

## 6. POTENTIAL FOR HUMAN EXPOSURE

### 6.1 OVERVIEW

PBDEs have not been identified in any of the 1,832 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (ATSDR 2015). However, the number of sites evaluated for PBDEs is not known.

The widespread use of PBDEs since the late 1970s has resulted in their presence in the environment. PBDEs were released into the environment from their manufacture and use as additive flame retardants in thermoplastics in a wide range of products (WHO 1994a). Waste containing PBDEs may be incinerated as municipal waste, deposited in landfills, discharged to municipal sewage-treatment plants, or emitted directly to the atmosphere as particulates (Darnerud et al. 2001).

Adsorption of PBDEs generally increases as bromination of PBDEs and organic carbon content of soil and sediment increase. As a result, most PBDEs have little or no mobility in soil and are not expected to leach (e.g., into groundwater). PBDEs, particularly lower BDE homologs (e.g., tri- and tetraBDE), have the potential for long-range transport in the atmosphere (Dodder et al. 2000). The detection of PBDEs in remote regions of the world suggests that long-range transport of these congeners is occurring (Dickhut et al. 2012; Hung et al. 2010). Biodegradation is a slow environmental fate process for PBDEs, but under certain conditions, some PBDEs compounds (e.g., decaBDE) may degrade by direct photolysis to form lower-brominated congeners. However, determining the rate and extent of degradation processes (e.g., biodegradation and photolysis) for PBDEs, such as decaBDE and pentaBDE commercial mixtures, is still an active area of research.

Studies of the biota indicate that lower-brominated congeners (e.g., BDE 47) are being preferentially bioconcentrated. Lower-brominated diphenyl ether (e.g., tetra- and pentaBDE) concentrations increase with respect to trophic level; thus, organisms that reside higher on food chains tend to have higher concentrations of these brominated diphenyl ethers (Shaw et al. 2009). Body-burden data indicate that the general population is exposed to PBDEs through a variety of pathways (CDC 2015; Lorber 2008; Trudel et al. 2011). The primary exposure pathway to PBDEs for residents of North America is through indoor dust contact (ingestion and dermal exposure) (EPA 2010; Lorber 2008). Dust contact is also the primary exposure pathway for BDE 209 in the United Kingdom. For Europeans, food consumption appears to be the primary exposure pathway for most congeners (Abdallah and Harrad 2014; Law et al. 2008; Trudel et al. 2011). Body burden data, as well as intake modeling, suggest that infants and toddlers

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have higher exposures to PBDEs as compared to older children or adults. PBDE levels increase from infant to toddler and then PBDE concentrations gradually decrease at older ages. Most studies indicate that concentrations of PBDEs in body fluids and tissues are a factor of 10–100-fold higher for individuals living in the United States compared to individuals living in other regions of the world (e.g., Europe).

## 6.2 RELEASES TO THE ENVIRONMENT

The Toxics Release Inventory (TRI) data should be used with caution because only certain types of facilities are required to report (EPA 2005). This is not an exhaustive list. Manufacturing and processing facilities are required to report information to the TRI only if they employ 10 or more full-time employees; if their facility is included in Standard Industrial Classification (SIC) Codes 10 (except 1011, 1081, and 1094), 12 (except 1241), 20–39, 4911 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4931 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4939 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4953 (limited to facilities regulated under RCRA Subtitle C, 42 U.S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited to facilities primarily engaged in solvents recovery services on a contract or fee basis); and if their facility produces, imports, or processes  $\geq 25,000$  pounds of any TRI chemical or otherwise uses  $>10,000$  pounds of a TRI chemical in a calendar year (EPA 2005).

The widespread use of PBDEs from the 1970s until 2013 resulted in their presence in the environment. PBDEs were released into the environment from their manufacture and use in a wide range of consumer products (WHO 1994a). PBDEs were used as additive flame retardants in thermoplastics. Additive flame retardants are physically, rather than chemically, combined with polymers. Thus, there is a possibility that some PBDE congeners may diffuse out of the treated materials to some extent (EU 2001). Although these substances are no longer manufactured in the United States and Europe, the disposal of consumer products that contain penta-, octa-, and decaBDE will result in their continued release to the environment. Waste from products containing PBDEs may be incinerated as municipal waste, deposited in landfills, or discharged to municipal sewage-treatment plants (Darnerud et al. 2001).

### 6.2.1 Air

Estimated releases of 2,180 pounds (~0.9 metric tons) of decaBDE to the atmosphere from 34 domestic manufacturing and processing facilities in 2014, accounted for about 1.5% of the estimated total

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environmental releases from facilities required to report to the TRI (TRI14 2016). These releases are summarized in Table 6-1. There are no TRI data for penta- or octaBDE.

The estimated release of decaBDE from the 2014 TRI continues to reflect a decreasing trend as production slowed and was eventually discontinued in 2013. The total on-site and off-site releases of decaBDE since 1998 are illustrated in Figure 6-1 (TRI14 2016).

No quantitative information was located on the releases of the pentaBDE technical mixtures to the atmosphere from its former production and use. However, the release of pentaBDE technical mixtures to air had the potential to occur during the curing phase, since the polyurethane foam was at elevated temperatures (e.g., up to 160°C) for several hours during this phase. Since pentaBDE technical mixtures were additive flame retardants, they are subject to volatilization or leaching from the polymer matrix during the lifetime of the use of the foam article. Losses of foam particles containing the substance (e.g., due to abrasion) may also occur. However, most congeners in pentaBDE technical mixtures have very low vapor pressures (see Table 4-3) and therefore, losses from polyurethane foam due to volatilization would be expected to be low. Migration of pentaBDE technical mixtures from consumer products may be a significant diffuse source of lower-brominated congeners of pentaBDE technical mixtures to the atmosphere. Although no studies were found that determined the migration rate of pentaBDE technical mixtures from polymers into the air, estimates have been made. The estimated migration rate for pentaBDE technical mixtures is 0.39% per year (Danish EPA 1999).

Similarly, no quantitative information is available on emissions of octaBDE technical mixtures to the atmosphere from production operations. The major sources of air emissions of octaBDE technical mixtures were thought to be a result of grinding and bagging operations.

The EPA National Center for Environmental Assessment (NCEA), Office of Research and Development completed a comprehensive exposure assessment of PBDEs (EPA 2010). A series of studies were summarized that estimated the release of PBDEs from various products under laboratory conditions. Two computer workstations manufactured after 2000 consisting of a monitor, computer, keyboard, mouse, and printer were used for 93 and 150 days and PBDE concentrations were monitored during their operation. BDE 47, BDE 100, BDE 99, and BDE 85 concentrations in surrounding air were <0.3 ng/m<sup>3</sup> for one of the workstations; however, concentrations of BDE 47, BDE 100, and BDE 99 were 150, 28, and 61 ng/m<sup>3</sup>, respectively, in air monitored for the second workstation. An emission test was summarized that used the back panel of a television set treated with octaBDE manufactured before 1979. Maximum

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**Table 6-1. Releases to the Environment from Facilities that Produce, Process, or Use Decabromodiphenyl Ether<sup>a</sup>**

State <sup>c</sup>	RF <sup>d</sup>	Reported amounts released in pounds per year <sup>b</sup>							Total release	
		Air <sup>e</sup>	Water <sup>f</sup>	UI <sup>g</sup>	Land <sup>h</sup>	Other <sup>i</sup>	On-site <sup>j</sup>	Off-site <sup>k</sup>	On- and off-site	
		AR	2	120	0	0	5,374	0	120	5,374
CA	1	1	0	0	139	0	1	139	140	
CT	1	0	0	0	0	1,747	0	1,747	1,747	
GA	3	347	0	0	2,917	0	347	2,917	3,264	
IL	3	0	0	0	5	0	0	5	5	
IN	1	5	0	0	0	0	5	0	5	
KS	1	NR	NR	NR	NR	NR	NR	NR	NR	
MA	2	18	5	0	84	1	23	85	108	
MS	1	28	0	0	0	0	28	0	28	
NH	1	NR	NR	NR	NR	NR	NR	NR	NR	
NJ	1	0	0	0	0	150	0	150	150	
NV	1	NR	NR	NR	NR	NR	NR	NR	NR	
NY	1	0	0	0	137	0	0	137	137	
OH	4	505	0	0	75	112,501	505	112,576	113,081	
PA	3	1,152	0	0	6,919	0	1,152	6,919	8,070	
SC	5	0	0	0	67	0	0	67	67	
TX	2	5	0	0	6,182	0	5	6,182	6,187	
VA	1	0	0	0	2,504	0	0	2,504	2,504	
Total	34	2,180	5	0	24,403	114,399	2,185	138,802	140,987	

<sup>a</sup>The TRI data should be used with caution since only certain types of facilities are required to report. This is not an exhaustive list. Data are rounded to nearest whole number.

<sup>b</sup>Data in TRI are maximum amounts released by each facility.

<sup>c</sup>Post office state abbreviations are used.

<sup>d</sup>Number of reporting facilities.

<sup>e</sup>The sum of fugitive and point source releases are included in releases to air by a given facility.

<sup>f</sup>Surface water discharges, waste water treatment-(metals only), and publicly owned treatment works (POTWs) (metal and metal compounds).

<sup>g</sup>Class I wells, Class II-V wells, and underground injection.

<sup>h</sup>Resource Conservation and Recovery Act (RCRA) subtitle C landfills; other onsite landfills, land treatment, surface impoundments, other land disposal, other landfills.

<sup>i</sup>Storage only, solidification/stabilization (metals only), other off-site management, transfers to waste broker for disposal, unknown

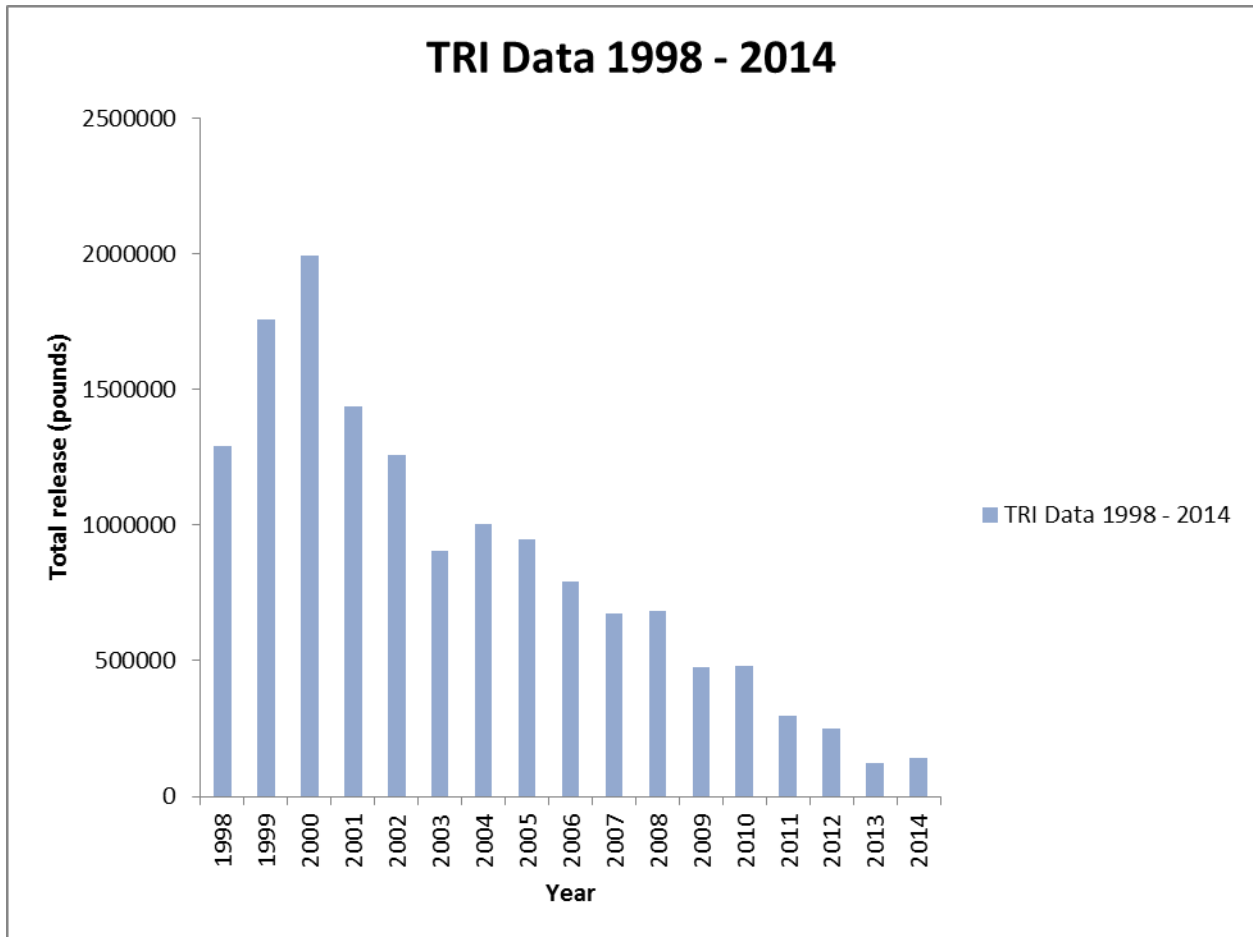
<sup>j</sup>The sum of all releases of the chemical to air, land, water, and underground injection wells.

<sup>k</sup>Total amount of chemical transferred off-site, including to POTWs.

NR = not reported; RF = reporting facilities; UI = underground injection

Source: TRI14 2016 (Data are from 2014)

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**Figure 6-1. Total On- and Off-Site Releases of Decabromodiphenyl Ether, 1998–2014**

Source: TRI14 (2016)

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concentrations of BDE 28, BDE 47, BDE 66, BDE 100, and BDE 99 were reported as 0.5, 8, 0.24, 0.27, and 0.84 ng/m<sup>3</sup>, respectively (EPA 2010). These data suggest that older treated consumer products may continue to release PBDEs long after they were originally treated.

More generalized approaches were summarized that could be used to estimate the possible total volatilization of PBDEs from treated plastic products (EPA 2010). The EU utilized a regression derived equation to estimate the percentage loss of PBDE that volatilizes from plastic components treated with PBDEs:

$$\text{Percent Volatilized} = 1.1 \times 10^6 \times VP \times SL$$

where VP is the vapor pressure in units of mm Hg and SL is the service life of the product, assumed to be 10 years

For instance, using a vapor pressure of  $3.47 \times 10^{-8}$  mm Hg for decaBDE, the volatilization loss after 10 years would be approximately 0.38% (EPA 2010). Since approximately 6,710 metric tons of decaBDE were used in plastics the EU before it was banned, the total loss to air over the assumed 10-year lifetime would be approximately 25.5 metric tons (EPA 2010).

Breivik et al. (2002) developed a regression-derived equation using the octanol-air partition coefficient ( $K_{OA}$ ) to estimate emission factors of PCBs from commercial sealants, which was also applied to estimate the emission factors of PBDEs

$$\log EF = -0.839 \times \log K_{OA} + 4.83$$

The emission factor (EF) is the ratio of the mass of PBDE emitted divided by the mass PBDE used per year. Both equations above were used to estimate total emissions of penta-, octa-, and decaBDE from products used in the United States (EPA 2010).

### 6.2.2 Water

Estimated releases of 5 pounds (~0.002 metric tons) of decaBDE to surface water from 34 domestic manufacturing and processing facilities in 2014, accounted for <1% of the estimated total environmental releases from facilities required to report to the TRI (TRI14 2016). These releases are summarized in Table 6-1.

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Industrial and urban effluents are sources of PBDEs to surface waters and sediments. Limited data on industrial and urban effluents were located for the United States. Hale et al. (2002) measured the concentration of PBDEs in soil and stream sediments collected near a polyurethane manufacturing plant (near the Dan River, Virginia). Summed concentrations of BDE 47, BDE 99, and BDE 100, the dominant congeners in these samples, ranged from <1 to 132  $\mu\text{g}/\text{kg}$  ( $\text{ng}/\text{g}$ ) dry weight. In 1995, sediment samples were collected up- and downstream near an area where the Swedish plastics industry used brominated flame retardants (Sellström and Jansson 1995; Sellström et al. 1998a). Samples were analyzed for tetraBDEs (50  $\text{ng}/\text{g}$  dry weight) and pentaBDEs (sum of three congeners, 2,300  $\text{ng}/\text{g}$  dry weight). These PBDEs were found in higher concentrations downstream of the plant than upstream, which indicates that the plastics industry was the most likely source of these compounds. Surficial sediment samples were collected at eight locations along River Viskan near several textile manufacturing facilities that used various brominated flame retardants in the production of textiles. The concentrations of BDE 47, BDE 99, BDE 100, and BDE 209 in sediments increased as samples were collected further downstream where additional industries were located (Sellström et al. 1998a). The lowest concentrations of PBDEs were found upstream of the textile industries. The combined concentration of BDE 47, BDE 99, and BDE 100 ranged from not detected to 120  $\text{ng}/\text{g}$  ( $\mu\text{g}/\text{kg}$ ) dry weight; the concentration of BDE 209 ranged from not detected to 16,000  $\text{ng}/\text{g}$  ( $\mu\text{g}/\text{kg}$ ) dry weight. Allchin et al. (1999) surveyed the concentrations of PBDEs in sediments from several rivers and estuaries in Great Britain. Sediments were collected upstream and downstream of suspected sources of pentaBDE and octaBDE, including a manufacturer, several industries, landfills, and a reference site. The highest concentrations of BDE 47, BDE 99, pentaBDE (as BDE 71), and octaBDE (as BDE 79) were in sediments near or downstream from a manufacturing site at Newton Aycliffe in River Skerne. The highest concentrations of decaBDE (as BDE 83) were found downstream of a sewage-treatment plant on River Calder. High concentrations were also detected on River Skerne downstream of a manufacturing site. BDE 99 concentrations were identical or slightly higher than BDE 47 in most sediments (Allchin et al. 1999). The sum of five pentaBDE congeners (BDE 47, BDE 99, BDE 100, BDE 153, and BDE 209) ranged from 0.07 to 10.6  $\text{ng}/\text{g}$  ( $\mu\text{g}/\text{kg}$ ) dry weight in freshwater sediments from Denmark (Christensen and Platz 2001). The highest concentrations were found in sediment close to populated areas.

A study conducted by the U.S. Geological Survey (USGS) analyzed waste water treatment plant (WWTP) effluent from nine cities located in Oregon and Washington for anthropogenic compounds, including PBDEs (USGS 2012). Detectable levels in the low  $\text{ng}/\text{L}$  range were observed at every WWTP, and the highest concentrations measured were for congeners BDE 47, BDE 99, and BDE 100. The greatest

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PBDE concentrations were observed in Richland and Portland, Oregon. The Portland PBDE values showed varying concentrations as a function of the time of the day that samples were obtained. The lowest PBDE levels were observed in the morning hours and afternoon and then increased 2–4 times by evening hours. Shreder and LaGuardia (2014) measured PBDEs in the effluent of laundry waste water from 20 residences located near the Columbia River in Washington state. BDE 47 and BDE 209 were detected in the laundry waste water effluent of all 20 homes at median levels of 1,230 and 140 ng/L, respectively. The median concentration of total PBDE (sum of BDE 28, 47, 66, 85, 99, 100, 153, 154, 183, 206, and 209) in the laundry waste water was reported as 2,550 ng/L. BDE 47, BDE 49, and BDE 209 were also detected in the influents of two WWTPs near the Columbia River, Washington that primarily serve residential households. The sum total levels of BDE 47, BDE 49, and BDE 209 in the influents of the WWTPs ranged from 35 to 206 ng/L. Effluent PBDE levels were below the detection limit at one WWTP and 28.2 ng/L at the other facility (Shreder and LaGuardia 2014).

Although the available information indicates that leaching of PBDEs from landfills is minimal, movement of polymer particles containing pentaBDE, octaBDE, and decaBDE commercial mixtures within the landfill could lead to entry into leachate water of groundwater. PBDEs have been detected in landfill leachate and landfill related aqueous samples (Daso et al. 2013; Kwan et al. 2013; Odusanya et al. 2009; Oliaei et al. 2010; Stubbings and Harrad 2014). The presence of hydrophobic compounds like PBDEs in leachate is expected to be a result of enhanced leachability due to the presence of other constituents present in the leachate (Stubbings and Harrad 2014). Mass transfer evaluation of PBDEs from e-waste found that lower pH conditions resulted in higher transfer of PBDEs to the aqueous phase, with the highest concentration of PBDEs detected at pH 5 (Danon-Schaffer et al. 2013). It is not currently possible to assess the significance of this type of process. Well-designed landfills already include measures to minimize leaching in general, and these measures would also be effective in minimizing leaching of any PBDEs present (EU 2002, 2003a).

### 6.2.3 Soil

Estimated releases of 24,403 pounds (~11.1 metric tons) of decaBDE to soils from 34 domestic manufacturing and processing facilities in 2014, accounted for about 17% of the estimated total environmental releases from facilities required to report to the TRI (TRI14 2016). These releases are summarized in Table 6-1.



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PBDEs are released to land (i.e., landfills) as waste from their manufacture (both raw material and polymer) and as municipal wastes with the disposal of consumer products. The disposal of consumer products containing PBDEs is likely to increase worldwide due to rapid obsolescence of plastic products.

PBDEs may be present in biosolids and may therefore be inadvertently released to soils from the use of biosolids as a nutrient amendment to agricultural soils. Biosolids are sewage sludge that has been treated to meet regulatory requirements for land application and must adhere to concentration limits and loading rates for chemical pollutants, treatment and use requirements for controlling and reducing pathogens and the attraction of vectors, and management practices (NRC 2002). PBDEs were detected in biosolids destined for land applications in four different regions of the United States (Hale et al. 2001c). The total concentrations of pentaBDE in biosolids ranged from 1,100 to 2,290  $\mu\text{g}/\text{kg}$  dry weight. The concentration of decaBDE (BDE 209) varied widely among biosolids from different regions; the concentration of BDE 209 ranged from 84.8 to 4,890  $\mu\text{g}/\text{kg}$  dry weight in the biosolid samples. Kim et al. (2013b) analyzed 288 samples of sludge and biosolids from 15 WWTPs in Canada. Total PBDE levels were 230–82,000, 530–8,800, and 420–6,000  $\mu\text{g}/\text{kg}$ , in primary sludge, waste biological sludge and treated biosolids respectively. BDE 209, BDE 99, and BDE 47 were reported as the predominant congeners. In the biosolids, these three congeners accounted for approximately 80% of the total. The median percentages of BDE 209, BDE 99, and BDE 47 were 47, 16, and 16%, respectively, of the total amount of all PBDE congeners in the biosolids.

### 6.3 ENVIRONMENTAL FATE

#### 6.3.1 Transport and Partitioning

PBDEs exist in both the vapor phase and the particulate phase in the atmosphere. Particulate-phase PBDEs will be removed from the atmosphere by wet and dry deposition. A vapor phase–particulate phase analysis of indoor air samples obtained from Birmingham, United Kingdom found that 66–86% of BDE 47, 54–65% of BDE 99, 63–74% of BDE 100, <20–48% of BDE 153, and 37–48% of BDE 154 existed in the vapor phase (Harrad et al. 2004). Strandberg et al. (2001) performed a vapor phase–particulate phase analysis of outdoor air samples obtained from the Great Lakes region and found that about 80% of BDE 47, 55–65% of BDE 100 and BDE 99, and 30% of BDE 154 and BDE 153 existed in the gas phase. Several PBDE congeners have been detected in Arctic regions, suggesting that these substances undergo aerosol-mediated, long-range atmospheric transport. BDE 47, BDE 99, BDE 100, and BDE 209 were measured in air, snow, and sea ice samples throughout western Antarctica between 2001 and 2007 (Dickhut et al. 2012). Fourteen PBDE congeners are monitored for, and have been

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detected at, Alert and Nuuk monitoring stations as part of the Arctic Monitoring and Assessment Programme (Hung et al. 2010), indicating the importance of long-range transport as an environmental fate process for these substances.

In water, PBDEs are expected to adsorb strongly to suspended solids and sediment, and bioconcentrate in aquatic organisms. The volatilization of PBDEs from water to air is expected to be attenuated by adsorption in the water column. In soil, PBDEs are adsorbed strongly and will be immobile. They are not likely to leach into groundwater. Volatilization of PBDEs from soil to air is limited by the low volatility of PBDEs and strong adsorption of PBDEs to soil. There is potential for PBDEs to volatilize from soil to air, particularly if the organic carbon content of the soil is low, as demonstrated by PBDEs being monitored in air as described in Section 6.4.1.

PBDEs adsorb strongly onto suspended solids and sediments in the water column. Volatilization of PBDEs from water surfaces will be attenuated by adsorption, and is thus not an important fate process. Sediment-water partition coefficients ( $K_p$ ) have been measured for several components of commercial pentaBDEs (Watanabe 1988).  $K_p$  values for tetra-, penta-, and hexaBDEs are 28,300, 49,200, and 62,700 L/kg, respectively, which suggest strong partitioning to sediment. Log organic carbon-water partition coefficients ( $K_{oc}$ ) were estimated for PBDEs: di- (4.11); tri- (4.35–4.41); tetra- (4.57–4.73); penta- (4.89–5.17); hexa- (5.11–5.69); octa- (5.92–6.22); and deca- (6.80) (Lyman et al. 1990).

DecaBDE and octaBDE commercial products do not bioconcentrate in fish to the same extent as congeners from the penta mixture. Monitoring data show that higher-brominated congeners such as BDE 209 are taken up in marine organisms. The reported bioconcentration factors (BCFs) for commercial decaBDE mixtures are typically <50 (Hardy 2002b). A single study on a mixed range of PBDEs, between hexaBDE and decaBDE, indicated little bioconcentration in carp (e.g., *Cyprinus carpio*) with a BCF of <4 after 8 weeks of exposure (WHO 1994a). A bioconcentration study was carried out with rainbow trout under static conditions. The concentration of  $^{14}\text{C}$ -decaBDE/L in water was 20  $\mu\text{g}$ . Fish were exposed to decaBDE for 0, 0.5, 1, 2, 4, 6, 12, 24, or 48 hours. For each of the exposure periods, there was no measurable accumulation of decaBDE in flesh, skin, or viscera (WHO 1994a). The bioconcentration of BDE 209 was studied by exposing zebrafish embryos to BDE 209 at concentrations of 0, 0.08, 0.38, and 1.92 mg/L until 14 days post-fertilization (Chen et al. 2012). BCFs of 29, 9, and 20 were calculated for exposure of 0.08, 0.38, and 1.92 mg/L, respectively. Several lower-brominated congeners were also detected in the larvae, with the main metabolite being nonaBDE. These results are

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consistent with other finding that indicate that BDE 209 was bioavailable and taken up by zebrafish larvae from spiked sediments (Garcia-Reyero et al. 2014).

An abundance of monitoring data illustrates the uptake of lower-brominated diphenyl ethers by aquatic organisms, which results in bioconcentration (see Section 6.4.4). The commercial pentaBDE product undergoes bioconcentration with a BCF of approximately 14,000 (Hardy 2002b). Congener components of pentaBDE commercial product bioconcentrate to different extents. For example, approximately 50–70% of PBDEs detected in fish is a single isomer (BDE 47). The next most prominent isomer is typically BDE 99 followed by BDE 100. In a laboratory study of Baltic blue mussels (*Mytilus edulis* L), BCFs from water absorption were found to be 1,300,000 for BDE 47, 1,400,000 for BDE 99, and 1,300,000 for BDE 153 (Gustafsson et al. 1999). At several sites along the coast and in the Schelde Estuary (the Netherlands), BCFs for blue mussels were determined (Booij et al. 2000). The maximum BCFs were  $1 \times 10^9$  for BDE 99 and BDE 100,  $\approx 2.5 \times 10^7$  for BDE 28,  $\approx 2.5 \times 10^8$  for BDE 47 and  $\approx 1.6 \times 10^8$  for BDE 153. Biomagnification of PBDE congeners BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 155 in the marine food chain was demonstrated by comparing concentrations in the blubber of harbor seals with their prey fish (Shaw et al. 2009). Biomagnification factors (BMFs) from fish to seals were 21.4–109, 17.9–213, 6.9–29.8, 148–700, 11.3–447, and 12.4–236 for BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 155, respectively (Shaw et al. 2009). BDE 209 was detected at measurable concentrations in fish and seal tissue, although it did not appear to biomagnify like the other congeners. A laboratory study was conducted using juvenile carp fed BDE 209 amended food over a 60-day period (Stapleton et al. 2004). BDE 209 was not highly accumulated in the carp; however, seven debrominated lower congeners not initially present (penta- to octaBDEs) were detected in whole fish samples and liver tissue, suggesting that while BDE 209 did not accumulate in the fish, it may be a source of lower-brominated metabolites in aquatic organisms.

Other studies have demonstrated the biomagnification of lower-brominated PBDE congeners. Haglund et al. (1997) examined the concentrations of tetra- to hexaBDEs in herring, salmon muscle, and gray and ringed seals collected along the Swedish coast of the Baltic Sea between 1981 and 1988. The concentrations of tetraBDEs (e.g., BDE 47) were found to increase with trophic level. Concentrations of PBDEs in herring and their predators, grey seal and guillemot, all collected at the same location of the Baltic Sea, have been compared to estimate potential biomagnification (de Wit 2002). The herring were caught in the autumn of the same year as guillemot egg collection (1987). BMFs for guillemot egg versus herring were 19, 17, and 7.1 for BDE 47, BDE 99, and BDE 100, respectively. Bureau et al. (2000) analyzed small herring and salmon from the Atlantic Ocean (near Iceland) for several PBDEs. The

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calculated biomagnification factors for Atlantic salmon versus small herring were 3.5, 3.8, and 6.0 for BDE 47, BDE 99, and BDE 100, respectively. These authors concluded that biomagnification was occurring for the lower-brominated congeners.

Biosolids from the Metropolitan Water Reclamation District of Greater Chicago, Stickney WWTP, collected between 2004 and 2007, were applied at two sites at a depth of 15–20 cm (Hale et al. 2012). Maximum total soil PBDE concentrations were 565 and 1,810 µg/kg for high clay soil and sandy soil, respectively. Corn grown at the two sites after the third year of annual biosolid application was evaluated for PBDEs using GC/MS with ENCI. PBDEs were not detected in the 46 grain, stover, or root samples examined, suggesting little uptake by corn from soils amended using biosolids. However, earthworms exposed to PBDE containing biosolids were shown to accumulate these substances (Gaylor et al. 2013). Earthworms were exposed to a Class B anaerobically digested biosolid (ADB) containing 5,560±440 µg/kg dry weight total penta congeners (BDE 47, 99, 100, 153, 154, and 183) and a composted biosolid (CB) containing 1,130±79 µg/kg dry weight total penta congeners over the course of a 28-day incubation period. Total penta PBDE body burdens in worms exposed to ADB amended soils were about 5 times greater than those in the substrate, and worms exposed to CB amended soils had body burdens about 4 times greater than in the substrate.

### 6.3.2 Transformation and Degradation

Photolysis appears to be the dominant transformation process for PBDEs. However, the importance of photochemical transformation reactions in the environment cannot be determined due to lack of information. Based on a very limited number of studies, biodegradation does not appear to be significant for PBDEs.

#### 6.3.2.1 Air

In air, PBDEs may undergo indirect photolysis with hydroxyl radicals or direct photolysis with sunlight. Vapor-phase PBDEs may be degraded in the atmosphere by reaction with photochemically produced hydroxyl radicals. The half-lives for this reaction in air are estimated to be 29, 140, and 476 days, respectively, for penta-, octa-, and decaBDE homologs, calculated using a structure estimation method (Meylan and Howard 1993). This estimation is calculated using an atmospheric concentration of  $5 \times 10^5$  hydroxyl radicals per  $\text{cm}^3$  and is based on a 24-hour day of sunlight. The half-lives of PBDEs that are expected to be present in the particulate phase in the air will be longer than the estimated half-lives

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calculated for the gas-phase reaction. Thus, for the higher-brominated PBDEs (e.g., octa- and decaBDEs), indirect photolysis with hydroxyl radicals will be less important.

In water, some PBDEs have been reported to undergo direct photolysis (Hua et al. 2003). Likewise, PBDEs present in the vapor phase (e.g., tetraBDE) or as particulates (e.g., decaBDE) may also undergo photolysis in the atmosphere. However, the rate and extent of the photolysis of PBDEs in air cannot be evaluated due the lack of information.

### 6.3.2.2 Water

PBDEs absorb light in the environmental spectrum. Hua et al. (2003) found that decaBDE and the commercial octaBDE absorbed light up to 325 nm, which indicates that these compounds may be susceptible to photodegradation at environmental wavelengths (Hua et al. 2003). Di- and tetraBDEs were reported to absorb minimal light at wavelengths >300 nm. This trend suggests that the lower-brominated diphenyl ethers (e.g., pentaBDE commercial mixtures) will be less susceptible to photolysis compared to octaBDE and decaBDE commercial mixtures.

PBDEs undergo debromination by direct photolysis in organic solvents and organic solvent:water mixtures. Laboratory studies of the photolytic breakdown of decaBDE in toluene have shown that it is debrominated by ultraviolet (UV) light to hexaBDE and that photolysis occurs very rapidly (Sellström et al. 1998b). The photolysis half-life in toluene was <15 minutes. However, the amounts of lower-brominated congeners appear to be small (EU 2002). The photolysis of decaBDE (and tetra-, penta-, hexa-, hepta-, and octaBDEs) was reported in an 80:20 mixture of methanol:water at wavelengths >290 nm (EU 2002). The rate of photodegradation was found to increase with increasing degree of bromination. DecaBDE was found to degrade with a half-life of around 30 minutes, while half-lives for tetra-, penta-, hexa-, hepta-, and octaBDEs were 12–16 days, 2.4 days, 1.2 days, 1.2 days, and 5 hours, respectively. The decomposition products of decaBDE were identified to be PBDEs (with >6 bromine atoms per molecule) and polybrominated furans (with <6 bromine atoms per molecule). Results of this study indicate that the photochemical stability of PBDEs increases with decreasing bromination (EU 2002). Rayne et al. (2003b) reported that BDE 15 photodegraded in organic (acetonitrile-methanol) and aqueous (H<sub>2</sub>O:acetonitrile; 1:1 v/v) solvent systems at a wavelength of 300 nm. Reductive bromination was reported to be much slower in the aqueous system (e.g., 73% remained after 300 minutes) compared to the organic system (where 51 and 41% remained after 30 minutes). However, these studies were conducted in the presence of organic solvents, which are not representative of conditions found in the

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environment. Organic solvents can act as hydrogen donors in photolysis reactions, which will potentially affect the distribution of products formed.

The photolysis of PBDEs was examined under environmentally relevant conditions. Hua et al. (2003) studied the degradation of decaBDE in several different experiments: (1) on humic acid-coated silica particles; (2) on glass surfaces in contact with aqueous humic acid solutions; and (3) on glass surfaces in contact with water. DecaBDE dissolved in toluene was deposited on the solid substrate under a stream of nitrogen (to evaporate the solvent) and then desiccated to remove any residual toluene. The adsorbed decaBDE on the solid substrate was then inundated with the aqueous test solution, followed by irradiation for the duration of the test period. In all experiments, natural sunlight (location, 40° 26' N, 86° 55' W) was used. The extent of degradation was determined using HPLC with UV detection or by GC/MS. In the first experiment, solar irradiation of decaBDE adsorbed onto humic acid-sand indicated that the photolysis of decaBDE was slow. After 96 hours of exposure to sunlight, 88% of initial decaBDE remained on the coated sand. There is some evidence that lower-brominated congeners (e.g., BDE 155) were formed in the experiment (EU 2002). In the second experiment, decaBDE was adsorbed on glass tubes containing a humic acid. In this study, the concentration of decaBDE decreased relatively quickly over the first 24 hours of exposure, after which, the concentration remained stable. Bromide ion accumulated at an almost linear rate from start to end of the 72-hour exposure period. Approximately 70% of the initial decaBDE remained after the 72-hour exposure. The difference in kinetics (for the disappearance of decaBDE vs. the appearance of bromide ion) suggests that after the initial degradation of decaBDE, bromide ion was generated by the degradation of lower-brominated diphenyl ether congener products (possibly octa- and nonaBDEs). Bromide ion mass balance for the system indicated that 70% of the total bromine present was accounted for by decaBDE or bromide, with the remaining 30% present as unidentified compounds. In the third experiment, Hua et al. (2003) investigated the photodegradation of decaBDE adsorbed on glass tubes, which were filled with aqueous solutions (without humic acid). The result of this test showed a much more rapid loss of decaBDE than found in the analogous test using humic acid solutions. Approximately 29% of the initial decaBDE present remained after 72 hours. The rate of decaBDE loss and bromide ion accumulation was relatively constant over the entire 72-hour test period. Mass balance indicated that approximately 50% of the total bromine was present as either decaBDE or bromide ion, while the remaining 50% was possibly unidentified nona- and octaBDE congeners. The difference between the tests using glass tubes with and without humic acid solution is possibly due to the absorption of light by humic acids, which may attenuate the degradation process. These studies indicate that adsorbed decaBDE may undergo photolysis forming octa- and nonaBDEs under somewhat environmentally relevant conditions. Lower-brominated diphenyl ether congeners are

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also formed although only to a minor extent. These tests do not provide evidence that lower-brominated diphenyl ethers (e.g., tetra- and pentaBDEs) are a major degradation product of decaBDE (EU 2002). There is also insufficient information from these studies to estimate the rate of photolysis or if intermediate degradation products build up after long-term exposures (EU 2002).

Söderström et al. (2004) examined the time course of photolysis of decaBDE (BDE 209) in toluene, on silica gel, sand, sediment, and soil using artificial sunlight and on the natural matrices (e.g., sediment, soil, and sand) using natural sunlight. On natural samples, BDE 209 was first dissolved in toluene and then deposited on the natural matrix. The toluene was allowed to evaporate, and then the sample was reconstituted with water to resemble natural conditions. BDE 209 was photolytically labile and formed debromination products in all matrixes studied. Nona- to tetraBDEs were formed as well as some PBDFs. The half-lives in toluene and on silica gel were <15 minutes, and half-lives on other matrices ranged from 40 to 200 hours. No differences were observed in the debromination patterns under different matrices or light conditions. These experiments show that photolytic debromination of BDE 209 is a possible pathway for the formation of more bioavailable, lower-brominated PBDEs. However, the most commonly found BDEs in environmental samples (e.g., BDE 47, BDE 99, and BDE 100) were only formed to a minor degree (Söderström et al. 2004).

Following the methodology described for decaBDE, photolysis experiments were conducted on BDE 47 (EU 2002). BDE 47 was adsorbed on glass tubes filled with an aqueous solution and exposed to natural sunlight. After 72-hours of exposure, 30% of the initial BDE 47 remained. The rate of disappearance of BDE 47 was comparable to that found for decaBDE under similar test conditions. Accumulation of bromide was initially slow with the rate increasing after 24 hours while the disappearance of BDE 47 was initial rapid over the first 24 hours. Using GC/MS, the authors concluded that 2,4,4'-triBDE was being formed during this reaction and that removal of bromine atoms *ortho* to the ether functionality may be a significant reaction pathway for removal of bromine atoms under the conditions of this study. This study suggests that adsorbed BDE congeners, like decaBDE, may undergo photolysis under somewhat environmentally relevant conditions (EU 2002).

PBDEs are not expected to undergo abiotic hydrolysis under environmental conditions due to the lack of hydrolysable functional groups (Wolfe and Jeffers 2000).

PBDEs are unlikely to biodegrade rapidly in the environment under aerobic conditions. PentaBDE did not undergo biodegradation (determined by CO<sub>2</sub> evolution) after 29 days in an Organisation for Economic

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Co-operation and Development (OECD) 301B ready biodegradation test (EU 2001). The substance tested was a composite sample from two producers with the following composition: 33.7% tetraBDE, 54.6% pentaBDE, and 11.7% hexaBDE. The test was extended to 93 days to allow sufficient opportunity for adaptation to occur. At the end of 93 days, 2.4% of the theoretical amount of CO<sub>2</sub> had been evolved. Thus, pentaBDE was determined to be not readily biodegradable. No degradation (as oxygen uptake) was seen for octaBDE after 28 days in an OECD 301D ready biodegradation test (EU 2003a). Thus, octaBDE was determined to be not readily biodegradable. The biodegradability of decaBDE has been studied under aerobic conditions using an activated sludge inoculum (EU 2002). DecaBDE at 100 mg/L was incubated with activated sludge (at 30 mg/L) over a 2-week period using a method similar to an OECD 301C MITI test. No degradation (as measured by biochemical oxygen demand) was observed. Thus, decaBDE was determined to be not readily biodegradable.

No data on biodegradation of pentaBDE and octaBDE commercial mixtures under anaerobic conditions are available. An anaerobic degradation study was carried out with BDE 47 using a mixture of <sup>14</sup>C-labeled and unlabeled compound (EU 2003a). The test was carried out using a sediment-water (Schuylkill River, Pennsylvania) inoculum. After 32 weeks, it appeared that no significant degradation of BDE 47 had occurred. However, the analytical method (i.e., HPLC using radiometric detection) used in this test indicated that some unidentified products had been formed in samples taken after 32 weeks. From these results, it is clear that BDE 47 has the potential to degrade slowly under anaerobic conditions (EU 2003a). Rayne et al. (2003b) reported that 4,4'-diBDE undergoes reductive debromination under anaerobic conditions. Debromination proceeds with replacement of a bromine atom by a hydrogen atom. The authors suggest that anaerobic debromination may sequentially debrominate BDE 15 to the parent diphenyl ether.

The anaerobic biodegradability of <sup>14</sup>C-labeled decaBDE was studied over a period of 32 weeks (EU 2002). The test chambers consisted of 500 mL bottles containing 300 mL of sediment (Schuylkill River, Pennsylvania) prepared under anaerobic conditions. The test chambers were incubated at 25°C and in the dark during the test. After the 32-week period, <1% of the total radioactivity added was found as <sup>14</sup>CO<sub>2</sub> and <sup>14</sup>CH<sub>4</sub> indicating that essentially no mineralization had occurred. GC/MS results showed no evidence for the formation of lower-brominated congeners from decaBDE under the conditions of this test (EU 2002).



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**6.3.2.3 Sediment and Soil**

Information on the transformation and degradation of PBDEs in soil is limited. The extent to which PBDEs undergo direct photolysis in soils and sediment is unknown. However, sunlight would only penetrate the uppermost few millimeters of soil and will not impact sediment. Photolysis of PBDEs is possibly important for land-applied sewage sludge contaminated with PBDEs. However, no information was available on this possibility. Based on studies in water, most PBDEs biodegrade slowly in soils or sediment under aerobic or anaerobic conditions. The anaerobic biodegradation of BDE 47, BDE 99, and BDE 209 was studied using microcosms prepared from loam sediment (pH 6.3, 16.4% organic carbon) obtained from a pond located in West Lafayette, Indiana (Tokarz et al. 2008). After an 8-month incubation period, microcosms containing BDE 47 showed variable losses (up to 30% of the initially applied amount) of the parent congener without concurrent increases in expected debromination products, suggesting that other degradation mechanisms other than reductive debromination may have occurred. Only about 3% degradation of BDE 99 was observed after 8 months, with BDE 28 being the most important debromination product. After 10 months, only slight decreases in the initial BDE 209 concentration was observed in six microcosms, and the half-life of this congener was estimated to range from 6 to 50 years; however, some aged microcosms exhibited greater degradation after 3.5 years, yielding nine degradation products (Tokarz et al. 2008).

**6.3.2.4 Other Media**

The bacteria, *Pseudomonas aeruginosa*, that was isolated from an electronic waste dismantling area was capable of degrading BDE 209 under aerobic conditions, especially in the presence of co-metabolic substrates such as glucose (Shi et al. 2013). Nonabromodiphenyl ethers (BDE 208, BDE 207), four octabromodiphenyl ethers (BDE 203, BDE 202, BDE 197, BDE 196), and one heptabromodiphenyl ethers (BDE 183) were noted as degradation products.

**6.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT**

Reliable evaluation of the potential for human exposure to PBDEs depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. Concentrations of PBDEs in unpolluted atmospheres and in pristine surface waters are often so low as to be near the limits of current analytical methods. In reviewing data on PBDEs levels monitored or estimated in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily

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equivalent to the amount that is bioavailable. The analytical methods available for monitoring PBDEs in a variety of environmental media are detailed in Chapter 7.

Monitoring studies indicate that PBDEs are transported globally. Atmospheric, water, and biota concentrations of PBDEs tend to be dominated by lower-brominated congeners (e.g., BDE 47). Sediments tend to be dominated by higher-brominated congeners (e.g., BDE 209). Biota monitoring studies indicate that PBDE concentrations have increased since the late 1970s, with lower-brominated congeners (e.g., BDE 47) being preferentially bioconcentrated. Studies indicate that PBDE concentrations increase with respect to trophic level; organisms that reside higher on the food chain tend to have higher concentrations of PBDEs.

#### 6.4.1 Air

PBDEs will exist in both the vapor and particulate phase in both indoor and outdoor air (Harrad et al. 2004). The higher-brominated congeners (hepta-deca) have lower vapor pressures and partition more to the particulate phase, while the lower-brominated substances have a greater tendency to partition to the vapor phase. Concentrations of PBDEs in outdoor air in the United States are typically in the range of 20–200 pg/m<sup>3</sup>, with BDE 47 and BDE 99 being the congeners most often detected (EPA 2010). Monitoring data from the 1990s showed infrequent detections of decaBDE; however, more recent monitoring data have shown an increase in the frequency of detection of this substance both in outdoor and indoor air samples (EPA 2010; Hoh and Hites 2005). Monitoring data from Europe and Asia suggest lower concentrations of PBDE in air samples as compared to data obtained in the United States.

Representative concentrations of PBDEs in outdoor air samples obtained at various locations are summarized in Table 6-2. Air samples obtained from urban (Chicago, Illinois), rural (Sleeping Bear Dunes, Michigan and Sturgeon Point, New York), and remote (Eagle Harbor, Michigan) shorelines of the U.S. Great Lakes all contained quantifiable concentrations of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 (Dodder et al. 2000; Strandberg et al. 2001). The most significant congeners were BDE 47 and BDE 99. Air measurements were averaged over a 3-year period between 1997 and 1999. The concentration of total PBDEs ranged from 5.5 pg/m<sup>3</sup> in rural environments to 52 pg/m<sup>3</sup> in urban air from Chicago, Illinois. The concentration of BDE 47 was 48 pg/m<sup>3</sup> observed near Chicago, Illinois. The average concentration of decaBDE at the remote and rural locations was <0.10 pg/m<sup>3</sup> for each of the years investigated. The average concentration of decaBDE in the particulate phase at the urban location ranged from 0.20 to 0.35 pg/m<sup>3</sup> (Strandberg et al. 2001).

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**Table 6-2. Concentrations (pg/m<sup>3</sup>) of Several Polybrominated Diphenyl Ethers (PBDEs) in Air Samples**

Location	BDE 47	BDE 99	BDE 100	BDE 209	ΣPBDEs <sup>a</sup>	Reference
Urban, United States	48	25	3.0	No data	77*	Dodder et al. 2000
Rural, United States	6.2–9.2	4.3–5.0	0.6–0.9	No data	2–4.8*	Dodder et al. 2000
Remote, United States	3.7	2.6	0.33	No data	6.9*	Dodder et al. 2000
Alert, Northwest Territories Canada	No data	No data	No data	No data	1–28	Alaee et al. 1999
Eagle Harbor, Wisconsin	2.9	2.1	0.28	<0.10	5.5*	Strandberg et al. 2001
Chicago, Illinois	3.9–42	2.4–15	0.68–3.3	1.5–878	13–980	Hoh and Hites 2005
Sleeping Bear Dunes, Michigan	0.51–27	0.32–23	0.030–5.1	<0.29–21	1.4–61*	Hoh and Hites 2005
Bloomington, Indiana	1.9–21	1.2–11	0.26–2.75	<0.29–21	6.4–44*	Hoh and Hites 2005
Rohwer, Arkansas	1.2–42	0.87–35	0.17–3.7	<0.10–135	2.7–165*	Hoh and Hites 2005
Cocodrie, Louisiana	2.0–24	0.89–11	0.21–2.7	<0.10–14	5.1–42*	Hoh and Hites 2005
Sturgeon Point, New York	3.8	2.8	0.39	<0.10	7.2*	Strandberg et al. 2001
Sleeping Bear Dunes, Michigan	8.4	5.3	0.80	<0.10	15*	Strandberg et al. 2001
Chicago, Illinois	33	16	2.0	0.20–0.35	52*	Strandberg et al. 2001
Ammarnäs, Sweden	6.3	1.6	0.4	No data	8.3	de Wit 2000, 2002
Hoburgen, Sweden	0.7	0.35	0.07	No data	1.1	de Wit 2000, 2002
Stoke Ferry, United Kingdom	4.7–50	5.5–13	1.1–3.9	No data	6.7–58	Peters et al. 1999
Hazelrigg, United Kingdom	3.2–61	3.1–22	0.62–5.4	No data	4.1–69	Peters et al. 1999
Dunai Island, Russia	No data	No data	No data	No data	1–8*	Alaee et al. 1999
Arctic	2.2–2.8	2.0–2.3	0.40–0.47	1.0–1.8	6.7–8.6*	Hung et al. 2010

<sup>a</sup>ΣPBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether

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PBDEs were monitored at five different locations (Chicago, Illinois; Sleeping Bear Dunes, Michigan; Bloomington, Indiana; Rohwer, Arkansas; and Cocodrie, Louisiana) across the United States from 2002 to 2003 (Hoh and Hites 2005). Total PBDE concentrations at the urban site (Chicago, Illinois) were 3–6 times greater than the other locations, with BDE 47, BDE 99, BDE 100, and BDE 209 being the most abundant congeners at all five locations. DecaBDE concentrations as high as 960 and 410  $\text{pg}/\text{m}^3$  were observed in Chicago on two dates in 2003.

Throughout the year of 1997, air samples were taken from a rural site in southwestern England called Stokes Ferry and a semirural site in northwestern England called Hazelrigg and analyzed for PBDEs (Peters et al. 1999). Tri- and heptaBDEs were detected; the combined concentrations of BDE 47, BDE 99, and BDE 100 ranged from 7 to 69  $\text{pg}/\text{m}^3$  at Hazelrigg and from 6 to 58  $\text{pg}/\text{m}^3$  at Stoke Ferry (de Wit 2002). PBDEs have also been measured in air samples taken from remote stations in the Arctic (e.g., Alert, Northwest Territories, Canada; Dunai Island, eastern Siberia, Russia) between January 1994 and January 1995 (de Wit 2002). The total concentration of several di- to hexaBDEs ranged from 1 to 4  $\text{pg}/\text{m}^3$  at Alert for the majority of the year; however, in July 1994, the concentration was 28  $\text{pg}/\text{m}^3$ . At Dunai, the major congeners found were BDE 47 and BDE 99. In Sweden during 1990–1991, air samples collected from Ammarnäs in the northern mountains and Hoburgen on the southern tip of Gotland in the Baltic Sea, had measurable amounts of BDE 47, BDE 99, and BDE 100 (de Wit 2002). Total PBDE concentrations were approximately 1 and 8  $\text{pg}/\text{m}^3$ , respectively. The concentration of BDE 47 was found to be highest in the gas phase, while BDE 99 and BDE 100 were highest in the particulate phase. No decaBDE was found, although the limit of detection limit for decaBDE is much higher than for the lower-brominated diphenyl ethers.

Indoor air concentrations of PBDEs vary depending upon potential sources such electronics or foams used in upholstery stuffing for furniture that were treated with PBDEs (EPA 2010; Harrad et al. 2006; Hazrati and Harrad 2006). Harrad et al. (2004) found a significant positive correlation between PBDE concentrations in indoor air and both the number of electrical appliances and the number of chairs containing FPUF. Concentrations of tetra- and pentabrominated congeners (BDE 47, BDE 99, and BDE 100) in indoor air were always higher than those detected in outdoor air. On average, indoor air concentrations were 150, 120, and 140 times higher than outdoor air for BDE 47, BDE 99 and BDE 100, respectively. Indoor air concentrations of PBDEs also tended to be higher in workplace environments as compared to domestic residences; however, these concentrations could vary substantially from one room to another (Harrad et al. 2004). Air samples collected from 31 homes, 33 offices, and 25 automobiles in the West Midlands, United Kingdom were analyzed for the presence of PBDEs (Harrad et al. 2006).

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Total PBDE concentrations in samples obtained from homes ranged from 4 to 245  $\text{pg}/\text{m}^3$  (24  $\text{pg}/\text{m}^3$ , median), while concentrations in offices and cars ranged from 10 to 1,416 (71  $\text{pg}/\text{m}^3$ , median) and from 4 to 1,416  $\text{pg}/\text{m}^3$  (41  $\text{pg}/\text{m}^3$ , median), respectively. PBDE congeners 47 and 99 were reported as the major contributors to the overall total (Harrad et al. 2006). Although the previous study reported a statistically significant positive correlation between the PBDE concentrations and the number of electrical devices and FPUF-containing chairs (Harrad et al. 2004), no clear statistical evidence of such correlations were reported in these indoor air environments (Harrad et al. 2006; Hazrati and Harrad 2006). PBDE concentrations in the air of one office did drop dramatically after an older computer was replaced by a relatively newer one and a statistically significant positive correlation was observed between automobile age and PBDE concentrations when two highly contaminated outliers were removed from the analysis. Further observations indicated a seasonal variation of the PBDE concentrations in indoor air, with higher concentrations being observed during the summer months (Hazrati and Harrad 2006). BDE 47, BDE 99, BDE 100, BDE 183, and BDE 209 were detected in 63, 22, 29, 32, and 42% of air samples taken aboard aircraft during routine flights (Allen et al. 2013). Concentrations ranged from below the detection limits to a maximum concentration of 2,100,000  $\text{pg}/\text{m}^3$  for BDE 209.

Concentrations of PBDEs were measured in floor dust, indoor air, ventilation filter dust and carpets in 10 buildings located in Michigan (Batterman et al. 2010). Median concentrations of total PBDEs were reported as 8,754  $\text{ng}/\text{g}$  in settled dust, 1,250  $\text{pg}/\text{m}^3$  vapor-phase air, and 155  $\text{pg}/\text{m}^3$  particulate-phase air. The highest concentrations of PBDEs were generally noted in rooms that contained sources such as computer servers. Monitoring data from one building that was built in 2006 indicated a very low concentration of PBDEs in settled dust at the time it was constructed (145  $\text{ng}/\text{g}$ ); however, these concentrations increased exponentially to >10,000  $\text{ng}/\text{g}$  5–8 months after the building was opened. Despite the voluntary phase-out of pentaBDE and octaBDE in 2004, high concentrations of congeners associated with these mixtures (BDE 47, BDE 99, BDE 100, BDE 153, and BDE 203) were detected in dust and air samples, suggesting that there are significant sources of these PBDEs in products that remain in the market and that were used in this building even after the phase out in 2004.

#### 6.4.2 Water

Due to the hydrophobic nature of PBDEs, this class of compounds is expected to be present in water at very low concentrations or at concentrations below the limit of detection of acceptable analytical methods. In 1999, the concentration of PBDEs in Lake Ontario surface waters ranged between 4 and 13  $\text{pg}/\text{L}$  with ~90% in the dissolved phase (Luckey et al. 2001). BDE 47 and BDE 99 were the most

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abundant congeners, together making up >70% of the total PBDEs. Streets et al. (2006) reported 18 and 3.1 pg/L average concentration of  $\Sigma$ PBDEs (BDE 47, BDE 66, BDE 100, BDE 99, BDE 85, BDE 154, and BDE 153 of pg/L) in Lake Michigan Water for the dissolved phase and particulate phase, respectively. In Japan, PBDEs were not detected in 75–200 water samples (ENVIRON 2003a). In nine English freshwater lakes grab samples were obtained from April 2008 to February 2012, average concentrations of the sum total of PBDEs, which included BDE 17, BDE 28, BDE 49, BDE 47, BDE 66, BDE 100, BDE 99, BDE 85, BDE 154, and BDE 153, ranged from 41.5 to 73.3 pg/L (Yang et al. 2014).

The EPA utilized data from the San Francisco Monitoring Program for Trace Substances in surface water to estimate possible concentrations of PBDEs that might be found in drinking water. Thirty-three water samples were obtained from the San Francisco Estuary, with total PBDEs concentrations ranging from 3 to 513 pg/L, and a mean concentration of 146.2 pg/L (Oros et al. 2005). It was reported that the most frequently detected congeners were BDE 47, BDE 99, and BDE 209. The source of these PBDEs was most likely effluent from municipal treatment plants. Concentrations of PBDEs monitored in the Spokane River in the state of Washington exhibited seasonal variation (Furl and Meredith 2010). Dissolved PBDE concentrations collected in the fall of 2005 were approximately 6 times greater (926 pg/L) as compared to concentrations observed in the spring of 2006 (146 pg/L). The variation in PBDE concentrations were likely a result of dilution of local sources in the spring from increased flow due to snowmelt in the upper watershed.

### 6.4.3 Sediment and Soil

Hale et al. (2002) reported the concentration of PBDEs in soil samples collected in the vicinity of a polyurethane foam-manufacturing facility. Concentrations in these soils are likely to be higher than those to be expected in rural and potentially urban areas of the United States. Total PBDE concentrations in these samples ranged from not detected to 76  $\mu\text{g}/\text{kg}$  dry weight. BDE 99 was the predominant congener in soil followed by BDE 47 and BDE 100. Concentrations of total PBDEs in soil obtained from a large automotive shredding and metal recycling facility in Brisbane, Australia ranged from 29 to 726 ng/g dry weight as compared to background concentrations at an uncontaminated site (0.2–2 ng/g dry weight) (Hearn et al. 2013). BDE 209 was the predominant congener in dust, soil, and air at the facility.

In Eastern China, near dismantling areas for waste electrical and electronic equipment that are considered a potential exposure source of PBDEs, 45 farmland soil samples were collected from 12 locations (Dong et al. 2014). The farmland soil PBDE concentrations ranged from 2.96 to 200 ng/g and air concentrations

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ranged from 884 to 2,791 pg/m<sup>3</sup>. The PBDE concentrations in high-mountain pasture soil, grass, and milk from grazing cows in the Italian Alps were analyzed (Parolini et al. 2012). Thirteen BDE congeners were investigated, including BDE 17, BDE 28, BDE 71, BDE 47, BDE 66, BDE 100, BDE 99, BDE 85, BDE 154, BDE 153, BDE 138, BDE 183, and BDE 190. Average ΣPBDE concentrations in soil from 0 to 7 cm soil depth were 0.43–1.55 ng/g dry weight, 1.7–8.2 ng/g dry weight in grass vegetation, and 0.659–1.576 ng/g dry weight in milk. The average total PBDE concentration in soil samples collected from the Ny-Alesund region of the Arctic was reported as 0.042 ng/g dry weight (Wang et al. 2015). BDE 99 was the predominant congener, with an average value of 0.0097 ng/g dry weight.

The EPA used 33 surface soil measurements taken from 15 states in the United States to estimate ingestion rates and dermal exposure rates of PBDEs from soil. The concentration of 30 total BDEs in the soils was reported to average 103 ng/g dry weight, with a geometric mean (GM) concentration of 5.3 ng/g (EPA 2010). BDE 47 (1.9 ng/g), BDE 99 (3.6), BDE 100 (0.4), BDE 153 (5.7), BDE 154 (4.8), BDE 183 (37.4), and BDE 209 (15.3) were included in this evaluation.

Sediment concentrations of PBDEs tend to be dominated by higher-brominated congeners (e.g., BDE 209) (deWit 2002). Temporal trends suggest that concentrations of PBDEs in sediments are increasing. Burdens of PBDEs in sediment appear to be a function of distance from the source and their organic carbon content (Hale et al. 2003). Representative concentrations of PBDEs in sediment samples are summarized in Table 6-3.

A 20-year field study regarding the land application of Class B biosolids to a site located in Arizona was discussed by Quanrud et al. (2011). Risk assessments were made based on the intake of compounds via inhalation, dermal sorption, or ingestion. PBDE concentrations were detected, primarily in the surface 30-cm depth sample, and surface accumulation of PBDEs occurred due to their hydrophobic nature, which resulted in sorption to colloids. The maximum amount of PBDE detected was 80 ng/g soil as congener BDE 209. A risk evaluation of PBDEs based on Hazard Indices indicated that the health risk to humans of PBDEs was negligible when all three routes of exposure were considered.

Li et al. (2006) collected 199 sediment samples from 16 locations of Lake Superior, Lake Michigan, Lake Huron, Lake Erie, and Lake Ontario and analyzed these samples for PBDE concentrations. The concentrations of PBDEs in surface sediment ranged from 0.5 to 6.7 ng/g dry weight tri- to heptaBDE congeners and from <4 to >240 ng/g dry weight for BDE 209. PBDEs were detected in 100% of sediment samples obtained from the San Francisco Bay with total PBDE concentrations ranging from

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**Table 6-3. Concentrations (ng/g) of Several Polybrominated Diphenyl Ethers (PBDEs) in Sediment and Suspended Particulate Samples**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs	BDE 209	Reference
Sediment	Lake Superior	No data	No data	No data	No data	12	Zhu and Hites 2005
Sediment	Lake Michigan	No data	No data	No data	2.6 <sup>a</sup>	320	Zhu and Hites 2005
Sediment	Lake Michigan	No data	No data	No data	1.67–3.97 <sup>a</sup>	43.9–95.6	Song et al. 2005
Sediment	Lake Huron	No data	No data	No data	1.02–1.87 <sup>a</sup>	21.5–36.0	Song et al. 2005
Sediment	Lake Erie	No data	No data	No data	1.1 <sup>a</sup>	40	Zhu and Hites 2005
Sediment	Lake Ontario	No data	No data	No data	2.8 <sup>a</sup>	14	Qui et al. 2007
Sediment	Hadley Lake, Indiana	16±2 (dw)	37±8 (dw)	7.1±1.5 (dw)	584 (dw) <sup>b</sup>	480±170 (dw)	Dodder et al. 2002
Sediment	Baltic Sea	ND–3.4	ND–2.4	ND–1.3	ND–5.4	ND	Nylund et al. 1992
Sediment	Upstream plastics plant, Sweden	3.7	8.8	1.6	14.1	No data	Sellström and Jansson 1995
Sediment	Downstream plastics plant, Sweden	780	1,200	270	2,250	No data	Sellström and Jansson 1995
Sediment	River Viskan (Sweden), upstream and downstream textile industries	<2–50	<1–53	<0.4–19	ND–120	ND–16,000	Sellström et al. 1998a
Sediment	22 European river mouths	<0.17–6.2 (dw)	<0.19–7.0 (dw)	No data	No data	<0.51–1,800	de Wit 2002
Sediment	Seven rivers, Great Britain	<0.3–368 (dw)	<0.6–898 (dw)	No data	No data	<0.6–3,190	Allchin et al. 1999
Sediment	Netherlands, several sites	0.3–7.1 (dw)	<0.2–9 (dw)	No data	No data	<4–510 (dw)	de Boer et al. 2000b
Suspended particulates	Netherlands, several sites	<2–9 (dw)	<0.1–23 (dw)	No data	No data	<9–4,600 (dw)	de Boer et al. 2000b

<sup>a</sup>Tri- to hepta-PBDE congeners.

<sup>b</sup>Includes sum of BDE 47, BDE 99, BDE 100, BDE 209, and other congeners (not specified).

BDE = brominated diphenyl ether; dw = dry weight; ND = not detected



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2.1 to 8.0 ng/g dry weight and a median concentration of 4.3 ng/g dry weight (Klosterhaus et al. 2012). BDE 209 was the primary congener of each sample, accounting for approximately 38–68% of the total PBDE amount.

In the United States, Dodder et al. (2002) analyzed four surficial sediments from Hadley Lake (Indiana). This lake is in the vicinity of a production point source. DecaBDE (BDE 209) was the major congener detected at concentration ranging from 19 to 36  $\mu\text{g}/\text{kg}$  (ng/g) dry weight. Other congeners detected (in decreasing order: BDE 99, BDE 153, BDE 154, BDE 47, and BDE 100) were  $<5$   $\mu\text{g}/\text{kg}$  (ng/g) dry weight. PBDEs were above the detection limit (i.e., 0.5  $\mu\text{g}/\text{kg}$  [ng/g] dry weight) in 22% of surficial sediment samples (from 133 sites) in freshwater tributaries of Virginia (Hale et al. 2001b). BDE 47 was the predominant congener followed by BDE 99 and BDE 100. The maximum concentration detected in sediment was 52.3  $\mu\text{g}/\text{kg}$  (ng/g) dry weight. Hale et al. (2002) reported that stream sediment adjacent to a former polyurethane foam production facility in North Carolina contained up to 132  $\mu\text{g}/\text{kg}$  (ng/g) dry weight of pentaBDE. Since the phase out of PBDE-containing flame retardants, levels of these substances have begun declining in sediment and other environmental media. Sutton et al. (2015) reported that levels of BDE 47 have declined by over one-third in sediment samples obtained from the San Francisco Bay from 2000 to 2012; however, no decline of BDE 209 was evident, presumably since decaBDE was not phased out until 2013.

In Japan, tetra-, penta-, hexa-, and decaBDEs have been detected in river sediments (Watanabe et al. 1986, 1987, 1995). The combined concentrations of tetra- and pentaBDEs ranged from 21 to 59 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight. The concentration of decaBDE (BDE 209) ranged from  $<25$  to 11,600 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight (deWit 2002). In 1999, sediment samples from several locations in the Netherlands contained BDE 47, BDE 99, and BDE 209 (de Boer et al. 2000b). Concentrations ranged from 0.3 to 7.1 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 47, not detected to 5.5 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 99, and not detected to 510 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 209. The concentration of PBDEs in suspended particulate matter ranged from not detected to 9 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 47, not detected to 23 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 99, and not detected to 4,600 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight for BDE 209 (de Boer et al. 2000b). The concentration of several brominated flame retardants was measured in sediments collected from the mouths of major European rivers (de Wit 2002). Elevated concentrations of BDE 47 and BDE 99 were found in Humber and Mersey rivers (Great Britain). In two rivers of the Netherlands, the sum of BDE 47 and BDE 99 ranged from 1.61 to 13.1 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight. The highest hexaBDE concentrations (as BDE 153) were found in the river Seine (France), three rivers in the Netherlands, and the rivers Schelde (Belgium), Forth (Great Britain) and Ems (Germany); the concentration of

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BDE 153 ranged from 0.013 to 0.056 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight in these sediments. The concentrations of decaBDE were highest in sediment from the Seine, ranging from 2.4 to 3.9 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight. The concentrations of decaBDE in River Mersey (Great Britain), Schelde, and River Liffey (Ireland) ranged from 34 to 1,800 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight. In the southern Baltic Sea (Bornholm Deep), the upper layer of sediment was analyzed for BDE 47, BDE 99, and BDE 100; the combined concentration of these three congeners was 0.52 ng/g ( $\mu\text{g}/\text{kg}$ ) dry weight (Nylund et al. 1992).

A well-studied sediment core collected from the southern part of the Baltic Sea proper was analyzed for PBDEs and a number of organochlorine contaminants (Nylund et al. 1992). The retrospective temporal trend from 1939 to 1987 showed that the PBDE concentrations (i.e., sum of BDE 47, BDE 99, and BDE 100) have increased with a sharp increase after 1980. The PBDE concentration in the sample from 1989 was 2.9 ng/g (Nylund et al. 1992). Measurable amounts of BDE 28, BDE 47, BDE 66, BDE 99, and BDE 100 were found in sediment cores from a freshwater lake in Germany, the Wadden Sea (the Netherlands), and Drammenfjord (Oslo Fjord, Norway) (Zegers et al. 2000). Samples from the Drammenfjord and freshwater lake also contained BDE 153 and BDE 154, and the Wadden Sea and freshwater lake samples contained BDE 209. The lower-brominated PBDEs appear in the 1960s, and BDE 209 appears about 10 years later. The Drammenfjord sediment core shows increasing concentrations of BDE 47 starting in the 1940s (range, 0.02–0.18 ng/g dry weight) and increasing concentrations of BDE 99 (range, 0.5–0.28 ng/g dry weight), BDE 100 (range, not detected–0.07 ng/g dry weight), and BDE 154 (range, not detected–0.06 ng/g dry weight) beginning in the 1950s up to 1999. In the sediment core from Lake Woserin, lower-brominated PBDE congeners were detected in the sediment horizons beginning in the late 1950s, and the concentrations increased until the late 1970s, and then leveled off when residues of BDE 209 first appeared. A similar leveling-off trend is also observed in the Wadden Sea core (Zegers et al. 2000). It is important to note that this study identified the presence of PBDEs compounds in sediments from the late 1950s and early 1960s. This is nearly a decade prior to any significant commercial production of these substances. The existence of PBDEs at these early dates may be a result of vertical mixing of sediment cores or blurring of core horizons through burrowing activity of benthic organisms, or may lend some credibility to the likelihood that either the substances identified in the environment as PBDEs are not necessarily PBDEs.

#### 6.4.4 Other Environmental Media

**Dust.** The predominant PBDE exposure pathway for the general population of the United States is from indoor dust (EPA 2010; Lorber 2008). Total PBDE (sum of BDE 28, 47, 66, 85, 99, 100, 153, 154, 183,

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206, and 209) levels in dust samples obtained from 20 residences located near the Columbia River in Washington state ranged from 311 to 19,700 ng/g, with BDE 209 being the predominant congener (Shreder and LaGuardia 2014). Total PBDE concentrations in settled dust from 10 office buildings located in Michigan ranged from 1,340 to 38,900 ng/g (median, 15,800 ng/g) (Batterman et al. 2010). BDE 47, BDE 99, and BDE 209 had the highest concentrations for the individual congeners, with a maximum value of 29,000 ng/g measured for BDE 209 in one of the office buildings. A study of eight office buildings in Boston, Massachusetts had even greater PBDE concentrations in settled dust. BDE 209 was detected in 100% of the samples at concentrations ranging from 912 to 106,204 ng/g (Watkins et al. 2011). Total pentaBDE congeners ranged from 141 to 61,264 ng/g in these office buildings. Lower concentrations of the lesser brominated congeners, tri-hexaBDE, were detected in dust samples in Europe as compared to the United States and Canada (Harrad et al. 2008). Concentrations of BDE 209 in dust samples are similar in the United States and the United Kingdom. Congener pattern analysis of indoor dust suggests that North American dusts are contaminated with decaBDE and pentaBDE commercial formulations, whereas U.K. dusts are predominantly contaminated with decaBDE. For example, the average concentration of total PBDE congeners in eight homes located in West Midlands, United Kingdom was about 215 ng/g, with a range of 16.2–625.4 ng/g (Harrad et al. 2006). Dust samples obtained from Spain and Belgium had total PBDE ranges of 2.9–380.2 and 6.2–384.8 ng/g, respectively (Harrad et al. 2006). Concentrations in North America also seem to be greater than even source-dominated areas in other parts of the world. The mean total concentration of 10 PBDE congeners in dust samples obtained from an office building located near a large automotive shredding and metal recycling facility in Brisbane, Australia was 2,014 ng/g (Hearn et al. 2013).

**Food.** Food ingestion typically accounts for <20% of the total PBDE intake for adults in North America (EPA 2010; Lorber 2008); however, it accounts for the majority of intake for the European population, with the exception of BDE 209 where dust exposure is the primary source (Abdallah and Harrad 2014; Trudel et al. 2011). Schecter et al. (2006) measured concentrations of 13 PBDE congeners in 62 food samples in the United States. Concentrations of total PBDEs ranged from 7.9 pg/g in milk to 3,762 pg/g in canned sardines. Fish, meat, and dairy products tended to have the highest concentrations of PBDEs. The results of these measurements for meat and fish are reproduced in Table 6-4. A survey of three categories of baby food (formula, cereal, and puree) from the United States and China found the median concentrations of total PBDE (sum of BDE 17, BDE 28, BDE 47, BDE 49, BDE 100, BDE 153, BDE 183, and BDE 209) were 21 and 36 pg/g in foods purchased in the United States and China, respectively (Liu et al. 2014).

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**Table 6-4. PBDE Concentrations (pg/g Wet Weight) in 18 U.S. Meat and Fish Samples<sup>a</sup>**

Sample	Lipid (%)	BDE 17	BDE 28	BDE 47	BDE 66	BDE 77	BDE 85	BDE 99	BDE 100	BDE 138	BDE 153	BDE 154	BDE 183	BDE 209	Total PBDEs <sup>b</sup>
U.S. meat samples															
Bacon A	52.3	ND (5.2)	ND (7.1)	ND (78.8)	ND (5.2)	ND (5.2)	ND (5.2)	ND (28.8)	ND (6.8)	ND (5.2)	ND (5.2)	ND (5.2)	ND (5.2)	ND (166.6)	165
Bacon B	43.4	ND (0.4)	ND (2.1)	ND (19.9)	ND (0.4)	ND (0.2)	NA	ND (15.6)	ND (2.8)	ND (0.4)	ND (1.1)	ND (0.9)	ND (1.7)	ND (32.8)	39
Bacon C	35.3	0.7	ND (2.0)	30.1	NA	NA	1.4	16.8	4.8	ND (0.7)	4.5	2.8	14.3	28.4	105
Beef (ground) A	30.7	ND (3.1)	59.7	87.5	ND (3.1)	ND (3.1)	ND (3.1)	35.5	6.2	ND (3.1)	6.8	4.6	ND (4.2)	ND (95.7)	258
Beef (ground) B	13.6	0.2	ND (0.7)	23.4	0.5	NA	NA	32.3	4.5	0.4	4.7	2.5	NA	9.7	79
Beef tenderloin	13.7	ND (1.4)	ND (1.5)	35.1	ND (1.4)	NA	1.7	40.3	6.9	ND (1.4)	4.9	3.7	3.8	ND (11.1)	105
Chicken breast	4.9	ND (0.04)	0.5	60.5	NA	NA	NA	128	17.1	2.2	12.0	10.8	3.2	48.5	283
Duck	75.1	ND (0.5)	ND (3.0)	286	2.7	ND (0.3)	15.2	609	122	7.3	52.3	42.9	31.6	113	1,283
Ground chicken	7.3	ND (0.7)	ND (1.5)	11.0	ND (0.7)	NA	ND (0.7)	18.9	4.6	ND (0.7)	4.1	2.6	5.8	80	129
Ground lamb	19.7	ND (2.0)	ND (2.1)	ND (23.0)	ND (2.0)	ND (2.0)	3.2	56.8	16.8	ND (2.0)	9.6	6.3	ND (2.0)	ND (150.6)	186
Ground pork	21.5	ND (2.2)	ND (3.5)	53.8	ND (2.2)	NA	3.1	74.2	12.9	4.3	18.7	15.0	19.9	ND (31.3)	221
Ground turkey	11.1	0.2	ND (0.5)	98	0.8	ND (0.1)	NA	217	54.4	3.9	32.9	24.1	36.8	245	713
Pork	8.9	0.1	ND (0.5)	6.9	NA	NA	NA	16.3	1.8	0.2	1.0	1.2	1.3	11.7	41
Pork sausage A	23.7	ND (1.3)	ND (6.9)	387	ND (1.0)	ND (0.3)	16.8	688	74.5	5.6	81.6	55.3	14.6	49.7	1,378
Pork sausage B	24.4	ND (2.4)	ND (3.4)	39.4	ND (2.4)	ND (2.4)	2.6	71.6	8.3	ND (2.4)	22.0	13.7	10.7	ND (139)	244
Sausage A	26.2	ND (2.6)	ND (5.5)	ND (34.8)	ND (2.6)	NA	3.1	40.1	6.4	ND (2.6)	5.9	4.9	6.9	ND (51.0)	1,426
Sausage B	28.5	ND (2.9)	ND (3.2)	94.1	ND (3.5)	ND (2.9)	ND (2.9)	43.7	8.3	ND (2.9)	8.5	9.2	ND (2.9)	ND (41.7)	195
Wieners	32.9	ND (0.3)	ND (1.5)	386	1.4	ND (0.2)	11.1	703	53.9	7.2	106	49.8	14.3	ND (28.7)	1,348
Mean	26.3	0.76	4.59	93.2	1.19	0.83	4.93	157	22.7	2.33	21.1	14	10.1	53.3	383
Median	24.1	0.66	1.03	39.4	1.08	0.57	2.62	42	7.57	1.37	7.68	5.63	5.83	38.1	190
Minimum	4.87	0.02	0.24	6.93	0.21	0.06	0.36	7.79	1.39	0.16	0.53	0.44	0.86	5.53	39
Maximum	75.1	2.62	59.7	387	2.74	2.62	16.8	703	121	7.28	106	55.3	36.8	245	1,426
U.S. fish samples															
Canned tuna A	0.3	0.1	0.6	5.1	0.2	NA	0.2	3.2	0.6	ND (0.0)	0.3	0.2	1.1	4.9	16.6
Canned tuna B	0.5	ND (0.1)	0.2	2.1	0.2	NA	ND (0.1)	1.1	0.4	ND (0.1)	0.2	0.3	2.1	8.8	15.5
Catfish A	11.1	4.6	6.4	372	4.3	NA	NA	589	116	5.1	37.1	39.6	7.3	1269	2,450
Catfish B	5.3	4.6	5.1	438	13.5	ND (0.1)	41.6	834	102	7.9	49.9	45.8	4.9	ND (15.9)	1,547
Catfish C	5.2	2.2	3.7	137	0.7	ND (0.5)	11.7	184	39.5	ND (2.7)	15.8	15.2	ND (1.6)	ND (49.4)	437

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**Table 6-4. PBDE Concentrations (pg/g Wet Weight) in 18 U.S. Meat and Fish Samples<sup>a</sup>**

Sample	Lipid (%)	BDE 17	BDE 28	BDE 47	BDE 66	BDE 77	BDE 85	BDE 99	BDE 100	BDE 138	BDE 153	BDE 154	BDE 183	BDE 209	Total PBDEs <sup>b</sup>
Catfish fillet (farm)	5.7	1.1	3.7	197	6.3	NA	16.4	282	53.0	ND (4.1)	18.4	21.3	3.8	22.7	627
Halibut	0.2	0.6	4.1	76.6	2.8	NA	ND (0.1)	10.6	12.4	ND (0.1)	1.1	2.6	1.8	11.4	124
Herring	9.1	4.1	56.3	2,072	69.4	3.6	ND (0.9)	267	221	ND (0.9)	29.3	69.9	2.5	ND (26.4)	2,809
Mahi	0.5	0.6	ND (2.0)	24.1	2.0	NA	0.6	13.0	5.1	ND (0.8)	1.4	4.9	4.3	ND (16.6)	66
Salmon A	8.0	79.2	92.6	1,222	30.6	ND (0.2)	NA	93.2	348	ND (0.2)	27.7	98.8	1.4	ND (9.0)	1,999
Salmon B	13.9	118	142	2,081	59.1	ND (0.1)	NA	147	353	ND (0.2)	36.6	142	ND (1.2)	ND (7.0)	3,082
Salmon C	10.3	18.4	49.4	1,103	35.3	ND (0.1)	ND (0.1)	239	217	ND (0.1)	18.3	45.1	ND (1.3)	ND (11.2)	1,732
Salmon D	6.3	1.4	5.2	94.7	5.2	ND (0.9)	ND (0.6)	15.4	7.1	ND (0.6)	1.4	5.0	ND (0.8)	ND (9.1)	141
Salmon E	12.3	1.7	20.4	356	ND (2.1)	ND (1.2)	ND (1.2)	84.4	84.2	ND (1.2)	10.1	29.8	ND (1.4)	ND (29.2)	605
Salmon fillet (farm) A	7.4	11.1	50.5	1,000	63.1	NA	7.9	410	210	ND (1.4)	37.4	104	3.7	20.5	1,919
Salmon fillet (farm) B	6.9	2.3	27.9	517	24.3	NA	ND (0.7)	168	115	ND (0.7)	16.0	35.8	1.7	681	1,590
Sardines	9.6	3.3	53.6	2,748	85.6	ND (5.0)	ND (1.0)	358	257	ND (1.0)	51.9	139	ND (3.2)	ND (51.4)	3,726
Shark	0.4	1.1	29.8	784	29.5	0.3	NA	57.8	608	0.4	112	291	2.0	5.4	1,920
Shrimp	0.6	0.3	3.6	75.6	NA	NA	NA	9.4	14.3	ND (0.1)	1.2	2.6	0.2	ND (1.3)	108
Tilapia	1.0	ND (0.1)	ND (0.7)	5.9	NA	NA	0.1	1.3	0.6	ND (0.1)	0.2	0.5	ND (0.2)	ND (4.0)	11
Trout A	4.2	4.8	22.2	320	NA	NA	ND (0.2)	79.8	66.5	0.2	11.8	26.3	4.4	ND (26.7)	549
Trout B	10.1	4.3	49.3	826	ND (5.6)	ND (1.0)	ND (1.0)	128	198	ND (1.0)	24.7	61.3	2.5	ND (42.9)	1,319
Tuna	0.2	ND (0.1)	ND (1.0)	16.6	0.7	NA	ND (0.0)	ND (4.6)	2.9	ND (0.1)	ND (0.4)	ND (1.0)	0.5	23.4	48
Wild perch	1.2	ND (0.1)	0.7	10.2	0.4	NA	ND (0.1)	2.3	2.1	ND (0.1)	0.7	2.4	0.6	5.9	25
Mean	5.43	11.01	26.19	603	20.8	0.78	4.29	166	126	0.89	21	49.3	2.08	91.8	1,120
Median	5.52	1.97	5.77	338	5.23	0.30	0.35	88.8	75.3	0.33	15.9	28.	1.68	10.1	616
Minimum	0.15	0.03	0.20	2.11	0.18	0.06	0.02	1.15	0.43	0.02	0.21	0.21	0.12	0.63	11.14
Maximum	13.9	118	142	2,748	85.6	3.60	41.6	834	608	7.94	112	291	7.32	1,269	3,726

<sup>a</sup>Limits of detection (LODs) are shown in parentheses. Total PBDE concentrations and statistics for each congener were calculated by assuming that nondetected concentrations were one-half the LOD; for calculations, these were treated as zero.

<sup>b</sup>Totals rounded to the nearest whole number for hundreds and to the nearest decimal place for tens.

NA = not available; ND = not detected

Source: Adapted from Schecter et al. (2006)

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Huwe et al. (2002a) reported total PBDE concentrations in farm chickens raised in two different regions of the United States. The total PBDE concentration of discrete samples of chickens raised in Arkansas was 39.4 ng/g whole weight, while one composite sample of chickens raised in North Dakota was 1.7 ng/g whole weight. In the United States, chickens fed ball clay (a sedimentary, kaolinite clay) and chickens bought in the grocery store were analyzed for total PBDEs (ENVIRON 2003b). BDE 99 was the dominant congener in all samples. Total PBDEs ranged between 4 and 35 ng/g lipid weight in chickens fed ball clay and 0.5 ng/g lipid weight in store-bought chicken. Ohta et al. (2002) determined the concentration of total PBDEs in vegetables and meat samples from Japan. The concentrations of PBDEs in spinach, potato, and carrot were 134, 47.6, and 38.4 pg/g fresh weight, respectively. The highest concentrations of total PBDEs and BDE 47 were found in spinach. Interestingly, different congener patterns were found among the vegetables analyzed. Compared to root vegetables, which had high concentrations of BDE 153, spinach (representing a leafy vegetable) might be strongly influenced by PBDE contamination in air. The concentrations of PBDEs in pork, beef, and chicken were 63.6, 16.2, and 6.25 pg/g fresh weight, respectively. PBDE concentrations were highest in pork samples; however, the reason for this is unknown (Ohta et al. 2002). Bocio et al. (2003) determined the concentrations of PBDEs in food samples from Catalonia, Spain during 2000. The highest concentration of total PBDEs was found in oils and fats (587.7–569.3 pg/g), followed by fish and shellfish (333.9–325.3 pg/g), meat and meat products (109.2–102.4 pg/g), and eggs (64.5–58.3 pg/g). In all of these food groups, a predominance of the tetra- and pentaBDE homologs, followed by hexaBDE, was observed in the sum total PBDEs. By contrast, PBDEs were not detected in the groups of fruits, cereals, or tubers. Four types of commercial fish oils sold in Sweden were found to contain PBDEs (0.2–28.1 ng/g lipid weight) (Haglund et al. 1997). The highest concentration of PBDEs was found in the cod liver oil. These oils were from products marketed as dietary supplements for humans. The concentrations of PBDEs in seafood from the Inland Sea of Japan were determined for samples collected in 1998 (Hori et al. 2000). BDE 28, BDE 47, BDE 66, BDE 99, BDE 100, BDE 153, and BDE 154 were detected in all analyzed seafood samples. BDE 47 was detected as the predominant congener, with concentrations ranging from 58 to 2,100 pg/g wet weight. Harrad et al. (2004) determined the concentrations of several PBDE congeners in omnivorous and vegan diet samples from the United Kingdom. Median concentrations of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 in omnivorous diet samples were 66.8, 63.8, 10, 20, and 20 pg/g dry weight, respectively. In vegetarian samples, median concentrations of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 were 47.2, 56.7, 10.0, 20, and 20 pg/g dry weight, respectively. Concentrations of BDE 47, BDE 99, and total PBDE were found to be statistically higher in omnivorous diet samples compared to vegetarian diet samples.

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***Biosolids and Effluents.*** The concentrations of PBDEs in biosolids (sewage sludge) and effluents are summarized in Table 6-5. PBDEs were detected in 11 biosolids obtained from Virginia, New York, Maryland, and California (Hale et al. 2001c). The total concentrations of pentaBDE in biosolids ranged from 1,100 to 2,290 ng/g dry weight. The concentration of decaBDE (BDE 209) varied widely among biosolids from the four states; the concentration of BDE 209 ranged from 84.8 to 4,890 ng/g dry weight in the biosolid samples.

Levels of PBDEs in biosolids were analyzed using samples from the EPA 2001 National Sewage Sludge Survey (NSSS) (Venkatesan and Halden 2014). Thirty-two PBDEs were detected in the samples analyzed. The total mean±standard deviation PBDE concentration detected in biosolids composites was 9,388±7,778 µg/kg dry weight. Deca, nona, and penta BDE congeners accounted for roughly 57, 18, and 13% of the total, respectively. Using these data and the estimated annual biosolids production and disposal figures in the United States, the annual mean loading rate of PBDEs was estimated to range from 47,900 to 60,100 kg/year. Analysis of samples collected between August 2006 and March 2007 (2–3 years after the voluntary phase-out of penta and octa PBDE formulations) indicated that the levels of 8 out of 11 major congeners in biosolids had declined approximately 10–57 % when compared to 2001 levels.

Sewage sludge in the vicinity of the Dan River (Virginia) were collected and analyzed for PBDEs (Hale et al. 2002). Congener patterns suggestive of both penta- and decaBDE commercial products were present at concentrations of 1,370 ng/g dry weight (sum of BDE 47 to BDE 154) and from 1,470 ng/g dry weight, respectively. While no known industrial source of pentaBDE discharged to this plant, the distribution pattern for lower-brominated congeners matched the pentaBDE commercial product.

Sewage sludge samples from 13 WWTPs in Germany were sampled (Hagenmaier et al. 1992). The mean concentration of tri- to heptaBDEs was 8.37 ng/g, with tri-, tetra-, penta-, hexa-, and heptaBDEs at concentrations of 0.65, 3.06, 3.02, 0.49, and 0.22 ng/g, respectively. Concentrations of penta- and hexaBDEs were highest in these samples. de Boer et al. (2000b) determined the concentration of PBDEs in sewage treatment plant effluents from the Netherlands. The concentration of total PBDEs (the sum of BDE 47, BDE 99, and BDE 100) ranged from 11 to 35 ng/g dry weight with the overwhelming majority as BDE 47, while the concentrations of BDE 209 ranged from 310 to 920 ng/g dry weight. Kohler et al. (2003) determined the concentrations of decaBDE in sewage sludge from Switzerland between 1993 and 2002. These authors reported that the average concentration of decaBDE increased with time from 220 to 1,100 ng/g dry weight, corresponding to an average increase of 560%.

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**Table 6-5. Concentrations (ng/g Dry Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Biosolids (Sewage Sludge) and Effluents**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Sewage sludge	Dan River, Virginia	No data	No data	No data	2,840*	1,470	Hale et al. 2002
Sewage sludge	11 biosolid samples from Virginia, New York, Maryland, and California	No data	No data	No data	1,100–2,290*	No data	Hale et al. 2001c
Sewage sludge	Gothenburg, Sweden	15	19	3.5	38	No data	Nylund et al. 1992
Sewage sludge	Klippan, Sweden	22	18	5.4	45.4	No data	Sellström et al. 1999; Sellström and Jansson 1995
Sewage Sludge	Rimbo, Sweden	53	53	13	119	No data	Sellström et al. 1999; Sellström and Jansson 1995
Sewage sludge	Three plants, Stockholm, Sweden	39–91	48–120	11–28	98–239	140–350	Sellström et al. 1999
Sewage sludge	Germany	No data	No data	No data	04–15*	No data	Hagenmaier et al. 1992
Sewage treatment plant effluents	Netherlands, several sites	11–35	<1	No data	11–35	310–920	de Boer et al. 2000b

<sup>a</sup>ΣPBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether



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**World Trade Center Site.** In 2001, PBDEs were detected in dust and smoke samples taken near the World Trade Center disaster site (Lioy 2002). The highest concentration was for decaBDE (i.e., BDE 209), which was present in thermoplastics (e.g., computers). Concentrations of PBDE congeners were 107–174 µg/kg dry weight basis for BDE 47, 51.1–74.1 µg/kg dry weight basis for BDE 100, 155–293 µg/kg dry weight basis for BDE 99, 42.0–53.5 µg/kg dry weight basis for BDE 153, 219–305 µg/kg dry weight basis for BDE 154, and 1,330–2,660 µg/kg dry weight basis for decaBDE (BDE 209). Concentrations of PBDEs were found to be similar to concentrations found in sewage sludge (Lioy 2002).

**Freshwater Fish.** Monitoring data indicated that the concentrations of PBDEs were historically increasing in freshwater organisms, with higher concentrations near point sources. The congener profiles show the highest concentrations for BDE 47. The presence of PBDEs in freshwater aquatic organisms taken from remote regions suggests that diffuse sources of PBDEs are also important. A sampling of concentrations of PBDEs in freshwater fish samples in the United States are summarized in Table 6-6. Fish were sampled from two U.S. lakes, Hadley Lake, Indiana near a possible PBDE point source, and Lake of the Ozarks, Missouri, with no known sources (Dodder et al. 2000). Mean total PBDE concentrations (sum of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154) were higher in crappie (*Pomoxis annularis*) and bluegill (*Lepomis macrochirus*) from Hadley Lake (1,500 and 1,900 ng/g lipid weight, respectively) than from Lake of the Ozarks (340 and 390 ng/g lipid, respectively). BDE 47, BDE 99, BDE 153, and BDE 154 were the primary congeners. From the Lake of Ozarks, BDE 47 was the dominant congener in fish. The total PBDE concentrations in smelt (*Osmerus mordax*) from Lakes Superior and Ontario were 150±9 and 240±30 ng/g lipid, respectively (Dodder et al. 2002). The dominant congeners in these fish were BDE 47 and BDE 99.

An analysis of fish tissue samples from selected locations in Washington State showed that total PBDE concentrations ranged from 29 ng/g lipid in rainbow trout from a remote spring-fed stream (Douglas Creek, Washington) to 19,000 ng/g lipid in rainbow trout from the urbanized Spokane River, Washington (Johnson and Olson 2001). The tetra- and pentaBDE isomers were the major compounds present.

TetraBDE to hexaBDE were found in carp (*Cyprinus carpio*) from the Buffalo River (New York), a polluted area around the Great Lakes (Loganathan et al. 1995). TetraBDEs dominated the congener pattern with 94–96% of total PBDEs. TetraBDE and pentaBDE concentrations ranged from 13 to 22 ng/g fresh weight. Asplund et al. (1999a) found tri- to hexaBDEs in steelhead trout (*Oncorhynchus mykiss*) sampled in 1995 from Lake Michigan. The combined concentration of BDE 47, BDE 99, BDE 100,

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**Table 6-6. Concentrations (ng/g Lipid Weight, Except as Noted) of Several Polybrominated Diphenyl Ethers (PBDEs) in Freshwater Fish Samples from the United States**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Lake trout	Lake Michigan	93–230	9.0–48	12–45	120–350*	No data	Streets et al. 2006
Lake trout	Lake Superior	22–79	8.5–53	5.2–19	39–180*	No data	Streets et al. 2006
Lake trout	Lake Huron	32–59	7.8–13	6.5–12	50–94*	No data	Streets et al. 2006
Lake trout	Lake Ontario	45–140	6.5–34	7.7–24	64–230*	No data	Streets et al. 2006
Lake Trout	Lake Ontario	4.3–114	59–680	7.4–1,285	269–3,339*	2.3–12	Ismail et al. 2009
Alewife	Grand Traverse Bay, Lake Michigan	16	No data	No data	36	No data	Stapleton and Baker 2003
Bloater chub	Grand Traverse Bay, Lake Michigan	11 (fw)	No data	No data	23 (fw)	No data	Stapleton and Baker 2003
Bluegill	Hadley Lake, Indiana	420	320	240	1,900	No data	Dodder et al. 2000
Bluegill	Lake of the Ozarks, Missouri	200	91	59	390	No data	Dodder et al. 2000
Burbot	Grand Traverse Bay, Lake Michigan	43 (fw)	No data	No data	86 (fw)	No data	Stapleton and Baker 2003
Carp	United States	No data	No data	No data	13–22* (fw)	No data	Loganathan et al. 1995
Carp	Detroit River, Grosse Isle, Michigan	3.0 (fw)	0.50 (fw)	0.48 (fw)	40.7*	No data	Rice et al. 2002
Carp	Des Plaines River, Joliet, Illinois	2.54 (fw)	0.5 (fw)	0.44 (fw)	281*	No data	Rice et al. 2002
Carp	Des Plaines River, Joliet, Illinois	1.34 (fw)	0.50 (fw)	0.49 (fw)	78.3*	No data	Rice et al. 2002
Carp (fillet)	Yakima River, Washington	No data	No data	No data	22 (fw)	No data	Johnson and Olson 2001
Crappie	Hadley Lake, Indiana	250	430	150	1,500*	No data	Dodder et al. 2000
Crappie	Lake of the Ozarks, Missouri	190	78	59	340*	No data	Dodder et al. 2000
Deepwater sculpin	Grand Traverse Bay, Lake Michigan	2.8 (fw)	No data	No data	3 (fw)	No data	Stapleton and Baker 2003
Lake trout	Grand Traverse Bay, Lake Michigan	75 (fw)	No data	No data	126 (fw)	No data	Stapleton and Baker 2003

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**Table 6-6. Concentrations (ng/g Lipid Weight, Except as Noted) of Several Polybrominated Diphenyl Ethers (PBDEs) in Freshwater Fish Samples from the United States**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Lake trout	Lake Ontario, United States	No data	No data	No data	540*	No data	Alaee et al. 1999
Lake trout	Lake Ontario, United States	58 (fw)	14 (fw)	5.7 (fw)	No data	No data	Luross et al. 2002
Lake trout	Lake Huron, United States	No data	No data	No data	240*	No data	Alaee et al. 1999
Lake trout	Lake Huron, United States	27 (fw)	7.7 (fw)	3.8 (fw)	No data	No data	Luross et al. 2002
Lake trout	Lake Superior, United States	No data	No data	No data	140*	No data	Alaee et al. 1999
Lake trout	Lake Superior, United States	29 (fw)	12 (fw)	4.1 (fw)	No data	No data	Luross et al. 2002
Lake trout	Lake Erie, United States	No data	No data	No data	117*	No data	Alaee et al. 1999
Lake trout	Lake Erie, United States	16 (fw)	2.0 (fw)	2.5 (fw)	No data	No data	Luross et al. 2002
Largescale sucker (whole)	Yakima River, Washington	No data	No data	No data	64 (fw)	No data	Johnson and Olson 2001
Largescale sucker (whole)	Spokane River, Washington	No data	No data	No data	105 (fw)	No data	Johnson and Olson 2001
Mountain whitefish (whole)	Spokane River, Washington	No data	No data	No data	1,250 (fw)	No data	Johnson and Olson 2001
Rainbow trout (whole)	Douglas Creek, Washington	No data	No data	No data	1.5 (fw)	No data	Johnson and Olson 2001
Rainbow trout	Spokane River, Washington	No data	No data	No data	20–174 (fw) (fillet)	No data	Johnson and Olson 2001
Northern pike minnow	Spokane River, Washington	59–160	0.3–<0.4	17–47	297 (fw) (whole) No data	Not detected	Furl and Meredith 2010
Mountain whitefish	Spokane River, Washington	127–942.6	81–942.6	26.3–368.1	No data	Not detected	Furl and Meredith 2010
Largescale sucker	Spokane River, Washington	87–270	0.2–<4.4	13–45	No data	Not detected	Furl and Meredith 2010
Salmon	Grand Traverse Bay, Lake Michigan	34 (fw)	No data	No data	95 (fw)	No data	Stapleton and Baker 2003
Salmon	Lake Michigan, United States	52.1 (fw)	9.3 (fw)	9.7 (fw)	2,440	No data	Manchester-Neesvig et al. 2001
Smelt	Lake Superior, United States	5.7 (fw)	1.8 (fw)	0.98 (fw)	150	<1.5 (fw)	Dodder et al. 2002

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**Table 6-6. Concentrations (ng/g Lipid Weight, Except as Noted) of Several Polybrominated Diphenyl Ethers (PBDEs) in Freshwater Fish Samples from the United States**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Smelt	Lake Ontario, United States	10 (fw)	5.3 (fw)	1.6 (fw)	240	<1.6 (fw)	Dodder et al. 2002
Starry flounder (whole)	Columbia River, Washington	No data	No data	No data	30 (fw)	No data	Johnson and Olson 2001
Steelhead trout	Lake Michigan, United States	1,700	600	360	3,000*	No data	Asplund et al. 1999b
Whitefish	Columbia River, United States	No data	No data	No data	72 (fw)	No data	Rayne et al. 2003a
Whitefish	Grand Traverse Bay, Lake Michigan	9.8 (fw)	No data	No data	18 (fw)	No data	Stapleton and Baker 2003

<sup>a</sup> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether; dw = dry weight; fw = fresh weight

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BDE 153, and BDE 154 was 3,000 ng/g lipid weight (Asplund et al. 1999b). Lake trout (*Salvelinus namaycush*) from Lakes Ontario, Huron, and Superior were also found to have di- to heptaBDEs with combined concentrations of 545, 237, and 135 ng/g lipid weight, respectively (Alaee et al. 1999).

Lake trout from Lake Erie had 117 ng/g lipid weight (Luross et al. 2000). Variations in local sources, combined with atmospheric transport, may explain differences that were seen in congener profiles for the different lakes. A retrospective temporal study for the years 1978, 1983, 1988, 1993, and 1998 using archived trout samples from Lake Ontario show a dramatic increase in total PBDE concentrations over time (Luross et al. 2000). At 50 freshwater sites in Virginia, muscle samples from 253 fish samples were collected and analyzed for PBDEs (Hale et al. 2000, 2001b). Approximately 85% of the samples contained BDE 47, the predominant congener, at measurable concentrations. Concentrations were >1,000 ng/g lipid weight at 9 of 50 sites. The highest combined PBDE concentrations (up to 57,000 ng/g lipid weight) were observed in carp downstream of textile and furniture facilities. BDE 47 concentrations were greater than 2,2',4,4',5,5'-hexachlorobiphenyl (PCB 153) concentrations in 58% of the samples analyzed. PBDEs were identified in fish collected from the Detroit River (Michigan) and Des Plaines Rivers (Illinois). In Detroit River fish (carp and largemouth bass), the congener patterns were dominated by BDE 47; however, in the Des Plaines River carp, the dominant congeners were heptaBDE congeners (BDE 181 and BDE 183), lesser amounts of BDE 190, and two hexaBDEs (BDE 154 and BDE 153). Possible sources for the heptaBDE congeners were not obvious Ozarks (340 and 390 ng/g lipid, respectively). BDE 47, BDE 99, BDE 153, and BDE 154 were the primary congeners. From the Lake of Ozarks, BDE 47 was the dominant congener in fish. The total PBDE concentrations in smelt (*O. mordax*) from Lakes Superior and Ontario were 150±9 and 240±30 ng/g lipid, respectively (Dodder et al. 2002). The dominant congeners in these fish were BDE 47 and BDE 99. An analysis of fish tissue samples from selected locations in Washington State showed that total PBDE concentrations ranged from 29 ng/g lipid in rainbow trout from a remote spring-fed stream (Douglas Creek, Washington) to 19,000 ng/g lipid in rainbow trout from the urbanized Spokane River, Washington (Johnson and Olson 2001). The tetra- and pentaBDE isomers were the major compounds present. TetraBDE to hexaBDE were found in carp (*C. carpio*) from the Buffalo River (New York), a polluted area around the Great Lakes (Loganathan et al. 1995). TetraBDEs dominated the congener pattern with 94–96% of total PBDEs. TetraBDE and pentaBDE concentrations ranged from 13 to 22 ng/g fresh weight. Asplund et al. (1999a) found tri- to hexaBDEs in steelhead trout (*O. mykiss*) sampled in 1995 from Lake Michigan. The combined concentration of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 was 3,000 ng/g lipid weight (Asplund et al. 1999b). Lake trout (*S. namaycush*) from Lakes Ontario, Huron, and Superior were also

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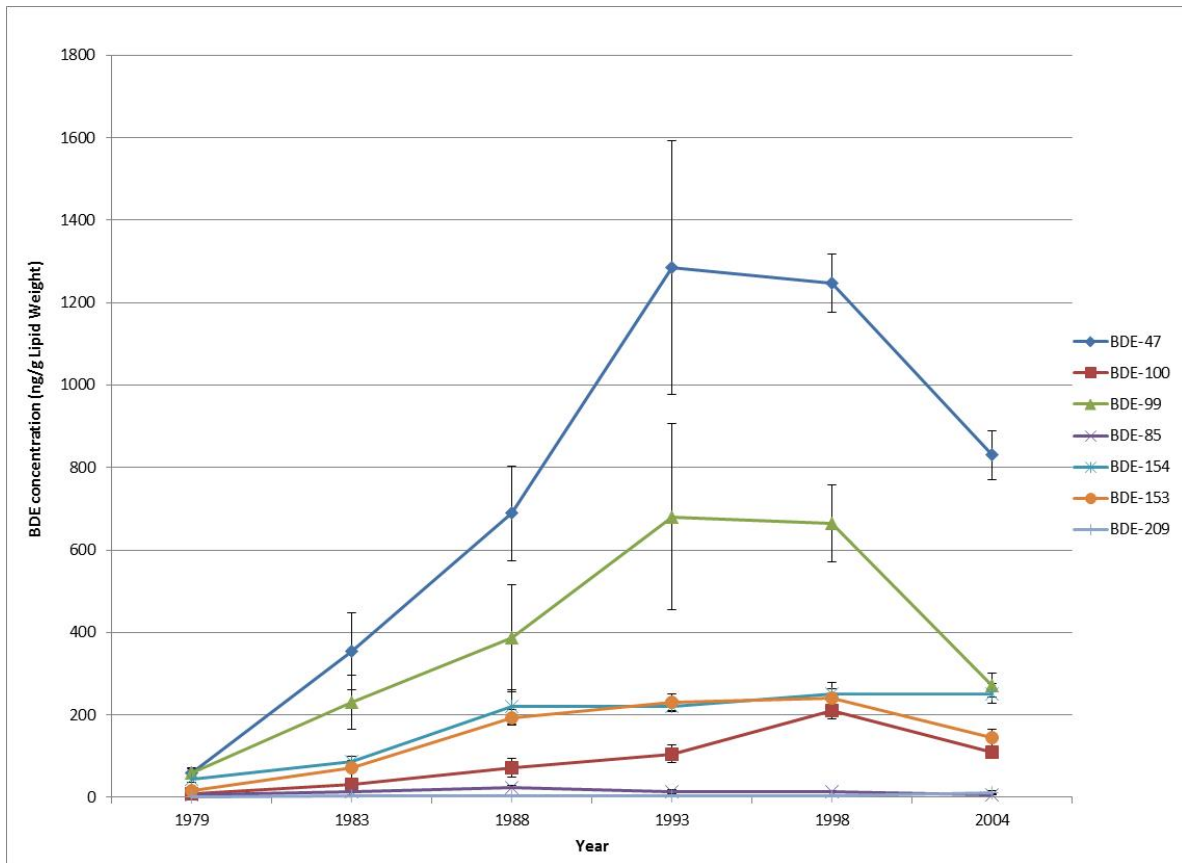
found to have di- to heptaBDEs with combined concentrations of 545, 237, and 135 ng/g lipid weight, respectively (Alaee et al. 1999).

The National Study of Chemical Residues in Lake Fish Tissue (1998–2009) is one of the statistically-based surveys conducted by EPA that analyzed the concentrations of PBDEs and other contaminants in fish from 500 lakes in the continental United States (EPA 2009c, 2013k). The most prevalent PBDE congeners detected in both predator and bottom dweller fish were reported as BDE 47, BDE 99, and BDE 100 (EPA 2013k).

Ismail et al. (2009) studied the temporal trends of PBDE congeners in trout obtained from Lake Ontario from 1979 to 2004. Concentrations of most PBDE congeners (BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154) increased dramatically from 1979 to the mid-1990s and then either leveled off or decreased from 1998 to 2004; average concentration and standard error values are presented in Figure 6-2. The temporal trend of BDE 209 was different than for the other congeners, however. Concentrations of BDE 209 in lake trout increased more slowly from 1979 until the mid-1990s, but its concentrations increased dramatically from the late 1990s ( $3.3 \pm 0.8$  ng/g in 1998) until 2004 ( $12 \pm 5.3$  ng/g in 2004), corresponding to its increased use in consumer products.

The concentrations of PBDEs in freshwater fish samples from Europe are summarized in Table 6-7. Between 1986 and 1988, concentrations of BDE 47, BDE 99, and BDE 100 were measured in whitefish (*Coregonus spp.*) from a remote mountain lake in Northern Sweden (Lake Storvindeln), in Arctic char (*Salvelinus alpinus*) from a heavily populated lake (Lake Vättern) in south-central Sweden with numerous municipal and industrial point sources, and in trout (*Salmo trutta*) and pike (*Esox lucius*) from several sites along Dalslands Canal in west central Sweden (Jansson et al. 1993). No point sources of PBDEs were identified from these sites. Whitefish from the remote lake contained the lowest concentrations (26 ng/g lipid weight) of PBDEs, whereas the Arctic char, from a heavily populated lake, contained 520 ng/g lipid weight PBDEs. In both samples, BDE 47 was the predominant congener. PBDE concentration in pike and trout from the Dalslands Canal ranged from 180 to 210 ng/g lipid weight and from 280 to 1,200 ng/g lipid weight, respectively. The congener pattern in these samples was similar to the technical mixture, Bromkal 70-5DE, with equal quantities of both BDE 47 and BDE 99. The concentrations in pike and trout are of the same order of magnitude as in the Arctic char, indicating the spread of PBDEs from diffuse sources (de Wit 2002). In 1979 and 1980, high concentrations of tri- to hexaBDEs (range, 950–27,000 ng/g lipid weight in muscle tissues) were measured in fish sampled along a river in Sweden (Viskan) where numerous textile industries are located (Andersson and Blomkvist

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**Figure 6-2. Temporal Trends in the Concentrations (ng/g Lipid Weight) of PBDE Congeners in Lake Trout from Lake Ontario, Canada**

Source: Ismail et al. 2009

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**Table 6-7. Concentrations (ng/g Lipid Weight Except as Noted) of Several Polybrominated Diphenyl Ethers (PBDEs) in Freshwater Fish Samples from Europe**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Freshwater fish	Pyrenees Mountains	0.40–0.51	0.24	0.16–0.18	No data	No data	Gallego et al. 2007
Freshwater fish	Tatras Mountains	0.20–0.26	0.17–0.21	0.043	No data	No data	Gallego et al. 2007
Arctic char	Lake Vättern, Sweden	400	64	51	520	No data	Sellström et al. 1993
Bream	Netherlands (several sites)	0.2–130 (dw)	Not detected	No data	No data	No data	de Boer et al. 2000b
Eels	Netherlands	<20–1,400	No data	No data	<50–1,700	No data	de Boer 1990
Osprey	Sweden	1,800	140	200	2,140	No data	Sellström et al. 1993
Pike	Dalslands canal, Sweden	94–98	60–79	25–36	180–210	No data	Sellström et al. 1993
Pike	River Viskan, Sweden, upstream and downstream	<46–2,000	<37–1,600	<14–1,000	<130–4,600	Trace	Sellström et al. 1998a
Several fish species	Germany	No data	No data	No data	19–983*	No data	Krüger 1988
Trout	Dalslands canal, Sweden	120–460	130–590	33–150	280–1,200	No data	Sellström et al. 1993
Whitefish	Lake Storvindeln, Sweden	15	7.2	3.9	26	No data	Sellström et al. 1993
Whitefish	Lake Geneva, Switzerland	26	13	2.5	44*	No data	Zennegg et al. 2003
Whitefish	Lake Greifen, Switzerland	96	52	9.1	165*	No data	Zennegg et al. 2003
Whitefish	Lake Biel, Switzerland	75.9	39	7.1	128*	No data	Zennegg et al. 2003
Whitefish	Lake Lucerne, Switzerland	56	46	10	121*	No data	Zennegg et al. 2003
Whitefish	Lake Zürich, Switzerland	56	25	4.5	89*	No data	Zennegg et al. 2003
Whitefish	Lake Nauchatel, Switzerland	41	20	4.0	68*	No data	Zennegg et al. 2003
Whitefish	Lake Constance, Switzerland	32	15	2.9	52*	No data	Zennegg et al. 2003



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**Table 6-7. Concentrations (ng/g Lipid Weight Except as Noted) of Several Polybrominated Diphenyl Ethers (PBDEs) in Freshwater Fish Samples from Europe**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Whitefish	Lake Thun, Switzerland	19	12	2.5	36*	No data	Zennegg et al. 2003

<sup>a</sup> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether

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1981). These textile industries have used PBDEs in the production of textiles. BDE 47 was the predominant congener at 70–80% of the total PBDEs. In 1977, the PBDEs were not detected in fish sampled at the same sites. The elevated concentrations of BDE 47, BDE 99, and BDE 100 were later confirmed in a follow-up study in which fish were caught from approximately the same locations (Sellström et al. 1993). In the current study, BDE 47 was the predominant congener at 65–96% of total PBDEs. Several fish species were sampled (pike, perch, bream, eel, tench, and sea trout) in these studies. In 1995, fresh samples of pike and sediments were collected at four of eight sites along River Viskan in order to search for point sources of contaminants. The combined concentrations of BDE 47, BDE 99, and BDE 100 ranged from not detected to 4,600 ng/g lipid weight; with BDE 47 being the predominant congener (50–90% of total). DecaBDE (BDE 209) was found in a few fish at trace amounts. The lowest concentrations of the PBDEs were found upstream of the industries. The concentrations of PBDEs increased further downstream as more industries were passed (Sellström et al. 1998a). Concentrations of BDE 47 ranged from <20 to 1,700 ng/g lipid in eels (*Anguilla anguilla*) from Dutch rivers and lakes (at 10 locations); BDE 47 comprised 70% of the total PBDEs (de Boer 1990). Bream (*Abramis brama*) sampled from several sites in the Netherlands had concentrations of BDE 47 ranging from 0.2 to 130 ng/g dry weight (de Boer et al. 2000b). BDE 99 was below the detection limits. BDE 153 ranged from <0.04 to 4.1 ng/g dry weight. Allchin et al. (1999) conducted a study of PBDEs in plaice (*Pleuronectes platessa*), flounder (*Platichthys flesus*), and dab (*Limanda limanda*) collected in the estuaries of rivers in the United Kingdom. Suspected sources of PBDEs in the estuaries include a manufacturer of pentaBDE and octaBDE, several industries using pentaBDE, and several landfills receiving wastes suspected to contain PBDEs. Concentrations of BDE 47, BDE 99, pentaBDE (as technical mixture DE-71), and octaBDE (as technical mixture DE-79) in fish ranged from not detected to 9,500 ng/g lipid weight, not detected to 370 ng/g lipid weight, 47–1,200 ng/g lipid weight, and not detected to 1,200 ng/g lipid weight. The highest concentrations were at Tees Bay downstream from a manufacturing plant on the River Tees. These results are similar to the situation found in Sweden along the River Viskan (Andersson and Blomkvist 1981; Sellström et al. 1993). Freshwater mussels (*Dreissena polymorpha*) were collected at several locations in the Netherlands and analyzed for BDE 47, BDE 99, BDE 153, and BDE 209 (de Boer et al. 2000b). Concentration ranges for the congeners were 0.7–17, 0.4–11, and <0.1–1.5 ng/g dry weight for BDE 47, BDE 99, and BDE 153, respectively; BDE 209 was below the detection limit. Poma et al. (2014) analyzed freshwater zebra mussels in Lake Maggiore, Northern Italy for the presence of PBDEs and other brominated flame retardants. Total tri- to heptaBDE concentrations ranged from 1.0 ng/g for samples obtained in May 2011 to 144.6 ng/g for samples collected in September 2012. The authors noted that even though penta-BDE was banned in Europe in 2004, increasing concentrations of tri- to heptaBDE congeners in mussels from 2011 to 2012 were observed in the samples obtained. Average

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values of hepta to decaBDE congeners in zebra mussels ranged from 88.2 to 182.8 ng/g and tended to be dominated by BDE 209, with average concentrations of 71.2–144.7 ng/g (Poma et al. 2014). The presence of the lower brominated octa and hepta congeners was likely due to metabolism of BDE 209 within the organism or the environmental debromination of BDE 209 followed by uptake by the zebra mussels.

***Saltwater Fish.*** Spatial trends show higher concentrations of lower-brominated BDE congeners found near human populated areas. The congener profiles show the highest concentrations for BDE 47. Representative concentrations of several PBDEs in marine aquatic species are summarized in Table 6-8. BDEs were detected in 10/10 white croakers and 8/8 shiner surfperch obtained from the San Francisco Bay at concentrations of (total PBDEs) 470–2, 260 and 730–3,930 ng/g lipid, respectively (Klosterhaus et al. 2012). Mean levels of total PBDEs in halibut, jack smelt, leopard shark, northern anchovy, shiner surfperch, striped bass white croaker, and white sturgeon collected from the San Francisco Bay in 2009 ranged from 1.5 to 8.3 ng/g wet weight (Sutton et al. 2015). In the year 2000, sole liver collected from five sites along the Canadian west coast (Crofton, Bamfield, Kitimat, Trincomali, and Vancouver) were analyzed for 14 BDE congeners (Ikonomou et al. 2002); the total PBDE concentrations were 64–340 ng/g lipid while the three highest congener concentrations were 27–160 ng/g lipid (BDE 47), 8.5–54 ng/g lipid (BDE 100), and 9.5–46 ng/g lipid (BDE 99). The highest concentrations were found in sole samples collected near Vancouver, Canada. DecaBDE was not detected in these samples at the level of procedural blank. Farmed salmon collected at two locations in Canada were analyzed for PBDE congeners (Easton et al. 2002). Forty-one congeners were detected with BDE 47 at the highest concentration (690 and 2,600 ng/g wet weight) followed by BDE 99 and BDE 100; total BDE congener concentrations were 1,188 and 4,147 ng/g wet weight for the two samples. Likewise, wild salmon from four locations in Canada were analyzed for BDE congeners. Concentrations were a factor of 10 lower for these samples compared to farmed salmon samples. The total PBDE concentration for the 41 detected congeners ranged from 38.7 to 485.2 ng/g wet weight. The concentration of the highest congener, BDE 47, ranged from 29 to 280 ng/g wet weight (Easton et al. 2002). PBDE concentrations in skipjack tuna from Asian offshore waters, off-Seychelles, off-Brazil, and open seas were determined for samples collected during 1996–2001 (Ueno et al. 2003). The concentration of total BDEs in muscles tissues ranged from not detected (<0.05 ng/g lipid) to 53 ng/g lipid. The concentration of the highest congener in muscle tissues, BDE 47, ranged from <0.1 to 15 ng/g lipid. BDE 99, BDE 100, BDE 153, and BDE 154 also were detected; BDE 209 was below the detection limit (<5.0 ng/g lipid) for these samples. Samples collected off the coast of the Seychelles (relatively pristine area) did not have detectable concentrations of any PBDEs, while samples collected in industrial areas of southeast Asia had the highest. Fall-caught

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**Table 6-8. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Aquatic Species**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
California halibut	San Francisco Bay	No data	No data	No data	1.8±0.5* (ww)	ND	Sutton et al.2015
Jack smelt	San Francisco Bay	No data	No data	No data	1.4±0.4* (ww)	ND	Sutton et al.2015
Leopard shark	San Francisco Bay	No data	No data	No data	5.0±1.2* (ww)	ND	Sutton et al.2015
Northern anchovy	San Francisco Bay	No data	No data	No data	7.9±2.9* (ww)	ND	Sutton et al.2015
Shiner surfperch	San Francisco Bay	No data	No data	No data	8.3±2.9* (ww)	ND	Sutton et al.2015
Striped bass	San Francisco Bay	No data	No data	No data	5.0±2.6* (ww)	ND	Sutton et al.2015
White croacker	San Francisco Bay	No data	No data	No data	4.3±2.5* (ww)	ND	Sutton et al.2015
White sturgeon	San Francisco Bay	No data	No data	No data	2.8±1.3* (ww)	ND	Sutton et al.2015
Winter Flounder	Northwest Atlantic	35	2.5	6.4	52*	ND	Shaw et al. 2009
Atlantic Herring	Northwest Atlantic	40	6.9	6.8	82*	ND	Shaw et al. 2009
American Plaice	Northwest Atlantic	42	4.0	7.0	69*	1.9	Shaw et al. 2009
White Hake	Northwest Atlantic	25	0.63	7.2	42*	0.91	Shaw et al. 2009
Alewife	Northwest Atlantic	8.3	3.6	1.7	18*	ND	Shaw et al. 2009
Atlantic Mackerel	Northwest Atlantic	20	7.5	4.1	69*	1.6	Shaw et al. 2009
Silver Hake	Northwest Atlantic	19	6.3	4.0	38*	ND	Shaw et al. 2009
Farmed Salmon	Canada	690; 2,600 (ww)	140; 390 (ww)	130; 470 (ww)	1,187; 4,147 (ww)	No data	Easton et al. 2002
Salmon (wild)	Canada	29–280 (ww)	ND–97 (ww)	4.2–43 (ww)	38.7–485.2 (ww)	No data	Easton et al. 2002

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**Table 6-8. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Aquatic Species**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Sole liver	West coast, Canada	27–160	9.5–46	8.5–54	64–340*	ND	Ikonomou et al. 2002
Skipjack tuna	Seychelles, Indian Ocean	<0.1	<0.05	<0.05	ND	<5.0	Ueno et al. 2003
Skipjack tuna	East China Sea	9.0–15	2.4–4.7	3.4–4.4	23–34	<5.0	Ueno et al. 2003
Skipjack tuna	Pacific Ocean	2.9–7.9	0.18–3.0	0.56–2.1	5.8–21	<5.0	Ueno et al. 2003
Herring	Baltic Sea	19–38	7.8–17	3.4–6	30–61	No data	de Wit 2002 Sellström et al. 1993
Herring	Baltic Sea	3.2–27	ND–2.9	1.3–1.9	3.2–32	No data	Haglund et al. 1997
Herring	Baltic Sea	7.6–24	4.3–3.9	No data	12.9–28.3*	No data	Strandman et al. 1999
Herring	Baltic Sea	6.3	0.6	0.8	12*	No data	Burreau et al. 1999
Herring	Kattegatt, Sweden	12	3.4	1.6	17	No data	de Wit 2002; Sellström et al. 1993
Herring	North Sea	8.4–100	No data	No data	No data	No data	de Boer 1990
Sprat (different age groups)	Baltic Sea	17.5–140.8	1.9–9.5	No data	21–149*	No data	Strandman et al. 1999
Sprat	Baltic Sea	4.3	0.7	0.8	8.4*	No data	Burreau et al. 1999
Cod liver	North Sea	170	No data	No data	1.9–360	No data	de Boer 1989
Salmon	Baltic Sea	167	52	44	220	No data	Haglund et al. 1997
Salmon	Baltic Sea	190	52	46	290	No data	Asplund et al. 1999b
Salmon	Baltic Sea	46	7.3	6.4	86*	No data	Burreau et al. 1999
Several fish species	Japan	No data	No data	No data	0.1–17*	No data	Watanabe et al. 1987
Yellowfin tuna	Japan	0.5	0.4	0.25	1.9*	No data	Ohta et al. 2000
Yellowtail	Japan	17	4.5	4.0	30.5*	No data	Ohta et al. 2000
Yellowtail (cultured)	Japan	29	3.3	5.3	44*	No data	Ohta et al. 2000
Salmon	Japan	22	8.1	5.3	46*	No data	Ohta et al. 2000

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**Table 6-8. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Aquatic Species**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Several flatfish	Seven river estuaries, Great Britain	73–9,500	16–790	No data	No data	ND	Allchin et al. 1999
Flounder	Netherlands, several sites (dw)	0.6–20	<0.01–4.6	No data	No data	No data	de Boer et al. 2000b

<sup>a</sup> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether; dw = dry weight; ND = not detected; ww = wet weight

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herring (*Clupea harengus*) muscle from five sites along the Swedish coast was analyzed for BDE 47, BDE 99, and BDE 100; the combined concentration of these three congeners ranged from 17 to 61 ng/g lipid, with BDE 47 being the dominant congener (Sellström et al. 1993). Likewise, the concentration of BDE 47 in Baltic herring ranged from 3.2 to 27 ng/g lipid in different age groups; the combined concentration of BDE 47, BDE 99, and BDE 100 ranged from 3.2 to 32 ng/g lipid (Haglund et al. 1997); 2-year-old herring had the lowest concentrations and 5-year-old herring had the highest concentrations. Similarly, Strandman et al. (1999) observed increasing concentrations with age of BDE 47, BDE 99, and BDE 153 in Baltic sprat (*Sprattus sprattus*, age 3–13 years). However, this trend was not evident for herring. BDE 47 was the primary congener with concentrations ranging from 7.6 to 24 ng/g lipid weight for 1–3-year-old sprat, 17–140 ng/g lipid weight for 3–13-year-old sprat, and 7.6–24 ng/g lipid weight in herring. The concentrations of BDE 47, BDE 99, and BDE 100 in whole-body composites of herring were 6.21, 0.62, and 0.81 ng/g lipid, respectively; in sprat, the concentrations were 4.32, 0.71, and 0.80 ng/g lipid, respectively (Burreau et al. 1999). Baltic sea herring had similar concentrations of BDE 47 (46.3 ng/g lipid) compared to 8.4–100 ng/g lipid of BDE 47 found by de Boer (1990) for herring collected from three regions of the North Sea. BDE 47, BDE 99, and BDE 153 concentrations in Baltic salmon (*Salmo salar*) muscle were 167, 52, and 4.2 ng/g lipid, respectively (Haglund et al. 1997). BDE 47, BDE 99, and BDE 100 concentrations were 47, 7.2, and 6.3 ng/g lipid, respectively, in whole-body composites (Burreau et al. 1999). In another study, the concentrations of BDE 47, BDE 99, and BDE 100 were determined in muscle, ripe eggs, and blood plasma from Baltic salmon (Asplund et al. 1999a). The mean concentrations of PBDEs in tissues from Baltic salmon (ng/g lipid weight) were as follows: BDE 47 (muscle, 190; ripe eggs, 64; blood, 190), BDE 99 (muscle, 52; ripe eggs, 16; blood, 55), and BDE 100 (muscle, 46; ripe eggs, 18; blood, 59). Cod (*Gadus morhua*) liver samples at three locations of the North Sea had combined concentrations of BDE 47 and BDE 99 of 1.9–360 ng/g lipid (de Boer 1989). BDE concentrations in flounder were 0.6–20 ng/g dry weight for BDE 47 and <0.01–4.6 ng/g dry weight for BDE 99 from several sites in the Netherlands (de Boer et al. 2000b). Concentrations of BDE 153 and BDE 209 were below the detection limit. In 1996, de Boer et al. (2001) measured the concentrations of two BDE congeners in flounder liver samples from the Amsterdam and Rotterdam harbors, and off the Dutch coast; BDE 47 and BDE 99 ranged from 15 to 280 and from <2 to 24 ng/g lipid weight, respectively. Olsson et al. (1999) detected BDE 47 in perch (*Perca fluviatilis*) from Latvia in a study examining environmental contamination in coastal areas of the former Soviet Union; the concentration of BDE 47 ranged from 6.4 to 10 ng/g lipid weight in the perch.

Watanabe et al. (1987) detected PBDEs in numerous marine fish and shell fish in Japan. TetraBDE and pentaBDE concentrations ranged from 0.1 and 17 ng/g fresh weight, with tetraBDE being the major

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congener. DecaBDE was also detected in a mussel sample from Osaka Bay (at 1.4 µg/kg wet weight). Japanese market fish were analyzed for PBDEs. The highest combined PBDE concentrations (BDE 28, BDE 47, BDE 66, BDE 99, BDE 100, BDE 153, and BDE 154) were in salmon, cultured yellowtail, and wild yellowtail muscle (46, 44, and 30.5 ng/g lipid weight, respectively) and lowest concentrations in yellowfin tuna (1.9 ng/g lipid weight) (Ohta et al. 2000). BDE 47 was major congener in all samples. In another study, several fish species from Japan were analyzed for 15 BDE congeners (Hori et al. 2000). The PBDE concentrations ranged from 0.00136 to 2.1 ng/g fresh weight, with BDE 47 as the predominant congener. Seven species of marine fish (conger eel, flounder, gray mullet, horse mackerel, red sea bream, sea bass, and yellowtail) were collected from the Inland Seas near Seto, Japan (Akutsu et al. 2001). Seven PBDEs (BDE 28, BDE 47, 2,3',4,4'-tetraBDE [BDE 66], BDE 99, BDE 100, BDE 153, and BDE 154) were detected in all samples, with BDE 47 being the most abundant congener. Concentrations of total PBDEs in gray mullets and yellowtails were 63 and 15 ng/g lipid weight, respectively.

**Marine Aquatic Organisms.** Marine mussels (*Mytilus edulis*) collected at several locations in the Netherlands and analyzed for BDE 47, BDE 99, BDE 153, and decaBDE (BDE 209) (de Boer et al. 2000b). Concentrations of BDE 47 and BDE 99 were 0.9–4.3 and 0.3–1.6 ng/g dry weight, respectively. BDE 153 and BDE 209 were not detected. Di- to heptaBDE were analyzed for in hepatopancreas samples from Dungenes crab from several sites on the Strait of Georgia, British Columbia, Canada (Ikonomou et al. 1999). The primary congener detected was BDE 47. The combined concentration of BDE 47 and BDE 99 was approximately 100–350 ng/g lipid weight.

**Marine Animals.** In marine animals, temporal trends show increasing concentrations of lower-brominated BDE congeners with higher concentrations found near human-populated areas. In all marine animal studies, the congener profiles show the highest concentrations for BDE 47. The concentrations of several PBDEs in marine animals are summarized in Table 6-9. Frouin et al. (2011) reported PBDE concentrations measured from serum and blubber samples obtained from six harbor seal pups (*Phoca vitulina*) live captured in May 2007, six harbor seal pups (*Phoca vitulina*) live captured in May 2008, six grey seal pups (*Halichoerus grypus*) live captured early January 2008, and six harp seal pups (*Phoca groenlandica*) live captured in March 2008 from the Gulf of St. Lawrence or the St. Lawrence Estuary. The ΣPBDEs in serum (BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 155) and blubber (BDE 28, BDE 47, BDE 49, BDE 66, BDE 99, BDE 100, BDE 153, BDE 154, BDE 155, and BDE 183) were strongly correlated. BDE 47 was detected in all serum samples and accounted for 66–73% of ΣPBDEs. ΣPBDE concentrations in lipid ranged from 21 to 530 ng/g lipid weight and from 34 to 600 ng/g lipid weight in serum. PBDEs have been detected in several species of seal from several



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**Table 6-9. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Animals**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Bottlenose dolphin	Gulf of Mexico	No data	No data	No data	8,000	No data	Kuehl and Haebler 1995
Harbor seal	Northwest Atlantic	904	134	49	1,385*	1.2	Shaw et al. 2009
Harbor seal	San Francisco Bay, California	No data	No data	No data	530–5,075*	No data	Klosterhaus et al. 2012
Cormorant eggs	San Francisco Bay, California	No data	No data	No data	3,425–5,550*	No data	Klosterhaus et al. 2012
Harbor seal	San Francisco Bay, California	46–6,682	17–303	No data	No data	No data	She et al. 2000
Cormorant eggs	Suisan Bay, CA (2002)	No data	No data	No data	19,000±4,000*	No data	Sutton et al. 2015
Cormorant eggs	Suisan Bay, California (2006)	No data	No data	No data	6,100±5,200*	No data	Sutton et al. 2015
Cormorant eggs	Suisan Bay, California (2009)	No data	No data	No data	440±170 <sup>8</sup>	No data	Sutton et al. 2015
Cormorant eggs	Suisan Bay California (2012)	No data	No data	No data	1,300±900*	No data	Sutton et al. 2015
Cormorant eggs	Central Bay, California (2002)	No data	No data	No data	9,100±2200	No data	Sutton et al. 2015
Cormorant eggs	Central Bay, California (2004)	No data	No data	No data	3,700±500	No data	Sutton et al. 2015
Cormorant eggs	Central Bay, California (2006)	No data	No data	No data	1,800±400	No data	Sutton et al. 2015
Cormorant eggs	Central Bay, California (2009)	No data	No data	No data	1,800±300	No data	Sutton et al. 2015
Cormorant eggs	Central Bay, California (2012)	No data	No data	No data	1,100±200	No data	Sutton et al. 2015
Cormorant eggs	South Bay California (2002)	No data	No data	No data	4,200±400	No data	Sutton et al. 2015
Cormorant eggs	South Bay California (2004)	No data	No data	No data	3,300±300	No data	Sutton et al. 2015

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**Table 6-9. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Animals**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Cormorant eggs	South Bay California (2006)	No data	No data	No data	4,400±2,100	No data	Sutton et al. 2015
Cormorant eggs	South Bay California (2009)	No data	No data	No data	2,100±1,200	No data	Sutton et al. 2015
Cormorant eggs	South Bay California (2012)	No data	No data	No data	1,100±100	No data	Sutton et al. 2015
Harbor seal (blubber)	San Francisco Bay, California	1,304	112	87.1	1,730*	No data	She et al. 2002
Herring Gull Eggs	Lake Superior, United States	253–323 (fw)	202–284 (fw)	83.6–113 (fw)	664–887 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Lake Michigan, United States	522–602 (fw)	323–459 (fw)	167–203 (fw)	1,366–1,400 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Lake Huron, United States and Canada	146–291 (fw)	74.6–161 (fw)	37.3–89.5 (fw)	308–652 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Detroit River, United States	322 (fw)	130 (fw)	92.6 (fw)	639 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Lake Erie, United States	70–163 (fw)	52–55.9 (fw)	24.6–51.8 (fw)	192–340 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Niagara River, United States	168 (fw)	111 (fw)	53 (fw)	432 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	Lake Ontario, Canada	220–401 (fw)	113–322 (fw)	66.5–102 (fw)	530–1,003 (fw)*	No data	Norstrom et al. 2002
Herring Gull Eggs	St. Lawrence River, United States	220 (fw)	89.8 (fw)	56.6 (fw)	453 (fw)*	No data	Norstrom et al. 2002
Harbor seal	St. Lawrence Estuary and Gulf of St. Lawrence	52–408	No data	No data	72–530*	No data	Fouin et al. 2011
Grey seal	St. Lawrence Estuary and Gulf of St. Lawrence	41	No data	No data	69*	No data	Fouin et al. 2011
Harp seal	St. Lawrence Estuary and Gulf of St. Lawrence	14	No data	No data	21*	No data	Fouin et al. 2011
Beluga whale	Canadian Arctic	No data	No data	No data	81–160*	No data	Alaee et al. 1999
Beluga whale	Southeast Baffin, Canada	10	0.9	1.6	15*	No data	Stern and Ikonomou 2000

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**Table 6-9. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Animals**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Bottlenose dolphin	South Atlantic Ocean	No data	No data	No data	180–220	No data	Kuehl et al. 1991
Brunnich's guillemot	Svalbard, Sweden	No data	No data	No data	130	No data	Jansson and Asplund 1987
Cormorant	England, United Kingdom	170–3,500	50–250	50–1,500	300–6,400*	No data	Allchin et al. 2000
Cormorant liver	Rhine delta, Germany	No data	No data	No data	28,000 (fw)	No data	de Boer 1990
Galaucous gull	Bear Island, Norway (Arctic)	290–634	160	No data	No data	No data	de Wit 2002
Grey seal	Baltic Sea	650	40	38	730	No data	de Wit 2002; Sellström et al. 1993
Grey seal	Baltic Sea	308	54	57	419	No data	Haglund et al. 1997
Grey seal	Baltic Sea	No data	No data	No data	208	No data	Andersson and Wartanian 1992
Harbor porpoise	British Columbia, Canada	50–1,200	No data	No data	350–2,300*	No data	Ikonomou et al. 2000
Harbor porpoise	England and Wales, United Kingdom	227–6,790	No data	No data	440–7,670	No data	Law et al. 2000
Harbor seal	Baltic Sea	No data	No data	No data	90	No data	Jansson and Asplund 1987
Harbor seal	Skagerrak, Norway and Sweden	No data	No data	No data	230	No data	Andersson and Wartanian 1992
Harbor seal	North Sea	390–4,900	42–660	25–450	600–6,000	No data	de Boer et al. 1998b
Long-finned pilot whale	Faeroe Islands	410–1,780	160–600	87–280	843–3,160*	No data	Lindström et al. 1999
Long-finned pilot whale	Faeroe Islands	66–860	24–170	12–98	126–1,250*	No data	van Bavel et al. 1999
Minke whale	Netherlands	630	160	79	870	No data	de Boer et al. 1998b
Ringed seal	Baltic sea	256	33	61	350	No data	Haglund et al. 1997
Ringed seal	Baltic sea	No data	No data	No data	320	No data	Andersson and Wartanian 1992
Ringed seal	Svalbard, Sweden	47	1.7	2.3	51	No data	de Wit 2002; Sellström et al. 1993

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**Table 6-9. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Marine Animals**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Ringed seal	Canadian Arctic	No data	No data	No data	25.8–50*	No data	Alaee et al. 1999
Ringed seal	Holman Island, Northwest Territories, Canada	2.8	No data	No data	2.4–4.9*	No data	Ikonomou et al. 2000
Sperm whale	Netherlands	130–250	32–64	21–35	187–349	No data	de Boer et al. 1998b
White-beaked dolphin	Netherlands	5,500	1,000	1,200	7,700	No data	de Boer et al. 1998b

<sup>a</sup> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

BDE = brominated diphenyl ether; fw = fresh weight

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different sites. In San Francisco Bay, California, the concentrations of PBDEs in harbor seals have increased dramatically based on samples obtained from 1989 to 1998. The samples from 1998 had PBDE concentrations among the highest reported for this species (She et al. 2002). The concentration of total PBDEs (sum of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154) in harbor seal blubber increased by over a factor of 50 from a concentration of 88 ng/g lipid for species samples collected in 1988 to a concentration of 2,985–8,325 ng/g lipid for species samples collected in 1998. The highest concentrations reported were for BDE 47, which increased from 45.6 ng/g lipid for blubber samples in 1989 to 2,343–6,682 ng/g lipid for blubber samples collected in 1998. The dominance of the tetraBDE congeners over other congeners may indicate that tetraBDEs bioaccumulate more than the higher-brominated congeners (She et al. 2002).

In the Baltic Sea, female grey seals (*Halichoerus grypus*) sampled in 1979–1985 contained 730 ng PBDE/g lipid in their blubber (sum of BDE 47, BDE 99, and BDE 100) (Jansson et al. 1993); male grey seals had 280 ng PBDE/g lipid weight (Andersson and Wartanian 1992). Male ringed seals (*Pusa hispida*) from the Baltic Sea had 320 ng PBDE/g lipid weight (Andersson and Wartanian 1992). Baltic gray and ringed seal blubber sampled between 1981 and 1988 contained 419 and 350 ng PBDEs/g lipid (total of BDE 47, BDE 99, and BDE 100), respectively (Haglund et al. 1997). In 1981, female ringed seals from Svalbard in the Swedish Arctic contained 40–51 ng PBDEs/g lipid in blubber (Jansson and Asplund 1987, Jansson et al. 1993; Sellström et al. 1993). Higher concentrations of PBDEs are generally evident in Baltic Sea ringed seals (320–350 ng/g lipid) (Andersson and Wartanian 1992; Haglund et al. 1997) compared to Arctic ringed seals (26–51 ng/g lipid) (Alaee et al. 1999; Jansson et al. 1987). The concentration of PBDEs in harbor seals from Skagerrak on the Swedish west coast was 230 ng PBDE/g lipid (Andersson and Wartanian 1992).

She et al. (2000) analyzed the concentration of BDE 47, BDE 99, and BDE 153 in harbor seals from the San Francisco Bay area. Mean concentrations for BDE 47, BDE 99, and BDE 153 were 1,124, 107, and 50 ng/g lipid weight, respectively. Alaee et al. (1999) found that ringed seals from the Canadian Arctic had mean PBDE concentrations (sum of di- to hexaBDEs) of 25.8 ng/g lipid weight (females) and 50.0 ng/g lipid weight (males). The lower concentrations in female seals suggest that PBDEs are transferred to young through breast milk. On Holman Island, Northwest Territory, Canada (Arctic) in 1996, ringed seals had total PBDE concentrations of 2.4–4.9 ng/g lipid for males. The concentrations of PBDEs were found to increase with age (Ikononou et al. 2000). In a temporal trend study, archived samples of blubber from ringed seals from Holman Island, Northwest Territory, Canada were analyzed for PBDE concentrations. The concentration of PBDE in samples collected between 1981 and 1996

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increased from approximately 0.3 ng/g lipid weight in 1981 to 3.6 ng/g lipid weight in 1996 (Ikonomou et al. 2000).

The concentrations of PBDEs have been determined in harbor porpoises (*Phocaena phocaena*) from British Columbia, Canada (Ikonomou et al. 2000) and from the coasts of England and Wales (Law et al. 2000). In British Columbia (Canada) samples, the total PBDE concentrations (sum of tri- to hepta-congeners) were 350–2,300 ng/g lipid weight; BDE 47 was found at the highest concentrations in these samples (range, 50–1,200 ng/g lipid weight) (Ikononmou et al. 2000). Concentrations of total PBDEs (sum of 13 congeners) along the coast of England and Wales, ranged from 450 to 7,670 ng/g lipid weight, with BDE 47 concentrations ranging from 227 to 6,790 ng/g lipid weight (Law et al. 2000).

During a mass mortality event on the south Atlantic coast in 1987–1988, blubber samples were collected from three bottlenose dolphins (*Tursiops truncatus*); these samples contained 180–220 ng PBDEs/g lipid (Kuehl et al. 1991). Blubber samples, taken from stranded bottlenose dolphins from several locations around the Gulf of Mexico in 1990, contained 3,110 ng PBDEs/g lipid (Kuehl and Haebler 1995). On the Dutch coast in early 1998, de Boer et al. (1998b) found PBDEs in blubber of one whitebeaked dolphin (*Lagenorhynchus albirostris*); the concentrations of BDE 47, BDE 99, and BDE 100 were 5,500, 1,000, and 1,200 ng/g lipid weight, respectively.

The concentration of 19 PBDEs was determined in long-finned pilot whale (*Globicephala melas*) from the Faeroe Islands in the north Atlantic (Lindström et al. 1999). Young males and females had the highest concentrations, ranging from 3,000 to 3,160 ng/g lipid; lower concentrations were observed for both adult females (840–1,050 ng/g lipid) and males (1,610 ng/g lipid). The predominant isomers in all samples were BDE 47 and BDE 99, accounting for 70% of the sum of 19 congeners. van Bavel et al. (1999) also studied the concentrations of PBDEs in long-finned pilot whales. They observed a similar trend with young animals having higher PBDE concentrations (740 ng/g lipid weight) and adult animals having lower concentrations (females, 230 ng/g lipid; males, 540 ng/g lipid). In Beluga whales sampled in 1997 from southeast Baffin (Cumberland Sound), the concentrations of total PBDEs and BDE 47 were 15 and 10 ng/g lipid weight, respectively (Stern and Ikonomou 2000). Between 1982 and 1997, total PBDE concentrations in archived blubber samples of beluga whales from southeast Baffin Canada increased significantly. For this time period, BDE 47, BDE 99, BDE 100, and BDE 154, and total PBDEs increased by factors of 6.5, 10.3, 7.9, 30.6, and 6.8, respectively (Stern and Ikonomou 2000). Three sperm whales (*Physeter macrocephalus*) and one minke whale (*Balaenoptera acutorostrata*) found stranded on the Dutch coast in early 1998 were analyzed for PBDEs (de Boer et al. 1998a, 1998b). Exposure to PBDEs for these animals occurred in the deep Atlantic through the food web. The

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concentrations of PBDEs in these marine mammals were as follows: sperm whale (BDE 47, 130–250 ng/g lipid weight; BDE 99, 32–64 ng/g lipid weight; and BDE 100, 21–35 ng/g lipid weight) and minke whale (BDE 47, 630 ng/g lipid weight; BDE 99, 160 ng/g lipid weight; BDE 100, 79 ng/g lipid weight); BDE 209 (decaBDE) was below detection limits in all samples.

PBDEs, methoxylated (MeO-) PBDEs, and hydroxylated (OH-) PBDEs were evaluated in whole blood samples collected from northern fur seal (*Callorhinus ursinus*), spotted seal (*Phoca largha*), Steller sea lion (*Eumetopias jubatus*) and ribbon seal (*Phoca fasciata*) (Nomiya et al. 2014), and harbor porpoise and Dall's porpoise (Ochiai et al. 2013) from northern Japanese coastal waters. The samples contained 3.9–280 pg/g median values of 6OH-BDE 47; <1.0–51 pg/g median values of 2' MeO-BDE 68; 2.9–1,020 pg/g median values of 6MeO-BDE 47; <1–18 pg/g median values of 6MeO-BDE 99; and <100–230 pg/g median values of total PBDEs.

**Marine Birds.** Increasing concentrations of PBDEs have been found in marine birds and eggs, with BDE 47 found at the highest concentrations. Di- and triBDE have been detected, but not quantified, in black skimmer (*Rynchops nigra*) tissues and eggs in the United States (Stafford 1983). In 2000, herring gull eggs collected from 15 locations around the Great Lakes (United States and Canada) were pooled and analyzed for PBDEs (Norstrom et al. 2002). A total of 25 di- to hepta-BDE congeners were identified in herring gull through the Great Lakes system. No mono-, octa-, nona-, or decaBDEs were found at the detection limit of the analysis (0.01–0.05 ng/g wet weight). Seven congeners, BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 184, constituted 97.5% of total PBDEs (192–1,400 ng/g wet weight). BDE 47 was the dominant congener (70–602 ng/g wet weight) followed by BDE 99 (52–459 ng/g wet weight). The highest concentrations (1,003–1,400 ng/g wet weight) were found in two Lake Michigan colonies and in Toronto Harbor, Lake Ontario (Norstrom et al. 2002). Muscle tissues from ospreys (*Pandion haliaetus*), found dead at various locations around Sweden, were pooled and analyzed for PBDEs (Jansson et al. 1993; Sellström et al. 1993). The ospreys' diet was freshwater fish. The combined concentration of BDE 47, BDE 99, and BDE 100 was 2,100 ng/g lipid in samples collected between 1982 and 1986; BDE 47 was the primary congener (86%) in these samples (n=35). High concentrations of PBDEs may reflect biomagnification and/or fish consumption along their migratory routes. The concentrations of PBDEs in common guillemots (*Uria aalge*) collected in 1979–1981 from the Baltic and North Seas were 370 and 80 ng/g lipid, respectively (Jansson and Asplund 1987). As part of the Swedish National Environmental Monitoring Program, guillemot eggs (St. Karlsö, Baltic Sea) are collected yearly and placed in the Swedish Natural History Museum's Environmental Specimens Bank. The concentrations of BDE 47, BDE 99, and BDE 100 in pooled egg samples from the specimen bank

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showed a significant increase from 1969 to the beginning of the 1990s, with highs of 1,100 ng/g for BDE 47 in 1984 and 190 ng/g for BDE 99 in 1990 (Sellström et al. 1993, 1999). Between 1992 and 1997, PBDE concentrations started to decrease statistically. In 1997, the PBDE concentration (sum of BDE 47, BDE 99, and BDE 100) was 190 ng/g lipid, with BDE 47 as the predominant congener. Cormorant eggs obtained from the San Francisco Bay regions had total PBDE concentrations ranging from 3,425 to 5,550 ng/g (median, 5,500 ng/g) and was dominated by the penta (BDE 47, BDE 99, BDE 100) congeners (Klosterhaus et al. 2012). Sutton et al. (20015) noted decreasing levels of PBDEs in cormorant eggs obtained from three locations in northern California. Eggs collected from Suisun, Central, and South Bays in 2012 had total PBDE levels 93, 88, and 74% lower, respectively, when compared to the levels in eggs collected in 2002 (see Table 6-9).

***Human Body Tissues and Fluids.*** The quantitative determination of the concentrations of PBDEs in body tissues and fluids is important in determining the human body burden of these chemicals. Increasing concentrations of lower-brominated PBDEs have been measured in blood and breast milk in temporal trend studies. Individuals who consumed fish had a somewhat higher concentration of total PBDEs in body fluids compared to individuals who ate less fish.

Lipid adjusted serum levels of 11 BDE congeners collected from the U.S. general population were reported in the Fourth National Report on Human Exposure to Environmental Chemicals, Updated Tables (CDC 2015). Serum levels for BDE 17, BDE 28, BDE 47, BDE 66, BDE 85, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183 were evaluated in samples collected between 2003 and 2008; BDE 209 was evaluated in samples collected from 2005 to 2008. In the NHANES 2003–2004 survey years, congener BDE 47 was detected at a concentration of 20.5 ng/g lipid (geometric mean), the highest concentration for all samples. BDE 28, BDE 99, BDE 47, BDE 100, and BDE 153 were in >60% of participants (Sjödin et al. 2008). BDE 17 was not detected above the limit of detection of 1.0 and 0.6 ng/g lipid in survey years 2003–2004 and 2005–2008, respectively. BDE 209 was not detected above the limit of detection, 6.0 and 5.8 ng/g lipid, in survey years 2005–2006 and 2007–2008, respectively.

BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 were detected in 98, 100, 100, 96, and 48%, respectively, of serum samples collected from 48 mothers participating in the California Childhood Leukemia Study from 2006 to 2007 (Whitehead et al. 2015a). Median serum levels ranged from below the detection limit for BDE 154 to 35 ng/g lipid for BDE 47. BDE 47 and BDE 153 were detected in whole blood of 61 and 85%, respectively, of 191 children participating in the California Childhood Leukemia Study from 1999 to 2007 (Whitehead et al. 2015b). The median, 75<sup>th</sup> percentile, 90<sup>th</sup>



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percentile, and maximum levels were 410, 820, 1,500, and 17,000 pg/mL, respectively, for BDE 47 and 130, 270, 460, and 6,500 pg/mL, respectively, for BDE 153.

Tables 6-10, 6-11, and 6-12 summarize representative concentrations of PBDEs found in blood (serum), adipose tissue, breast milk, and other body tissues or fluids, respectively. These studies indicate that concentrations of lower-brominated BDEs in body fluids are a factor of 10–100-fold higher for individuals living in the United States compared to individuals living in other regions of the world (e.g., Europe). Serum samples collected from 12 U.S. blood donors in 1988 were analyzed for PBDEs, and BDE 47, BDE 153, BDE 183, and BDE 209 were detected (Patterson et al. 2000; Sjödin et al. 2001b). Concentrations of these congeners were similar to those found in the Sjödin et al. (1999b) study for the control group. The median concentrations and ranges of BDE 47, BDE 153, BDE 183, BDE 209, and total PBDEs (sum of four congeners) were 0.63 (<0.4–24); 0.35 (0.08–2.0); 0.17 (0.09–1.3); <1 (<1–34); and 2.2 ng/g lipid weight, respectively (Sjödin et al. 2001b). DecaBDE was found at concentrations above the limit of quantification (1 pmol/g lipid) in 5 of 12 serum samples (Patterson et al. 2000).

Schechter et al. (2005) provided a summary of PBDE (BDE 17, BDE 28, BDE 47, BDE 66, BDE 77, BDE 85, BDE 99, BDE 100, BDE 138, BDE 153, BDE 154, BDE 183, and BDE 209) concentrations in blood from 29 adults residing in Mississippi and 10 adults in New York City. These blood samples were obtained in 2003. These data were then compared to archived blood samples from 100 individuals obtained in 1973 from the Dallas, Texas area and stored at the University of Texas Southwestern Medical Center. The 13 PBDE congeners were not detected in the blood samples from the 100 individual collected in 1973, but all congeners were detected in at least one of the blood samples obtained from the 29 residents of Mississippi collected in 2003. BDE 28, BDE 47, BDE 77, BDE 99, BDE 100, BDE 138, BDE 153, BDE 154, and BDE 183 were all detected in at least one of the blood samples obtained in 2003 from New York City residents. BDE 47 followed by BDE 99 were the predominant congeners detected in both the Mississippi and New York residents blood samples collected in 2003.

Rawn et al. (2014) analyzed data from nearly 5,000 serum samples collected from the Canadian Health Measures Survey (CHMS) from 2007 to 2009. PBDE congeners were detected in all samples tested, with a range of values of 27–130 ng/g lipid (total PBDEs) and a GM of 46 ng/g lipid. BDE 47 was the predominant congener with a GM of 22 ng/g lipid followed by BDE 153 (GM=9.4 ng/g lipid), BDE 99 (GM=4.6 ng/g lipid), BDE 100 (GM=4.1 ng/g lipid), and BDE 209 (GM 1.1 ng/g lipid) (Rawn et al. 2014).

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**Table 6-10. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Human Blood Samples<sup>a</sup>**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Human blood	United States (in 1988)	0.63	0.32	0.17	2.2*	<0.1	Sjodin et al. 2001b
Human blood	Mississippi and New York City (in 2003)	25 (mean)	11.1 (mean)	4.7 (mean)	52.6 (mean)	2.7 (mean)	Schechter et al. 2010
Maternal serum	Indiana	28 (9.2–310)	5.7 (2.4–68)	4.2 (1.9–110)	37 (15–580)	No data	Mazdai et al. 2003
Fetal serum	Indiana	25 (8.4–210)	7.1 (2.2–54)	4.1 (1.8–91)	39 (14–460)	No data	Mazdai et al. 2003
Maternal serum	Texas	14.9	3.0	2.8	27.8*		Schechter et al. 2010
Human serum	United States (in 2003–2004)	20.5 (geometric mean)	No data	3.93 (geometric mean)	No data	No data	CDC 2015
Human serum	United States (in 2005–2006)	21.2–74.9 (weighted arithmetic mean)	4.16–23.3 (weighted arithmetic mean)	4.06–14.2 (weighted arithmetic mean)	No data	No data	CDC 2015
Human serum	United States (in 2007–2008)	20.5–53.1 (weighted arithmetic mean)	4.30–10.7 (weighted arithmetic mean)	4.12–11.0 (weighted arithmetic mean)	No data	No data	CDC 2015
Human serum	Australia	2.6–55.1	0.9–24.2	0.6–14.1	24 (newborn); 31 (0–2 years); 41 (2–6 years); 26 (7–12 years); 20 (13–30 years); 9.4 (>31 years) (mean)	No data	Toms et al. 2009
Maternal serum	France	2.831	1.939	0.365	No data	5.783	Antignac et al. 2009
Cord serum	France	No data	7.434	1.467	No data	27.110	Antignac et al. 2009
Maternal serum	Spain	2.3	0.35	No data	9.6	<0.7	Vizcaino et al. 2011
Cord serum	Spain	2.3	1.5	No data	9.6	<1.2	Vizcaino et al. 2011

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**Table 6-10. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Human Blood Samples<sup>a</sup>**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Human Serum	Canada	23 (mean)	5.4 (mean)	4.4 (mean)	48 (mean)	1.9 (mean)	Rawn et al. 2014
Maternal serum	China	0.75 (mean)	No data	No data	29.26 (mean)	2.12 (mean)	Li et al. 2013a
Cord serum	China	0.62 (mean)	No data	No data	41.12 (mean)	1.33 (mean)	Li et al. 2013a
Human blood	Sweden	No data	No data	No data	2.1*	No data	Klasson Wehler et al. 1997
Human blood	Japan	0.001	<LOQ	No data	No data	No data	Fujii et al. 2014
Human blood	Sweden, computer dis-assembly workers	2.9 (median)	No data	No data	26*	4.8	Sjödin et al. 1999a
Human blood	Sweden, cleaning personnel/office workers	1.5–1.6	No data	No data	3.3–4.1*	<0.7	Sjödin et al. 1999a
Human blood	Sweden, high fish intake	2.1	No data	No data	No data	No data	Bergman et al. 1999; Sjödin et al. 2000
Human blood	Sweden, no fish intake	0.40	No data	No data	No data	No data	Bergman et al. 1999; Sjödin et al. 2000
Maternal blood	Sweden	0.83 (0.3–5.1)	0.19 (<0.01–1.43)	0.17 (<0.01–0.52)	2.07 (0.71–8.39)	No data	Meironyte Guvenius et al. 2003
Cord blood	Sweden	0.98 (0.33–3.28)	0.07	0.07	0.46–4.28	No data	Meironyte Guvenius et al. 2003
Human blood	Germany	3.9	No data	No data	5.6*	No data	Schröter-Kermani et al. 2000

<sup>a</sup>ΣPBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*). All values are median values unless stated otherwise.

BDE = brominated diphenyl ether; LOQ = limit of quantitation

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**Table 6-11. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Human Adipose Tissue Samples**

Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Northern California	7.0–28 (18, mean)	3.1–7.3 (4.9 mean)	No data	No data	No data	She et al. 2000
San Francisco, California	16.5 (5.2–196)	No data	No data	No data	No data	Petreas et al. 2003
Sweden	8.8	1.1	1.8	11.7	No data	Haglund et al. 1997
Sweden	2.2 (mean)	1.6 (mean)	0.1 (mean)	5* (mean)	No data	Meironyté Guvenius and Norén 1999
Finland	7.3 (mean)	2.3 (mean)	No data	6.2–22*	No data	Strandman et al. 1999
Finland	0.55	0.74	No data	No data	No data	Smeds and Saukko 2003
Spain	1.36 (mean)	0.42 (mean)		No data	No data	Meneses et al. 1999
Japan	0.459	0.118	0.250	1.288*	No data	Choi et al. 2003
France	0.651	0.166	0.168		0.752	Antignac et al. 2009

<sup>a</sup> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*). All values are median values unless otherwise stated.

BDE = brominated diphenyl ether; ND = not detected

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**Table 6-12. Concentrations (ng/g Lipid Weight) of Several Polybrominated Diphenyl Ethers (PBDEs) in Human Breast Milk Samples**

Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Texas	18.4	5.7	2.9	34.0	8.24 (maximum)	Schechter et al. 2003
Texas	24.0	4.3	3.5	39.7*	No data	Schechter et al. 2010
California	29.7	6.40	5.65	54.5	1.41	Park et al. 2011
Pennsylvania	26	No data	4	No data	No data	LaKind et al. 2009
Philippines (mean)	0.9	0.22	0.19	1.8*	<0.05	Malarvannan et al. 2013
France	1.152	0.527	0.226		1.615	Antignac et al. 2009
Uppsala County, Sweden	1.78	0.43	0.27	3.15	No data	Lind et al. 2003
Sweden	1.8	0.442	0.340	3.373*	No data	Darnerud et al. 1998
Finland	0.85	0.35			No data	Strandman et al. 2000
Birmingham, United Kingdom	2.80	0.69	0.45	5.00*	0.25	Abdallah and Harrad 2014
Japan	0.18–0.57	0.09–0.13	0.07–0.18	0.65–1.48*		Ohta et al. 2000
Japan	No data	No data	No data	0.66–2.8*	No data	Ohta et al. 2002
Japan	0.57	0.33	No data	No data	No data	Fujii et al. 2014
Sweden	No data	No data	No data	0.07* (1972); 0.28 (1976); 0.48 (1980); 0.72 (1984-5); 1.21 (1990) 2.15 (1994); 3.11 (1996); 4.01 (1997)	No data	Norén and Meironyté 2000

<sup>a</sup>ΣPBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*). Concentrations are median concentrations unless stated otherwise

BDE = brominated diphenyl ether

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Serum samples were collected from a group of 50 Laotian immigrants (aged 19–40) participating in a reproductive outcome study in the San Francisco Bay area (Petreas et al. 2002). Participants were recruited and sampled in the late 1990s. The mean concentration of BDE 47 in serum was approximately 95 ng/g lipid. The contemporary samples were compared to serum samples taken from a group of over 400 women from the San Francisco Bay in the 1960s. Concentrations of BDE 47 in all archived samples were below the limit of detection. Petreas et al. (2003) expanded their investigation to include a diverse group of local women from the San Francisco Bay area sampled in the late 1990s. Their results confirmed earlier findings reported in Petreas et al. (2002). Mean concentrations of BDE 47 in serum samples taken from California women ranged from 5 to 510 ng/g lipid, with a median (16.5 ng/g lipid) 3–10 times higher than those reported from Europe (Petreas et al. 2003). In 2001, Mazdai et al. (2003) determined the concentration of six PBDE congeners (BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183) and total PBDEs in maternal and fetal blood samples taken from subjects in Indianapolis, Indiana. Median concentrations of total PBDE (sum of six congeners) were 39 and 37 ng/g lipid for fetal and maternal serum, respectively. BDE 47 was the predominant congener reported at median concentrations of 25 and 28 ng/g lipid for fetal and maternal serum samples, respectively. When compared with serum PBDE concentrations for a similar population of Swedish mothers and newborns, the concentrations for the Indiana population were 20–69-fold higher for maternal blood and 30–106-fold higher for fetal blood. In fact, the median blood concentrations for this study were comparable to Swedish workers considered to have direct work-related exposures. These observations indicated that women in some areas of North America are exposed to much higher concentrations of lower-brominated BDEs (i.e., BDE 47) than European women. In general, the PBDE congener profile found in human serum was similar to that detected in environmental samples, except that there was an apparent decrease in the proportion of BDE 99. BDE 183 was detected in <17% of the samples even though it is the primary congener in octaBDE commercial mixtures (Mazdai et al. 2003). The conclusion that PBDE concentrations are higher in North America than in Europe is further supported by a study conducted in the Netherlands that analyzed maternal serum from 90 female volunteers collected at the 35<sup>th</sup> week of pregnancy, and in cord serum of a number of their infants (Meijer et al. 2008). Median concentrations of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 in maternal serum were reported as 0.8, 0.2, 0.2, 1.6, and 0.5 ng/g lipid weight, respectively. Median concentrations of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 in cord serum were 0.5, 0.1, 0.1, 0.9, and 0.3 ng/g lipid weight, respectively (Meijer et al. 2008). These concentrations are on the same order of magnitude as reported in other areas of Europe and much lower than concentrations typically detected in the United States (Vizcaino et al. 2011).

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Six PBDE congeners (BDE 28, BDE 47, BDE 66, BDE 99, BDE 100, and BDE 153) were quantified in 40 human blood-plasma samples from Sweden. The highest concentrations in plasma were for BDE 47 and BDE 99; these congeners made up 70% of the total PBDE concentration. The mean concentration of total PBDE was  $2.1 \pm 1.4$  ng/g lipid weight (Klasson Wehler et al. 1997). Whole-blood samples from a German environmental specimen bank, collected in 1985, 1990, 1995, and 1999, contained measurable quantities of BDE 28, BDE 47, BDE 66, BDE 85, BDE 99, BDE 100, BDE 153, and BDE 154. An increasing temporal trend was also observed; the mean total PBDE concentration (sum of eight congeners) increased from 3.9 ng/g lipid weight in 1985 to 5.6 ng/g lipid weight in 1999. For the 1999 sample, BDE 47 was the major congener found, with a mean concentration of 3.9 ng/g lipid weight. The total PBDE concentrations were significantly lower in female blood samples (Schröter-Kermani et al. 2000). In a study of the influence of diet on concentrations of PBDEs, BDE 47 was measured in blood serum from persons with high fish intake and no fish intake (Bergman et al. 1999; Sjödin et al. 2000). High-fish-intake groups of Swedish and Latvian men had median BDE 47 concentrations of 2.2 and 2.4 ng/g lipid weight, respectively, whereas the no-fish-intake groups had median concentrations of 0.4 and 0.26 ng/g lipid weight, respectively (Sjödin et al. 2000).

Serum samples collected in 2006–2007 and analyzed for different age groups in Australia suggest that PBDE concentrations increase from infant to toddler and then gradually decrease over time (Toms et al. 2009). Mean total PBDE (sum of BDE 47, BDE 99, BDE 100, and BDE 153) concentrations in cord blood of 0–2 year olds, 2–6 year olds, 7–12 year olds, 13–30 year olds, and >31 year olds were 24, 31, 41, 26, 20, and 9.4 ng/g lipid, respectively. The peak mean concentration was observed in toddlers 2.6–3 years of age (51 ng/g lipid), which is later than when breastfeeding usually ceases, suggesting a lower capacity to eliminate PBDEs or greater exposure through unique exposure pathways more common for this age group (e.g., ingestion or dermal exposure of contaminated dust particles in carpeting).

BDE 47, BDE 153, BDE 154, BDE 183, and BDE 209 were measurable in blood plasma from three groups of workers (i.e., workers at a computer-disassembly plant, workers in a computerized office, and a control group) (Sjödin et al. 1999a). The median concentrations (sum of five congeners) were highest for the computer-disassembly plant workers (26 ng/g lipid weight); the office workers had a median concentration of 4.1 ng/g lipid weight and the control group had a median concentration of 3.3 ng/g lipid weight. The congener patterns for the control group and office workers were similar, with BDE 47 having the highest concentrations. For the computer disassembly plant workers, the median concentrations of BDE 183, BDE 153, BDE 154, BDE 47, and BDE 209 were 7.8, 4.5, 1.2, 2.9, and

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4.8 ng/g lipid weight, respectively. Blood serum samples from 19 full-time computer technicians were analyzed (Hagmar et al. 2000a). The serum concentrations of BDE 153, BDE 183, and BDE 209 in these samples were found to be approximately 5 times higher than the control and office workers in the Sjödin et al. (1999a) study. The median concentration for total PBDEs (for the sum of five congeners) was 10.6 pmol/g (7.0 ng/g) lipid weight. The highest concentrations were of BDE 153. Two octaBDE congeners and one nonaBDE congener were also detected. Connections were observed between fish consumption and serum concentrations for congeners BDE 47, BDE 153, and BDE 183, and between worktime at the computer and congeners BDE 153 and BDE 183.

DecaBDE, as well as hexa- through nonaBDE, has been found in composite samples from the 1987 National Human Adipose Tissue Survey repository (Cramer et al. 1990; Stanley et al. 1991). The concentrations ranged from not detected to 1 ng/g fat for hexaBDE, 0.001–2 ng/g fat for heptaBDE and not detected to 8 ng/g fat for octaBDE. NonaBDE concentrations were estimated to be >1 ng/g fat; decaBDE was estimated to range between not detected and 0.7 ng/g fat. In the late 1990s, breast adipose samples collected in northern California contained quantifiable amounts of BDE 47, BDE 99, and BDE 153 (She et al. 2000). Mean concentrations were 18 ng/g lipid weight for BDE 47, 4.9 ng/g lipid weight for BDE 99, and 2.2 ng/g lipid weight for BDE 153. Average total PBDEs concentrations (86 ng/g lipid) were the highest human concentrations reported to date. Petreas et al. (2003) expanded their investigation to include a diverse group of local women from the San Francisco Bay area sampled in the late 1990s. Their results confirmed earlier findings reported in She et al. (2000). Mean concentrations of BDE 47 in adipose tissues samples taken from California women were 28.9 ng/g lipid. In the adipose tissue of a 74-year-old Swedish male, the BDE 47 concentration was 8.8 ng/g lipid weight (Haglund et al. 1997).

Adipose and liver tissue from two Swedish males were examined for several PBDEs (BDE 28, BDE 47, BDE 85, BDE 99, BDE 100, BDE 153, and BDE 154) (Meironyté Guvenius and Norén 1999). The distribution of congener concentrations in the adipose and liver tissues for each individual were similar. BDE 47, BDE 99, and BDE 153 were the predominant congeners with adipose BDE 47 concentrations ranging from 2 to 2.4 ng/g lipid weight, BDE 99 concentrations of 1.6 ng/g lipid weight, BDE 100 concentrations of 0.1 ng/g lipid weight, and BDE 153 concentrations ranging from 1 to 1.3 ng/g lipid weight. The total PBDE concentration (i.e., the sum of the seven congeners) in adipose tissue was 5 ng/g lipid weight. Human liver and adipose tissues from one woman and four men autopsied in Sweden in 1994 were analyzed for PBDEs containing 3–6 bromine atoms (Meironyté Guvenius and Norén 2001). PBDEs were found in all of the tissue samples. The sums of nine congeners (BDE 17, BDE 28, BDE 47,



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BDE 66, BDE 100, BDE 99, BDE 85, BDE 154, and BDE 153) were 5–18 and 4–8 ng/g lipids in liver and adipose tissue, respectively. The PBDE congeners BDE 47, BDE 99, and BDE 153 occurred at the highest concentrations and constituted 87–96 and 84–94% of the total sum in liver and adipose tissue, respectively. Strandman et al. (1999) measured the concentration of BDE 47, BDE 99, and BDE 153 in adipose tissue samples from 10 randomly selected individuals in Finland. Mean concentrations were 7.3 ng/g fat for BDE 47, 2.2 ng/g fat for BDE 99, and 2.3 ng/g fat for BDE 153. Concentrations of PBDEs were measured in adipose tissue samples from 13 individuals (3 women, 10 men) from Tarragona, Spain; the mean concentrations of BDE 47, BDE 99, and BDE 153 were 1.36, 0.42, and 1.83 ng/g lipid weight, respectively. The mean concentrations of pentaBDE and hexaBDE were 0.93 and 1.83 ng/g lipid weight, respectively (Meneses et al. 1999).

**Human Milk.** Schechter et al. (2003) reported the first findings on concentrations of PBDEs congeners in human milk from individuals in the United States. Forty-seven individual milk samples were analyzed from nursing mothers, 20–41 years age, from a milk bank in Austin, Texas, and a community health clinic in Dallas, Texas, both in the year 2001. The median concentration of the sum of PBDE congeners was 34.0 ng/g lipid. The predominant congener was BDE 47 (18.4 ng/g lipid); other congeners detected were BDE 17, BDE 28, BDE 66, 2, BDE 85, BDE 99, BDE 100, BDE 138, BDE 153, BDE 154, and BDE 183 at median concentrations of 0.01, 1.2, 0.14, 0.41, 5.7, 2.9, 0.09, 2.0, 0.22, and 0.07 ng/g lipid, respectively. DecaBDE was detected in 7 out of 47 samples with a maximum concentration of 8.24 ng/g lipid. PBDE concentrations in breast milk from this study were similar to concentrations found in U.S. blood and adipose tissue lipid from California and Indiana and are 10–100 times greater than human tissue concentrations in Europe (Schechter et al. 2003). These data have been updated to include 2001–2004 samples and are provided in Table 6-13 (Schechter et al. 2005).

Median concentrations of BDE 47, BDE 100, and BDE 153 were 26, 4, and 3.5 ng/g lipid, respectively, in breast milk samples collected from 10 mothers in Pennsylvania (LaKind et al. 2009). The detection frequency was 100% for each congener in all 35 samples collected.

Norén and Meironyté (1998, 2000) examined the temporal trends of PBDE concentrations in pooled breast milk samples from mothers in Stockholm, Sweden. Between 1972 and 1997, the concentration of PBDEs in human breast milk increased, with a doubling rate of 5 years. In the 1997 sample, the concentration of PBDEs (sum of eight congeners) was 4 ng/g lipid, whereas the 1972 sample contained 0.07 ng/g lipids (Meironyté et al. 1999). The authors suggest that the current exposure of humans to PBDEs may not be only diet; other exposure routes may result from the presence of PBDE in both work

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**Table 6-13. Concentrations of PBDE Congeners in Human Milk from Nursing Mothers in the United States (2001–2004) (ng/g Lipid)**

Female	Lipid (%)	Age (years)	Nursing (weeks)	BDE congener													Sum
				17	28	47	66	77	85	99	100	138	153	154	183	209	
1 <sup>a</sup>	4.8	31	3	ND	0.2	2.9	0.02	ND	0.08	0.7	0.7	ND	1.5	0.06	ND	ND	6.2
2 <sup>a</sup>	1.3	29	3	ND	0.3	3.5	ND	ND	0.08	0.7	0.5	ND	0.9	0.06	0.06	ND	6.2
3 <sup>a</sup>	2.1	23	74	ND	0.2	3.9	0.06	ND	0.11	1.5	0.6	ND	0.4	0.08	ND	ND	6.9
4 <sup>b</sup>	4.8	32	21	—	0.3	3.5	0.14	—	0.08	1.6	0.7	0.09	1.4	0.09	0.04	—	8
5 <sup>a</sup>	2.6	22	40	0.01	0.3	6.3	0.05	ND	0.23	2.8	1.2	ND	0.7	0.2	0.05	ND	11.8
6 <sup>a</sup>	3.6	36	109	ND	0.4	7.8	0.09	ND	0.23	2.4	1.1	0.01	0.4	0.11	ND	ND	12.5
7 <sup>a</sup>	1.9	32	20	ND	0.7	8.2	0.04	ND	0.22	1.3	1.7	ND	0.9	0.12	0.08	ND	13.3
8 <sup>a</sup>	6.3	25	2	ND	0.4	7.9	0.02	ND	0.38	2.3	2.7	ND	0.8	0.15	0.06	ND	14.7
9 <sup>b</sup>	2.1	35	29	—	0.7	8.8	0.19	—	0.17	1.5	1.7	0.16	2	0.06	0.03	—	15.2
10 <sup>a</sup>	5.5	32	30	ND	0.4	8	0.01	ND	0.44	2.9	2	ND	0.9	0.14	0.24	0.48	15.6
11 <sup>b</sup>	5	20	2	0.01	1.1	10.9	0.05	ND	0.18	2	2.4	ND	1.3	0.17	0.04	ND	18.1
12 <sup>a</sup>	3.4	23	3	0.01	0.4	8	0.03	ND	0.35	3.1	2.7	ND	2	0.21	0.61	0.93	18.3
13 <sup>b</sup>	1.3	32	16	—	0.8	10.5	ND	—	0.35	2.5	2.2	0.19	2	0.12	0.03	—	18.6
14 <sup>a</sup>	3.4	25	NA	0.02	1.1	12	0.13	ND	0.23	2.5	1.8	ND	1.3	0.15	0.08	1.85	21.1
15 <sup>a</sup>	2.9	21	29	0.03	0.5	10.7	0.09	ND	0.27	5.5	2.1	ND	0.9	0.35	0.07	2.74	22.4
16 <sup>b</sup>	3.5	30	30	—	0.7	6.9	ND	—	0.12	1.3	4.6	0.41	8.5	0.19	0.06	—	22.8
17 <sup>a</sup>	1	23	2	ND	0.9	14.2	0.11	ND	0.37	3.7	2.6	ND	1.3	0.24	0.09	ND	23.5
18 <sup>b</sup>	3.7	23	19	—	0.7	13.2	0.57	—	0.29	3.7	2.5	0.29	1.9	0.17	0.06	—	23.5
19 <sup>a</sup>	3.2	26	2	ND	1.3	17.4	0.19	ND	0.35	4	2.1	ND	0.7	0.18	ND	ND	26.2
20 <sup>b</sup>	3.5	34	22	—	1	14.3	0.29	—	0.46	5.7	3.6	0.25	1.4	0.2	0.1	—	27.3
21 <sup>b</sup>	3.1	33	60	—	1.4	18.4	ND	—	0.25	4.1	1.8	0.09	2.1	0.16	0.06	—	28.3
22 <sup>b</sup>	4.9	38	26	—	1.2	17.4	ND	—	0.34	7.1	2.3	0.14	0.6	0.3	0.12	—	29.6
23 <sup>a</sup>	3.4	30	2	0.01	0.7	15.2	0.06	ND	0.42	4.2	2.3	ND	3	0.22	0.03	3.97	30.1
24 <sup>a</sup>	5.1	28	53	0.01	1.1	20	0.18	ND	0.53	5.1	3.9	0.01	2.7	0.32	0.11	ND	34
25 <sup>b</sup>	4.7	35	NA	—	1.3	20.9	0.56	—	0.31	6.3	2.9	0.14	1.2	0.22	0.17	—	34.1
26 <sup>a</sup>	1.1	41	38	0.02	1.5	19.5	0.11	ND	0.41	3.4	3.3	ND	7.7	0.18	ND	ND	36.1
27 <sup>b</sup>	6.1	37	25	—	7.6	17.2	1.19	—	0.35	6.1	2.3	0.18	1.7	0.27	0.05	—	36.8
28 <sup>b</sup>	3	27	51	—	1.4	28.2	ND	—	0.51	7.5	2.9	0.25	0.7	0.2	0.75	—	42.4
29 <sup>b</sup>	4.8	25	NA	—	1.8	21.6	0.94	—	0.5	9.4	4.4	0.47	5.8	0.6	0.06	—	45.5
30 <sup>b</sup>	2.2	39	11	—	1.1	26.8	ND	—	0.75	8.9	5.3	0.58	2	0.45	0.1	—	46
31 <sup>b</sup>	5.6	34	NA	—	2.7	31.8	ND	—	0.42	7.8	3.1	0.09	0.8	0.22	0.1	—	47
32 <sup>b</sup>	3.4	27	10	—	2.6	30.1	0.75	—	0.57	5.9	6.5	0.32	2.5	0.34	0.09	—	49.6
33 <sup>a</sup>	2.8	20	13	0.02	1.1	31.3	0.17	ND	0.13	10.2	5.9	0.02	1.5	0.48	0.11	2.96	53.9
34 <sup>b</sup>	4	20	13	—	3.4	33.5	2.32	—	0.49	5.8	5.8	0.27	2.6	0.32	0.06	—	54.6
35 <sup>b</sup>	3.3	26	17	—	1.4	32.3	0.7	—	0.66	9.6	5.7	0.46	12.4	0.51	0.05	—	63.8
36 <sup>b</sup>	2.2	20	16	—	1.4	25.5	0.75	—	0.76	8	18.3	1.75	18.3	0.94	0.08	—	75.8
37 <sup>a</sup>	1.1	22	51	0.04	2.2	44.3	0.55	0.03	0.64	10.8	8.1	0.02	14.7	0.56	ND	ND	81.9
38 <sup>b</sup>	4.3	29	38	—	5.2	34.8	0.61	—	1.94	9.8	29.2	1.2	14.5	0.96	0.1	—	98.2

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**Table 6-13. Concentrations of PBDE Congeners in Human Milk from Nursing Mothers in the United States (2001–2004) (ng/g Lipid)**

Female	Lipid (%)	Age (years)	Nursing (weeks)	BDE congener													Sum
				17	28	47	66	77	85	99	100	138	153	154	183	209	
39 <sup>a</sup>	1	26	22	0.02	1.7	54.7	0.54	ND	1.63	23.6	10	ND	4.8	1.15	0.07	ND	98.2
40 <sup>b</sup>	4.9	32	38	—	3.4	49.7	1.21	—	1.2	7.7	21.1	1.4	16.3	0.93	0.15	—	103.1
41 <sup>b</sup>	3.4	30	9	—	3.9	63.1	3.13	—	2.81	30.1	16.2	3.29	17.2	1.94	0.08	—	141.6
42 <sup>b</sup>	1.2	21	2	0.1	10.1	120.9	1.68	0.06	2.64	30.3	20.1	0.13	16.4	2.07	ND	ND	204.3
43 <sup>b</sup>	1.2	33	15	—	8	139.6	ND	—	4.12	44.6	23	4.47	21.8	2.76	0.18	—	248.5
44 <sup>a</sup>	1	23	2	0.06	3.6	172.4	1.14	ND	6.28	69.8	31.9	0.08	8.4	3.07	0.16	ND	296.9
45 <sup>b</sup>	2.1	34	13	—	7.5	199.6	6.67	—	7.73	108.5	31.7	4.12	6.9	3.62	0.36	—	376.7
46 <sup>a</sup>	1.7	33	47	0.18	6.1	196.2	2.07	0.16	6.46	111	31	0.27	15.5	7.21	1.32	8.24	385.5
47 <sup>b</sup>	5.1	29	28	—	16.1	271.5	3.16	—	6.29	50.4	47.4	6.86	14.1	2.87	0.12	—	418.8
48	3.18	NA		0.011	0.6	9.9	0.058	NA	NA	2.7	1.3	0.022	0.62	0.13	0.072	2.4	17.81
49	3.28	NA		0.016	1.2	25	0.12	NA	NA	11	4.5	0.092	3.1	0.57	0.092	0	45.69
50	3.9	NA		0.016	4.4	41	0.098	NA	1.3	11	11	0.15	6.1	0.71	0.14	0	75.91
51	3.12	NA		0.015	2.3	37	0.11	NA	0.49	6.7	6.9	0.086	11	0.39	0.11	0	65.10
52	6.31	NA		0.01	1.8	14	0.084	NA	NA	2.9	2.9	0.033	2.1	0.17	0.1	0	24.1
53	7.04	37		0.02	0.71	16	0.13	NA	0.46	5.8	2.5	0.03	2.2	0.28	0.057	0.02	28.21
54	3.29	33		0.014	0.76	9	0.056	NA	0.14	1.5	2.1	0.01	2.6	0.094	0.051	0.05	16.37
55	2.44	38		0.02	1.2	19	0.18	NA	0.35	5	2.7	0.02	2.4	0.27	0.051	2.5	33.69
56	6.5	NA		0.005	0.9	6.1	0.088	NA	0.082	1	1.2	0.01	6.7	0.077	0.029	0.03	16.22
57	5.55	NA		0.005	0.9	6.1	0.088	NA	0.082	1	1.2	0.01	3.4	0.077	0.029	0.03	12.92
58	6.75	NA		0.046	1.5	24	0.18	NA	0.38	4.8	2.7	0.02	2.1	0.22	0.57	0.05	36.57
59	3.08	NA		0.016	1.2	10	0.086	NA	0.18	2	1.3	0.01	4	0.15	0.096	0.1	19.14

<sup>a</sup>Austin milk bank sample.<sup>b</sup>Dallas milk bank sample.

NA = not available; ND = not detected

Source: Schecter et al. 2005

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and home environments. PBDE concentrations were studied in breast milk obtained from mothers pregnant for the first time (n=39, ages 22–36 years old) from Uppsala County, Sweden (Darnerud et al. 1998). The mean value of total PBDEs (sum of eight congeners) was 4.4 ng/g fat; the major congener was BDE 47, contains ca. 55% of the total PBDEs. Lind et al. (2003) reported concentrations of PBDEs in human breast milk sampled from Uppsala County, Sweden. Total PBDEs, BDE 47, BDE 99, and BDE 100 concentrations were 4.01, 2.35, 0.62, and 0.38 ng/g lipid, respectively. In human breast milk from 25 German mothers, the concentrations of PBDEs ranged from 0.6 to 11 ng/g lipid (de Wit 2002). In 1992, the mean concentration of total PBDEs (sum of BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, and BDE 183) was 5.8 ng/g lipid weight for samples (n=6) from mothers from Ontario and Quebec, Canada (Ryan and Patry 2000). Combined samples from 1992 representing four regions of Canada and one representing all Canadian provinces had total PBDE concentrations ranging from 2.6 to 19 ng/g lipid weight; the highest concentrations were observed in the New Brunswick, Nova Scotia, and Prince Edward Island. Breast milk samples from Finland, collected between 1994 and 1998, had concentrations of total PBDEs (sum of BDE 28, BDE 47, BDE 99, and BDE 153) ranging from 0.88 to 5.9 ng/g lipid weight (Strandman et al. 2000). In Japan, breast milk samples had total PBDE concentrations (sum of BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154) ranging from 0.66 to 2.8 ng/g lipid weight (Ohta et al. 2002). Women who consumed fish had a somewhat higher concentration of total PBDEs (range, 1.4–2.8 ng/g lipid weight) compared to women who ate less fish (range, 0.67–0.87 ng/g lipid weight). BDE 47 was the major congener in most of the samples; BDE 153 concentrations were analogous to BDE 47 concentrations in some samples (Ohta et al. 2002).

***Hydroxy- and Methoxy- Derivatives in Biota.*** Hydroxy- and methoxy- derivatives of PBDEs have been identified in biota. However, their origin in the environment has not yet been explained. Anthropogenic sources of these compounds have not been found. Tetra- and pentabrominated methoxy (MeO) BDEs were found in herring, salmon, grey seal, ringed seal, and white-tailed sea eagle from the Baltic region (Asplund et al. 1999a; Haglund et al. 1997) as well as beluga whale from Svalbard and pilot whale from the Faroe Islands (van Bavel et al. 2001). The concentrations of hydroxy- and methoxy- derivatives were of the same order of magnitude as PBDEs present in the samples. Biogenic production via metabolism of PBDEs or natural production via biobromination has been suggested as the origin for these compounds. Naturally produced methoxy-tetrabrominated diphenyl ethers have been reported in tropical marine sponges (*sp. Dysidea*) as well as in green algae (*sp. Cladophora*) collected in Japan (Kierkegaard et al. 2004). Kierkegaard et al. (2004) found that the concentrations of 6-methoxy-2,2',4,4'-tetraBDE in herring from five locations along the Swedish coast increased from south to north in the Baltic Sea. No

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correlation between the concentrations of BDE congeners and methoxy-brominated diphenyl ethers was observed, indicating sources other than PBDEs for these compounds.

***Biomonitoring Historical Trends and Future Projections.*** Concentrations of PBDE in human and animal tissues have increased since their development and widespread use as flame retardants in commercial products. Blood samples collected from U.S. residents in 1973 did not contain measurable concentrations of PBDE congeners; however, many congeners have been identified at varying concentrations in U.S. blood samples since the widespread use of PBDEs as flame retardants (Schechter et al. 2005). For example, the CDC reported BDE congener concentration in serum collected in 2003–2004 from the general U.S. population (CDC 2015). The total geometric mean concentrations ranged from below the limit of detection for BDE 17 to 20.5 ng/g lipid for BDE 47. In general, body burden concentrations of PBDEs in North America are higher than in Europe due to higher historical demand and usage. Since all production and use of penta-, octa-, and now decaBDE have ceased in the United States, future biomonitoring results will likely show a gradual decline in body burden concentrations of these substances in U.S. residents as products containing PBDEs ultimately become rare. Age-dependent data on PBDE levels indicate several sources of human exposure. Serum samples collected in Australia suggest that PBDE levels increase from infant to toddler and then gradually decrease over time (Toms et al. 2009). Peak average concentrations were observed in toddlers 2.6–3 years of age (51 ng/g lipid), which is later than when breastfeeding usually ceases, suggesting a lower capacity to eliminate PBDEs or greater exposure through unique exposure pathways more common for this age group (e.g., ingestion or dermal exposure of contaminated dust particles in carpeting). The EPA Exposure Assessment of Polybrominated Diphenyl Ethers published in May of 2010, summarizes many other biomonitoring studies not discussed here and the reader is encouraged to consult this assessment for additional analysis of the environmental fate and biomonitoring of PBDEs (EPA 2010).

## 6.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

Humans are exposed to PBDEs by a wide variety of routes including ingestion of contaminated foods, inhalation of air, ingestion of contaminated dusts/soils, and dermal exposure routes. The EPA published an exposure assessment of the U.S. population to PBDEs and determined that the overall weight-of-evidence suggested that bulk of U.S. exposures occur in indoor environments through ingestion and contact with house dust. It concluded that house dust accounts for between 80 and 90% of total exposures of the general population, with the remainder due primarily to food ingestion. Watkins et al. (2011)

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determined that regular hand washing decreases the mass of PBDEs on hands from dust samples and is thus expected to reduce intake from hand-to-mouth activities.

Intake doses of BDE 47, BDE 99, BDE 100, and BDE 153 from all exposure pathways for the North American population were modeled by Wong et al. (2013). The model assumed intake of PBDEs increased exponentially to a peak in 2004, and has since exponentially declined. The intakes of BDE 47, BDE 99, BDE 100, and BDE 153 were estimated as 3.88–54, 1.59–2.39, 1.17–2.98, and 1.37–2.44 ng/kg body weight/day depending upon how the intakes were fit to measured body burden data using different elimination half-lives. Trudel et al. (2011) used eight different exposure pathways (oral uptake of food, dust, soil, and organic films; inhalation of air; and dermal uptake of dust, soil, and organic films) to model intakes of PBDEs for different age/gender groups in North American and European populations. The mean intakes for total PBDEs in the North American population were 210.0, 80.0, 79.0, 69.0, 43.0, 28.0, and 22.0 ng/kg body weight/day for infants, toddlers, children, female teenagers, male teenagers, female adults, and male adults, respectively. These concentrations are about 3–8 times greater than the estimated intakes for European populations. Lorber (2008) also estimated PBDE intake of the U.S. population through similar exposure routes. The adult intake of total PBDEs was estimated as 7.7 ng/kg body weight/day, while the intake of children aged 1–5 years was 49.3 ng/kg body weight/day. The intakes for 6–11 year olds and 12–19 year olds were estimated as 14.4 and 9.1 ng/kg body weight/day, respectively (Lorber 2008). Exposure from indoor house dust accounted for about 82% of the intake (66% from soil/dust ingestion, 16% from soil/dust dermal contact) of total PBDEs, while inhalation and ingestion of food and water accounted for <20% of the total intake. BDE 47, BDE 99, BDE 100, and BDE 209 were the predominant congeners, accounting for 26, 28, 11, and 27%, respectively, of the total intake (Lorber 2008). The EPA 2010 Exposure Assessment of Polybrominated Diphenyl Ethers calculated the adult intake dose of total PBDEs to be 7.1 ng/kg body weight/day (EPA 2010). The largest source contributing to PBDE exposure in the United States was reported to be house dust (ingestion and dermal exposure), contributing about 90% of the overall estimated intakes. The EPA exposure assessment estimated children intakes as 47.2 ng/kg body weight/day for 1–5 year olds, 13.0 ng/kg body weight/day for 6–11 year olds, and 8.3 ng/kg body weight/day for 12–19 year olds. Intake modeling using a breastfeeding pathway, which used measured milk concentrations and infant ingestion rates of human milk, led to estimated infant intakes of 141 ng/kg body weight/day (EPA 2010). While exposure to dust appears to be the predominant exposure pathway for the general population of North American residents, PBDE exposure through dietary routes appears to be more important for European communities (Abdallah and Harrad 2014; Law et al. 2008).

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Breast adipose samples collected in northern California in the late 1990s contained quantifiable amounts of BDE 47, BDE 99, and BDE 153 (She et al. 2000). Mean concentrations were 18 ng/g lipid weight for BDE 47, 4.9 ng/g lipid weight for BDE 99, and 2.2 ng/g lipid weight for BDE 153. In studies of the general populations of other countries, it has also been shown that exposure to lower-brominated PBDE congeners by the general population is widespread (see Section 6.4.4; Haglund et al. 1997; Meneses et al. 1999). In general, concentrations of decaBDE in human tissues and body fluids are lower than for the lower-brominated congeners, presumably due to a more rapid elimination half-life (Trudel et al. 2011).

Consumption of fish has been associated with elevated concentrations of PBDEs in tissues from the Swedish population (Bergman et al. 1999). In Sweden, fish consumption is about 30 g/day; this translates to an estimated 0.1 µg of pentaBDE and 0.3 µg of total PBDEs from fish that is ingested by humans daily (WHO 1994a). The fish of greatest concern to humans are bottom feeders like carp and catfish. Harrad et al. (2004) estimated the daily dietary intakes of PBDEs in omnivorous and vegan diet samples from the United Kingdom. In this study, the median lower bound estimates of dietary exposure (i.e., where a congener is below the detection limit, the concentration is assumed to be zero) for BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and total PBDEs were 46.4, 42.6, 0, 0, 0, and 90.5 ng/day, respectively (Harrad et al. 2004). The International Polar Year Inuit Health Survey in 2007–2008 evaluated PBDE blood concentration for 2,172 Inuit adults in Canada (Laird et al. 2013). The sum concentration of BDE 47, BDE 99, and BDE 100 in the general population ranged from 0.04 to 10.6 µg/L in blood plasma. Like PCBs, there may be a higher risk of exposure to PBDEs in Native Americans who reside in the Arctic region and consume whale and seal blubber (Jaret 2000).

Workers involved in the production and manufacture of PBDE-containing plastics and plastic products are exposed to PBDEs. Body burden data indicate higher concentrations for workers exposed to PBDEs than for the general population. Occupational exposure to PBDEs also occurs in workers at plants that dismantle electronic equipment, computer monitor repair technicians, and automobile drivers, as well as other professions (Lindström 1999). Occupational exposure occurs primarily by inhalation. Inhalation of vapor-phase PBDEs is expected to be low due to the low vapor pressures of PBDEs (see Table 4-4); however, the inhalation of particulate phase PBDEs is possible during plastic reprocessing where grinding or shredding of polymers with PBDEs occurs. Occupational exposure may also likely involve oral exposure to particulate PBDEs as a result of hand-to-mouth activity.

Air samples were taken from an electronics dismantling plant, an office with computers, and outdoors and then analyzed for PBDEs (Sjödín et al. 1999a, 2001a). The electronics dismantling plant had the highest

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concentrations of PBDEs, with mean concentrations of 2.5 pmol/m<sup>3</sup> (1.25 ng/m<sup>3</sup>) for BDE 47, 4.6 pmol/m<sup>3</sup> (2.6 ng/m<sup>3</sup>) for BDE 99, 6.1 pmol/m<sup>3</sup> (3.93 ng/m<sup>3</sup>) for BDE 153, 26 pmol/m<sup>3</sup> (18.8 ng/m<sup>3</sup>) for BDE 183, and 38 pmol/m<sup>3</sup> (36.5 ng/m<sup>3</sup>) for decaBDE (BDE 209) (Sjödin et al. 1999a, 2001a). Air samples were found to be 4–10 times higher in PBDE concentrations near a plastic shredder when compared to other locations in the plant (range, 0.42–200 ng/m<sup>3</sup>). Concentrations of PBDEs in the office (range, <0.002–0.09 ng/m<sup>3</sup>) were 400–4,000 times lower than in the plant, and PBDEs were not detected in outside air (Sjödin et al. 1999a, 2001a).

Lipid adjusted serum levels of 11 BDE congeners collected from the US general population were reported in the Fourth National Report on Human Exposure to Environmental Chemicals, Updated Tables (CDC 2015; also see <http://www.cdc.gov/biomonitoring/>). Serum levels for BDE 17, BDE 28, BDE 47, BDE 66, BDE 85, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183 were evaluated in samples collected between 2003 and 2008; BDE 209 was evaluated in samples collected from 2005 to 2008. In the NHANES 2003–2004 survey years, congener BDE 47 was detected at a concentration of 20.5 ng/g lipid (geometric mean), the highest concentration for all samples. BDE-153 had the second highest geometric mean concentration of 5.7 ng/g lipid. BDE 28, BDE 99, BDE 47, BDE 100, and BDE 153 were in ≥60% of participants (Sjödin et al. 2008). The serum levels of BDE 47, BDE 99, and BDE 153 were highest in the youngest age group (12–19 years old) and decreased for the older age groups (from 20–39 to 40–59 years old) and then increased in the ≥60 years old age group.

BDE 47, BDE 153, BDE 154, BDE 183, and decaBDE (BDE 209) were measurable in blood plasma from three groups of workers (i.e., workers at a computer disassembly plant, workers in a computerized office, and a control group) (Sjödin et al. 1999b). The median concentrations (sum of five congeners) were highest for the computer disassembly plant workers (26 ng/g lipid weight); the office workers had a median concentration of 4.1 ng/g lipid weight and the control group had a median concentration of 3.3 ng/g lipid weight. The congener patterns for the control group and office workers were similar, with BDE 47 having the highest concentrations. For the computer disassembly plant workers, the median concentrations of BDE 183, BDE 153, BDE 154, BDE 47, and BDE 209 were 7.8, 4.5, 1.2, 2.9, and 4.8 ng/g lipid weight, respectively. Blood serum samples from 19 full-time computer technicians were analyzed (Hagmar et al. 2000a). The serum concentrations of BDE 153, BDE 183, and BDE 209 in these samples were found to be approximately 5 times higher than the control and office workers in the Sjödin et al. (1999b) study. The median concentration for total PBDEs (for the sum of five congeners) was 10.6 pmol/g (7.0 ng/g) lipid weight. The highest concentrations were for BDE 153. Two octaBDE congeners and one nonaBDE congener were also detected.



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**6.6 EXPOSURES OF CHILDREN**

This section focuses on exposures from conception to maturity at 18 years in humans. Differences from adults in susceptibility to hazardous substances are discussed in Section 3.7, Children's Susceptibility.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways. Children drink more fluids, eat more food, breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor, put things in their mouths, sometimes eat inappropriate things (such as dirt or paint chips), and spend more time outdoors. Children also are closer to the ground, and they do not use the judgment of adults to avoid hazards (NRC 1993).

Body burden data, as well as intake modeling, suggest that infants and toddlers have higher exposures to PBDEs as compared to older children or adults. PBDE concentrations increase from infant to toddler and then gradually decrease over time. PBDE intake values for children have been estimated using several models (EPA 2010; Lorber 2008; Trudel et al. 2011; Wong et al. 2013). Using the model developed by the EPA, total PBDE intakes for children residing in the United States were estimated as 47.2 ng/kg body weight/day for 1–5 year olds, 13.0 ng/kg body weight/day for 6–11 year olds, and 8.3 ng/kg body weight/day for 12–19 year olds. Data from fetal tissue and several studies, including measurements of PBDE congeners from umbilical cord blood, indicate that the fetus is exposed to PBDEs through the mother.

Schechter et al. (2003, 2005) reported the first findings on concentrations of PBDEs congeners in human milk from individuals in the United States. The median concentration of the sum of PBDE congeners was 34.0 ng/g lipid with BDE 47 (18.4 ng/g lipid) as the predominant congener. DecaBDE was detected in 7 out of 47 samples with a maximum concentration of 8.24 ng/g lipid. The concentrations of PBDEs in breast milk from this study were 10–100 times greater than human tissue concentrations in Europe (Schechter et al. 2003). Guo et al. (2016) found that PBDE levels in breast milk samples declined in females from California over two sampling periods (2003–2005 and 2009–2012). The geometric mean of total PBDE congeners (sum of BDE 28, 47, 99, 100, 153, and 154) in breast milk was 67.8 ng/g lipid in the 2003–2005 sampling period (n=82) and 45.7 ng/g lipid in the 2009–2012 sampling period (n=66).

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PBDE levels were shown to increase in human milk samples collected from different regions of Canada from 1992 to 2002, but declined slightly from 2002 to 2005 (Ryan and Rawn 2014). The median and geometric mean levels of total PBDEs (sum of BDE 28, 47, 99, 100, 153, 154, and 183) in collected milk samples were 2.992 and 3.536 ng/g lipids in 1992, respectively, and 22.104 and 25.162 ng/g lipid, respectively, in 2002. The median and geometric mean of total PBDEs in milk samples collected in March and April 2005 were 19.948 and 21.082 ng/g lipid, respectively.

PBDEs were detected in human placental tissues (n=102) collected between 2010 and 2011 in Durham County, North Carolina (Leonetti et al. 2016). The geometric mean concentration of total PBDE (sum of BDE 47, 99, 100, 153, 154, and 209) was 17.6 ng/g lipid. The detection frequencies of the individual congeners were: BDE 47, 91.2%; BDE 99, 68.6%; BDE 100, 88.2%, BDE 153, 93.1%; BDE 154, 83.3%; and BDE 209, 52.9%.

PBDEs were detected at a median concentration of 53,000 ng/g in a variety of toys such as hard plastic toys (racing cars, vehicles, toy weapons, etc.), foam toys, rubber/soft plastic toys (dolls and teething), and stuffed toys that were purchased in China from 2007 to 2008 (Chen et al. 2009). These findings suggest additional possible exposure routes to children and toddlers through mouthing activities and dermal contact with toys.

PBDE congeners (predominantly BDE 47, 99, and 209) were detected in 100% of dust samples collected from 40 California daycare and preschool centers (Bradman et al. 2014). The mean and median total PBDE (BDE 47, 99, 100, 118, 153, 154, 183, 190, 197, 203, 205, 206, 207, and 209) levels in dust samples were 7,956.6 and 4,225 ng/g, respectively. Individual congeners (BDE 47, 99, 100, 153, 154, and 209) were detected in indoor air samples at the facilities at mean levels ranging from 0.001 to 1.63 ng/m<sup>3</sup>.

Hoffman et al. (2016) analyzed serum levels and handwipe samples from 83 children aged 12–36 months residing in the state of North Carolina. Correlations between serum and handwipe levels of PBDE congeners and behavioral patterns were observed. For example, increased age and increased activity was positively correlated to levels of PBDEs in serum and handwipe samples, while time spent sleeping (a measure of inactivity) was negatively correlated with PBDE levels in serum. It was reported that for each additional hour of sleep, BDE 47, BDE 99, BDE 100, and BDE 153 serum levels decreased by 12, 15, 9, and 10%, respectively, in the children. BDE 47, BDE 99, BDE 100, and BDE 153 levels in handwipes decreased 30, 31, 30, and 23% for each additional hour of sleep.

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**6.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES**

Workers who were involved in the production and manufacture of PBDE-containing plastics and plastic products were exposed to higher concentrations of PBDEs than the general population. Body burden data indicate higher concentrations for workers exposed to PBDEs than for the general population.

Occupational exposure to PBDEs also occurs in workers at plants that dismantle electronic equipment, computer monitor repair technicians, and automobile drivers, as well as other professions (Lindström 1999). Occupational exposure occurs primarily by inhalation and ingestion of dust containing PBDEs.

Stapleton et al. (2008) examined PBDE serum concentrations in workers involved with foam recycling and carpet installation in the United States. Serum PBDE concentrations in foam recyclers (median, 160 ng/g lipid) and carpet installers (median, 178 ng/g lipid) were significantly greater than a non-occupationally exposed control group (median, 19.3 ng/g lipid).

Firefighters appear to have higher exposure potential to PBDEs and other types of flame retardants because they are exposed to the combustion products of the flame retardants as well as the original forms of the chemicals. In a study conducted to examine exposure to PBDEs in 101 firefighters from Southern California, the median and geometric mean for total PBDEs (sum of BDE 28, 47, 99, 100, and 153) in the serum of the firefighters were 59.1 and 66.2 ng/g lipid, respectively (Park et al. 2015). These levels are approximately 40% greater when compared to the general population. The median and geometric mean for the same total PBDE congeners obtained from a subset of the NHANES survey with similar age and gender as the firefighters were 36 and 40.8 ng/g lipid, respectively.

**6.8 ADEQUACY OF THE DATABASE**

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of PBDEs is available. Where adequate information is not available, ATSDR, in conjunction with NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of PBDEs.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean

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that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

### 6.8.1 Identification of Data Needs

**Physical and Chemical Properties.** Many of the relevant physical and chemical properties of the PBDEs are available (see Tables 4-3 and 4-4). Very limited data are available on the physical and chemical properties for the individual congeners (Braekevelt et al. 2003; Tittlemier et al. 2002). Important data, such as  $K_{ow}$ ,  $K_{oc}$ , vapor pressure, and Henry's Law constant, are necessary for the prediction of the environmental fate and transport of PBDEs.

**Production, Import/Export, Use, Release, and Disposal.** According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit substance release and off-site transfer information to the EPA. The TRI, which contains this information for 2014, became available in March 2016. This database is updated yearly and should provide a list of industrial production facilities and emissions.

There are no current manufacturers of technical PBDEs in the United States. PentaBDE and octaBDE mixtures were voluntarily withdrawn from the U.S. marketplace by their manufacturers at the end of 2004, leaving only decaBDE being marketed for use in commercial products in the United States (EPA 2010). In December of 2009, the two remaining U.S. producers of decaBDE, Albemarle Corporation and Chemtura Corporation (formerly known as the Great Lakes Chemical Corporation), and the largest U.S. importer, ICL Industrial Products, Inc., announced commitments to phase out manufacture and importation of decaBDE for most uses in the United States by December 31, 2012, and to end manufacture and import for all uses by the end of 2013 (EPA 2013j). Many consumer goods enter the United States from other countries such as China. OctaBDE was never produced in China, and manufacture of the commercial pentaBDE mixture stopped in 2004; however, there are currently no restrictions on the use of decaBDE, which had a production volume of 20,500 metric tons in 2011 (Ni et al. 2013). It is unclear if items being treated with decaBDE are still entering U.S. markets from other parts of the world.

Given the importance assigned to dust ingestion as an exposure pathway to Americans, more data are needed on human bioavailability of PBDEs from external matrices, such as dust, as well as food for

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exposure characterization. Characterization of the quantity of dust containing PBDEs, ingested by humans, and in particular young children, would improve exposure estimates.

**Environmental Fate.** Based on limited data, photolysis appears to be the dominant transformation process for some PBDEs (e.g., decaBDE) (Hua et al. 2003). PBDEs absorb light in the environmental spectrum. Hua et al. (2003) found that decaBDE and the commercial octaBDE absorbed light up to 325 nm, which indicates that these compounds may be susceptible to photodegradation at environmental wavelengths. However, the importance of photochemical transformation reactions in the environment cannot be determined due to lack of quantitative rate information (EU 2002, 2003a). Better data on degradation via hydroxyl radical reaction and photolysis are needed. Based on a very limited number of studies, biodegradation does not appear to be significant for commercial mixtures of PBDEs (EU 2002, 2003a). Limited studies have been done on biodegradation of PBDEs in the environment under both aerobic and anaerobic conditions, especially studies investigating dehalogenation mechanisms (EU 2002, 2003a). More studies are needed to determine conclusively if commercial PBDE mixtures, such as decaBDE, are degraded to lower-brominated congeners (e.g., BDE 47), which appear to bioaccumulate in fish, animals, and humans (see Section 6.4). Additional data on degradation via hydroxyl radical reaction and photolysis are needed. Since the toxicity and the environmental fate of PBDEs depend on specific PBDEs congeners, development of more data regarding congener-specific fate and transport of PBDEs in the environment are needed.

**Bioavailability from Environmental Media.** The absorption and distribution of PBDEs as a result of inhalation, ingestion, and dermal exposure are discussed in Sections 3.4.1 and 3.4.2. PBDEs will exist in both the vapor and particulate phase in both indoor and outdoor air, and more data are needed regarding the bioavailability of these substances in these two phases (Harrad et al. 2004) and the bioavailability of PBDEs from PBDE-contaminated toys (Chen et al. 2009).

**Food Chain Bioaccumulation.** An abundance of monitoring data illustrates the uptake of lower-brominated diphenyl ethers by aquatic organisms, which results in bioconcentration (see Section 6.4.4; Hardy 2002b). Congener components of the pentaBDE commercial product tend to bioconcentrate to different extents. DecaBDE and octaBDE commercial products do not bioconcentrate to the extent of the penta mixtures; however, monitoring data clearly show that even the higher-brominated congeners are taken up. More information on bioaccumulation and biomagnification of PBDE and its congeners is needed in assessing human health risks.

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**Exposure Levels in Environmental Media.** Reliable monitoring data for the levels of PBDEs in contaminated media at hazardous waste sites are needed so that the information obtained on levels of PBDEs in the environment can be used in combination with the known body burden of PBDEs to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

More monitoring data on the concentrations of total PBDEs and PBDE congeners in air in remote, rural, and urban areas, as well as areas near hazardous waste sites and incinerators, are needed. Although concentrations are predicted to be low, monitoring data on PBDE concentrations in finished drinking water nationwide would be helpful. Sediment concentrations of PBDEs tend to be dominated by higher-brominated congeners (e.g., decaBDE or BDE 209) (deWit 2002; Dodder et al. 2002; Hale et al. 2001b, 2002). Monitoring data indicated that the concentrations of PBDEs are increasing in aquatic organisms with higher concentrations near point sources (Alaee et al. 1999; Dodder et al. 2000; Johnson and Olson 2001; Loganathan et al. 1995; Luross et al. 2000). Additional monitoring data on environmental concentrations of PBDEs would be useful to determine the extent of contamination in environmental media, and also the mechanisms of human exposure to this class of chemicals.

**Exposure Levels in Humans.** Body-burden data indicate that there are low-level exposures to lower-brominated PBDEs for the general population. Information about the average daily intake of PBDEs is available (Bergman et al. 1999; EPA 2010; Lindström 1999; Lorber 2008; WHO 1994a). PBDE concentrations are reported in the current literature for serum, blood, breast milk, and adipose tissue of the general population and occupationally exposed individuals (CDC 2015; EPA 2010; WHO 1994a). Additional data regarding the concentrations of PBDEs in body fluids or tissues of people who reside near hazardous waste sites are needed. This information is necessary for assessing the need to conduct health studies on these populations.

**Exposures of Children.** The most important exposure pathway for infants to PBDEs likely occurs through ingestion of breast milk (EPA 2010). PBDE intakes for infants and young children are typically greater than older children and adults (EPA 2010; Lorber 2008; Trudel et al. 2011; Wong et al. 2013). Since children tend to spend more time playing in carpeting, this leads to the potential to greater exposure to PBDEs through indoor dust. PBDEs have been detected in a variety of toys such as hard plastic toys (racing cars, vehicles, toy weapons, etc.), foam toys, rubber/soft plastic toys (dolls and teething rings), and stuffed toys that were purchased in China from 2007 to 2008 (Chen et al. 2009). These findings indicate additional possible exposure routes to children and toddlers through mouthing activities and dermal contact with toys. Although there has been a gradual phase-out of pentaBDE, octaBDE, and now

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decaBDE, products are still in households that contain these substances. Therefore, continued biomonitoring data of infants and children is needed.

Child health data needs relating to susceptibility are discussed in Section 3.12.2, Identification of Data Needs: Children's Susceptibility.

**Exposure Registries.** No exposure registries for PBDEs were located. This substance is not currently one of the compounds for which a sub-registry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for sub-registries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to exposure to this substance.

### 6.8.2 Ongoing Studies

A prospective study of 316 mothers enrolled during pregnancy being conducted at the Icahn School of Medicine at Mount Sinai, New York will examine prenatal exposure to complex mixtures of endocrine-disrupting compounds including PBDEs (RePORTER 2016).

A study being conducted at the University of California, Berkeley is designed to examine exposures and health effects in vulnerable populations, such as pregnant women and children living in California, where stricter flammability standards have resulted in very high flame retardant exposures (RePORTER 2016).

The Center for Children's Environmental Health Research at the University of California, Berkeley will examine novel methods of examining prenatal exposure to PBDEs and other compounds using shed deciduous teeth and geographic information system (GIS) methods with remote sensing (RePORTER 2016).

A study at the University of Cincinnati is investigating two groups of persistent organic chemicals for their associations with adverse effects in child neurobehavior: PBDEs and perfluoroalkyl chemicals (including perfluorooctane sulfonic acid [PFOS] and perfluorooctanoic acid [PFOA]). The research project will provide novel information to the public about the developmental neurotoxicity of these chemicals. It will also generate new data regarding PBDE exposure routes to aid in future prevention initiatives (RePORTER 2016).

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A study is being conducted at the University of California, San Diego that is collecting and analyzing sediment and biota sample in the in the Southern California Bight for PBDEs (RePORTER 2016).