of PBDEs in areas as remote as the Arctic and Antarctica (Law et al. 2014; Vecchiato et al. 2015; Khairy et al. 2016; Markham et al. 2018). Being hydrophobic, PBDEs tend to attach to particulate matter and therefore accumulate in sediment and soil, both serving as PBDEs environmental sinks (Law et al. 2006; Anh et al. 2017; Mcgrath et al. 2017; Pei et al. 2018; Tiwari et al. 2018; Ma et al. 2019).

Aquatic environments are exposed to PBDEs not only through atmospheric deposition but also through effluent and sewage sludge from wastewater treatment plants and landfill leaches (Aigars et al. 2017; Tombesi et al. 2017; Liu et al. 2018; Pei et al. 2018). With their low vapor pressure, very low water solubility, and high octanol/water partition coefficient (log K_{ow}), PBDEs in aquatic environments adsorb onto the organic fraction of sediments, suspended particulate matter, or enter aquatic organisms. There, they bioaccumulate in lipid-rich tissues of organisms and biomagnify along food chains (Webster et al. 2010; Govaerts et al. 2018). This is why aquatic organisms tend to be highly burdened with PBDEs.

Terrestrial animals, in turn, are much less exposed to bioavailable PBDEs, and—to our knowledge—only three studies have reported PBDE levels in them over the last five years: two in carnivore species from the USA (Boyles and Nielsen 2017; Boyles et al. 2017) and one in two herbivore and one omnivore species from Latvia (Zacs et al. 2018).

Humans are exposed to PBDEs through diet, inhalation, accidental ingestion of dust, and dermal contact. According to the European Food Safety Authority (EFSA 2011), the main source of exposure would be food of animal origin with higher fat content (fish, meat, and dairy products), in which PBDEs tend to accumulate due to their lipophilicity. One of the first reports that found a strong association between the consumption of contaminated fish and elevated PBDE levels in human serum comes from Sweden. Median PBDE level in the serum of Swedes who did not consume fish was 0.4 ng/g lipid weight (lw) (< 0.1–2.5; 10–90 percentile) compared to 2.2 ng/g lw (0.96–5.7; 10–90 percentile) in consumers who ate between 12 and 20 meals of fatty Baltic Sea fish per month (Sjödin et al. 2000). Another recent study from the USA also suggests that people who eat over 10 servings of seafood per week have a significantly higher ∑PBDE in serum than those who eat less than 1 serving per week (Kuo et al. 2019).

Martellini et al. (2016) reported PBDEs in various foodstuffs commonly consumed in Italy (meat, eggs, milk, cheese, fish, fish oil, and mussels). The highest mass fraction of total PBDEs was measured in dairy products (18,537 pg/g ww), meat (12,672 pg/g ww), and eggs (9729 pg/g ww). PentaBDEs were dominant in fish oil, while BDE 209 dominated in other food groups. The authors also showed that exposure to PBDEs through food varied considerably with region and personal food habits.

Lorber (2008) pointed out much higher PBDE levels found in a US population than the rest of the world, even though PBDE levels in US foodstuffs were not. These findings

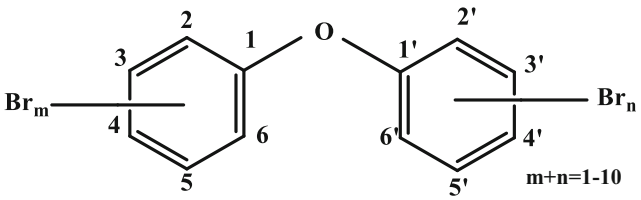

Fig. 1 General structure of the PBDE congeners

pointed to dust inhalation and/or ingestion as one of the most significant routes of human exposure to PBDEs, especially considering the dominantly indoor lifestyle (house, car, school/kindergarten, office/working place). That PBDE exposure through dust inhalation/ingestion is even greater than from food has been confirmed by several reports (Schecter et al. 2006; Wu et al. 2007; Stapleton et al. 2008; Lorber 2008; Fraser et al. 2009; Johnson-Restrepo and Kannan 2009; Wei et al. 2009). Indoor environments are contaminated with PBDEs from flame retardants in a wide range of consumer products that remain in use for a long time, such as polyurethane foam in furniture and automobile seats, textiles, and electrical and electronic equipment. From these products, PBDEs are released into the ambient air through volatilization, mechanical abrasion, and/or sorption to dust particles. Higher PBDE concentrations in dust from the USA than from Europe and Asia seems to directly correlate with the historical usage of PBDEs (Whitehead et al. 2011). Similarly, higher PBDE human body burdens found in California (Zota et al. 2008; Rose et al. 2010) and the UK (Bramwell et al. 2017) than in the rest of the USA and Europe seem to be related to very strict fire protection regulations in California and the UK.

Regardless of the exposure routes, the most vulnerable group to PBDE exposure are children. Numerous studies found higher PBDE levels in children's blood than in blood of their mothers (US EPA 2010; Lunder et al. 2010; Shin et al. 2016; Terry et al. 2017), parents (Fischer et al. 2006; Wu et al. 2015), or generally of the adult population (Toms et al. 2009; Rose et al. 2010).

Similar to other lipophilic contaminants, PBDEs enter the organism as early as in the prenatal period, as evidenced by their findings in the umbilical cord blood (Herbstman et al. 2008; Zota et al. 2018), placenta (Dassanayake et al. 2009), fetal blood (Mazdai et al. 2003), and fetal liver (Zota et al. 2018). Breastfeeding, however, is the period of life when PBDE intake is at its peak (Jones-Otazo et al. 2005; Johnson-Restrepo and Kannan 2009). PBDEs accumulated in breast milk are directly transferred to infants, who receive very high doses per the unit of mass. Exposure to PBDEs continues through early childhood, but the dominant exposure route is now dust ingestion and inhalation associated with their frequent hand-to-mouth activity and the extensive contact with floors, carpets, and other dusty surfaces (EPA 2008; Stapleton et al. 2008). An interesting survey conducted by Hoffman et al. (2017) on children's blood and hand wipe samples showed that toddlers who licked their fingers while eating, who played more with plastic toys, and who were more active in general had higher PBDE levels on their hands and in their serum.

Toxicity

Knowledge about the mechanisms of toxic PBDE action and effects on human health is still quite limited. Toxicity has mostly been investigated in animal studies, and several recent studies have evaluated the associations between PBDE concentrations in human tissues (e.g., blood, breast milk) and various health effects (ATSDR 2017).

Animal models report adverse effects at low doses of pentaBDEs and octaBDEs (from 0.6 and 2 mg/kg body weight, respectively) and much higher doses of decaBDEs (80 mg/kg body weight) and include effects on neurobehavioral development and thyroid hormone levels for pentaBDEs, fetal toxicity/teratogenicity for octaBDEs, and morphological effects in the thyroid, liver, and kidney of adult animals for decaBDEs (Darnerud 2003). Carcinogenicity studies have for now been limited to decaBDEs and show some effects only at very high doses, which is probably why the International Agency for Research on Cancer (IARC) still has not classified decaBDEs in respect to its carcinogenicity to humans (Darnerud 2003).

Glazer et al. (2018) reported short- and long-term behavioral impairments in zebrafish embryos exposed to low concentrations of BDE 47 (0.01–0.3 μM) and BDE 99 (0.003–20 μM). They also found that exposure to very low concentrations had no visible effects on larval activity but adult behavior was still strongly affected.

Epidemiological studies point to an association between prenatal PBDE exposure and lower birth weight, lower levels of thyroid-stimulating hormone (TSH), lower intelligence quotient (IQ), increased incidence of hyperactivity disorder, and impaired cognitive, motor, and behavioral neurodevelopment (Gibson et al. 2018). Postnatal exposure is associated with similar effects, including lower IQ and increased incidence of hyperactive or aggressive behavior.

One possible explanation for the observed neurological impairments might be related to changes in the thyroid hormone status. The development of the nervous system highly depends on thyroid hormones, thyroxine (T4) in particular, and is the most sensitive to environmental effects from the last trimester of pregnancy to two years of age. In vitro evidence suggests that PBDEs may disrupt thyroid hormone production by binding to thyroid hormone receptors, because PBDEs and T4 have a similar stereochemical structure (Marchesini et al. 2008). Animal and human studies indicate that PBDEs may alter the circulating levels of thyroid hormones (Costa et al. 2008; Turyk et al. 2008; van der Ven et al. 2008; Meeker et al. 2009; Chevrier et al. 2010).

Chemical analysis

Analytical methods for PBDE determination are generally similar to those used for determining polychlorinated biphenyls, but de Boer and Cofino (2002) pointed that improvements are still needed, especially when it comes to the analysis of BDE 209, which requires a different approach.

PBDE analysis in environmental and human samples takes several steps (sample pretreatment, extraction, extract clean-up, and final instrumental analysis), and we will mention only frequently used techniques in this review, without going into detail.

Sample pretreatment means moisture removal from samples where is necessary and/or convenient. Extraction will highly depend on sample type and available laboratory equipment. The most common extraction methods from solid samples include Soxhlet extraction, ultrasound extraction, accelerated solvent extraction (ASE), microwave-assisted extraction (MAE), and supercritical fluid extraction (SFE) (de la Cal et al. 2003; Regueiro et al. 2006; Wang et al. 2010; Annunciação et al. 2017). For liquid samples, the most common method is liquid-liquid (LLE) (Byczkiewicz and Jabłoński 2015; Darrow et al. 2017; Pei et al. 2018; Kuo et al. 2019) and solid phase extraction (SPE) (Thomsen et al. 2002; Covaci et al. 2003; Thomsen et al. 2007). Extract clean-up depends on the matrix. Sediments and soils may require sulfur removal, whereas biota require lipid removal, which can be done with sulfuric acid treatment, gel permeation chromatography (GPC), or column adsorption chromatography on sorbents like silica, alumina, or Florisil (Boyles et al. 2017; Giulivo et al. 2017; Novak et al. 2017; Persson et al. 2019). Instrumental analysis is based on gas-chromatographic (GC) separation on nonpolar or semi-polar capillary columns with mass spectrometric (MS) detection (Vecchiato et al. 2015; Newton et al. 2015; Martellini et al. 2016; Mcgrath et al. 2016; Pozo et al. 2016; Kademoglou et al. 2017; Liu et al. 2018; Pei et al. 2018; Ohajinwa et al. 2019; Wu et al. 2019).

There are many articles on the determination of PBDEs in a variety of environmental samples, but the determination of one frequently reported congener, BDE 209, is particularly demanding because (1) it is not stable at high temperatures in the GC injector and GC column; (2) it is sensitive to degradation by UV light; (3) in the MS source it behaves differently than chlorinated and lower-brominated compounds (de Boer and Cofino 2002); and (4) it may easily adsorb onto small dust particles in the laboratory, which may result in sample contamination (Covaci et al. 2003). Thermal decomposition of BDE 209 can be avoided by using a short GC column and a thermally inert GC injection port (Beser et al. 2014), but this means that it should be analyzed separately from other PBDEs. Another way to address the difficulties with BDE 209 determination is to use liquid chromatography–tandem mass spectrometry (LC-MS/MS) (Abdallah et al. 2009).

Levels and distribution

To avoid repeating data presented in previous review articles covering massive amounts of earlier data (de Wit 2002; Law et al. 2006, 2014; Mcgrath et al. 2017; Tang and Zhai 2017), we limited our literature search to the last five years of research (2015 to September 2019). This is currently a very active research area, and the number of articles reporting PBDE levels in environmental samples is constantly increasing. Since there is no list of key toxic PBDE congeners to be monitored, reports vary from just one to more than 10 congeners. However, as certain congeners dominated in commercial formulations, they were also more frequently detected in environmental and biota samples than others, which ultimately led to narrowing the range of research to the following congeners: BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 as representatives of the penta formulation (occasionally including BDE 28 and BDE 138 as well), and BDE 183 and BDE 209 as representatives of the octa and deca formulations, respectively. We decided to take in consideration studies that include at least six congeners. Studies not reporting data on BDE 209 were not excluded from this review, as we are well aware of the difficulties involved in BDE 209 analysis, which has narrowed down the possibility of accurate measurements to a limited number of laboratories.

To make comparison easier between studies, we compared the sums of the mass fractions of all analyzed PBDE congeners in specific research (Σ_xPBDE), unless indicated otherwise. For the same reason, we also did our best to present reported data as uniformly as possible. PBDE mass fractions/concentrations in soil and sediment samples were mostly expressed in ng/g dry weight (dw), in air samples in pg/m^3, and in dust samples in ng/g of dust. In human and biota samples, mass fractions have been lipid-normalized and reported in ng/g of lipid weight, because these contaminants are highly lipophilic and accumulate in lipids. Some authors (Sühring et al. 2016; Aigars et al. 2017; Novak et al. 2017; Trabalón et al. 2017), however, report mass fractions in biota in ng/g of wet weight (ww), which facilitates assessment of human intake and comparison with Environmental Quality Standards (EQS) in biota set in the EU Directive 2013/39/EU for the evaluation of potential ecotoxicological risk of certain pollutants in aquatic environments. The EQS in biota refers to the sum of mass fractions of six PBDE congeners—BDE 28, 47, 99, 100, 153, and 154—and is set to 0.0085 ng/g ww. This is the limit mass fraction below which no harmful effects are expected in wildlife or humans.

Aquatic environment

Aquatic organisms are highly susceptible to bioaccumulation and biomagnification of organic pollutants, which can cause serious health problems, especially in species at the top of food chains. Accordingly, consumption of fish has been recognized as one of the main sources of human exposure to organic pollutants through food, and in these terms, fish is

probably the most investigated sample for PBDE contamination (Sühring et al. 2016; Anh et al. 2017; Govaerts et al. 2018).

Many studies have investigated PBDE levels and distribution in aquatic environments all over the world (Sühring et al. 2016; Aigars et al. 2017; Annunciação et al. 2017; Novak et al. 2017; Markham et al. 2018; Pei et al. 2018). Eljarrat and Barceló (2018) summarized literature data on PBDE levels in river fish samples and compared them to the corresponding EQS limits to see if they were exceeded. Taking into account even the best case scenario, the vast majority of fish samples from Europe exceeded the EQS limits at least hundred times over. Reports for Asia and North America were even worse. Authors also discuss controversy around EQS, implying that it is set too low and that it should be revised as soon as new toxicological data will be available.

Table 1 shows PBDE mass fractions in fish reported on wet weight basis. The highest levels of 11 ng/g ww were found in Norway (Govaerts et al. 2018). If we exclude this extreme, the highest level of 1.3 ng/g ww is two orders of magnitude over the EQS. Moreover, even the lowest reported level (0.03 ng/g ww) is 3.5 times higher than the EQS set for biota.

Recently, Giulivo et al. (2017) compared PBDE levels in sediment and biota samples from three European rivers, one continental (the Sava), one Mediterranean (the Evrotas in Greece), and one Alpine (the Adige, Italy). The biota samples contained all of the analyzed PBDE congeners (BDE 28, 47, 99, 100, 153, 154, 183, and 209), while none of the sediment samples contained BDE 153, 154, or 183. Furthermore, the biota samples had significantly higher sum PBDEs. BDE 209 was the most abundant congener in the continental and Alpine river sediments, while BDE 47 was the most abundant congener in the Mediterranean river sediments as well as fish samples. These findings suggest lower use of decaBDE in commercial mixtures in the Mediterranean.

The samples along the continental, Sava river were collected in four countries through which it passes, including four locations in Croatia. There, the $\sum_8$PBDE ranged from below the limit of detection (< LOD) to 16.7 ng/g dw in sediment, and from 11.9 to 461 ng/g lw in fish, which was higher than in the Mediterranean or the Alpine river. The conversion of the PBDE concentrations in fish samples from the Sava from the lipid to wet weight basis showed that all samples exceeded the EQS threshold. In general, however, all PBDE levels, regardless of the river, were comparable to other European countries. The Sava and the Adige PBDE sediment levels were also comparable to one Australian report (Anim et al. 2017). Giulivo et al. (2017) also reported that BDE 209 contributed with more than 90% to the sum of congeners. In contrast, lower PBDE levels were reported in sediment samples collected from five rivers and eight lakes in Latvia (Aigars et al. 2017). In that study, BDE 209 was not detected in any of the sediment samples but was dominant in fish samples. For comparison, several orders of magnitude higher PBDE levels were detected in sediment samples collected around a flame retardant manufacturing plant in China (Song et al. 2016), along the second largest Chinese Yellow River (Huang He) (Pei et al. 2018), and in the vicinity of a sewage treatment plant on the south coast of Korea (Lee et al. 2018).

Air and soil

According to literature, atmospheric levels of PBDEs depend on deposition processes, meteorological conditions, long-range atmospheric transport, and the vicinity of PBDE sources to the sampling site (urban/industrial vs. background locations). Between 2011 and 2014, Degrendele et al. (2018) monitored atmospheric PBDE levels (BDE 28, 47, 85, 99, 100, 153, 154, 183, and 209) at a background site in central Europe, which is known to have no sources of PBDEs. Their findings indicated an increase in lower-brominated PBDE congeners in the atmosphere on the global scale, most likely because of debromination of higher brominated congeners by photolysis. As if to confirm that assumption, higher brominated PBDE levels (BDE 99, 100, 153, and 209) decreased over the same period. A similar PBDE profile was reported in background air in Europe by Besis et al. (2017). Pozo et al. (2016), in turn, reported lower air PBDE levels for coastal areas of Sicily. In addition, only three (BDE 47, 99, and 100) of the 26 analyzed PBDEs were detected routinely. To estimate the main factors affecting PBDE air mass concentrations in Spain, Roscales et al. (2018) collected a larger number of seasonal air samples at five urban and seven background sites between 2008 and 2015. The analysis included 14 PBDEs (BDE 28, 47, 66, 85, 99, 100, 153, 154, 183, 184, 191, 196, 197, and 209). Urban air samples had higher median $\sum_{14}$PBDE than the background samples, but the single highest levels were measured in two samples collected at the same background site in 2014. In both cases, this finding was owed to a rise in BDE 209. This congener also dominated the rest of the air samples, regardless of the sampling site. The authors also reported no significant reduction in PBDE levels in Spanish air over the observed period, despite the European ban and global regulations limiting the production and usage of PBDEs. In comparison to Spain, a heavily industrialized region in Turkey had much higher air PBDE levels (Cetin et al. 2019). Air samples were collected from 23 sites every month for one year, and eight PBDE congeners were analyzed (BDE 28, 47, 99, 100, 153, 154, 183, and 209). Again, BDE 209 was the dominant congener. Air samples collected at industrial/urban locations had the highest PBDE levels, followed by urban, suburban, and rural locations. Seasonal variations were also observed: summer had higher average PBDE air levels (118.5 ± 98.7 pg/m^3) than winter (79.7 ± 59.1 pg/m^3). This may be owed to greater volatilization from

Table 1 ΣPBDEs (number of analyzed congeners in parenthesis) in aquatic environment (sediment, fish, water), air and soil analyzed worldwide

	Location	Sample type			Reference
Aquatic environment		Sediment [ng/g dw]	Fish [[a]ng/g ww or [b]ng/g lw]	Water [ng/L]	
	Antarctica (Terra Nova Bay)	0.19–1.68 (14)		0.06–0.15 (14)	Vecchiato et al. 2015
	German Bight (North Sea)	n.d. – 0.20 (8)	0.058–0.216 [a] (8)		Sühring et al. 2016
	China (Shandong province)	10.07–69,300 (9)			Song et al. 2016
	Latvia (3 rivers and 7 lakes)	0.01–0.13 (8)	0.13–0.82 [a] (8)	<LOD (8)	Aigars et al. 2017
	Croatia, Slovenia (northern Adriatic Sea, the Sava river)		0.28–0.83 [a] (7)		Novak et al. 2017
	Spain (Tarragona County)		0.4–1.3 [a] (8)		Trabalón et al. 2017
	Atlantic, Pacific and Indian Ocean		0.03–1.06 [a] (9)		Nicklisch et al. 2017
	Italy (the Adige River) Greece (the Evrotas River)	0.26–10.8 (8) <LOD – 4.52 (8)	18.7–187 [b] (8) 9.32–116 [b] (8)		Giulivo et al. 2017
	Slovenia, Croatia, Bosnia and Herzegovina, Serbia (the Sava River)	<LOD – 16.7 (8)	11.9–461 [b] (8)		
	Australia (the Brisbane River)	0.01–12.4 (8)			Anim et al. 2017
	Argentina (Buenos Aires)	0.16–2.02 (10)			Tombesi et al. 2017
	Vietnam (e-waste recycling area)	1.31–1710	21.6–1380 [b] (8)		Anh et al. 2017
	Africa (the Ga-Selati River) Norway (the Laagen and Rena river)	0.042–0.077 (7) 0.033–0.406 (7)	<LOQ – 0.29 [a] (7) 0.19–11 [a] (7)		Govaerts et al. 2018
	India (Mumbai, Thane creek)	15.98–132.72 (15)			Tiwari et al. 2018
	USA (South Carolina)		0.1–0.99 [a] (8)		Fair et al. 2018
	Korea (Tongyeong Bay)	2.18–307 (19)		1.58–6.94 (19)	Lee et al. 2018
	China (the Yellow River)	0.035–23.4 (14)		0.49–17.4 (14)	Pei et al. 2018
		Soil [ng/g dw]	Air (gas) [pg/m^3]	Air (gas + particles) [pg/m^3]	
Air and soil	Arctic (Ny-Ålesund)	0.011–0.089 (12)			Wang et al. 2015
	Antarctica (Terra Nova Bay)	0.40–33 (14)			Vecchiato et al. 2015
	Italy (Sicily Island)		>LOD – 2.5 (26)		Pozo et al. 2016
	UK (Birmingham)	2.3–49* (13)		100–490 (13)	Drage et al. 2016
	Australia (Melbourne)	<LOD - 13,200 (8)			Mcgrath et al. 2016
	Japan; China; South Korea; Vietnam; India	1.0–1000; 0.46 – 800; 0.46–94; 0.20–2.4; 0.06–13 (23)			Li et al. 2016
	Greece, Turkey			0.807–18.8 [c] (8), 0.949–4.72 [d]	Besis et al. 2017
	Turkey (Istanbul)		110 and 620 (12)		Kurt-Karakus et al. 2017
	Argentina (Buenos Aires)	0.04–10.7 (10)			Tombesi et al. 2017
	China (Qingyuan)	31.3–2130	11–431		Y. Wang et al. 2017

Table 1 (continued)

Location	Sample type		Reference
Czech Republic (Košetice Observatory)		0.084–6.08 [c] (9) <LOQ – 4.72 [d]	Degrendele et al. 2018
France (Greater Paris)	0.3–13 (8)		Gaspéri et al. 2018
Spain (5 urban sites)		4.09–25.7 (14)	Roscales et al. 2018
Spain (7 background sites)		2.63–12.8 (14)	
Turkey (Dilovasi)	0.15–286 (8)	5.73–520 (8)	Cetin et al. 2019
Arctic (Ny–Ålesund)	0.011–0.09 (12)	0.79–2.19 (12)	Ma et al. 2019
Antarctica (King Georgia Island)	0.003–0.052 (12)	0.27–0.71 (12)	
China (Changzhou)	2.21–18,451 (13)		Xu et al. 2019

dw, dry weight; *ww*, wet weight; *lw*, lipid weight * ng/g organic matter

[c] sum of mass concentrations of all detected PBDE congeners except BDE 209 [d] mass concentration of BDE 209

nearby sources at higher temperatures. Another Turkish study of Istanbul air (Kurt-Karakus et al. 2017) reported similar PBDE levels (BDE 17, 28, 47, 66, 85, 99, 100, 138, 153, 154, 183, and 209) for urban (620 pg/m^3), suburban (280 pg/m^3), and rural (110 pg/m^3) neighborhoods.

Air-to-soil transfer is one of the main sources of soil PBDE contamination. Higher brominated PBDE congeners (> 5 bromine atoms) create strong bonds with organic matter in soil. Soils are therefore important secondary sources of pollution (Law et al. 2006; Ma et al. 2019), especially in the vicinity of industrial activities involving PBDEs (Newton et al. 2015; Cetin et al. 2019; Ma et al. 2019; Xu et al. 2019), including e-waste recycling (Wang et al. 2017; Ohajinwa et al. 2019). This was confirmed by Li et al. (2016), who investigated PBDE levels in soil samples collected at urban, rural, background, and industrial/e-waste recycling locations in Japan, China, South Korea, Vietnam, and India. BDE 209 dominated among 23 congeners in the majority of samples, and its mass fraction was more than 20 times higher than that of the sum of the remaining 22 congeners at some sampling locations. Urban locations had the second highest $\sum_{23}$PBDE, followed by rural, and background locations. By country, soil $\sum_{23}$PBDE followed this order: Japan > China > South Korea > India > Vietnam in urban (450 > 75 > 39 > 3.4 > 1.1 ng/g, respectively) and rural (161 > 28 > 10 > 0.79 > 0.75 ng/g, respectively) locations. As expected, the highest PBDE levels were observed at e-waste recycling and BFR industrial sites. This industrial impact on PBDE levels has also been reported by Xu et al. (2019). $\sum_{13}$PBDE in soils collected at three plastic manufacture plants and their vicinity ranged from 2.21 to 18,451 ng/g dw, with an overall mean of 1004 ng/g dw. These levels decreased with the distance from the contaminated area. Again, BDE 209 was the most common and also the most abundant congener.

Significantly lower mass fractions were detected in soils sampled across Azerbaijan (Aliyeva et al. 2018). Only six out of ten analyzed PBDE congeners were detected, and the mean and median of Σ_6PBDE were 167 and 91.1 pg/g dw, respectively. Curiously enough, mean Σ_6PBDE in soils from industrial sites were not significantly different from mean Σ_6PBDE in soils from non-industrial areas. The authors suggested that in their case, PBDE levels were affected by wider regional sources instead of significant point sources inside the country.

There are only a few PBDE soil level studies outside Asia. Eight PBDE congeners of environmental concern (BDE 28, 47, 99, 100, 153, 154, 183 and 209) were analyzed in soils collected in the UK (Drage et al. 2016), France (Gaspéri et al. 2018), and Australia (Mcgrath et al. 2016). In the UK and France, PBDE levels were associated with urbanization. BDE 209 was dominant across all these studies, with the highest levels measured in e-waste recycling sites (Mcgrath et al. 2016). The domination of BDE 209 was also reported by

Tombesi et al. (2017) in soils sampled from different locations of Bahía Blanca city and the surrounding region (Argentina).

Due to long-range atmospheric transport and persistence, PBDEs were also detected in soil samples collected at as remote sites as the Arctic and Antarctica. Their levels measured kept mainly at the pg/g level (Wang et al. 2015; Ma et al. 2019), but Vecchiato et al. (2015) suggested that research stations in Antarctica could be significant sources of PBDEs and similar compounds, where they found soil levels as high as 33 ng/g dw.

Indoor dust

Dust as a significant source of human exposure to PBDEs has attracted a lot of attention over the recent years, especially in terms of PBDE burden in a variety of indoor environments (Zhu et al. 2015; Civan and Kara 2016; Anh et al. 2017; Kurt-Karakus et al. 2017; Muenhor and Harrad 2018; Ohajinwa et al. 2019; Kuo et al. 2019; Rantakokko et al. 2019; Tao et al. 2019). Table 2 summarizes the data published over the last five years. Comparisons between studies can be challenging because of the differences in dust sampling methods, size, and type of the area that has been vacuumed (specific room or entire household), presence of specific products which could increase PBDE levels, and the timing of sampling. Allgood et al. (2017) found that dust settled at elevated surfaces and dust settled on the floor has a different PBDE profile, and that human exposure assessments will much depend on the place from which the accumulated dust is sampled. Furthermore, PBDE levels in dust vary by season. In university laboratories in China, winter recorded the highest levels, and the variations coincided with changes in BDE 209 levels (Jin et al. 2018). Studies from different parts of the world compared PBDE dust levels in a variety of indoor environments, such as households, offices, stores, classrooms, cars, and theaters (Zhu et al. 2015; Sun et al. 2016; Cristale et al. 2016; Kademoglou et al. 2017; Kurt-Karakus et al. 2017; Muenhor and Harrad 2018). Just like in air and soil, their distribution in dust samples also depended on the level of urbanization, i.e., they were higher in urban/industrialized areas than in rural areas. Some studies also suggest that the presence of electronic devices in the indoor environment increases PBDE levels in dust (Sun et al. 2016; Allgood et al. 2017; Muenhor and Harrad 2018; Sugeng et al. 2018).

Historically higher use of PBDEs in the USA and Canada has also reflected on much higher PBDE levels in indoor dust than in Europe and Asia. Allgood et al. (2017) reported very high median mass fractions of Σ_{10}PBDE in dust samples (up to 23,508 ng/g) collected at various locations of the University of California campus in Irvine, USA. In Europe, dust samples collected across UK had PBDE mass fractions nearly as high as those reported in the USA and Canada. This may be related to stringent fire safety regulations in the UK and the widespread use of carpets (Tao et al. 2016; Kademoglou et al. 2017). In other European countries (Turkey, Norway, Sweden, Finland, and Spain) the highest PBDE levels in dust were at least one order of magnitude lower (Civan and Kara 2016; Cristale et al. 2016; Kademoglou et al. 2017; Kurt-Karakus et al. 2017; Rantakokko et al. 2019; Tao et al. 2019).

Venier et al. (2016) compared PBDE indoor levels of three countries: the USA, Canada, and the Czech Republic. Median $\sum_{10}$PBDE in dust, air, and window films followed this order: USA > Canada > Czech Republic (3650 > 1770 > 163 ng/g; 148 > 60 > 3 pg/m^3, and 7.0 > 6.5 > 0.98 ng/m^2, respectively). In contrast, Wong et al. (2017) found no significant differences in total PBDE dust levels between Australia, UK, Canada, Sweden, and China.

As concerns PBDE profiles in dust, a number of studies singled out BDE 209 as the dominant congener (Zhu et al. 2015; Cristale et al. 2016; Kim et al. 2016; Venier et al. 2016; Kademoglou et al. 2017; Korcz et al. 2017; Kurt-Karakus et al. 2017; Rantakokko et al. 2019; Tao et al. 2016, 2019). In dust from Australia, the UK, Sweden, and China, BDE 209 dominated in the congener profile, ranging from 50 to 70% of total PBDEs, while in Canada, it accounted for only 20% of total PBDEs (Wong et al. 2017). BDE 209 in house dusts was also reported to vary a lot, especially in studies with a large number of samples. For example, in a study of PBDE levels in 129 house dusts collected in Warsaw, Poland, BDE 209 ranged from 36 to 336,000 ng/g (median 270 ng/g) (Korcz et al. 2017). In indoor dust samples collected in China, BDE 209 ranged from 8.36 to 37,400 ng/g (median 1090 ng/g) (Zhu et al. 2015). Although the components of the decaBDE mixture taking over the dominance in indoor air and dust due to the phase-out of commercial pentaBDE (Björklund et al. 2012), this is not yet true for areas with high historical use of the pentaBDE mixture.

PBDEs in humans

Like other POPs, PBDEs have a tendency to accumulate in lipid-rich compartments. This is why breast milk is the most common and practical tool used for human biomonitoring. It is an ideal bioindicator not only of infant but also of human exposure and the sampling method is not invasive. In a large systematic review, Zhang et al. (2017) summarized global research data on PBDE concentrations in human breast milk specimens collected from 2000 to 2015. The most commonly reported PBDE congeners were BDE 28, 47, 99, 100, 153, 154, and 183. The medians of total PBDE mass fractions ranged from 19.9 to 54.5 ng/g lw in North America, from 0.4 to 6.3 ng/g lw in Europe, and from 1.5 to 11.5 ng/g lw in Asia. In other words, PBDEs were about 20 times higher in North America than in Europe or Asia. These findings

Table 2 ΣPBDEs (number of analyzed congeners in parenthesis) in dust samples (expressed as ng/g dust) and human samples (expressed as ng/g lipid weight (lw)) analyzed worldwide

	Location	Sample type			Reference
Dust		House dust	Office dust	Industry dust	
	China (different provinces)	0.445–939 (13)[a,*] 8.36-37,400[b,*]			Zhu et al. 2015
	Turkey (Kocaeli)	29.32 - 4790 (14)			Civan and Kara 2016
	Spain (Barcelona)	3.0–184 [a] (7)* 1103–14,990 [b] *			Cristale et al. 2016
	Korea (9 cities)	99.5–4878 (36)			Kim et al. 2016
	UK (Birmingham)	180–370,000 (8)	270–110,000 (8)		Tao et al. 2016
	USA (Bloomington, Indiana)	47–4410 [a] (10) 75–7450 [b] (10)			Venier et al. 2016
	Canada (Toronto)	53–8360 [a] (10) 223–4860 [b] (10)			
	Czech Republic (Brno)	0.05–193 [a] (10) 16–788 [b] (10)			
	China (Hangzhou)	56.5–2207 (8)	188–4357 (8)		Sun et al. 2016
	South Korea (Ansan, Jeju, Pyungchon, Seoul)	645.8–6360.1 (21)			Shin et al.2016
	Turkey (Istanbul)	400–12,500 (12)	330–32,200 (12)		Kurt-Karakus et al. 2017
	Vietnam	25.6–69 (8)	250–300 (8)	8–8740 (8)	Anh et al. 2017
	UK UK	25.5–7325 (17)[a] 33.0–107,000 (17)[b]	16.6–1500 (17)[a] 728–40,000 (17)[b]		Bramwell et al. 2017
	Thailand (different provinces)	0.59–260 (10)			Muenhor and Harrad 2018
	Sweden (Stockholm)		82–1400 (17)		Tao et al. 2019
	China (Guangzhou)	118–27,980 (20)			Tang et al. 2019
		Blood	Human milk	Umbilical cord	
Humans	Uppsala (Sweden)	1.0–11 (10)	0.53–6.6 (10)		Darnerud et al. 2015
	Denmark Finland		1.22–111.1 (7) 1.47–19.02 (7)		Antignac et al. 2016
	France		0.45–15.27 (7)		
	USA (California; 2003–2005) USA (California; 2009–2012)		11.2–435 (19) 10.1–1310 (19)		Guo et al. 2016
	South Korea (Ansan, Jeju, Pyungchon, Seoul)	<LOQ – 13.6 (21)	0.3–19.8 (21)	<LOQ – 49 (21)	Shin et al. 2016
	North America Asia		44.7–88.9 (7) 2.4–3.3 (7)		Zhang et al. 2017
	Europe		2.2–3.1 (7)		
	USA (Atlanta)	6.5–937.1 (7)**			Darrow et al. 2017
	USA (Knoxville)	9.94–61.8 (35)		17.3–120 (35)	Terry et al. 2017
	UK	0.78–12.6 (17)[a] < 1.13–19.8 (17)[b]	1.33–21.0 (17)[a] < 0.19–1.04 (17)[b]		Bramwell et al. 2017
	USA (New York)	0.45–20.20 (11)**		0.29–11.59 (11)	Cowell et al. 2018
	China (Shanghai)	0.280–12.330 (8)			Xu et al. 2018
	Australia (Queensland)	0.88–26 (7)**			Drage et al. 2019
	USA (Puget Sound region, WA)	< 2.5–310 (12)			Kuo et al. 2019
	China (Beijing)		0.288–22.2 (8)		Chen et al. 2019
	Uganda (Nakaseke, Kampala)		0.59–8.11 (12)		Matovu et al. 2019

* dust samples collected in private houses and public places (e.g., school, university, theater) ** children blood samples

LOQ, limit of quantification [a] sum of mass concentrations of all detected PBDE congeners except BDE 209 [b] mass concentration of BDE 209

strongly suggest that this population is still exposed to some PBDE sources. Furthermore, significantly higher PBDE levels were reported in the breast milk of women living near e-waste recycling plants for more than 20 years than in the breast milk of women living there for less than three years, which confirms that e-waste recycling is an important source of PBDEs (Li et al. 2017).

Many investigations have focused on children as the most vulnerable group. Darrow et al. (2017) reported higher serum PBDE levels in children aged between 1 and 6 years than Cowell et al. (2018) did in children aged between 2 and 9 years. Both studies were conducted in the USA, and in both, BDE 47 was the most abundant congener. Cowell et al. (2018) reported the highest mean concentrations of BDE 47, 99, and 100 at the age of two years, which confirmed that toddler are the most vulnerable group in terms of exposure. They also showed that the levels of PBDE congeners from the pentaBDE mixture significantly decreased over the study period, which is consistent with the fact that its use ceased in the meantime. A similar significant decrease in serum PBDE levels was reported in Australian children after the ban of commercial penta and octaBDE mixtures (Drage et al. 2019). The only research of PBDE levels in children from Europe is the one by Drobná et al. (2019) in 6-year-olds from Slovakia. Their data for individual PBDE congeners (as they did not report the sum) showed relatively low levels compared to other countries. Even so, the authors found an association between PBDE exposure and poor preschool maturity test results.

Information about BDE 209 levels in humans is scarce, because it was not analyzed by most of the human studies. Those few that did include it in analysis did not detect it or the detection rate was very low, mostly because BDE 209 has a much higher limit of detection than other congeners (Darrow et al. 2017; Cowell et al. 2018; Drobná et al. 2019; Kuo et al. 2019). Darnerud et al. (2015) investigated the time trend of BDE 209, 47, 99, 100, and 153, in pooled blood serum samples collected from Swedish first-time mothers between 1996 and 2010. They detected only BDE 153 and 209 in all samples (BDE 209 had the highest mean mass fraction of 1.27 ng/g lw), while other congeners were below the quantitation limit (LOQ) in more than 70% of the samples. Linear regression analysis showed that the levels of BDE 47, 99 and 100 decreased significantly in serum during the study period, BDE 153 showed an increasing trend, while there was no significant trend for BDE 209. In addition, the authors compared serum with breast milk PBDEs in matched samples and found significant correlations between levels of seven BDE congeners (28, 47, 100, 153, 154, 183, and 209). These correlations were weaker for higher brominated BDE congeners (Darnerud et al. 2015). In an investigation of serum PBDE levels of the residents of Washington, median Σ_{11}PBDE (without BDE 209) was 28.70 ng/g lw (Kuo et al. 2019). The highest mass fraction had BDE 209 (617.07 ng/g lw), but it was detected in only 13% of the samples, probably because of its relatively high LOD. The same study reported high levels of BDE 209 in dust samples collected at the workplace of the same people whose serum was analyzed.

Final remarks

PBDE findings reported in the last five years confirm that measures taken to minimize the use of these contaminants did not result in lower PBDE levels in the environment. Research has also showed that PBDE levels always reflect the proximity of a contamination sources, such as manufacturing or sewage treatment plants, landfills, and/or e-waste recycling industries. By far, the highest mass fractions of PBDEs, especially of BDE 209, have been reported in soil samples collected at industry locations in China (up to 19 μg/g dw).

Indoor dust PBDE levels are far higher in North America and the UK than in the rest of the world as a consequence of intensive BFR use in the past, and although this trend is declining, human exposure is still notable. Exposure through contaminated food has also been evidenced. Most of the fish analyzed in the last five years contained PBDE levels above the EU limit. In addition, the levels of lower-brominated PBDE congeners seem to persist or even increase in the environment as a result of the degradation of higher brominated congeners, mainly BDE 209 (Zeng et al. 2010).

All these findings indicate that the efforts made so far to eliminate PBDEs from the environment and reduce their negative impact on humans are not sufficient and that PBDEs shall remain a significant global environmental problem for many years to come.

This is why new measures need to be taken to remove the remaining greatest source of contamination, solid waste, and e-waste in particular. These measures should define efficient means to achieve a sustainable waste management goal. Informal recycling, one of the major issues of waste management in developing countries, needs to be integrated into the formal waste management sector. This is crucial for the health of people involved in recycling, but also for the environment and mankind in general.

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