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# Human exposure to polybrominated diphenyl ethers (PBDEs) through the diet: An update of the scientific literature

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame-retardants (BFRs). As for other persistent organic pollutants, dietary intake (followed by dust inhalation) is the main route of human exposure to PBDEs. In 2012, we reviewed the scientific literature on the concentrations of PBDEs in foodstuffs and their dietary exposure. The current review is aimed at updating the results of recent studies (2012–2022) focused on determining the levels of PBDEs in food samples, as well as the dietary intake of these compounds. We have revised studies conducted over the world. The current information on the concentrations of PBDEs in food and their dietary intake is now much more notable than that available in our previous review, being China the country contributing with the highest number of studies. Because of the important differences in materials and methods used in the available studies, the comparison of results is certainly complicated. However, there seems to be a general trend towards a decrease in the levels of PBDEs in foods, and consequently, in the dietary intake of these contaminants. The lack of tolerable daily intakes of PBDEs is an issue that needs to be solved for assessing human health risks of these BFRs.

## 1. Introduction

Flame retardants (FRs) are chemical substances applied to a number of different materials such as furnishings, electronics and electrical devices, building and construction materials, and transportation products (among others), in order to prevent the start (or to slow) the growth of a fire. Although the number of FRs is considerable, they are usually grouped into a few categories according to their chemical structures and properties. Among these categories, until recent years, the most used -and also studied-has been made up by the polybrominated diphenyl ethers (PBDEs). Due to their persistence and bioaccumulation, as well as their potential adverse/toxic effects on the environment and human health, various traditional FRs, including most congeners of PBDEs, have been already banned or strictly phased out (Xiong et al., 2019; Wang et al., 2020a; Symthe et al., 2022).

Artificially synthesized PBDEs, which have been widely applied as additive brominated flame retardants (BFRs) in consumer products since 1970s, have consequently led to an extensive release into the environment (Xu et al., 2019; Wang et al., 2020b; Hou et al., 2021; Ma et al., 2022; Turner, 2022). Since the characteristics of persistence, bioaccumulation, long-distance transportation, and adverse health effects of PBDEs, penta-BDEs, octa-BDEs and deca-BDEs were listed as persistent organic pollutants (POPs) in the United Nations Stockholm Convention in 2009 (UNEP, 2009; Hou et al., 2021; Sharkey et al., 2020; Wang et al., 2020a). In addition, the analogues of PBDEs, hydroxylated (OH-) and methoxylated (MeO-) PBDEs have also received public attention (Liu et al., 2018).

As a direct consequence of the environmental distribution of BFRs in general, and PBDEs in particular, in recent years a number of studies have been conducted in relation to human exposure to these environmental pollutants, as well as their potential adverse health effects. Most studies have concluded that food is a major pathway of exposure to BFRs -including PBDEs- for non-occupationally exposed individuals (Johnson-Restrepo and Kannan, 2009; Frederiksen et al., 2009; Bramwell et al., 2016; Shi et al., 2018; Tay et al., 2019; Liu et al., 2020; Esplugas et al., 2022). Human exposure to BFRs -including PBDEs- is still an issue of great concern. Consequently, the biomonitoring of PBDEs body burden has been carried out in various populations over the world (Petreas et al., 2011; Linares et al., 2015; Tao et al., 2017; He et al., 2018; Cowell et al., 2019; Liu et al., 2020; Orta et al., 2020; Wu et al., 2020; Yu et al., 2020). For our part, in addition to the periodical measurements of the levels of PBDEs in foodstuffs conducted in our

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laboratory, we have also determined the concentrations of PBDEs in human milk and adipose tissue (Meneses et al., 1999; Schuhmacher et al., 2007, 2009, 2013) in order to establish the potential correlations between human exposure to these pollutants and their body burden levels.

In 2000–2002, we performed -to the best of our knowledge the first up to that date-a Total Diet Study (TDS) aimed at assessing the dietary exposure to PBDEs by the population of Catalonia, Spain (Bocio et al., 2003). Information about the dietary intake of PBDEs was very scarce. In the scientific literature, at that time we only found four studies on that topic (Bocio et al., 2003). In a subsequent survey, we determined the concentrations of PBDEs in 14 species of fish and shellfish, which were selected among the most consumed by the population of Catalonia (Domingo et al., 2006). We also investigated the cooking-induced changes in the levels of PBDEs in various foodstuffs (Perelló et al., 2009). In the last two decades, we have periodically reviewed the scientific literature available worldwide on the levels of PBDEs in food and the human exposure through the diet (Domingo, 2004, 2012 Domingo et al., 2008).

Following this same line, in order to update the information available on this subject, we have prepared this new review, which covers the period between our last paper (Domingo, 2012) and the present (June 2022). Our review is again based only on studies available in the scientific literature. Reports and/or data of studies about risk assessment of PBDEs, conducted by regional, national or international agencies and/or food safety authorities are not included in the present review. The databases Pubmed (https://pubmed.ncbi.nlm.nih.gov/) and Scopus (https://www.scopus.com/) have been used with the terms for the search: "food consumption", "dietary intake", "polybrominated diphenyl ethers" and "PBDEs". Information on the available studies and their results are next summarized. It is presented as classified into continents and specific countries, and chronologically ordered. However, the year of publication of the reviewed studies does not necessarily correspond with the year in which the study was performed or the samples were collected.

## 2. Levels of PBDEs in food and human dietary exposure

### 2.1. Asian countries

#### 2.1.1. China

In recent years, China has been the country with the largest number of papers reporting the concentrations of PBDEs in foods and the human dietary intake of these compounds. We next summarize the results of the studies carried out in China here revised, which are presented following the order of publication.

Lei et al. (2015) assessed the human daily intake and uptake of multiple environmental contaminants -including 10 PBDEs-, which were estimated based on 18 animal-based foods collected from markets in Shanghai. The health risks due to food consumption were also assessed. The estimated intake and uptake considering the contaminant bioaccessibility via single food consumption were: 9.4–399 and 4.2–282 ng/kg body weight (bw)/day for adults, and 10.8–458 and 4.8–323 ng/kg bw/day for children, respectively. In turn, taking into account multiple food consumption, the intake and uptake values were 0.2–104 and 0.05–58.1 ng/kg bw/day, and 0.2–119 and 0.06–66.6 ng/kg bw/day for adults and children, respectively. Regarding only PBDEs, the total mean intake was estimated in 0.2 ng/kg body weight/day for both evaluated population groups, adults and children. Gong et al. (2015) determined the concentrations of PBDEs (BDE-28, 47, 99, 100, 153, 154, and 183) in 206 samples of animal-derived foods (pork, egg, fish, and milk samples) widely consumed by the population of Hubei and the middle reaches of the Yangtze River. The dietary intake of PBDEs was subsequently estimated. The highest levels of ∑PBDEs were detected in chicken eggs (0.191 ng/g wet weight (ww)), followed by duck eggs (0.176 ng/g ww), pork (0.050 ng/g ww), carps (0.047 ng/g ww), and cow milk (0.013 ng/g ww). The highest concentration of ∑PBDEs (4.830 ng/g ww) was found in one sample of pork, while PBDEs were not detected in a sample of milk. The mean levels of ∑PBDE ranged from 0.013 ng/g ww to 0.191 ng/g ww in milk and chicken eggs, respectively. Chicken eggs (65.9%) and pork (23.4%) were the most important contributors to the dietary intake of ∑PBDEs through the analyzed animal-derived foods, being 157.5 pg/kg bw/day the estimated dietary intake of ΣPBDEs for a standard adult of 60 kg.

Li et al. (2015) determined the levels of various BFRs (BDE-209 was included among them) in 30 food samples: local fish and shellfish, 5 types of vegetables, and 3 types of meat, in addition to air (14 samples) and indoor dust samples (13), collected in Weifang City (Shandong Province), which is an important industrial zone in North China. Human exposure to BFRs was also estimated. BDE-209 (together with DBDPE) was the dominant BFRs in all analyzed samples. The mean levels of BDE-209 (1400 pg/g ww) in meats were of the same order than those found in fish and shellfish (1800 pg/g ww), and the levels detected in vegetables (1300 pg/g ww). The dietary exposure to BDE-209 for male adults was estimated to be 7400 pg/kg bw/day. This PBDE congener contributed 85% to the total BFR intake in the area under evaluation. Shi et al. (2016) reported the concentrations in food composites and the dietary exposure to novel BFRs (NBFRs), among which, interestingly, PBDEs were already excluded. That study was based on the 5th Chinese TDS, conducted in 2011. In a previous study (the 4th Chinese TDS, 2007), PBDEs had been still included. Four food groups were analyzed for NBFRs: eggs and egg products, aquatic foods, milk and milk products, and meat and meat products. The mean estimated daily intake of total NBFRs via food consumption for a standard Chinese man was 4.77 ng/kg bw. Levels of DBDPE were several orders of magnitude higher than those of other NBFRs and were higher than or similar to those of legacy BFRs, including PBDEs. The authors remarked that this would suggest an evident shift in the consumption pattern between PBDEs and DBDPE in China. The same research group (Shi et al., 2017), and also based on the 5th Chinese TDS, assessed the dietary exposure to 3 widely used BFRs, including BDE-209. The estimated mean daily intake of BDE-209 via food consumption for a standard Chinese man was 0.96 ng/kg bw/day. Meat and meat products were the main contributor to the daily dietary intake to the three evaluated BFRs, including BDE-209. In relation to meat and meat products, in various countries -including China-viscera organs such as liver and intestines are widely consumed. Specifically, regarding chicken, a frequent practice is also to use chicken offal to feed other livestock, which could eventually cause human exposure to certain contaminants. Thus, to assess human health effects, to know the distribution patterns of pollutants such as PBDEs in chicken tissues at different growth stages is of considerable relevance. In this sense, Wang et al. (2017) measured the concentrations of ∑PBDEs (sum of BDE-28, 47, 99, 100, 153, 154, 183 and 209) in chicken tissues of different growth stages, and estimated the dietary intake of these PBDEs and its main metabolites via consumption of chicken. Tissue concentrations of ∑PBDEs followed the sequence: liver > blood > skin > intestine > stomach > leg meat > breast meat. The average dietary intakes via the consumption of chicken tissues of ∑PBDE were estimated to be 319 and 1380 ng/day for liver, 211 and 632 ng/day for leg meat, and 104 and 311 ng/day for breast meat, for adults and children, respectively. As analogues of PBDEs, hydroxylated (OH-) and methoxylated (MeO-) PBDEs have also been a subject of interest. Regarding this, Liu et al. (2018) analyzed in marine algae, invertebrates and fish species, collected from Dalian (near Chinese Bohai Sea), the concentrations of OH-PBDEs, MeO-PBDEs and PBDEs. In fish muscle, the total amounts of PBDEs and MeO-PBDEs accounted for over 96%. In contrast, the amount of OH-PBDEs was very low. With respect to the intake of these compounds, it was found that the contribution of OH-PBDEs, MeO-PBDEs and PBDEs via seafood consumption meant that 6-MeO-BDE-47 was the predominant chemical contributing to the dietary intake, with a value of 0.4 ng/kg/day, followed by BDE-47 (0.3 ng/kg/day) and 2′-MeO-BDE-68 (0.2 ng/kg/day). Fish contributed to 70%, 35% and

53% of the dietary intake of MeO-PBDEs, OH-PBDEs and PBDEs, respectively. Human exposure to PBDEs associated with the consumption of hens, has been also assessed by Cai et al. (2018) in Guangzhou. Samples of muscle, liver, fat, yolk, and ingluvies tissues were analyzed for 13 PBDE congeners (BDE-17, BDE−28, BDE−47, BDE−66, BDE−71, BDE−85, BDE−99, BDE−100, BDE−138, BDE−153, BDE−154, BDE−183 and BDE−190). The highest median concentrations of ∑PBDEs were found in the ingluvies (5.30 ng/g), followed by muscle (2.53 ng/g). In contrast, the lowest levels corresponded to the yolk (0.09 ng/g). The estimated intake of PBDEs was estimated to range from 0.08 to 0.31 ng/kg/day, with median and mean values of 0.15 and 0.16 ng/kg/day, respectively.

Huang et al. (2018) analyzed the levels of various halogenated FRs (HFRs) (including 18 congeners of PBDEs: BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-138, BDE-153, BDE-154, BDE-183, BDE-196, BDE-197, BDE-202, BDE-203, BDE-206, BDE-207, BDE-208 and BDE-209) in home-produced chicken eggs from Baihe village (Longtang, South China) 3 and 6 years after a previous study conducted by the same group in 2010 (Zheng et al., 2012). Longtang is known as "the renewable copper city of China", having been the largest *e*-waste recycling centers in Qingyuan, Guangdong Province. Human dietary exposure to HFRs via consumption of chicken eggs was also assessed. Mean PBDE levels in home-produced eggs were 14,100, 4736 and 4741 ng/g in 2010, 2013 and 2016, respectively. The concentrations of PBDEs and the rest of HFRs decreased significantly between 2010 and 2013–2016. The estimated dietary intakes for PBDEs via consumption of home-produced eggs by the adult population of the zone were 317, 120 and 129 ng/kg bw/day in 2010, 2013 and 2016, respectively. In order to provide specific recommendations for better managing the environmental pollution, as well as the human health risks from PBDEs, Yang et al. (2018) assessed human multiple exposure (soils, dust, outdoor and indoor air, human milk and foods) to these environmental contaminants. The levels of PBDEs in samples of various regions of China were obtained from the literature for the period January 2005–December 2016. Food samples included meat, fish, eggs, vegetables, milk and rice. According to the published data, total concentrations of PBDEs (sum of BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) in food ranged between 0.03 ng/g in vegetable samples of Shanghai, and 5.9 ng/g in meat samples from Guangdong. BDE-209 was the primary congener, accounting for 38–99% of the estimated daily intake. The results of that survey indicated that for infants, the dominant exposure pathway to PBDEs was human milk, while dust ingestion was a higher contributing factor for toddlers, children, teenagers and adults.

Wang et al. (2019a) analyzed the concentrations of 8 PBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) in 53 samples of chicken, duck, pig, and fish feeds, which were purchased from Guangxi, Hubei, Anhui, and Guangdong Provinces. BDE-209 was detected in all the animal feeds and raw materials. The remaining 7 PBDE congeners were detected at a frequency range between 11 and 79%. The median $\sum_8$PBDE levels ranged from 0.34 to 1.2 ng/g dry weight (dw). In that study, chicken was used as a representative animal to assess exposure risks of PBDEs via food intake. For adults, the HQ values of $\sum_8$PBDEs via intake of chicken leg and breast meat were estimated to be $9.6 \times 10^{-4}$ and $8.2 \times 10^{-4}$, respectively. In turn, Wang et al. (2019b) determined the dietary exposure of adults to PBDEs through commonly consumed animal- and plant-based foodstuffs from Beijing. The median levels of total PBDEs in animal-based foodstuffs were between 3.22 and 13.7 ng/g, with BDE-209 being the most abundant congener (at least 85% of total PBDEs). The dietary intake of PBDEs for adults was 9.77 ng/kg bw/day, being meat consumption the primary source of dietary intake. On the other hand, Jian et al. (2020) analyzed the concentrations and distribution of 27 legacy PBDEs in 9 food categories (105 different foods), which were purchased from three markets in Nanjing. The congeners BDE-7, BDE-15, BDE-17, BDE-28, BDE-49, BDE-71, BDE-47, BDE-77, BDE-99, BDE-126, BDE-153, BDE-196 and BDE-209 were detected in at least one of the 105 food samples. The detection frequencies of ΣPBDEs in the 9 food categories ranged from 16.7% (in nuts and fruits) to 100% (in meat), being BDE-17 the most detected congener in all food categories. The mean concentrations of ΣPBDEs in the 9 food categories ranked as follows: aquatic food (0.834 ng/g ww), meat (0.694 ng/g ww), poultry (0.223 ng/g ww), dairy products (0.146 ng/g ww), vegetables (0.098 ng/g ww), eggs (0.072 ng/g ww), nuts and fruits (0.068 ng/g ww), cereals (0.042 ng/g ww), and sugar (0.020 ng/g ww). The mean and 95th percentile levels of total exposure through the diet to ΣPBDEs were 154.5 ng/day and 553.4 ng/day, respectively.

The human dietary exposure to PBDEs was also assessed by means of a 3-days duplicate diet study (Wang et al., 2020b). For it, a 20 nursing-mother cohort with the assistance of the Beijing Children's Hospital participated in the survey. Duplicate diet samples (including snacks, fruits and beverages) were collected at home during 3 consecutive days. The mean and median concentrations of ΣPBDEs (sum of the congeners BDE-28, 47, 99, 100, 153, 154, 183 and 209) in the 60 duplicate diet samples were 449 and 306 pg/g ww, respectively, with BDE-209 showing the highest contribution. The mean and median intakes of ∑PBDEs were 4.61 and 3.69 ng/kg bw/day, respectively. On the other hand, Fan et al. (2021) reported the conclusions of a study aimed at determining the exposure levels and temporal trends of a number of POPs in various foodstuffs, as well as their associated daily intake in China. PBDEs were among the examined POPs. The authors used 270 reports to establish their conclusions. It was found that the concentrations of PBDEs in food decreased significantly at a rate of 0.26 ± 0.11 *Ln* (ng/kg)/year since 2005, a decrease relatively higher than that observed for most other POPs. The daily intake of PBDEs for adults was 3.3 ng/kg. Recently, Pei et al. (2022) has published the conclusions obtained by means of the 6th Total Diet Study (China) regarding exposure to PBDEs. The levels of $\sum_7$PBDEs, were dominated by aquatic products with 39.85 pg/g ww, followed by meats and eggs: 29.75 and 22.19 pg/g ww, respectively. By contrast, the lowest levels corresponded to dairy products and plant origin food samples. For Chinese adults, the mean dietary intake of $\sum_7$PBDEs was 0.24 ng/kg bw/day, which was lower than those obtained in the 4th and 5th TDS.

### 2.1.2. Pakistan

Mahmood et al. (2015) measured the concentrations of 8 PBDEs congeners (BDE-28, BDE-35, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-183) in samples of food crops (wheat and rice), collected in January 2013 and June 2013, respectively, from urban/industrial, peri-urban/industrial and rural areas of Gujranwala division (Punjab Province). ∑PBDEs concentration ranged from 0.30 to 1.43 ng/g, and from 0.07 to 46.0 ng/g, with mean values of 0.70 and 4.50 ng/g for wheat and rice, respectively. The estimated daily intakes for wheat and rice ranged between 0.002 and 0.035 pg/kg bw/day, and 0.033 and 0.680 pg/kg bw/day, respectively. In a more recent study of the same research group (Mahmood et al., 2020), the levels of 8 PBDE congeners (BDE- 28, 35, 47, 99, 100, 153, 154 and 183) were determined in composite samples of wheat grain, collected at 13 sampling sites across the Grand Trunk (GT) Road from Lahore to Peshawar. The mean concentrations of ∑PBDEs was 0.49 ng/g (range: ND-3.47 ng/g), with BDE-47, BDE-99 and BDE-100 being the predominant congeners. It was concluded that potential risks to human health through the dietary intake of wheat were marginal.

### 2.1.3. Japan

Using the 24-h duplicate-diet approach, Fujii et al. (2021) evaluated the dietary exposure to various classes of organohalogenated compounds -including 7 congeners of PBDEs: BDE- 28, 47, 99, 100, 154, 153 and 183- among 46 Japanese infants (<2 years old). For this, 46 whole-day meals of infants (7–24-months old) were collected during 2017 in Fukuoka. Only BDE-47 was detected, with estimated median and maximum intake values of 0.11 and 4.5 ng/day, respectively.

#### 2.1.4. India

Until recently, systematic assessments on the exposure and health risks of endocrine-disrupting chemicals (EDCs) such as PBDEs, had not been conducted in India. Sharma et al. (2021) assessed human dietary exposure to various EDCs (including PBDEs) in India, and compared the results with those corresponding to human dietary exposure in European countries. The most frequently consumed food items (fruits, vegetables, pulses, cereals, dairies, egg, fish, and meats) in the country were selected based on data from a survey conducted in 2018 by the Quality Council of India. Foodstuffs were collected in Delhi (urban zone) and Dehradun city (rural zone) (Uttarakhand State, Himalayan region). Ten congeners of PBDEs (BDE- 28, 47, 66, 85, 99, 100, 153, 154, 183 and 209) were analyzed. An ubiquitous occurrence of several PBDE congeners was found in the Delhi food basket, particularly in fish, eggs, poultry and goat meat. The dietary intake of PBDEs for adults was estimated in 0.7 ng/kg bw/day comparable with data from previous European surveys.

#### 2.1.5. Taiwan

To assess the dietary exposure to PBDEs by the population of Taiwan, Chang et al. (2017) conducted, between 2010 and 2013, a TDS including 600 samples of a number of foodstuffs (meats and poultry, livestock, eggs, fish and other seafood, dairy products, meats, vegetables, rice, oils and infant foods). The highest mean levels of the sum of the 24 congeners of PBDEs analyzed in that study was found in oils (1418 pg/g fresh weight), followed by eggs (960 pg/g fw) and livestock (950 pg/g fw). Subsequently, the dietary intake of PBDEs was estimated for various population groups, being the highest mean intake found in 0-3-year-olds (9.38 ng/kg bw/day), while the lowest one corresponded to 16-18-year--old girls (3.35 ng/kg bw/day). Studies conducted in other Asian countries (Pakistan, Korea, Taiwan and India) are also now available.

#### 2.1.6. Korea

Na et al. (2013) measured the concentrations of 19 congeners of PBDEs (BDE- 15, 17, 28, 33, 47, 49, 66, 71, 85, 99, 100, 119, 126, 138, 153, 154, 183 and 209) in various groups of foodstuffs (cereals, meats, eggs, dairy products, fish and shellfish) purchased from five Korean cities. PBDE intake was estimated for the Korean population according to the geographical location and age. The mean dietary intake of total PBDEs corresponding to four large cities and one rural city was 72.30 ng/day, being the largest contribution to this intake due to the consumption of fish and shellfish (48.96 ng/day), while those from meats and cereals were 16.05 ng/day and 5.27 ng/day, respectively. In turn, Nguyen et al. (2014) assessed the levels of 24 PBDEs in Korean foods by means of a progressive TDS, in which all analyzed samples (meats, fish and shellfish, cereals, vegetables, fruits, dairy products, beverages, and some other miscellaneous items) were prepared as table-ready, either to be cooked or raw edible. The congeners BDE-47, 99 and 153 were detected in almost all samples, with ratios depending on the matrix composition. The levels of BDE-154, 206 and 207 were relevant in some samples of meats, fish and shellfish and dairy products. The mean dietary intake of PBDEs was estimated in 63 ng/day, corresponding the highest value to the 19–39-year old group, and gradually decreasing with the age. Recently, Choi and Lee (2021) reported the results of a survey (2015–2017) aimed at investigating the temporal trend of the levels and the total dietary exposure to PBDEs through the same seafood groups (fish, crustaceans, cephalopods and bivalves) that had been analyzed in 2010–2011. The levels of 19 congeners of PBDEs (BDE-17, -28, −47, −49, −66, −71, −77, −85, −99, −100, −119, −126, −138, −153, −154, −156, −183, −184 and −191) in seafood and their dietary intake were measured, being then the data compared with those of the previous survey. The mean concentration of $\Sigma_{19}$PBDE was 0.35 ng/g ww (0.01–1.60 ng/g ww). Herring (1.60 ng/g ww), followed by Spanish mackerel (1.00 ng/g ww) and tuna (0.80 ng/g ww) were the species showing the highest PBDE level. The total PBDE dietary intakes was 9.86 ng/day (0.17 ng/kg bw/day). The rate of decrease during the 7-year period was 17% for the mean concentrations of PBDEs, while that corresponding to the dietary intake of PBDEs during the same period was 58%.

On the other hand, during the period covered by the current review, two reviews on the levels of PBDEs in foodstuffs in their dietary intake have been published. That of Mackintosh et al. (2015) was aimed at reviewing the occurrence and profiles of PBDEs in the Philippines, while the review of Kaw and Kannan (2017) was focused on the levels of PBDEs in South Asia, with a special attention to Malaysia.

### 2.2. USA

In USA, most studies on the levels of PBDEs in food and the dietary intake of these compounds were conducted in the 10 first years of the current century, being the Arnold Schecther's group the most active (see review by Domingo, 2012). The results of recent studies of the topic are next discussed. Chen et al. (2017) measured the levels of 12 PBDE congeners (BDE-17, 28, 47, 49, 66, 85, 95, 99, 100, 153, 154 and 183) in store-bought bovine milk distributed by eight commercial producers in the state of California, between August and September 2014. The most prevalent PBDE congeners in whole milk were BDE-47, BDE-99 and BDE-49: 18, 9.9 and 6.0 pg/ml (geometric means), respectively. The sum of PBDEs in milk samples suggested close proximity to industrial emissions. The intake of PBDEs through cows' milk was not estimated in that study. In turn, Lupton and Hakk (2017) analyzed the concentrations of 7 PBDE congeners in samples of beef, pork, chicken, and turkey collected in 2012–2013. The mean ΣPBDE concentrations were 0.40, 0.36, 0.19 and 0.76 ng/g lipid weight (lw), respectively, being BDE-47 and BDE-99 the main contributors. The estimate daily intake of PBDEs from meat and poultry for an US consumer was 6.42 ng.

Taking into account that some OH- and MeO-PBDEs, produced by simple marine organisms, are bioaccumulated by marine shell- and finfish, Cade et al. (2018) conducted an initial survey of PBDE, OH-PBDE and MeO-PBDE content in commonly consumed seafood items available to residents living in the Puget Sound region (Washington State). Samples of sole, trout, tuna (canned), pollock, sablefish, salmon, rockfish, clams, mussels, shrimp, calamari and scallops were analyzed for a number of congeners of PBDEs and OH- and MeO-PBDEs. BDE-47 and BDE-99 were the most common congeners detected. Clams and mussels had much higher levels of OH- and MeO-PBDEs than other types of seafood, being 6′–OH–BDE-47 and 2′-MeO-BDE-68 the most common OH- and MeO- congeners, respectively. The intake rates for Washington State residents were estimated between 34 and 644 ng PBDEs/day, depending on the specific species consumed.

### 2.3. Europe

#### 2.3.1. Spain

Santín et al. (2013) determined the occurrence and levels of PBDEs in 48 fish samples collected during 2010 from four river Spanish basins. Seven PBDE congeners were detected: BDE-28, BDE-47, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209. Total PBDE concentrations ranged from ND to 520 ng/g lw. The highest levels were found in the Llobregat river basin (55.9–520 ng/g lw). An estimation of the intake of PBDEs was not done because these fish species are not commercialized, and therefore, there were no reliable data on the potential consumption. In our laboratory, we determined the occurrence and levels of 8 PBDEs and 8 MeO-PBDEs in samples from 10 species of fish and shellfish that are widely consumed by the population of Tarragona County (Catalonia, Spain) (Trabalón et al., 2017). BDE-47 was the congener with the highest contribution to the ΣPBDEs, while BDE-100, BDE-183 and BDE-209 were not detected in any sample. The highest levels of ΣPBDEs corresponded to salmon (1.3 ng/g ww), with sole, tuna, cod and hake also presenting relatively high concentrations of ΣPBDEs (1.2, 0.8, 0.8 and 0.9 ng/g ww). In turn, mussel was the species with the highest level of MeO-PBDEs (1.5 ng/g ww). The estimated exposure to ΣPBDEs through the consumption of the fish and shellfish species analyzed was

0.45 ng/kg bw/day for a male adult.

In the Region of Valencia, Pardo et al. (2014) estimated the intake of PBDEs resulting from the consumption of fish and seafood over the period 2007–2012. Twenty-five species were selected based on the consuming habits of Valencia. The congeners BDE-28, BDE-47, BDE-49, BDE-66, BDE-99, BDE-100, BDE-119, BDE-139, BDE-153, BDE-154, BDE-155 and BDE-183 were analyzed. The mean ∑PBDEs was 803.4 (lower bound, lb) and 3790.2 (upper bound, ub) pg/g ww, being BDE-47 the congener showing the highest level (356.1 and 581.4 pg/g ww, lower and upper bound, respectively). The estimated mean intakes of PBDEs were 0.093 (lb) and 0.178 (ub) ng/kg bw/day for adults and 0.101 (lb) and 0.196 (ub) ng/kg bw/day for children. In a subsequent study conducted by the same research group (Pardo et al., 2020) the occurrence and levels of PBDEs were determined in foods of animal origin and in vegetable oils from the Region of Valencia. Thirty-two food items from 5 categories (vegetable oils, meat and meat products, eggs, milk and dairy products, and fish and seafood) were analyzed. Vegetable oils, and fish and seafood were the food groups showing the highest content of PBDEs, with mean levels of 503 and 464 pg/g ww for total PBDEs, respectively, in the ub). BDE-47 was the predominant in fish and seafood, meat and meat products, and vegetable oils, while BDE-99 dominated the categories of eggs, and milk and dairy products. The mean intakes (ub scenario) were 1.443 and 3.456 ng/kg bw/day for adults and young people (6–15 years old), respectively.

### 2.3.2. Norway

Xu et al. (2017) assessed the human exposure to PBDEs (BDE- 28, 47, 66, 85, 100, 153, 154, 183 and 209) through dietary intake. The participants (n = 61) in the study provided one duplicate diet sample corresponding to foods and drinks ingested over a 24-h period (November 2013–May 2014). BDE-209 was the major PBDE congener with a median of 0.045 ng/g ww, followed by BDE-47 (median 0.010 ng/g ww). Fish was the major dietary route for PBDE exposure. The estimated median dietary intake of ΣPBDEs was 1.3 ng/kg bw/day. In turn, the levels of PBDEs (BDE- 28, 47, 99, 100, 153, 154 and 183) were determined in the main commercial fish species harvested in and near Norwegian waters by Nøstbakken et al. (2018). The levels of ∑PBDEs in individual fillet samples of fish ranged from the Σub limit of quantifications (LOQs) in Atlantic cod fillet to 39.5 μg/kg in Atlantic halibut. The main contributor to ∑PBDEs in all 20 analyzed species was BDE-47 (about 65% of ∑PBDEs). Although the dietary intake of ∑PBDEs through fish consumption was not estimated, the margins of exposure (MOE) of BDE-47, BDE-99 and BDE-153 were evaluated. The lowest MOE was 1.0 for BDE-99 calculated for consumption of North Sea herring and Atlantic mackerel, using the European dietary surveys. Recently, Ho et al. (2021) reported the results on an investigation focused on establishing patterns of co-occurrence and geographical variation of seafood elements and contaminants including total arsenic, mercury and selenium, and selected POPs (dioxins, PCBs and PBDEs) in a wide array of marine fish taxa from the North East Atlantic Ocean (NEAO). The survey used seafood datasets (>25,000 subjects) and comprised 12 commercially important fish species collected between 2006 and 2019 in the NEAO. However, no specific information for PBDEs was given as the main goal of that survey was focused on the co-occurrence of contaminants in marine fish, without estimating the dietary intake of the specific pollutants.

### 2.3.3. Italy

Martellini et al. (2016) determined the levels of a number of PBDEs (BDE-7, BDE-15, BDE-17, BDE-28, BDE-49, BDE-71, BDE-47, BDE-66, BDE-77, BDE-100, BDE-119, BDE-99, BDE-85, BDE-126, BDE-154, BDE-153, BDE-138, BDE-156, BDE-184, BDE-183 and BDE-191) in various groups of foodstuffs (samples of meats, eggs, milk, cheese, fish, fish oil, mussels and clams collected in 2011–2012) and estimated the hazard indices (HI) of PBDEs by Italians, through the analyzed foodstuffs. PBDEs were detected in all the analyzed samples. The concentrations ranged from 0.11 pg/g fw (BDE-153, in fish) to 18,537 pg/g fw (BDE-209, in dairy products). The highest levels of total PBDEs were detected in dairy products (18,537 pg/g fw), meat (12,672 pg/g fw), and eggs (9729 pg/g fw). The HIs of the sum of 4 PBDEs (BDE-47, BDE-99, BDE-153 and BDE-209) ranged between 1.80E-04 for eggs consumption and 5.43E-03 for dairy products. For Italy, there are also recent data on the concentrations of 15 PBDEs (congeners: BDE- 28, 47, 49, 66, 77, 85, 99, 100, 138, 153, 154, 183, 197, 206 and 209) in 5 freshwater species (fish and crustacean) from the Lake Trasimeno (Umbria region), the largest lake in Central Italy (Tavoloni et al., 2021). Mean ∑PBDEs were not estimated in 4 species (perch, tench, carp and red swamp crayfish) because all analyzed congeners were below the LOQs in almost all samples, while in eel, ∑PBDEs (15 congeners) ranged from 0.269 to 0.916 ng/g ww. In another study carried out with commercially exploited freshwater fishes and crayfish also of Lake Trasimeno, Roila et al. (2021) estimated the dietary exposure to PBDEs through the consumption of species caught in that Lake. A total of 74 freshwater fishes and 16 crayfish pools were analyzed for BDEs −28, −47, −49, −66, −77, −85, −99, −100, −138, −153, −154, −183, −197, −206 and −209. The intake of total PBDEs was found to be low, with values of 0.229 pg/kg bw/day (range: 0.000–0.206 pg/kg bw/day) and 1.113 pg/kg bw/day (range: 0.146–0.339 pg/kg bw/day), for lb and ub, respectively. The highest contribution corresponded to BDE-47 (0.112 pg/kg bw/day).

### 2.3.4. Poland

Pajurek et al. (2019) studied the role of poultry eggs (126 samples collected from these layer chicken rearing systems: free range, organic, barn and battery cage) as a source of human exposure to PBDEs (BDE-28, 47, 49, 99, 100, 138, 153, 154, 183 and 209). BDE-209 was the predominant congener regardless of the production system, being the contribution of the remaining congeners to $\sum_9$PBDEs between 22.5 and 32.9%. The mean concentrations of PBDEs ranged from 0.52 to 1.08 ng/g fat. In addition to BDE-209, BDE-47, BDE-99 and BDE-153 also made up a significant proportion, particularly in organic eggs. The same research group (Pietron et al., 2019) also determined human exposure to 11 congeners of PBDEs (BDE-28, 47, 49, 99, 100, 138, 139, 153, 154, 183 and 209) associated with meat consumption. For it, 199 muscles from sheep, cow, pigs, chicken, turkey, horse, ostrich, rabbit and farm deer from different regions of Poland were collected during 2015–2017. The mean ∑PBDEs concentrations in farm animal muscle ranged between 12.1 pg/g ww for farm deer, and 97.6 pg/g ww for horse. The estimated dietary intakes for the ∑PBDEs were 0.049 and 0.074 ng/kg bw for adults and children, respectively. Since these values are low, it was concluded that meat consumption did not pose a risk for the health of the Polish consumers. On the other hand, Falandysz et al. (2019) investigated the historical human exposure to PBDEs resulting from the supplementary intake of historical cod liver oils produced in Iceland, Norway and Poland during 1972–2017. The dietary intake of PBDEs from canned liver products retailed in Poland, of Baltic Sea origins, was also estimated. For the ΣPBDE (17 analyzed congeners), the concentrations ranged between 9.9 and 415 ng/g for oils, and between 10.5 and 13 ng/g for canned liver products. For all samples, BDE-47 was the dominant congener. The estimated weekly intakes of ΣPBDE from canned cod liver was 17.6–25.1, 10-9-15.7, and 11.7–16.7 ng/kg bw for adults, teenagers and children, respectively.

### 2.3.5. United Kingdom

Bramwell et al. (2017a) compared two methods for estimating human dietary exposure to POPs during 2011–2012: a) the 2012 Total Diet Study (TDS) conducted by the UK Food Standards Agency (FSA), and b) a 24-h duplicate diet (DD) study of 20 adults from the North East of England. PBDEs were included into the analyzed POPs. Sugar and preserves, followed by fish and seafood where the food groups with the highest lipid weight ∑PBDE concentrations. The highest TDS concentrations (ww) corresponded to BDEs-47, -153, and −99 and −209 in fish and seafood, fats and oils, and sugar and preserves, respectively. The adult dietary exposures to PBDEs as determined by 24-h duplicate diet were 274 and 292 pg/kg bw/day, for the BL and UL, respectively. The same research group (Bramwell et al., 2017b) investigated in a cohort of 20 adults, the major dust and diet sources of PBDEs in a north east England cohort (2011–2012) and the potential health risks. Two of the main goals of that study were to evaluate the relative importance of PBDE exposure via indoor dust versus dietary PBDE exposure, and to compare intake estimates with reference health values. $BDEs_{3–7}$ were detected in all of the 24-h duplicate diet samples, with BDE-209 detected in 79% of them (median and mean values of 0.73 and 0.85 ng/g lw, respectively). The 24 h duplicate diet PBDE concentrations were converted to daily dietary intake estimates which ranged from 82 to 1320 pg/kg bw for $\sum BDEs_{3–7}$ and <0.8–1860 pg/kg bw for BDE-209, which made up a median of 73% of the total PBDE dietary exposure. Diet was the main source of intake of $BDE_{3–7}$ congeners for most individuals of this cohort, with meat consumption showing the highest positive association between diet type and serum $BDEs_{3–7}$ concentrations. In turn, dust was the cohort's primary source of BDE-209.

### 2.3.6. Other european countries

In Sweden, Sahlström et al. (2015) measured the levels of tri-decaBDEs, and other BFRs in the two most important external exposure media, diet and house dust for a mother–toddler cohort. Market basket samples (fish, meat, vegetable oils, dairy products and eggs) were collected in 2010. Eight tri-decaBDEs (BDE-28, -47, −153, −197, −206, −207, −208 and −209) were detected in the Swedish market basket samples, with a mean concentration of individual compounds ranging from 0.27 to 290 pg/g fw. BDE-47 was the predominant congener in fish and meat: 290 and 7.6 pg/g fw, respectively, while BDE-209 was found in all food items originating from the terrestrial food chain (dairy products, vegetable oils, meat, eggs), with the highest concentrations found in vegetable oils, and eggs (13 and 42 pg/g fw, respectively). The mean dietary intake of ΣPBDEs was 22 (range 13–36) ng/day. In turn, Coelho et al. (2016) determined the concentrations of various POPs (including PBDEs) and emerging BFRs in duplicate diet samples from a group of volunteers (n = 21) studying or working in the University of Aveiro, Portugal, and estimated the daily intakes of the analyzed compounds. Portions of the products consumed in all meals for 7 consecutive days were collected. The estimated daily intakes for PBDEs (sum of BDE-28, 47, 99, 100, 153, 154, 183, 209) ranged between 0 and 440 ng/day, and from 560 to 1200 ng/day (LB and UB estimations, respectively).

In Turkey, Aydin et al. (2019) analyzed the concentrations of PBDEs (congeners BDE- 47, 99, 100, 153 and 154) in 15 samples of raw cow's milk and 15 commercial brands of ultra-high-temperature (UHT) cow's milk. The mean ∑PBDEs was 9.51 and 6.99 ng/g lw, for raw and UHT samples, respectively. The estimated daily intakes of ∑PBDEs were 0.64 (adults) and 1.68 μg/kg bw (children), for raw samples, and 0.56 (adults) and 1.48 (children) μg/kg bw for UHT samples. In Switzerland, Bedi et al. (2020) estimated the dietary intake of PBDEs through internationally traded seafood consumed by the Swiss population using two different approaches, trade and survey data. Regarding specific PBDE exposures based on trade data, the highest one corresponded to shrimp/prawn: 0.4914 ng/kg bw/day (origin Vietnam), followed at a notable distance by salmon from Norway: 0.0306 ng/kg bw/day. In contrast, the lowest daily intakes corresponded to seabream (Italy): 0.0054 ng/kg bw/day, and mussels (the Netherlands): 0.0052 ng/kg bw/day. The estimated PBDE exposures by surveyed fish consumers ranged between 0.011 and 43.42 ng/kg bw/day, with a median of 0.68 ng/kg bw/day. In turn, the calculated origin-specific trade-data based exposure was 0.65 ng/kg bw/day, being both values very close. Moreover, these authors also reported that the 95th percentile of the surveyed Swiss population is exposed to PBDE levels as high as 8.5 ng/kg bw/day. In Latvia, Zacs et al. (2021) measured the concentrations and profiles of several FRs, including PBDEs (congeners BDE-17, 28, 47,49,99, 100, 138, 139, 153, 154, 155, 183 and 209), in a number of samples (meat, milk and dairy products, fish and fish products, plant origin, and eggs) belonging to the most frequently consumed groups foodstuffs in that Baltic country. Among the analyzed food product groups, samples of fish and cod liver showed the highest PBDE levels, with mean concentrations of ∑PBDEs: 620 and 3484 ng/kg, respectively. The mean estimated daily intake of ∑PBDEs for the Latvian general population was estimated to be: 1.24 ng/kg bw, with 0.29 ng/kg bw (about 25% of the total) corresponding to PBDE-209.

On the other hand, Aznar-Alemany et al. (2017) assessed the presence of a series of halogenated FRs (including PBDEs and MeO-PBDEs) in various species of seafood from all around Europe. The effects of cooking on the concentrations of these contaminants were also evaluated, as well as the exposure and risk for humans caused by consuming seafood. A total of 42 samples of seafood were collected (2014–2015), including species from the Mediterranean Sea, the North Sea, and the north-east Atlantic Ocean. BDE- 28, 47 and 99 were the most occurring congeners (45.2–78.6%). In contrast, BDE-209 was not detected. With respect to the concentrations of naturally-occurring MeO-PBDEs, they ranged between ND and 305 ng/g lw. However only 11.9% of the samples showed more than 60 ng/g lw. It was observed that cooking processes concentrated PBDEs. The same research group also investigated the occurrence of PBDEs in sediment, mussels and water from different European fish and shellfish farming sites (Aznar-Alemany et al., 2018). PBDEs were detected in 94% of the mussels mostly below the LOQ (range: < LOQ-5.42 ng/g lw). BDE-28, BDE-47 and BDE-100 were the most occurring congeners, which were found in 65%, 59% and 53% of the samples, respectively. In all analyzed samples, the concentrations of MeO-PBDEs were < LOQ.

## 2.4. Data from other countries over the world

In Australia, Toms et al. (2016) determined the concentrations of various POPs (including PBDEs) in baby foods and assessed the contribution of food intake to estimate total exposure. Baby and toddler foods representing fruit-, vegetable-, meat/vegetable- and dairy-based foods were purchased in April 2013 from three supermarkets of Brisbane. These congeners were analyzed: BDEs −17, −28, −47, −49, −66, −71, −77, −85, −99, −100, −119, −126, −138, −153, −154 and −183. PBDEs were only detected in 3/33 analyzed samples, being BDEs −47 and −100 the congeners detected, ranging from below the LOD to 31.7 pg/g fw. The highest concentration: BDE-100 (31.7 pg/g fw) corresponded to poultry-based food. Consumption of a 140 g meal would result in an intake ranging from 4.4 ng/day for PBDEs. In Nigeria, Babalola and Adeyi (2018) analyzed the levels of 8 PBDEs congeners (BDE-17, -28, −47, −99, −100, −153, −154 and −183) in various foods commonly consumed by the adult population of the Southwest the country (meat products, aquatic foods, dairy products, edible oil, eggs, fruits, vegetables and cereals). The dietary intake of PBDEs was also estimated. The highest mean levels of the ΣPBDEs were 34.5, 223, 11.4, 30.5, 13.7, 7.06, 4.17 and 2.11 pg/g in meat products, aquatic foods, dairy products, edible oil, chicken eggs, fruits, vegetables and cereals, respectively. The mean dietary intake of PBDEs by an adult was estimated to be 131 pg/kg bw/day. In Brazil, Souza et al. (2021) measured the concentrations of 7 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, 183) in food samples of animal origin (eggs, different species of fish and seafood, and cow milk) purchased from markets of Sao Paulo. The ∑PBDEs were 2.29, 1.98, 1.91 and 4.42 ng/g ww in eggs, fish, seafood, and milk, respectively, being BDE-47 the most abundant congener. Based on these results, the intake of ∑PBDEs was 3.25 (range 0.02–2.19) ng/kg bw per day.

## 3. Discussion and conclusions

It has been demonstrated that dust ingestion is an important source of human exposure to PBDEs (Fromme et al., 2014; Malliari and

**Table 1**
A summary of recent publications on the occurrence of PBDEs in food and their dietary intake by the Asian population.

| Country | Samples collected | Sampling year | Congeners | Intake of PBDEs (ng/kg/day)[a] | Remarks | Reference |
|---|---|---|---|---|---|---|
| China | 175 animal-based food samples including 18 food items | 2008–2009 | Not specified | 0.2 | The cancer and non-cancer risks caused by PBDEs were negligible. | Lei et al. (2015) |
| China | 206 samples from local markets | 2010 | 28, 47, 99, 100, 153, 154, and 183 | 0.1575 | Chicken eggs (65.9%) and pork (23.4%) were the largest contributors. | Gong et al. (2015) |
| China | 30 samples | 2013 | BDE-209 | 7.400 | BDE209 contributed 85% of the total intake of brominated flame retardants in the study area. | Li et al. (2015) |
| China | 80 composite samples from four animal-origin food groups | 2011 | BDE-209 | 0.96 | Data comparison indicated an obvious shift in the industrial production and usage pattern between PBDE and non-PBDE BFRs in China. | Shi et al. (2017) |
| China | Chicken tissues | n.a. | 28, 47, 99, 100, 153, 154, 183 and 209 | 319, 211 and 104 ng/day for liver, leg meat and breast meat, respectively. | Liver clearly poses the highest exposure risk for human consumption, particularly if chickens are fed with contaminated feed. | Wang et al. (2017) |
| China | Marine biological samples from 20 species. | 2012 | 28, 47, 66, 85, 99, 153, 154 | 0.8 | Coastal residents were in higher exposure risks to OH-PBDEs and MeO-PBDEs via the massive seafood consumption | Liu et al. (2018) |
| China | 25 samples of hens from different markets | 2016 | 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, and 190 | 0.16 | The health risk of PBDEs for the general population, through the consumption of market hens in Guangzhou, was generally low. | Cai et al. (2018) |
| China | 62 home-produced eggs laid by free-range chickens | 2013 and 2016 | 28, 47, 66, 85, 99, 100, 138, 153, 154, 183, 196, 197, 202, 203, 206, 207, 208, 209 | 120–129 | The margin of exposure (MOE) values represent a significant potential health concern due to the adverse impacts of PBDEs on human neurodevelopment and fertility. | Huang et al. (2018) |
| China | 90 samples of foodstuffs from local markets | 2013 | 28, 47, 99, 100, 153, 154, 183 and 209 | 9.77 | Meat consumption was found to be the primary source of dietary intake to PBDEs. | Wang et al. (2019a) |
| China | 105 samples from 9 food categories | 2018 | 7, 15, 17, 28, 49, 71, 47, 77, 99, 126, 153, 196, 209 | 2.63 | Meat and aquatic foods were the primary sources of PBDEs to the total local dietary intake. | Jian et al. (2020) |
| China | 60 duplicate diet samples, including all foods consumed over a 3-day period. | 2016 | 28, 47, 99, 100, 153, 154, 183 and 209 | 4.61 | The EDIs obtained for both mothers and their babies disclosed a low health risk to this mother-infant cohort. | Wang et al. (2020b) |
| China | 48 composite samples from 12 food categories | 2016–2019 | 47, 99, 153, 154, 175, 183, and 209 | 0.02–1.96 | The dietary intake level decreased sharply compared with the two previous TDS. | Pei et al. (2022) |
| Pakistan | Rice and wheat grain samples | 2013 | 28, 35, 47, 99, 100, 153, 154, and 183 | 0.000625 and 0.000035 for rice and wheat, respectively. | BDE-99 was the predominant congener over others. | Mahmood et al. (2015) |
| Pakistan | Composite samples of wheat grains | n.a. | 28, 35, 47, 99, 100, 153, 154 and 183 | 1.9 | Potential hazardous risks to human health across the study area via the dietary intake of cereal foods were deemed trifling | Mahmood et al. (2020) |
| Japan | 46 duplicate diet samples | 2017 | 28, 47, 99, 100, 154, 153 and 183 | 0.11 ng/day | PBDEs were the group of POPs with the lowest dietary intake. | Fujii et al. (2021) |
| India | 96 aggregated samples from four areas | 2018–2019 | 28, 47, 66, 85, 99, 100, 153, 154, 183, and 209 | 0.7 | Dietary exposure to PBDEs by the Indian population was comparable/lower than for Europeans. | Sharma et al. (2021) |
| Taiwan | 600 samples from 15 food categories | 2010–2013 | 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207, and 209 | 4.79 and 4.06 for men and women, respectively. | ΣPBDEs dietary intake decreased with the age. | Chang et al. (2017) |
| Korea | Twenty individual types of food samples | n.a. | 15, 17, 28, 33, 47, 49, 66, 71, 85, 99, 100, 119, 126, 138, 153, 154, 183, and 209 | 72.30 ng/day | PBDEs levels were highest in infants and decreased with increasing age. | Na et al. (2013) |
| Korea | 96 types of food (n = 250) regularly consumed | 2012–2013 | 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, and 207 | 1.00 | The TDS approach using foods in the table-ready form should be used for a better estimation. | Nguyen et al. (2014) |
| Korea | 230 samples from 31 marine species | 2015–2017 | 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, and 191 | 0.17 | The human exposure to PBDEs from seafood consumption are expected to continue to decrease. | Choi and Lee (2021) |

n.a.: not available.
[a] Unless specified.

Kalantzi, 2017). However, diet is the main source of exposure to these environmental pollutants (Linares et al., 2015; Fromme et al., 2016; Wu et al., 2020). In recent years, a number of studies have investigated the toxicity of different PBDEs congeners. Experimental studies in rodents have shown that PBDEs exert effects on the liver, the nervous system and other organs, while they are associated with possible endocrine-disrupting effects, and also affect thyroid hormone homeostasis (Costa et al., 2008; Linares et al., 2015; Dorman et al., 2018). In spite of the potential toxic effects of PBDEs, tolerable dietary intakes of these contaminants are not available yet. According to the web of the European Food Safety Agency (EFSA), they are working on an update of the EFSA scientific opinions on BFRs, taking into account new occurrence data and any newly available scientific information. Notwithstanding, the last update regarding PBDEs corresponds to May 2011, when the EFSA considered 8 congeners of PBDEs (BDEs: 28, 47, 99, 100, 153, 154, 183 and 209) of primary interest (EFSA, 2011). Considering

**Table 2**
A summary of recent publications on the occurrence of PBDEs in food and their dietary intake in European countries.

| Country | Samples collected | Sampling year | Congeners | Intake of PBDEs (ng/kg/day)[a] | Remarks | Reference |
|---|---|---|---|---|---|---|
| Spain | 48 fish samples from 4 river basins | 2010 | 28, 47, 100, 153, 154, 183, and 209 | n.a. | The bioaccumulation of halogenated norbornenes is lower than that of PBDEs. | Santín et al. (2013) |
| Spain | Samples of 10 edible marine species widely consumed | 2016 | 28, 47, 99, 100, 153, 154, 183, and 209 | 0.45 | BDE47 and 28 were the congeners with the highest levels in the analyzed samples. | Trabalón et al. (2017) |
| Spain | 206 samples of 25 fish and seafood species | 2007–2012 | 28, 47, 49, 66, 99, 100, 119, 139, 153, 154, 155, and 183 | 0.093–0.178 | Salmon, mackerel and swordfish were the most contaminated species. | Pardo et al. (2014) |
| Spain | 320 composite samples of different fatty foods | 2010 | 28, 47, 49, 66, 99, 100, 138, 139, 153, 154, 155, 183, and 209 | 0.253–1.443 | The current dietary exposure to these substances does not pose a risk to human health. | Pardo et al. (2020) |
| Norway | 61 duplicate diet samples | 2013–2014 | 28, 47, 66, 85, 100, 153, 154, 183, and 209 | 1.3 | The dietary exposure for the participants were below RfDs for PBDEs. | Xu et al. (2017) |
| Norway | 9211 marine samples including both wild and farmed fish, fish feed and fish feed ingredients | 2006–2016 | 28, 47, 99, 100, 153, 154, and 183 | n.a. | A risk of BDE 99 for toddlers was observed when all exposure sources were included at upper bound levels | Nøstbakken et al. (2018) |
| North East Atlantic Ocean (Norway and other countries) | 25,631 individual samples from 12 commercially important marine teleost species | 2006–2019 | 28, 47, 99, 100, 153, 154, and 183 | n.a. | Concentrations of PBDEs increased from North to South. | Ho et al. (2021) |
| Italy | 93 samples from 9 different food groups | 2011–2012 | 15, 17, 28, 49, 71, 47, 66, 77, 100, 119, 99, 85, 126, 154, 153, 138, 156, 184, 183, and 191 | 8.43 | BDE-209 was the most abundant congener. | Martellini et al. (2016) |
| Italy | 86 samples from 5 freshwater species | 2018–2020 | 28, 47, 49, 66, 77, 85, 99, 100, 138, 153, 154, 183, 197, 206 and 209. | n.a. | PBDEs were detected only in eels and red swamp crayfish. | Tavoloni et al. (2021) |
| Italy | 90 samples of the most represented edible fishes and crayfish | 2018–2021 | 28, 47, 49, 66, 77, 85, 99, 100, 138, 153, 154, 183, 197, 206 and 209 | 0.000138–0.001113 | The data show no health risks for the central Italian population consuming freshwater fish products from Lake Trasimeno in relation to exposure to PBDE. | Roila et al. (2021) |
| Poland | 126 samples of poultry eggs | n.a. | 28, 47, 49, 99, 100, 138, 153, 154, 183, and 209 | n.a. | The husbandry systems influenced contaminant accumulation. | Pajurek et al. (2019) |
| Poland | 199 meat samples | 2015–2017 | 28, 47, 49, 99, 100, 138, 139, 153, 154, 183 and 209 | 0.049 | The highest and the lowest PBDE levels were in sheep and deer meats, respectively. | Pietron et al. (2019) |
| Poland | Cod liver oils and canned liver products | 1972–2017 | 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 183, 209 | 2.2–284.8, 4.2–17 and 1.7–6.8 for Baltic, Norwegian and Icelandic cod liver oils, respectively. | Concentrations in the oils were highest during the period from 1993 to 2001. | Falandysz et al. (2019) |
| United Kingdom | 24 duplicate diet samples | 2011–2012 | 47, 99, 153, 183, and 209 | 82 to 1320 for ∑BDEs 3–7, and <0.8–1860 pg/kg bw for BDE-209. | Diet was the major source of tri-hepta BDEs, while dust was the major source of BDE-209. | Bramwell et al. (2017b) |
| Sweden | Market basket samples | 2010 | 28, 47, 153, 197, 206, 207, 208 and 209 | 22 ng/day | Dietary intake is an important exposure route for lower brominated BDEs. | Sahlström et al. (2015) |
| Portugal | 21 duplicate diet samples | n.a. | 28, 47, 99, 100, 153, 154, 183, and 209 | 1.6–12 | Low levels of flame retardants were found. | Coelho et al. (2016) |
| Turkey | 15 samples of raw milk and 15 commercial brands of UHT milk | n.a. | 47, 99, 100, 153 and 154 | 640 and 560 ng/kg/day for raw and UHT cow's milk, respectively | EDI values for people consuming raw or UHT milk exceeded maximum residue limits of some organohalogenated pollutants. | Aydin et al. (2019) |
| Latvia | 144 food samples | 2016–2019 | 17, 28, 47,49,99, 100,138, 139, 153, 154, 155, 183 and 209 | 1.24 ng kg−1 b.w. | The estimated dietary exposures are unlikely to be of significant health concern | Zacs et al. (2021) |
| Several European countries | 42 samples from 10 seafood species | 2014–2015 | 28, 47, 99, 100, 153, 154, 183, 209 | 5.8 × 10−4 μg/(kg bw)/day in Portugal and | The cooking process concentrates PBDEs. | Aznar-Alemany et al. (2017) |

n.a.: not available.
[a] Unless specified.

the effects of PBDEs on neurodevelopment as the critical endpoint, the EFSA identifies derived benchmark doses (BMDs) and their corresponding lower 95% confidence limit for a benchmark response of 10%, $BMDL_{10}$s, for a few PBDE congeners: BDE-47, 309 μg/kg bw, BDE-99, 12 μg/kg bw, BDE-153, 83 μg/kg bw and BDE-209, 1700 μg/kg bw (EFSA, 2011). On the other hand, the US EPA has established oral reference doses (RfDs) for some PBDEs: 7, 3 and 2 μg/kg/day for decaBDE, octaBDE and pentaBDE, respectively (US EPA, 2017). In turn, the US Agency for Toxic Substances and Disease Registry (ATDSR) has set a minimal risk level for intermediate (14–364 days) oral exposure of 10, 000 μg/kg/day for decaBDE (ATSDR, 2015). Only with this available information, it is not currently possible to establish a safety exposure to PBDEs through the diet, as the above reviewed papers indicate. Regarding the carcinogenicity of PBDEs, the IARC classified these compounds as a Group 3 carcinogen (not classifiable as to its carcinogenicity to humans) based on inadequate evidence of carcinogenicity in humans, and inadequate or limited evidence in experimental animals (ATSDR, 2105). Nevertheless, the US EPA assigned a classification of "suggestive evidence of carcinogenic potential" for decaBDE (US EPA, 2017).

Since our previous review (Domingo, 2012), a considerable number of studies aimed at determining the levels of PBDEs in food and the human exposure through the diet have been carried out over the world. Stands out by far the important number of data belonging to Chinese studies. When the previous review was prepared, only 5 papers corresponding to that country (plus 2 performed in Hong Kong) were found. In contrast, 17 papers of the current review belong to studies conducted in China, which allow us to have now a much more complete view of the dietary exposure to PBDEs by the Chinese population, and consequently, the potential health risks. A summary of the most relevant results from studies conducted in China and other Asian countries are shown in Table 1. In turn, Table 2 summarizes data from studies carried out in European countries, in which an important number of surveys have been also performed in the last 10 years. However, although the current information on the concentrations of PBDEs in food and their intake through the diet is now much more notable than that available in our previous review (Domingo, 2012), the problems in drawing clear conclusions that can help to prevent potential adverse effects from dietary exposure to PBDEs remain similar. Thus, we have again observed important differences among those countries for which results have been published. This is due to the different dietary habits between regions and countries, as well as the different food items included in the respective surveys. Another important issue is the number of individual PBDE congeners analyzed in each of those studies, which are different, not only in specificity of the analyzed BDEs, but also in their number. Anyway, while the comparison of results is complicated, there seems to be a general trend towards a decrease in the levels of PBDEs in the analyzed foods, and consequently, in the dietary intake of these contaminants. This could be expected. as in recent years the environmental levels of PBDEs have followed a continuous reduction in most industrialized countries, since most congeners have been banned and/or phased out. Because of these important environmental decreases, the dietary intake of PBDEs has also diminished. Consequently, the continued reductions in the environmental concentrations of PBDEs should mean a lower human exposure to PBDEs from all sources (mainly dust and food) in the coming years. In turn, this would point to a very probable further decline in the concentrations of PBDEs in biological tissues, and therefore, also on potential health adverse effects. In agreement with these findings, we have observed a similar decreasing trend for other POPs, especially dioxins and furans (Schuhmacher et al., 1999; Marquès and Domingo, 2019; González and Domingo, 2021).

## CRediT authorship contribution statement

**Montse Marquès:** Data curation, Formal analysis, Data collection, Data analysis and interpretation, Critical revision of the article, Final approval of the version to be published. **Martí Nadal:** Data curation, Formal analysis, Data collection, Data analysis and interpretation, Critical revision of the article, Final approval of the version to be published. **José L. Domingo:** Conceptualization, Conception or design of the work, Data curation, Formal analysis, Writing – original draft, Data collection, Data analysis and interpretation, Drafting the article, Critical revision of the article, Final approval of the version to be published.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

This is a REVIEW-Article

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