

## 8. POTENTIAL FOR HUMAN EXPOSURE

### 8.1 OVERVIEW

***Polybrominated Biphenyls.*** PBBs have been identified in at least 9 of the 1,647 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 2004). However, the number of sites evaluated for PBBs is not known. The frequency of these sites can be seen in Figure 8-1. Of these sites, all are located within the United States and none are located in the Commonwealth of Puerto Rico (not shown).

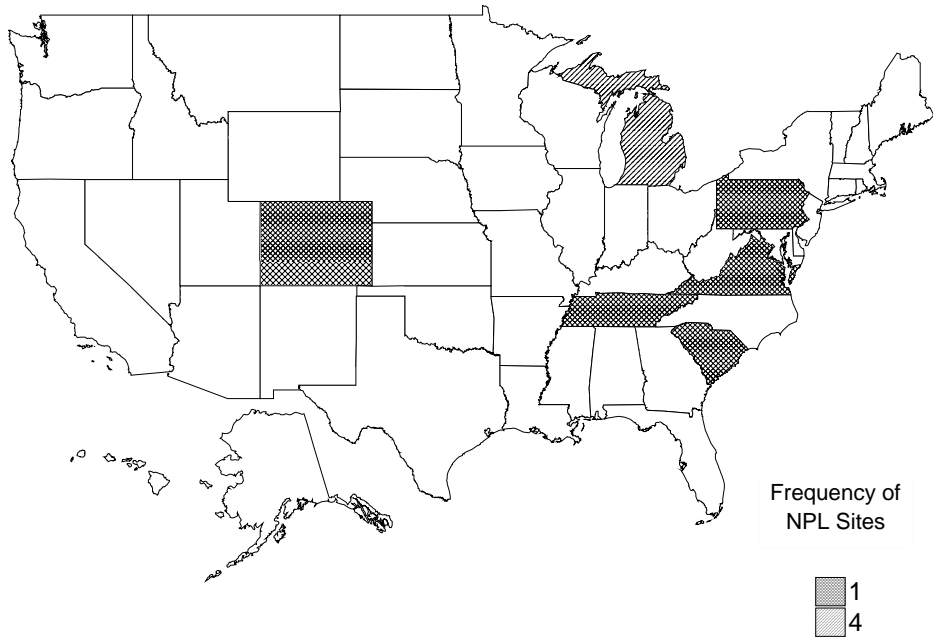
The production of PBBs in the United States ceased in 1979 (IARC 1986). In the past, PBBs were released to the environment during the manufacture of these compounds and disposal of commercial and consumer products containing these compounds (Hesse and Powers 1978; Neufeld et al. 1977). One of the significant sources of environmental contamination occurred as a result of the accidental mixup of FireMaster BP-6 with cattle feed in a number of farms in the lower peninsula in Michigan (see Section 5.2 for additional details concerning this incident). By June 1975, 412 farms had been quarantined. Disposal of contaminated feed, animal carcasses (poultry, dairy cattle, swine), and animal products (dairy, meat, eggs) contributed to environmental contamination (Dunckel 1975; Kay 1977). No information was located on the current levels of contamination at these locations.

PBBs can exist as 209 different congeners, but only about 42 have been synthesized (Sundstrom et al. 1976b). Environmental contamination of PBBs is likely to have occurred mainly from the two commercial products, FireMaster BP-6 and FireMaster FF-1. The principal component in both of these commercial products was 2,2',4,4',5,5'-hexabromobiphenyl or BB-153 (Robertson et al. 1983b).

PBBs are strongly adsorbed to soil and sediment (Filonow et al. 1976; Hesse and Powers 1978) and usually persist in the environment (Jacobs et al. 1978). Adsorption of PBBs generally increases as bromination of the PBBs and organic carbon content of soil and sediment increase (Filonow et al. 1976; Griffin and Chou 1981a, 1981b). As a result, the leaching of commercial mixtures of PBBs from soil is slow. Leaching studies with four Michigan soils mixed with 100 mg/kg 2,2',4,4',5,5'-hexabromobiphenyl showed that <0.6% of the compound leached through soils after a 19-day period. Leachate quantities in this study were equivalent to 20 times the average annual rainfall in Michigan (Filonow et al. 1976). The PBBs in commercial mixtures resist both chemical and biological degradation (Jacobs et al. 1978;

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**Figure 8-1. Frequency of NPL Sites with PBB Contamination**



Derived from HazDat 2004

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Kawasaki 1980; Shelton and Tiedje 1981), although biotic debromination to lower brominated products may occur in anaerobic zones of contaminated sediment and soil (Morris et al. 1992).

PBBs with six or fewer bromine substitutions bioconcentrate in aquatic organisms such as fish, but the octabromo- and decabromobiphenyls do not bioconcentrate significantly in fish (Gobas et al. 1989; Norris et al. 1973; Opperhuizen et al. 1985; Veith et al. 1979; Zitko 1979; Zitko and Hutzinger 1976). Orchard grass, alfalfa, corn, and tops of carrots grown in soil contaminated with PBBs showed no uptake of PBBs, and only minor uptake occurred on carrot roots (Jacobs et al. 1976, 1978). Although PBBs were detected in fish-eating birds and predatory animals that had consumed PBB-contaminated food (Heinz et al. 1983, 1985), the biomagnification potential of PBBs in predators resulting from such consumption remains unknown.

PBBs were detected in air, water, sediment, and soil in the vicinity of the manufacturing plants and in groundwater from a landfill site (DeCarlo 1979; Hesse and Powers 1978; Shah 1978). PBBs were also detected in soil near the contaminated farms in Lower Michigan (Fries and Jacobs 1980). The distribution of PBBs was limited to the environment in the vicinity of production sites and the contaminated farm sites. Recent studies have identified PBBs in marine mammals from coastal seas and the Atlantic Ocean (de Boer et al. 1998). Data regarding the current levels of PBBs in ambient air, drinking water, or food were not located.

No estimate on PBB intake by the general population from air, water, and food was located in the literature. Current intake of PBBs for the general population is expected to be zero or very small. Populations near the contaminated farms in Lower Michigan may still have low exposures from air, water, and food. The level of PBBs in human tissue and body fluids in the exposed population of Michigan has been extensively studied (Brilliant et al. 1978; Cordle et al. 1978; Eyster et al. 1983; Humphrey and Hayner 1975; Lambert et al. 1990; Landrigan et al. 1979; Wolff et al. 1979a, 1982). The finding that PBBs are stored in fatty tissues of the human body and are very slowly excreted (Eyster et al. 1983) indicates a slow decline in the body burden for exposed individuals.

***Polybrominated Biphenyl Ethers.*** The widespread use of PBDEs over the past 30 years has resulted in the presence of some lower-brominated congeners in the environment (e.g., 2,2',4,4'-tetrabromodiphenyl ether [BDE 47]). However, highly brominated congeners (e.g., decaBDE) are typically detected only near point sources. PBDEs are released into the environment from their manufacture and use as additive flame retardants in thermoplastics in a wide range of products (WHO 1994a). PBDEs containing waste

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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). In the future, the disposal of plastic consumables containing PBDEs is likely to increase in the United States (NSC 1999).

PBDEs are strongly adsorbed to soil and sediment and persist in the environment. 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). Lower BDE homologs (e.g., tri- and tetraBDE), which may exist partially in the vapor phase, have the potential for long-range transport in the atmosphere. The detection of lower brominated PBDEs (e.g., 2,2',4,4'-tetraBDE [BDE 47]) in remote regions of the world suggests that long-range transport of these congeners is occurring. Higher BDE homologs (e.g., decaBDE) will primarily exist near point sources. Biodegradation will not be significant 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.

Monitoring studies indicate that lower brominated PBDEs, for example, 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), are transported globally. Atmospheric, water, and biota levels of PBDEs also tend to be dominated by lower brominated congeners (e.g., BDE 47, BDE 99). Environmental concentrations of lower brominated PBDEs (i.e., tetraBDE and pentaBDE) appear to be leveling off in Europe, but appear to be increasing in certain areas in Canada and the United States, although data are too sparse to make broad statements regarding trends. In the environment, higher brominated commercial mixtures (e.g., decaBDE) are concentrated in soils and sediment near industrial point sources.

Studies of the biota indicate that lower brominated congeners (e.g., 2,2',4,4'-tetraBDE [BDE 47]) are being preferentially bioconcentrated. Lower brominated diphenyl ether (e.g., tetra- and penta-) 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. Body-burden data indicate that the general population is exposed to low levels of lower brominated (e.g., tetra- and penta-) BDEs. In general, decaBDE is not found in body burden data.

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Humans are primarily exposed to lower brominated (e.g., tetra- and pentaBDEs) BDEs by inhalation of ambient or contaminated air and ingestion of contaminated food. Levels of PBDEs in body tissues and fluids from individuals living in the United States have recently been determined. Most studies indicate that levels of lower brominated BDEs (e.g., 2,2',4,4'-tetraBDE [BDE 47]) 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). In general, decaBDE is not detectable in body fluids and tissues. Occupational exposure to PBDEs occurs primarily by inhalation of air containing PBDEs.

PBDEs have not been identified in any of the 1,647 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 2004). However, the number of sites evaluated for PBDEs is not known.

## 8.2 RELEASES TO THE ENVIRONMENT

***Polybrominated Biphenyls.*** The production of PBBs in the United States ceased in 1979 (IARC 1986). In the past, PBBs were released to the environment during the manufacture of these compounds and disposal of commercial and consumer products containing these compounds (Hesse and Powers 1978; Neufeld et al. 1977). One of the significant sources of environmental contamination occurred as a result of the accidental mixup of FireMaster BP-6 with cattle feed in a number of farms in the lower peninsula in Michigan (see Section 5.2 for additional details concerning this incident). By June 1975, 412 farms had been quarantined. Disposal of contaminated feed, animal carcasses (poultry, cattle, swine), and animal products (meat, milk, eggs) contributed to environmental contamination (Dunckel 1975; Kay 1977).

***Polybrominated Diphenyl Ethers.*** The widespread use of PBDEs over the past 30 years has resulted in the presence of lower brominated diphenyl ether congeners, for example, 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), in the environment. Higher brominated congeners (e.g., octabromodiphenyl ether and decabromodiphenyl) tend to concentrate near point sources (Wania and Dugani 2002). The commercial production of PBDEs began in late 1970s (WHO 1994a). In 2001, the total market demand for PBDEs in the Americas was 33,100 metric tons (BSEF 2003). Technical decabromodiphenyl ether constituted about 24,500 metric tons (74%), while technical mixtures of octa- and pentabromodiphenyl ethers were 1,500 and 7,100 metric tons (4 and 22% of this total), respectively (BSEF 2003). PBDEs may be released into the environment from their manufacture and use in a wide range of consumer products (WHO 1994a). PBDEs are used as additive

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flame retardants in thermoplastics. Additive flame retardants are physically, rather than chemically, combined with polymers. Thus, there is a possibility that some PBDEs congeners (e.g., lower brominated congeners) may diffuse out of the treated materials to some extent (EU 2001). Waste from products containing PBDEs may be either incinerated as municipal waste, deposited in landfills, or discharged to municipal sewage-treatment plants (Darnerud et al. 2001). In the future, the disposal of plastic consumables containing PBDEs to landfills is likely to increase in the United States and elsewhere in the world.

The widespread use of pentaBDE over the past 30 years has resulted in the rapid increase in concentrations of lower brominated congeners, for example, 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), and their presence in the environment. No quantitative information was located on the releases of the commercial pentaBDE product from its production and use in the United States. The commercial pentaBDE product is used predominantly (95–98%) as an additive flame retardant in flexible polyurethane foam (FPUF), which has applications as cushioning in bed mattresses and upholstered products (see Section 7.3). No quantitative information was located on the releases of pentaBDE from the production of flexible polyurethane foams. However, the main source of release for liquid flame retardant additives (e.g., pentaBDE) is associated with the handling of the raw material (e.g., splashes and spills) prior to the foaming process, where releases are to waste water (EU 2001). There is a potential release to air during the curing phase since the polyurethane foam is heated at elevated temperatures (e.g., up to 160 °C) for several hours. Foam scrap will be disposed of to a landfill or possibly incinerated (EU 2001). Since pentaBDE is an additive flame retardant, it may be subject to volatilization or leaching from the polymer matrix during the lifetime of the use of an article. Losses of foam particles containing the substance (e.g., due to abrasion) may also occur. However, pentaBDE has a very low vapor pressure (see Table 6-6) and volatilization losses from polyurethane foam would be expected to be low. Given that the major use of pentaBDE is as a component of polyurethane foam for furniture/seating/automobile use, the potential for leaching of pentaBDE during use will be minimal. This is because, although it is likely that foam coverings may be washed during the lifetime of use, it is very unlikely that the foam cushioning containing pentaBDE will be washed as well (EU 2001). Residual 'waste' pentaBDE in the environment will be particles of polymer (foam) products that contain pentaBDE (e.g., 2,2',4,4'-tetraBDE). Polyurethane foam has an open cell structure, which presents a large surface area to the environment and therefore, the potential for release is likely to be greater than the hard dense plastics where octaBDE and decaBDE are used. In addition, this foam may become friable and crumble with age. Thus, small particles thereby released could move into the environment and disperse its components (i.e., commercial pentaBDE mixture) (BFRIP 2002). These foam particles are released to

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the urban/industrial soil compartment, but may also end up in sediment or air (EU 2001). Movement of polymer (foam) particles containing pentaBDE within the landfill could provide a transport mechanism leading to entry into leachate water or groundwater. However, well-designed landfills, in general, already include measures to minimize leaching and these measures would also be effective in preventing any leaching of pentaBDE as well (EU 2001).

Commercial octaBDE is used almost exclusively to flame retard acrylonitrile-butadiene-styrene (ABS) terpolymers used in computer casings and monitors (ENVIRON 2003b). No quantitative information was available on emissions of octaBDE from production operations. The major sources of air emissions are thought to be as a result of grinding and bagging operations. The emission to air of octaBDE vapor from production can be considered to be negligible (EU 2003a). The most likely way in which octaBDE may reach water from its production is due to washing out of equipment. The major source of octaBDE waste is from filter waste and reject material (EU 2003a). Much of the loss from polymer applications (e.g., acrylonitrile-butadiene-styrene) is likely to be in the form of dust. It is expected that much of this dust will be collected for reuse or disposed of to landfill/incineration. Some of this may end up in waste water as a result of cleaning floors and equipment (EU 2003a). Given that the major use of plastics containing octaBDE appears to be in electrical applications and that the substance has very low water solubility, the potential for leaching of octaBDE from the products during uses appears to be small. Waste remaining in the environment can be considered to be particles (or dust) of polymer product, or dust generated from polymer products, that contain octaBDE. These particles are primarily released to urban/industrial soil, but may also end up in sediment or air. Plastics containing octaBDE will usually be disposed of either to landfills or by incineration. No experiments have been carried out on the leachability of octaBDE from polymers in landfills. However, octaBDE is not expected to leach to a significant extent from polymers, unless the polymer itself undergoes some form of degradation, thus releasing octaBDE. It is expected that emissions of octaBDE from incineration processes will be near zero (EU 2003a).

Commercial decaBDE product is an additive flame retardant used in a variety of polymer applications (EU 2002). The major application for decaBDE is in high-impact polystyrene (HIPS), which is used in the television industry for cabinet backs. No quantitative information is available on emissions of decaBDE from production operations. Since the major use of plastics containing decaBDE is in electrical/electronic applications and the substance has very low water solubility, the potential for leaching of decaBDE from plastic products during use appears to be small (EU 2003a). A study of the leaching of decaBDE (DOW FR-300-BA; 77.4% deca-, 21.8% nona-, and 0.8% octaBDE) from pellets of acrylonitrile-butadiene-styrene (ABS) polymer and polystyrene (both containing 10% decaBDE) was

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undertaken. The lack of increase in bromine concentration with time was observed. In static extractions of ABS containing 4.25% decaBDE (Dow FR-300-BA) with water, acetic acid, and cottonseed oil at elevated temperatures, little or no leaching of the decaBDE was evident. No decaBDE was detected in the water and acetic acid and only 0.03% of the total decaBDE was extracted by cottonseed oil over 7 days at elevated temperatures (EU 2002).

Releases of decaBDEs are required to be reported under the Superfund Amendments and Reauthorization Act (SARA) Section 313; consequently, data are available for this compound in the Toxics Release Inventory (TRI) (EPA 1995). According to the TRI, a total of 649,541 pounds (294,627 kg) of decaBDE were released to the environment in 2001. In addition, an estimated 787,927 pounds (357,398 kg) were transferred off-site, including to publicly-owned treatment works (POTWs) (TRI01 2004). The TRI data should be used with caution because only certain types of facilities are required to report. This is not an exhaustive list. Facilities are only required to report to the TRI if they manufacture or process more than 25,000 pounds of a TRI listed chemical during the year, or otherwise use more than 10,000 pounds, and have the equivalent of more than 10 full-time employees. According to the EPA, TRI data have certain limitations. TRI data reflect releases and other waste management of chemicals and not exposures of the public to those chemicals. TRI data alone are not sufficient to determine exposure or calculate potential adverse effects on human health and the environment.

On-site land releases dominate decaBDE releases on-site from its manufacture and use, and are predominantly associated with disposal of decaBDE in either a manufacturer's on-site landfill in Arkansas or in a commercial chemical landfill in Louisiana. On-site air and water releases make up only a small fraction of total on-site releases. For example, on-site air emissions in 2000 were ~6.6% of the total; on-site surface water discharges also make up only a small fraction of the total (~0.2% of the 2000 total) and are nearly all associated with operations that apply decaBDE to upholstery textiles. DecaBDE releases off-site for disposal from its manufacture and use are typically larger than those on-site to land; decaBDE-waste transfers for further waste management are dominated by transfers to POTWs from the textile industry, although a significant rise in recycling has occurred in recent years. The plastics industry released minimal amounts to POTWs. The largest releases of decaBDE or decaBDE-waste to water occur in Maryland, North Carolina, and South Carolina and result from its use in textiles (TRI01 2004).



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**8.2.1 Air**

***Polybrominated Biphenyls.*** In the past, PBBs were released into the air during the manufacture of these compounds in three areas: through the vents of the hydrogen bromide recovery system, from the centrifugation area for recovering PBBs from slurries produced by bromination, and from the drying, pulverizing, and bagging area of the finished product (Di Carlo et al. 1978). An estimated 0.07 pounds/million pounds of the PBBs produced were lost from the hydrogen bromide-recovery vent (Di Carlo et al. 1978). No data are available for the air pollution factor (amount released/million pounds produced) at the centrifugation site. The concentrations of FireMaster BP-6 in the Michigan Chemical Corporation bagging area were 0.016–0.032 mg/L of air during the bagging operation and 0.003 mg/L of air after the completion of bagging (Di Carlo et al. 1978). In 1977, the maximum air losses of PBBs at production sites were estimated to total 1,125 pounds of PBBs for every 1 million pounds of PBBs produced (Di Carlo et al. 1978).

Another process that could release lower levels of brominated biphenyls in the air is the incineration of PBBs. Pyrolysis of hexabromobiphenyl in the absence and presence of air has produced small amounts of lower brominated biphenyls (Thoma and Hutzinger 1987). No data are available on the importance of this source for the release of PBBs in the air during the incineration of PBBs. However, since the vast majority of products containing PBBs are expected to be out of circulation after more than 25 years since the voluntary ban, incineration will not be a significant source of PBBs to air.

PBBs have been identified in 1 air sample, collected from 1,647 NPL hazardous waste sites, where they were detected in some environmental media (HazDat 2004).

***Polybrominated Diphenyl Ethers.*** The widespread use of pentaBDE technical mixtures over the past 30 years has resulted in the increasing concentrations of lower brominated congeners in air, e.g., 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99). No quantitative information was located on the releases of the pentaBPE technical mixtures to the atmosphere from its production and use. However, the release of pentaBDE technical mixtures to air has the potential to occur during the curing phase, since the polyurethane foam is at elevated temperatures (e.g., up to 160 °C) for several hours during this phase. Since pentaBDE technical mixtures are additive flame retardants, they may be 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 6-6) and therefore, losses from polyurethane foam due to volatilization would be

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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).

In the regions surrounding metal-recycling plants in Taiwan and Japan, a variety of tri-, tetra-, and hexabromodiphenyl ethers (BDEs) were measured in air (Watanabe et al. 1992). The concentrations in Taiwan and Japan were 23–53 and 7.1–21 pg/m<sup>3</sup>, respectively.

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 are thought to be a result of grinding and bagging operations. However, the emission to air of vapors from production of octaBDE technical mixtures may be considered to be negligible (EU 2003a). OctaBDE technical mixtures are used almost exclusively to flame retard ABS terpolymers used in computer casings and monitors (ENVIRON 2003b). No quantitative information was available on emissions of octaBDE technical mixtures to the atmosphere from ABS applications. However, much of the loss from polymer applications is likely to be in the form of dust. Losses of octaBDE technical mixture powders (particle sizes >40 µm) have been estimated as 0.21% during handling of raw materials. These losses will initially be to the atmosphere, but it is expected that this dust will rapidly settle and may be disposed of to landfills. OctaBDE technical mixtures in the compounding stage are also susceptible to dust generation. However, losses at this stage are thought to be lower than during the handling of raw materials (EU 2003a). Plastics containing octaBDE technical mixtures will usually be disposed of either to landfill or by incineration. It is expected that emissions of PBDE congeners resulting from incineration of plastics containing octaBDE technical mixtures will be negligible (EU 2003a).

No quantitative information is available on emissions of decaBDE technical mixtures to the atmosphere from production operations. In 1979, decaBDE was found in the atmosphere as particulate matter in the vicinity of plants manufacturing brominated flame retardants (Zweidinger et al. 1979); the concentration of decaBDE ranged from not detected to 72 ng/m<sup>3</sup>. The estimated releases of decaBDE vapor to air during production are low, typically  $1.1 \times 10^{-5}$  mg/metric tons of production (EU 2002). The major source of air emissions is thought to be a result of grinding and bagging operations. Losses of powders during the handling of raw materials will initially be to the atmosphere, but it is expected that this dust will rapidly settle within the production facility and therefore, these losses will be mainly to solid waste (EU

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2002). The compounding stage is also susceptible to dust generation, but losses are thought to be lower than during handling of raw materials (i.e., an order of magnitude lower). Releases will again be initially to the atmosphere, but the particles would be expected to settle within the compounding facility and will ultimately be to solid waste or waste water. In addition to particulates, emissions to the atmosphere due to the volatility of the flame retardant at the elevated processing temperatures may be possible. However, due to the high vapor pressure of decaBDE (see Table 6-6), volatilization is expected to be negligible.

The commercial decaBDE product is an additive flame retardant used in a variety of polymer applications (EU 2002). The major application for decaBDE is in high-impact polystyrene (HIPS), which is used in the television industry for cabinet backs. Thus, decaBDE may be released during the polymer processing of HIPS. Waste remaining in the environment can be considered to be small particles of polymer product or dust generated from polymer products containing decaBDE (EU 2002). These particles are primarily released to soil, but may also end up in air. End products with outdoor uses are most likely to be atmospheric sources of decaBDE dust. Releases from these products occur over the lifetime of the product due to weathering and wear. Waste of this type may also be generated during disposal of a variety of plastic articles, in particular, where these articles are dismantled or subject to other mechanical processes. Ultimately, plastic articles containing technical decaBDE will be disposed of either to landfill or by incineration. Emissions of decaBDE from controlled incineration processes will be near zero (EU 2002). The concentration of decaBDE was determined in bottom ash from two municipal incinerators in Finland. With a method detection limit of 0.02–0.06 µg/kg, decaBDE was not detected in either ash sample (EU 2002). The low vapor pressure of decaBDE (see Table 6-6) limits its volatility to the atmosphere when disposed of in a landfill (EU 2002).

The estimated release of 97,198 pounds (44,088 kg) of decaBDE to the atmosphere from 54 manufacturing, processing, and waste disposal facilities in 2001 accounted for about 15.0% of the estimated total on-site environmental releases (TRI01 2004). These releases are summarized in Table 8-1. The data from the TRI listed in Table 8-1 should be used with caution, however, since only certain types of facilities are required to report (EPA 1995). This is not a comprehensive list.

DecaBDE was not identified in air samples collected from 1,647 NPL hazardous waste sites (HazDat 2004).

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**Table 8-1. Releases to the Environment from Facilities that Produce, Process, or Use Decabromodiphenyl Ether<sup>a</sup>**

State <sup>c</sup>	Number of facilities	Reported amounts released in pounds per year <sup>b</sup>						
		Air <sup>d</sup>	Water	Under-ground injection	Land	Total on-site release <sup>e</sup>	Total off-site release <sup>f</sup>	Total on and off-site release
AL	1	10	5	0	0	15	0	15
AR	5	81,717	0	0	130,000	211,717	9,902	221,619
CA	5	750	0	0	0	750	100,023	100,773
CT	3	1,024	No data	0	0	1,024	24,149	25,173
FL	2	0	No data	0	0	0	19,040	19,040
GA	7	755	0	0	0	755	11,225	11,980
IL	2	32	No data	0	0	32	13,700	13,732
IN	6	20	0	0	0	20	3,987	4,007
KY	2	555	No data	0	0	555	17,478	18,033
LA	1	0	No data	0	270,000	270,000	0	270,000
MA	13	292	8	0	0	300	30,658	30,958
MD	1	0	No data	0	0	0	0	0
MI	6	3,741	0	0	23,600	27,341	26,503	53,844
MN	3	0	No data	0	0	0	8,507	8,507
MO	1	500	No data	0	0	500	1,189	1,689
MS	2	106	No data	0	0	106	30,933	31,039
NC	14	2,818	4,237	0	99,090	106,145	73,151	179,296
NE	2	0	No data	0	0	0	0	0
NH	1	2	5	0	0	7	285	292
NJ	5	97	No data	0	0	97	84,846	84,943
NV	1	No data	No data	No data	No data	No data	No data	0
NY	4	0	5	0	990	995	36,837	37,832
OH	9	322	0	0	750	1,072	77,277	78,349
PA	7	1,996	109	0	0	2,105	156,439	158,544
RI	1	13	No data	0	0	13	1,766	1,779
SC	12	0	2,055	0	40	2,095	20,758	22,853
TN	7	786	42	0	18,137	18,965	13,644	32,609
TX	8	1,390	5	0	3,265	4,660	5,712	10,372
VA	5	270	No data	0	0	270	15,087	15,357
VT	1	0	No data	0	0	0	0	0

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**Table 8-1. Releases to the Environment from Facilities that Produce, Process, or Use Decabromodiphenyl Ether<sup>a</sup>**

State <sup>c</sup>	Number of facilities	Reported amounts released in pounds per year <sup>b</sup>						Total on and off-site release
		Air <sup>d</sup>	Water	Under-ground injection	Land	Total on-site release <sup>e</sup>	Total off-site release <sup>f</sup>	
WA	1	0	No data	0	0	0	0	0
WI	3	2	No data	0	0	2	4,831	4,833
Total	141	97,198	6,471	0	545,872	649,541	787,927	1,437,468

Source: TRI01 2004

<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>The sum of fugitive and stack releases are included in releases to air by a given facility.

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

<sup>f</sup>Total amount of chemical transferred off-site, including to publicly owned treatment works (POTW).

## 8. POTENTIAL FOR HUMAN EXPOSURE

**8.2.2 Water**

***Polybrominated Biphenyls.*** In the past, PBBs were released to water during the manufacturing process. An estimated 0.0046 pounds were lost to sewers for every 1,000,000 pounds of PBBs produced at manufacturing sites (Neufeld et al. 1977). To manufacture PBBs, water was added to the reaction mixture when the desired extent of bromination was achieved. Ultimately, this water was discharged as effluent into surface water. Samples of effluents from the Michigan Chemical Corporation contained PBB concentrations 503 ppm (Di Carlo et al. 1978). Runoff water from the manufacturing plants containing PBBs also contaminated surface water (Di Carlo et al. 1978). Landfill sites used to dispose of wastes from PBB production can also be a source of PBBs in water. Concentrations of PBBs in groundwater from one such landfill in St. Louis, Michigan were low (0.1–0.2 ppb), but those in water from a drainage ditch and catch basin were much higher (0.35–1.2 ppm) (Di Carlo et al. 1978).

PBBs have been identified in 2 and 5 surface water and groundwater samples, respectively, collected from 1,647 NPL hazardous waste sites (HazDat 2004).

***Polybrominated Diphenyl Ethers.*** Industrial and urban effluents are significant 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 soil and stream sediments collected near a polyurethane manufacturing plant (near the Dan River, Virginia). Summed concentrations of 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE (BDE 99), and 2,2',4,4',6-pentaBDE (BDE 100), the dominant congeners in these samples, ranged from <1 to 132 µg/kg (ng/g) dry weight. In 1995, sediment samples were collected up- and downstream near an area where the Swedish plastics industry uses brominated flame retardants (Sellström and Jansson 1995; Sellström et al. 1998a). Samples were analyzed for tetraBDEs (50 ng/g dry weight) and pentaBDEs (sum of three congeners, 2,300 ng/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 levels 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 ng/g (µg/kg) dry weight; the concentration of BDE 209 ranged from not detected to 16,000 ng/g (µg/kg) dry weight. Allchin et al. (1999) surveyed the

## 8. POTENTIAL FOR HUMAN EXPOSURE

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 2,3',4',6-tetrabromodiphenyl ether or BDE 71), and octaBDE (as 3,3',4,5'-tetrabromodiphenyl ether or BDE 79) were in sediments near or downstream from a manufacturing site at Newton Aycliffe in River Skerne. The highest concentrations of decaBDE (as 2,2',3,3',5-pentabromodiphenyl ether or 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 [i.e., 2,2',4,4',5,5'-hexabromodiphenyl ether], and BDE 209) ranged from 0.07 to 10.6 ng/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.

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 or groundwater. However, 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, 2003).

The estimated release of 6,471 pounds (2,935 kg) of decaBDE to water from nine domestic manufacturing and processing facilities in 2001 accounted for about 0.45% of the estimated total environmental releases (TRI01 2004). An additional 787,927 pounds (357,398 kg) were transferred off-site, including to POTWs (TRI01 2004). These releases are summarized in Table 8-1. The data from the TRI listed in Table 8-1 should be used with caution, however, since only certain types of facilities are required to report (EPA 1995). This is not a comprehensive list.

PBDEs were not identified in water samples collected from 1,647 NPL hazardous waste sites (HazDat 2004).

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**8.2.3 Soil**

***Polybrominated Biphenyls.*** The important former sources of PBBs in soil are manufacturing operations, disposal of PBB-containing finished products, and agricultural operations contaminated in the original episode in 1973–1974. The concentrations of PBBs in soils from bagging and loading areas of the Michigan Chemical Corporation were 3,500 and 2,500 mg/kg, respectively (Di Carlo et al. 1978). Similarly, soil from sites adjacent to the Hexcel Corp and the White Chemical Company, the manufacturers of octabromo- and decabromobiphenyl, contained decabromobiphenyl and other lower brominated biphenyls down to hexabromobiphenyl (Di Carlo et al. 1978). The disposal into landfills of solid wastes generated during the production of PBBs was another important source of PBBs in soil (Neufeld et al. 1977). Photodecomposition of FireMaster BP-6 in soil could also be a source of lower brominated biphenyls (Ruzo and Zabik 1975; Trotter 1977) in soil.

Approximately 11.8 million pounds (5,350,000 kg) of hexabromobiphenyl was used in commercial and consumer products in the United States, mostly in the production of plastic products. Since the cessation of production of hexabromobiphenyl, all of these products, such as TV cabinet and business-machine housings, with a usable life of 5–10 years must have been disposed of by landfilling or incineration (Neufeld et al. 1977). Disposal of these plastic materials in waste-disposal sites is an important source of PBBs in soil. The migration of plastic-incorporated PBBs to soil would be very low since PBBs would be tightly bound into the plastic (Neufeld et al. 1977).

The indirect source of PBBs in soil was the contaminated farms in Michigan. Approximately 650 pounds (290 kg) of PBBs was mixed in cattle feeds that were delivered to Michigan farms during 1973–1974 (Fries 1985b). About 50% of this amount was excreted in the feces of the exposed animals and remained on the farms in places of fecal deposition and manure disposal (Fries 1985b). Soil in fields that received contaminated manure contained as high as 300 µg/kg PBBs, whereas soil in resurfaced cattle-exercise lots contained as high as 1,000–2,000 µg/kg of PBBs (Fries 1985b).

PBBs have been identified in 5 soil and 3 sediment samples collected from 1,647 NPL hazardous waste sites (HazDat 2004).

***Polybrominated Diphenyl Ethers.*** 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. Solid waste from commercial production of octaBDE is typically disposed in landfills (EPA 1995). The disposal of consumer products containing PBDEs is likely to increase worldwide due to rapid



## 8. POTENTIAL FOR HUMAN EXPOSURE

obsolescence of plastic products. For example, between 1997 and 2004, the number of obsolete computers containing flame retardants is projected to be 315 million (NSC 1999). Based on a monitor weight of 30 pounds (14 kg), an estimated 350 million pounds (160,000,000 kg) of brominated flame retardants will be released to landfills (NSC 1999). Although PBDEs will only be a fraction of this total, the amount of PBDEs released to the environment by disposal will still be significant.

PBDEs are released to farmland with their disposal as biosolids (i.e., sewage sludge). PBDEs were detected in biosolids destined for land applications in four different regions of the United States (Pardini et al. 2001). The total concentrations of pentaBDE in biosolids ranged from 1,100 to 2,290 µg/kg dry weight; the levels of pentaBDE were high and consistent, regardless of the region of origin. 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 µg/kg dry weight in the biosolid samples.

In 2001, 545,872 pounds (247,603 kg) of decaBDE was released to land from 8 domestic manufacturing, processing, and waste-disposal facilities reporting releases of the compound to the environment (TRI01 2004). No releases (0 pounds) of decaBDE occurred via underground injection (TRI01 2004). Releases to the environment from facilities that produce, process, or use decaBDE are summarized in Table 8-1. The data from the TRI should be used with caution since only certain types of facilities are required to report (EPA 1995). This is not a comprehensive list.

PBDEs were not identified in soil and sediment samples collected from 1,647 NPL hazardous waste sites (HazDat 2004).

### 8.3 ENVIRONMENTAL FATE

#### 8.3.1 Transport and Partitioning

***Polybrominated Biphenyls.*** PBBs exist predominantly in the particulate phase in the atmosphere. Particulate phase PBBs are removed from the atmosphere by wet and dry deposition and should not travel long distances in the environment. In water, PBBs are expected to adsorb strongly to suspended solids and sediment, and may bioconcentrate in aquatic organisms. The volatilization of PBBs from water to air is not expected to be important due to attenuation by adsorption in the water column. In soil, PBBs are adsorbed strongly and will be immobile. Volatilization of PBBs from soil to air is not important due to the low volatility of PBBs and strong adsorption of PBBs to soil.

## 8. POTENTIAL FOR HUMAN EXPOSURE

Organic compounds with vapor pressures  $>10^{-4}$  mm Hg should exist almost entirely in the vapor phase in the atmosphere, while organic compounds with vapor pressures  $<10^{-8}$  mmHg should exist almost entirely in the particulate phase (Eisenreich et al. 1981). The estimated vapor pressure of FireMaster BP-6 is  $5.2 \times 10^{-8}$  mm Hg at 25 °C (Jacobs et al. 1976). The vapor pressure of octabromobiphenyl is  $7.0 \times 10^{-11}$  mm Hg at 28 °C (Waritz et al. 1977). Although no data are available, the vapor pressures of decabromobiphenyl at ambient temperatures should be lower than octabromobiphenyl. Thus, PBBs produced in the 1970s should exist predominantly in the particulate phase in the atmosphere. Since the particulate phase PBBs would precipitate out by dry deposition and wet deposition due to washout (Atlas and Giam 1987), PBBs would not be expected to be transported long distances in the atmosphere.

There are limited data regarding the transport and partitioning of PBBs in water. Based on an estimated Henry's law constant of  $3.9 \times 10^{-6}$  atm-m<sup>3</sup>/mol (where Henry's law constant = vapor pressure/water solubility) and an estimation method (Thomas 1990), the estimated volatilization half-life of hexabromobiphenyl is 23 days. Therefore, the transport of PBBs from water to the atmosphere by volatilization is not expected to be important. This is consistent with a fish bioconcentration study in which losses of octabromobiphenyl and decabromobiphenyl from water to air were found to be insignificant (Norris et al. 1973). Soil-mobility studies have shown that PBBs are strongly adsorbed by soil materials (Filonow et al. 1976; Griffin and Chou 1981a, 1981b). Therefore, sorption of water-bound PBBs to particulate matter and sediment is a major transport process for PBBs in water. The detection of at least a 1,000-fold higher concentration of PBBs in Pine River sediment (where effluent from Michigan Chemical Corporation was discharged) compared with the level of PBBs in the river water confirms the importance of this transport process (Hesse and Powers 1978).

PBBs may also be transported from water to aquatic organisms in which bioconcentration may take place. Data from different laboratories on the bioconcentration of PBBs in fish show wide variation. The experimentally determined bioconcentration factor (BCF; the BCF is the concentration of the chemical in fish tissues over concentration of chemical in water) for hexabromobiphenyl (mixtures of unspecified congeners) in the whole body of fathead minnows (*Pimephales promelas*) was 18,100 in a 32-day exposure (Veith et al. 1979). In fillet of fathead minnow, the estimated BCF was  $>10,000$  (Hesse and Powers 1978). The lipid weight-based BCF values of 4,4'-dibromobiphenyl, 2,4,6-tribromobiphenyl, 2,2',5,5'-tetrabromobiphenyl, and 2,2',4,4',6,6'-hexabromobiphenyl in guppies (*Poecilia reticulata*) were 269,000; 115,000; 1,440,000; and 708,000; respectively (Gobas et al. 1989). BCF values for mono- to tetra- bromobiphenyl congeners tend to increase with higher degrees of bromination while BCF values for tetra- and higher congeners tend to decrease with higher degrees of bromination. A similar trend in BCF

## 8. POTENTIAL FOR HUMAN EXPOSURE

values for various PBBs was also observed in juvenile Atlantic salmon (*Salmo salar*). For example, the whole body BCF values determined for 2,6-dibromobiphenyl, 2,4-dibromobiphenyl, 3,4-dibromobiphenyl, 2,5,4'-tribromobiphenyl, 2,2',4,5'-tetrabromobiphenyl, 2,3',4',5-tetrabromobiphenyl, hexabromobiphenyl (unspecified congener), and octabromobiphenyl were 1,267, 1,343, 63, 425, 314, 111, 2–48, and 0.02, respectively (Zitko 1979; Zitko and Hutzinger 1976). The BCF values determined for 2,2',3,3',4,4'-hexabromobiphenyl and decabromobiphenyl in whole body guppies (*P. reticulata*) were 10 and 0, respectively (Opperhuizen et al. 1985). The BCF value for octabromobiphenyl in filleted rainbow trout (*Salmo gairdneri*) was 0 (Norris et al. 1973). The lack of accumulation for the higher brominated compounds is most likely because they have very limited water solubility and are therefore not available to penetrate membranes (Zitko 1979).

PBBs are adsorbed strongly to soil, and the adsorption increases with an increase in the organic carbon content of soil (Filonow et al. 1976; Griffin and Chou 1981a, 1981b). Neither clay content nor pH of soil correlated with adsorption of hexabromobiphenyl to soil (Filonow et al. 1976). PBBs present in soil-water solution will partition to the soil solids by adsorption. The presence of certain types of dissolved organic carbon in natural water (e.g., leachate from a landfill) may decrease the adsorption of PBBs in sediments (Simmons and Kotz 1982). Because of the strong adsorption, PBBs will have low mobility in soil, and the leaching of PBBs from soil to groundwater will generally be insignificant (Filonow et al. 1976; Griffin and Chou 1981a, 1981b). However, the mobility of PBBs may greatly increase if methanol or other organic solvents (capable of solubilizing PBBs) are present at significant concentrations in soil as would happen at some contaminated sites (Griffin and Chou 1981b). This phenomenon is commonly called “co-solvency.” The transport of PBBs from soil to the atmosphere by volatilization is not important due to the low volatility and strong adsorption of PBBs (Jacobs et al. 1976). The transport of PBBs from soil to surface water or another land area via eroded soil contained in runoff water is possible (Jacobs et al. 1976). Orchard grass and tops of carrots grown in soil contaminated with PBBs showed no uptake, and carrot roots showed only minor uptake of PBBs (Jacobs et al. 1976, 1978). Therefore, the transport of PBBs from soil to plants via translocation is insignificant.

***Polybrominated Diphenyl Ethers.*** In air, PBDE commercial mixtures, which have low vapor pressures and exist in the particulate phase, will be removed from the atmosphere by wet and dry deposition. Thus, in general, PBDEs are not expected to travel long distances in the environment. However, some congeners in pentaBDE commercial mixtures, 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), have been found in Arctic regions. It has not been definitively explained how these lower brominated congeners have been transported such long distances

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from areas of emission. However, it is likely that they have been transported to remote regions as dust particles rather than in the vapor phase. 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 not expected to be important due to attenuation 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 not important due to the low volatility of PBDEs and strong adsorption of PBDEs to soil.

PBDEs with vapor pressures between  $10^{-4}$  and  $10^{-8}$  mm Hg (di- to hexa- bromodiphenyl ether) should exist in both the vapor and particulate phase in the atmosphere, while PBDEs with vapor pressures  $<10^{-8}$  mm Hg (hexa- to decaBDE) should exist almost entirely in the particulate phase in the atmosphere (Bidleman 1988; Eisenreich et al. 1981). PBDEs have low vapor pressures, with vapor pressure tending to decrease with increasing bromination. Watanabe and Tatsukawa (1990) determined the vapor pressures for a range of brominated PBDEs as follows (mm Hg at 25 °C): di- ( $9.8 \times 10^{-5}$ – $1.4 \times 10^{-4}$ ); tri- ( $1.2 \times 10^{-5}$ – $2.0 \times 10^{-5}$ ); tetra- ( $1.8 \times 10^{-6}$ – $2.5 \times 10^{-6}$ ); penta- ( $2.2 \times 10^{-7}$ – $5.5 \times 10^{-7}$ ); hexa- ( $3.2 \times 10^{-8}$ – $7.1 \times 10^{-8}$ ); and octa- ( $9.0 \times 10^{-10}$ – $1.7 \times 10^{-9}$ ). Vapor pressures have also been determined for commercial PBDE mixtures, such as pentaBDE ( $2.2 \times 10^{-7}$ – $5.5 \times 10^{-7}$  mm Hg) and decaBDE ( $3.2 \times 10^{-8}$  mm Hg at 25 °C) (EU 2001; NRC 2000). Since particulate phase PBDEs will precipitate out by wet and dry deposition, these PBDEs would not be expected to be transported long distances in the atmosphere (Atlas and Giam 1987). Thus, highly brominated PBDEs (e.g., octa- through decaBDEs), which have low vapor pressures and exist solely in the particulate phase, will not be transported long distances. Although no information was located in the literature, moderately brominated PBDEs (e.g., pentaBDEs and hexaBDEs) may have the potential to be transported and would likely be found at higher concentration close to PBDE point sources. Lower BDE homologs (e.g., tetraBDE), which exist partially in the vapor phase, have the potential for long-range transport in the atmosphere (Dodder et al. 2000a). For example, 2,2',4,4'-tetrabromodiphenyl ether (BDE47), 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), and 2,2',4,4',5,5'-hexabromodiphenyl ether (BDE 153) were detected in air samples from urban, rural, and remote areas of the Great Lakes region in the United States (Dodder et al. 2000a).

Concentrations of PBDEs in water are expected to be low due to the low water solubility of PBDEs. For example, the solubilities of the commercial mixtures of pentaBDE and decaBDE are 13.3 and  $<0.1$  µg/L, respectively (EU 2001; Hardy 2002b). As consequence of their low water solubility, they have not detected PBDEs in environmental waters (see Section 8.4.2).

## 8. POTENTIAL FOR HUMAN EXPOSURE

PBDEs adsorb strongly onto suspended solids and sediments in the water column. Volatilization of PBDEs from water surfaces will be attenuated by adsorption, and thus is 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. High log  $K_{ow}$  values have been measured for PBDEs as follows (Watanabe and Tatsukawa 1990): di- (5.03); tri- (5.47–5.58); tetra- (5.87–6.16); penta- (6.46–6.97); hexa- (6.86–7.92); octa- (8.35–8.90); and deca- (9.97). Using these log  $K_{ow}$  values, 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. The reported BCFs for these commercial mixtures are typically less than 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 bioconcentration factor of <4 after 8 weeks of exposure (WHO 1994a). A bioconcentration study was carried out with rainbow trout under static conditions. The concentration in the water was 20  $\mu\text{g}$   $^{14}\text{C}$ -decaBDE per liter. 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).

An abundance of monitoring data illustrates the uptake of lower brominated diphenyl ethers by aquatic organisms, which results in bioconcentration (see Section 8.4.4). The commercial pentaBDE product undergoes bioconcentration with a BCF of approximately 14,000 (Hardy 2002b). Congener components of the pentaBDE commercial product bioconcentrate to different extents. For example, approximately 50–70% of PBDEs detected in fish is a single isomer (2,2',4,4'-tetrabromodiphenyl ether [BDE 47]). The next most prominent isomer is typically 2,2',4,4',5-pentabromodiphenyl ether (BDE 99) followed by 2,2',4,4',6-pentabromodiphenyl ether (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 2,2',4,4',5,5'-hexabromodiphenyl ether (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, and  $\approx 2.5 \times 10^7$  for BDE 28,  $\approx 2.5 \times 10^8$  for BDE 47, and  $\approx 1.6 \times 10^8$  for BDE 153.

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Bioaccumulation of PBDEs in the aquatic food web is inversely related to the degree of bromination (Burreau et al. 2000; Jansson et al. 1993). Higher brominated congeners (e.g., decaBDE) are rarely detected in biota. This is a result of their low solubility, high log  $K_{ow}$  values, and sorption to soil and sediment (Hardy 2000). In contrast, tetraBDE to hexaBDE homologs are more frequently detected in biota (Burreau et al. 1997; Hale et al. 2003), which would be expected due to their greater water solubility and relatively high  $K_{ow}$  values.

Concentrations of lower brominated diphenyl ethers in biota are related to the trophic level of the species. For example, Haglund et al. (1997) examined the concentrations of tetra- to hexa- bromodiphenyl ethers 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 tetrabromodiphenyl ethers (e.g., 2,2',4,4'-tetrabromodiphenyl ether, 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). Biomagnification factors for guillemot egg versus herring were 19, 17, and 7.1 for BDE 47, 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), and 2,2',4,4',6-pentabromodiphenyl ether (BDE 100), respectively. Burreau et al. (2000) analyzed small herring and salmon from the Atlantic Ocean (near Iceland) for several PBDEs. The 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.

PBDEs will be strongly adsorbed to soils based on log  $K_{ow}$  values ranging from 5.03 to 9.97 (Watanabe and Tatsukawa 1990). Thus, PBDEs present in soil-pore water will bind to soil organic matter. Because PBDEs adsorb strongly to soil, they will have very low mobility (Swann et al. 1983), and leaching of PBDEs from soil to groundwater will be insignificant. Like PBBs, the presence of dissolved organic carbon in natural water may increase the mobility of PBDEs. The transport of PBDEs from soil to surface water via eroded soil contained in runoff water is also possible. Volatilization of PBDEs from moist soil surfaces will be attenuated by adsorption and is not expected to be an important fate process. Volatilization of PBDEs from dry soil will not be important due to the low volatility of PBDEs (see Table 6-6).

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**8.3.2 Transformation and Degradation**

Photolysis appears to be the dominant transformation process for PBBs and 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 either PBBs or PBDEs.

**8.3.2.1 Air**

***Polybrominated Biphenyls.*** In air, the two processes that may result in significant degradation or transformation of PBBs are photooxidation by hydroxyl (OH) radicals and direct photolysis. The estimated half-life of pentachlorobiphenyl in air due to reaction with hydroxyl radicals is 41.6–83.2 days (Atkinson 1987a). Based on a structure-activity relationship for the estimation of half-lives for the gas-phase reactions of hydroxyl radicals with organic compounds (Atkinson 1987b), the estimated half-lives of hexabromobiphenyl and decabromobiphenyl due to reaction with OH radicals are 182 and 2,448 days, respectively. These half-lives are consistent with the half-life of pentachlorobiphenyl due to reaction with OH radicals. However, the half-lives of brominated biphenyls expected to be present in the particulate phase in the air may be even longer than the estimated half-lives due to gas phase reaction. Therefore, the transformation of the hexa- and other higher brominated PBBs in the atmosphere due to reaction with OH radicals are probably not important.

Hexa- and other higher brominated biphenyls are expected to be present in the particle-adsorbed state in the atmosphere. These PBBs photolyze in solution and in soil (Hill et al. 1982; Ruzo and Zabik 1975; Trotter 1977). Since PBBs present in surface soil are known to photolyze, particle-sorbed PBBs present in the atmosphere may also undergo photolysis. The importance of the photochemical reaction under sunlight illumination conditions for the degradation/transformation of PBBs in air cannot be evaluated due the lack of information.

***Polybrominated Diphenyl Ethers.*** 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 deca- bromodiphenyl ether 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

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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 calculated for the gas-phase reaction. Thus, for the higher brominated PBDEs (e.g., octa- and decaBDEs), indirect photolysis with hydroxyl radicals will not be 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., decabromodiphenyl ether) 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.

### 8.3.2.2 Water

***Polybrominated Biphenyls.*** The photolytic degradation of PBBs in solution has been the subject of several studies. Available data in the literature indicate that brominated biphenyls photodegrade by reduction in solvents capable of proton transfer with the formation of lower brominated biphenyls. For example, the irradiation of FireMaster BP-6 and 2,2',4,4',5,5'-hexabromobiphenyl in methanol at wavelengths >286 nm produced mainly penta- and tetrabromobiphenyl (Ruzo and Zabik 1975). FireMaster BP-6 photolyzed 7 times faster than its chlorinated counterpart, 2,2',4,4',5,5'-hexachlorobiphenyl (Ruzo and Zabik 1975). Although an earlier study tentatively identified dimethoxy tetrabromobiphenyl as a photolysis product of FireMaster BP-6 (Ruzo and Zabik 1975), later work did not detect this compound (Ruzo et al. 1976). Earlier studies indicated that the debromination usually occurs with the stepwise preferential loss of bromine from the *ortho* and *para* positions of the biphenyl ring (i.e., 2, 2', 6, and 6' positions) (De Kok et al. 1977; Ruzo and Zabik 1975; Ruzo et al. 1976; Trotter 1977). Thus, the photolysis of 2,2',4,4',5,5'-hexachlorobiphenyl, the major component of FireMaster BP-6, would be expected to produce 2,3',4,4',5-pentabromobiphenyl and subsequently 3,3',4,4'-tetrabromobiphenyl. More recent work indicates that although photolysis mainly produces debromination products, unlike in the case of an individual PBB congener, reductive debromination of *ortho* substituents is not the predominant photolytic degradation pathway for FireMaster BP-6 (Robertson et al. 1983b).

The study of photolysis of PBBs in the aqueous phase is more relevant to natural environmental situations than photolysis in proton-donating organic solvents. It was suggested that the photolysis of PBBs in aqueous solution would proceed by oxidative process of photohydroxylation, leading to the formation of phenolic compounds (Norris et al. 1973). However, photolysis of 2,4-dibromo- and 2,3',4',5-tetrabromobiphenyl in acetonitrile-water solution showed that debromination was the major reaction (Ruzo et al.



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1976). No evidence of the formation of hydroxylated species (phenolic products) was found (Ruzo et al. 1976).

PBBs are not expected to undergo abiotic hydrolysis under environmental conditions due to the lack of hydrolysable functional groups (Boethling and Mackay 2000).

Several investigators assessed the biodegradation potential of PBBs under aerobic conditions, with activated sludge or pure cultures of microorganisms as microbial inoculum, and concluded that although the lower substituted biphenyls might biodegrade in aerobic water and sediment (Kong and Sayler 1983; Sugiura 1992; Yagi and Sudo 1980), the higher substituted biphenyls are resistant to aerobic biodegradation (Kawasaki 1980; Sasaki 1978; Shelton and Tiedje 1981). This is consistent with biodegradation studies in soil (see Section 8.3.2.3). It has been proposed that complete mineralization of 4-bromobiphenyl to carbon dioxide occurs via a 4-bromobenzoate intermediate by mixed bacterial cultures obtained from PBB-contaminated river sediment (Kong and Sayler 1983). However, complete mineralization was not observed for 2- and 3-bromobenzoate (Kong and Sayler 1984).

Although higher brominated biphenyls do not biodegrade in water or sediment under aerobic conditions, it has been shown that anaerobic microorganisms in river sediments obtained from populated areas can biodegrade higher substituted PBBs, including FireMaster mixtures (Morris et al. 1992). The biodegradation involved debromination at the *meta* and *para* positions, and no *ortho* bromine removal was observed (Morris et al. 1992). However, the possibility of *ortho* bromine removal from higher brominated biphenyls with certain inoculations (e.g., microorganisms from polluted river sediment repeatedly transferred on a pyruvate medium amended with Aroclor 1242) has been suggested (Morris et al. 1992).

***Polybrominated Diphenyl Ethers.*** 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 tetrabromodiphenyl ethers 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

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successively debrominated by 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 octabromodiphenyl ethers) was recently reported in a 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 octabromodiphenyl ethers 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) recently reported that 4,4'-dibromodiphenyl (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 environment. Organic solvents can act as hydrogen donors in photolysis reactions, which will potentially affect the distribution of products formed.

In a recent study, 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 high-performance liquid chromatography (HPLC) with ultraviolet detection (UV) detection or by gas chromatography-mass spectrometry (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., 2,2',4,4',6,6'-hexabromodiphenyl ether [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,

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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 nonabromodiphenyl ethers). 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 octabromodiphenyl ether 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 nonabromodiphenyl ethers under somewhat environmentally relevant conditions. Lower brominated diphenyl ether congeners are also formed although only to a minor extent. These tests do not provide evidence that lower brominated diphenyl ethers (e.g., tetra- and pentabromodiphenyl ethers) 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 tetra- BDEs were formed as well as some PBDFs. The half-lives in toluene and on silica gel were less than 15 minutes, and half-lives on other matrixes ranged from 40 to 200 hours. No differences were observed in the debromination patterns under different matrixes 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., 2,2',4,4'-tetrabromodiphenyl ether

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[BDE 47], 2,2',4,4',5-pentabromodiphenyl ether [BDE 99], and 2,2',4,4',6-pentabromodiphenyl ether [BDE 100]) were only formed to a minor degree (Söderström et al. 2004).

Following the methodology described for decaBDE, photolysis experiments were conducted on 2,2',4,4'-tetraBDE (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'-tribromodiphenyl ether 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 pentabromodiphenyl ether 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 (Boethling and Mackay 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 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 OCED 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 2,2',4,4'-tetrabromodiphenyl ether

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(BDE 47) using a mixture of  $^{14}\text{C}$  labeled and unlabeled compound (EU 2003a). The test was carried out using a sediment-water (Schuylkill River, PA) 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) recently reported that 4,4'-dibromodiphenyl undergoes reductive debromination under anaerobic conditions. Debromination proceeds with replacement of a bromine (Br) atom by a hydrogen (H) atom. The authors suggest that anaerobic debromination may sequentially debrominate BDE 15 to the parent diphenyl ether.

The anaerobic biodegradability of  $^{14}\text{C}$ -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  $^{14}\text{CO}_2$  and  $^{14}\text{CH}_4$  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).

### 8.3.2.3 Sediment and Soil

***Polybrominated Biphenyls.*** Information on the fate of PBBs in soil is limited. A pure culture of microorganism isolated from soil biodegraded 2-bromobiphenyl via the 2-bromobenzoic acid pathway (Takase et al. 1986). There is little evidence that the higher brominated biphenyls biodegrade in soil under aerobic conditions during an incubation period of  $\leq 1$  year (Griffin and Chou 1981a, 1981b; Jacobs et al. 1976). Some degradation of an undefined congener of pentabromobiphenyl was observed when incubated in soil, but this degradation could not be definitely attributed to biodegradation (Jacobs et al. 1976). As discussed in Section 8.3.2.2, higher brominated PBBs may biodegrade in an anaerobic region of river sediment and possibly soil polluted with PCBs and PBBs to form lower brominated products. Biodegradation of the photolysis products of hexa- and heptabromobiphenyl in soil (which produces lower brominated products) was only minor ( $\approx 3\%$  in 1 year) since the photodegradation products were bound to soil and light does not penetrate far into soil (Jacobs et al. 1978)

Degradation of PBBs present in a contaminated soil from a manufacturing site in Michigan was significant (Hill et al. 1982). For example, 2,2',4,4',5,5'-hexabromobiphenyl, the principal component of

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FireMaster (54–68% in FireMaster) was reduced to 26% of the total PBBs when exposed to a field soil for several years. In two other soils, in which the original concentrations of PBBs were much lower, the rate of degradation was much lower. Principal degradation products were 2,3',4,4',5-pentabromobiphenyl, 2,2',4,4',5-pentabromobiphenyl, and two unidentified tetrabromobiphenyls. The degradation was attributed to photochemical reactions. On the other hand, no significant photodegradation of FireMaster was observed after 1 year in contaminated manure spread in field soil from Michigan (Jacobs et al. 1978). The authors provided no explanation for the difference in photoreactivity of PBBs in soils with and without manure. It is important to point out that, due to attenuation and scattering of light, sunlight will not penetrate most soil beyond the surface layer. Therefore, it can be concluded from these studies that although photolysis may be the only viable degradative process for PBBs in soil, photolysis will be limited to the surface layer of soil, and the rate of photolysis will be very slow. PBBs incorporated into thermoplastics which were eventually buried at waste sites are not likely to absorb much light and undergo photolytic degradation.

Analysis (Morris et al. 1993) of sediments from a PBB-contaminated river in Michigan (Pine River) indicates that little degradation of PBBs has occurred since the 1970s. Although microorganisms capable of debrominating PBBs were not present in regions of highest contamination, they were found in sediments downstream from the area of highest contamination. The investigators (Morris et al. 1993) suggest that high levels of contaminants including PBBs may be inhibiting the microbial degradation of PBBs in this river.

***Polybrominated Diphenyl Ethers.*** 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 are unlikely to biodegrade in soils or sediment under aerobic or anaerobic conditions. Lower brominated diphenyl ether congeners (e.g., 2,2',4,4'-tetrabromodiphenyl ether [BDE 47]) may slowly biodegrade under anaerobic conditions in sediment (EU 2003a). However, information on the rate or extent of anaerobic biodegradation of BDE 47 has not been found. No other information was found in the literature about the transformation and degradation processes for PBDEs in soils and sediment.

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**8.3.2.4 Other Media**

No other information was found in the literature about the transformation and degradation processes for PBBs or PBDEs in other media.

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

Evaluation of the potential for human exposure to PBBs or PBDEs depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. Comparisons among various studies are complicated by the fact that authors may report PBB or PBDE concentrations as technical mixtures, as homologs, or as congeners. For PBDEs, it is common to determine the concentration of individual congeners. However, only a limited number of standards are available. Total PBDEs, the definition of what constitutes total PBDEs (i.e., how many and which congeners are summed), is often not the same in the various studies. Chemical analysis procedures are discussed in greater detail in Chapter 9. Recent monitoring data for PBBs are very limited. Historical monitoring data indicate that environmental PBB concentrations are confined to areas near former manufacturing facilities and regions of Michigan effected by the farm catastrophe of the early 1970's (see Section 8.1).

Monitoring studies indicate that PBDEs are transported globally. Atmospheric, water, and biota levels 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.

**8.4.1 Air**

***Polybrominated Biphenyls.*** Historically, PBBs were released to the atmosphere during three stages of the manufacturing process, and an estimate of the maximum amount of PBBs expected to be lost to the air during the manufacture of PBBs in the United States is available (see Section 8.2.1) (Neufeld et al. 1977). Monitoring data on the ambient air levels of PBBs are very limited. The concentration of hexabromobiphenyl in air samples collected downwind and crosswind from the White Chemical Company plant in Bayonne, New Jersey was 0.06 ng/m<sup>3</sup> (DeCarlo 1979).

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***Polybrominated Diphenyl Ethers.*** In general, limited information is available in the literature reporting the levels of PBDEs in ambient air. The concentrations of PBDEs in air samples are summarized in Table 8-2. Atmospheric concentrations of PBDEs tend to be dominated by lower brominated congeners (e.g., 2,2',4,4'-tetrabromodiphenyl ether [BDE 47], 2,2',4,4',5-pentabromodiphenyl ether [BDE 99], 2,2',4,4',6-pentabromodiphenyl ether [BDE 100], 2,2',4,4',5,5'-hexabromodiphenyl ether [BDE 153], and 2,2',4,4',5,6'-hexabromodiphenyl ether [BDE 154]). DecaBDE (BDE 209) has only been detected in the particulate phase in air near point sources. Air samples sampled 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 levels of BDE 47, BDE 99, BDE 100, BDE 153, and BDE 154 (Dodder et al. 2000a; 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).

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 heptabromodiphenyl ethers were detected; the combined concentrations of 2,2',4,4'-tetrabromodiphenyl ether (BDE 47), 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), and 2,2',4,4',6-pentabromodiphenyl ether (BDE 100) ranged from 7 to 69 pg/m<sup>3</sup> at Hazelrigg and from 6 to 58 pg/m<sup>3</sup> 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 hexabromodiphenyl ethers ranged from 1 to 4 pg/m<sup>3</sup> at Alert for the majority of the year; however, in July 1994, the concentration was 28 pg/m<sup>3</sup>. 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 levels were approximately 1 and 8 pg/m<sup>3</sup>, 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.



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**Table 8-2. Concentrations (pg/m<sup>3</sup>) of Several PBDEs in Air Samples**

Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	Reference
Urban, United States	48	25	3.0	77*	Dodder et al. 2000
Rural, United States	6.2–9.2	4.3–5.0	0.6–0.9	2–4.8*	Dodder et al. 2000
Remote, United States	3.7	2.6	0.33	6.9*	Dodder et al. 2000
Alert, Northwest Territories Canada	No data	No data	No data	1–28	Alaee et al. 1999
Eagle Harbor, Wisconsin	2.9	2.1	0.28	5.5*	Strandberg et al. 2001
Sturgeon Point, New York	3.8	2.8	0.39	7.2*	Strandberg et al. 2001
Sleeping Bear Dunes, Michigan	8.4	5.3	0.80	15*	Strandberg et al. 2001
Chicago, Illinois	33	16	2.0	52*	Strandberg et al. 2001
Ammarnäs, Sweden	6.3	1.6	0.4	8.3	de Wit 2000, 2002
Hoburgen, Sweden	0.7	0.35	0.07	1.1	de Wit 2000, 2002
Stoke Ferry, United Kingdom	4.7–50	5.5–13	1.1–3.9	6.7–58	Peters et al. 1999
Hazelrigg, United Kingdom	3.2–61	3.1–22	0.62–5.4	4.1–69	Peters et al. 1999
Dunai Island, Russia	No data	No data	No data	1–8*	Alaee et al. 1999

<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 (\*).

Sources: de Wit 2002; Lee et al. 2002; Strandberg et al. 2001

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**8.4.2 Water**

***Polybrominated Biphenyls.*** Recent information on the concentrations of PBBs is not available. The concentrations of PBBs in effluents discharged from the Michigan Chemical Corporation plant in St. Louis, Michigan, to the Pine River during 1974–1977 ranged from <0.01 to 150 µg/L (Hesse and Powers 1978). The concentrations of PBBs in effluents from White Chemical Company, Bayonne, New Jersey, and Hexcel Chemical Corporation, Sayerville, New Jersey, ranged from <0.2 to 210 µg/L (DeCarlo 1979). The concentrations of PBBs in the Pine River ≤12 miles downstream from the Michigan Chemical Corporation plant in 1974 were 0.01–3.2 µg/L (Hesse and Powers 1978; Neufeld et al. 1977). 2,2',5,5'-Tetrabromobiphenyl and 3,3',5,5'-tetrabromobiphenyl were qualitatively detected in water from Lake Ontario, and hexabromobiphenyl (unspecified congeners) was qualitatively detected in water from Lakes Ontario and Huron (Great Lakes Water Quality Board 1983). The concentrations of PBBs in test wells within a PBB landfill site in Michigan ranged from 0.4 to 26.0 µg/L, although the concentrations of PBBs in test wells outside the landfill ranged from <0.1 to 4.4 µg/L (Shah 1978). No other information was located about the concentrations of PBBs in water.

***Polybrominated Diphenyl Ethers.*** Due to the hydrophobic nature of PBDEs, this class of compounds has not been detected in water to any significant extent. In 1999, the concentration of PBDEs in Lake Ontario surface waters ranged between 4 and 13 pg/L with ~90% in the dissolved phase (Luckey et al. 2001). 2,2',4,4'-Tetrabromodiphenyl ether (BDE 47) and 2,2',4,4',5-pentabromodiphenyl ether (BDE 99) were the most abundant congeners, together making up >70% of the total PBDEs. In Japan, nondetectable levels of PBDEs were reported in 75–200 water samples (ENVIRON 2003a). No other information was located about the concentrations of PBDEs in water.

**8.4.3 Sediment and Soil**

***Polybrominated Biphenyls.*** Soil samples from the bagging and loading areas of the Michigan Chemical Corporation plant in St. Louis, Michigan, contained PBBs at concentrations of 3,500 and 2,500 mg/kg, respectively (Di Carlo et al. 1978). PBBs (mostly decabromobiphenyl, but some lower brominated biphenyls down to hexabromobiphenyl) in soil near the Hexcel Chemical Corporation plant in New Jersey and the White Chemical Company plant in New Jersey ranged from 40 to 4.6 mg/kg and from 1.14 to 4.25 mg/kg, respectively (DeCarlo 1979). PBB levels in surface soil samples from seven dairy farms in Michigan that spread contaminated manure on the fields ranged from 35 to 1,260 µg/kg, while the

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concentrations in surface soil of control farms (that did not use contaminated manure) were  $<25 \mu\text{g}/\text{kg}$  (Fries and Jacobs 1980).

Concentrations of PBBs in sediments upstream from the Michigan Chemical Corporation plant were below the detection limit ( $100 \mu\text{g}/\text{kg}$ ) with the exception of one sample (Hesse and Powers 1978). The concentration of PBBs in sediment from one upstream sample was  $350 \mu\text{g}/\text{kg}$ . Hesse and Powers (1978) explained that this higher value was due to contamination by upstream currents during periods of water-level regulation at the St. Louis dam. The concentrations of PBBs in near shore sediment near the Michigan Chemical Corporation plant sewer outfall were  $\leq 77.0 \text{ mg}/\text{kg}$ . PBB concentrations in Pine River sediments downstream from the plant showed a gradual decrease from a maximum value of  $9.2 \text{ mg}/\text{kg}$  to a value of  $0.1 \text{ mg}/\text{kg}$  at a location 29 miles downstream from the plant outfall (Hesse and Powers 1978). Similarly, PBB concentrations in sediment samples from swamps and marshes adjacent to the White Chemical Company and Hexcel Chemical Corporation plants in New Jersey ranged from  $<10 \mu\text{g}/\text{kg}$  to  $4.6 \text{ mg}/\text{kg}$  (DeCarlo 1979). A sludge sample from the discharge treatment plant of the White Chemical Company contained  $431 \text{ mg}/\text{kg}$  of PBBs (DeCarlo 1979).

***Polybrominated Diphenyl Ethers.*** No information was located on the ambient environmental concentrations of PBDEs in soils in the United States or other parts of the world. Hale et al. (2002) reported the concentration of PBDEs in soil samples collected in the vicinity of a polyurethane foam-manufacturing facility. Levels in these soils are likely to be higher than to be expected in rural and potentially urban areas of the United States. Total PBDE levels in these samples ranged from not detected to  $76 \mu\text{g}/\text{kg}$  dry weight. 2,2',4,4',5-Pentabromodiphenyl ether (BDE 99) was the predominant congener in soil followed by 2,2',4,4'-tetrabromodiphenyl ether (BDE 47), and 2,2',4,4',6-pentabromodiphenyl ether (BDE 100).

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). The concentrations of PBDEs in sediment samples are summarized in Table 8-3.

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  $36 \mu\text{g}/\text{kg}$  ( $\text{ng}/\text{g}$ ) dry weight. Other congeners detected (in

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**Table 8-3. Concentrations (ng/g) of Several PBDEs in Sediment and Suspended Particulate Samples**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs	BDE 209	Reference
Sediment	Hadley Lake, Indiana	16±2 (dw)	37±8 (dw)	7.1±1.5 (dw)	584 (dw) <sup>a</sup>	480±170 (dw)	Dodder et al. 2002
Sediment	Japan	No data	No data	No data	21–59 (dw)	<25–11,600	Watanabe et al. 1986, 1987, 1995
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>includes sum of BDE 47, BDE 99, BDE 100, BDE 209, and other congeners (not specified)

dw = dry weight; ND = not detected

Sources: de Wit 2002; Hale et al. 2003

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decreasing order: 2,2',4,4',5-tetrabromodiphenyl ether [BDE 99]; 2,2',4,4',5,5'-hexabromodiphenyl ether [BDE 153]; 2,2',4,4',5,6'-hexabromodiphenyl ether [BDE 154]; 2,2',4,4'-tetrabromodiphenyl ether [BDE 47]; and 2,2',4,4',6-pentabromodiphenyl ether [BDE 100]) were less than 5 µg/kg (ng/g) dry weight. PBDEs were above the detection limit (i.e., 0.5 µg/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 µg/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 µg/kg (ng/g) dry weight of pentaBDE.

In Japan, tetra-, penta-, hexa-, and decabromodiphenyl ethers have been observed in river sediments (Watanabe et al. 1986, 1987, 1995). The combined concentrations of tetra- and pentabromodiphenyl ethers ranged from 21–59 ng/g (µg/kg) dry weight. The concentration of decaBDE (BDE 209) ranged from <25 to 11,600 ng/g (µg/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. 2000). Concentrations ranged from 0.3 to 7.1 ng/g (µg/kg) dry weight for BDE 47, not detected to 5.5 ng/g (µg/kg) dry weight for BDE 99, and not detected to 510 ng/g (µg/kg) dry weight for BDE 209. The concentration of PBDEs in suspended particulate matter ranged from not detected to 9 ng/g (µg/kg) dry weight for BDE 47, not detected to 23 ng/g (µg/kg) dry weight for BDE 99, and not detected to 4,600 ng/g (µg/kg) dry weight for BDE 209 (de Boer et al. 2000). The concentration of several brominated flame retardants was measured in sediments collected from the mouths of major European rivers (de Wit 2002). Elevated levels 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 1.61 to 13.1 ng/g (µg/kg) dry weight. The highest hexaBDE levels (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 BDE 153 ranged from 0.013 to 0.056 ng/g (µg/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 (µg/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 (µg/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 (µg/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

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trend from 1939 to 1987 showed that the PBDE levels (i.e., sum of BDE 47, BDE 99, and BDE 100) have increased with a sharp increase after 1980. The PBDE level 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 levels of BDE 47 starting in the 1940s (range, 0.02–0.18 ng/g dry weight) and increasing levels 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 BDE congeners are detected beginning in the late 1950s, increase until the late 1970s, and then level off when BDE 209 first appears. 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 lends some credibility to the likelihood that either the substances identified in the environment as PBDEs are not necessarily PBDEs or that there are yet unknown sources of PBDEs produced in nature.

#### 8.4.4 Other Environmental Media

##### *Polybrominated Biphenyls.*

**Food.** Although the agriculture episode in Michigan involving contaminated feed occurred in May 1973, PBBs were not identified as the causative factor until April 1974 (Fries 1985b). PBB-containing meats, milk, butter, eggs, and cheese entered the human food chain for almost a year before the PBBs were identified. Concentrations of PBBs (on a fat basis) in milk samples collected from contaminated farms soon after PBB was identified ranged from 2.8 to 595 mg/kg (Cordle et al. 1978; Kay 1977).

Concentrations of PBBs in other products processed from the contaminated milk were as follows: butter, 1–2 mg/kg; cheese, 1.4–15.0 mg/kg; and canned milk, 1.2–1.6 mg/kg (Cordle et al. 1978). In 1974, the levels of PBBs in eggs from contaminated farm premises were as high as 59.7 mg/kg (Kay 1977). The levels of PBBs in poultry and cattle tissues from the contaminated farm collected in 1974 were 4,600 mg/kg and up to 2,700 mg/kg, respectively (Kay 1977). With the seizure and destruction of the contaminated farm animals and products, the levels of PBBs in consumer products showed a steady

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decline. For example, in 1975, among 18 milk samples, 13 cheese samples, and 14 butter samples taken in Michigan, only 3 butter samples exceeded the FDA guidelines of 0.3 mg/kg fat (Di Carlo et al. 1978). In 1975, PBBs were detected in 245/2,040 meat samples collected in Michigan, with only 24 samples containing levels >0.3 mg/kg fat (Di Carlo et al. 1978). Although 95% of 1,430 meat samples collected in Michigan in 1976 contained detectable PBBs, only 1 sample contained >0.6 mg/kg, and a market basket survey in Michigan showed detectable PBBs in only 1/102 meat samples (Di Carlo et al. 1978).

**Fish.** No PBBs were detected in several varieties of fish (carp, white sucker, Northern pike, bullhead, and bass) from the Alma Reservoir, which is upstream from the Michigan Chemical Corporation plant and above a dam that prevents fish from moving upstream (Hesse and Powers 1978). On the other hand, tissue samples from fish collected from the Pine River,  $\leq 29$  miles downstream from the plant, contained up to 1.33 mg PBBs/kg (wet weight in skinless fillets). There was no apparent change in PBB concentrations in fish between 1974 and 1976 (Hesse and Powers 1978). PBBs could be detected in fish from Pine River and other embayments and tributaries of Lake Huron in 1983. PBB concentrations in carp and other sedentary fish from embayments and tributaries of Lake Huron (including Pine River) and Lake Superior were determined (Great Lakes Water Quality Board 1989; Jaffe et al. 1985). PBBs were detected in the concentration range of 15–15,000  $\mu\text{g}/\text{kg}$  (fat basis) in fish from embayments and tributaries of Lake Huron, but not from Lake Superior. Recently, Luross et al. (2002) determined the concentrations of several PBB congeners in lake trout from Lakes Huron, Superior, Erie, and Ontario. 2,2',4,4',5,5'-Hexabromobiphenyl (BB-153) and 2,2',4,5,5'-pentabromobiphenyl (BB-101) were found at the highest levels at concentrations ranging from 189 to 2,083 pg/g wet weight and from 42 to 633 pg/g wet weight, respectively. Several other congeners were also detected in these lake trout samples (see Table 8-4).

In German rivers, elevated levels of nona- and octaBBs were present in fish. HexaBB was predominant in fish from the North Sea and Baltic Sea. 3,3',4,4',5,5'-Hexabromobiphenyl (BB-169) was found at a maximum concentration of 36 mg/kg ( $\mu\text{g}/\text{g}$ ) fat in samples from the Baltic Sea. However, BB-169 was not found in waters from the North Sea or rivers. In Baltic marine fish, the concentrations of 2,2',4,4',5,5'-hexabromobiphenyl (BB-153) ranged from 0.2 to 4.2 mg/kg ( $\mu\text{g}/\text{g}$ ) lipid (de Boer et al. 2000a).

**Animals.** PBB concentrations in whole (with skin) and skinless ducks collected within 2 miles of the Michigan Chemical Corporation plant in 1974–1977 ranged from not detected to 2.70 mg/kg ( $\mu\text{g}/\text{g}$ ) and not detected to 1.8 mg/kg ( $\mu\text{g}/\text{g}$ ), respectively (Hesse and Powers 1978). Three bottlenose dolphins

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**Table 8-4. Mean Concentrations of Nine PBB Congeners in Lake Trout from the Great Lakes (pg/g Wet Weight)**

Congener	Lake Superior	Lake Huron	Lake Erie	Lake Ontario
BB-26/29	<1.3	5.2±2.3	<1.3	<1.3
BB-31	<1.8	5.2±1.8	<1.8	<1.8
BB-49	6.8±1.7	125±43	20±7.6	14±4.9
BB-52	8.4±3.6	191±77	24±9.5	11±4.5
BB-80	<3.8	<3.8	<3.8	<3.8
BB-101	42±18	633±359	71±20	109±50
BB-103	<1.5	4.4±1.9	<1.5	<1.5
BB-153	189±105	2,083±1,282	220±47	1,008±513
BB-155	1.0±0.78	5.8±3.4	<0.98	1.1±0.43

Source: Luross et al. 2002



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(*Tursiops truncatus*) collected during 1987–1988 from the U.S. mid-Atlantic contained PBBs at concentrations of 14–20 µg/kg (ng/g) lipid basis (Kuehl et al. 1991). The source of the PBBs in the dolphins was not given. The median concentrations of PBBs in 10 specimens of carcass and brain of bald eagles (*Haliaeetus leucocephalus*) collected from 29 states in 1977 were 0.07 and 0.05 mg/kg (µg/g), respectively (Kaiser et al. 1980). Twenty-two other specimens did not contain detectable levels (<0.03 mg/kg [µg/g]) of PBBs. The concentrations of PBBs in eggs of fish-eating birds (common tern, little gull, herring gull, and red-breasted mergansers) collected during 1975–1980 from nesting islands in northwestern Lake Michigan and Green Bay contained PBBs in the concentration range of 0.02–0.25 mg/kg (µg/g) wet weight (Heinz et al. 1983, 1985).

White-tailed sea eagles collected from the Baltic Sea contained 280 ng PBBs/g lipid weight (Jansson et al. 1987). The concentration of PBBs in common guillemots (*Uria aalge*) collected in 1979–1981 from the Baltic Sea was 160 ng/g lipid (Jansson et al. 1987). Brunnich's guillemot (*Uria lomvia*), collected from Svalbard in the Arctic, contained 50 ng PBBs/g lipid (Jansson et al. 1987).

In 1981, female ringed seals from Svalbard in the Swedish Arctic contained 4 ng PBBs/g lipid (Jansson et al. 1987). The level of PBBs in Baltic Sea harbor seal (*Phoca vitulina*) was 20 ng/g lipid; North Sea harbor seal contained 3 ng PBBs/g lipid (Jansson et al. 1987). The concentration of hexaBB ranged from 13–61 µg/kg (ng/g) wet weight from harbor seals collected from the North Sea (decaBB <1 µg/kg [ng/g] wet weight). In whitebeaked dolphins from the North Sea, the concentration of hexa-, penta-, and deca-BBs were 13, 8.3, and <0.9 µg/kg (ng/g) wet weight, respectively. Tetra-, penta-, and deca-BBs concentration ranges were 1.1–1.9, 0.4–0.9, and <0.5 µg/kg (ng/g) wet weight, respectively, in sperm whales from the Atlantic Ocean (de Boer et al. 1999).

**Human Tissues and Body Fluids.** The quantitative determination of the concentrations of PBBs in blood, serum, adipose tissue, milk, and other body tissues or fluids is important in determining the human body burden of these chemicals. Fat is the largest repository of PBBs in the body, and concentrations in fat can provide an index of body burdens and exposure. It is simpler and less invasive to collect samples of serum or breast milk than body fat. However, the collection of milk and serum for the estimation of possible body burden has limitations. Breast milk can be obtained from limited segments of the population. Also the concentration of PBBs in breast milk can show considerable fluctuations because the breast is emptied only periodically (Brilliant et al. 1978; Willett et al. 1988). Serum, however, has lower PBB concentrations than body fat (see Section 5.5.1).

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Hexabromobiphenyl was detected (detection limit 6.6 µg/kg [ng/g]) at a frequency of 8–57% in human adipose tissue samples from six Canadian Great Lakes municipalities in 1984 (Williams et al. 1988). The concentration of 2,2',4,4',5,5'-hexabromobiphenyl in adipose tissue samples pooled from tissues of the general population of the conterminous United States ranged from 1 to 2 µg/kg (ng/g) (Lewis and Sovocool 1982). PBB levels in the adipose tissues of 15 quarantined dairy farm residents in mid-Michigan (where the mix-up involving FireMaster BP-6 occurred) ranged from 0.104 to 174 mg/kg (µg/g) (Humphrey and Hayner 1975).

In the fall of 1993, the serum levels of BB-167 (2,2',4,4',5,5'-hexabromobiphenyl) in 32 subjects, approximately 10 of whom consumed sport fish from the Great Lakes, were measured (Anderson et al. 1998). When the data were stratified by lake, on average, the Lake Huron fish consumers had the highest levels of PBBs (0.6 ppb [ng/g]) and Lake Erie fish consumers had the lowest (0.2 ppb [ng/g]). When the data were then stratified by state of residence, on average, Great Lakes sport fish consumers who live in Michigan had the highest PBB level (0.7 ppb [ng/g]) and residents of Wisconsin had the lowest level (0.05 ppb [ng/g]).

In Michigan after the agriculture contamination episode in 1973–1974, the median PBB concentrations in blood of exposed adults and children in farms were 0.014 and 0.035 mg/kg (14 and 35 ppb [ng/g]), respectively, compared to corresponding median concentrations of 0.003 and 0.006 mg/kg (3 and 6 ppb [ng/g]) in a control group (Humphrey and Hayner 1975). PBB levels in the blood of quarantined farm workers in Michigan were also higher than in nonquarantined farm residents and the general population of Michigan (Cordle et al. 1978; Kimbrough 1987; Lambert et al. 1990; Landrigan et al. 1979). The concentration ratio of PBBs in adipose tissue over blood plasma for 13 paired specimens was 175 to 1 (Humphrey and Hayner 1975).

A cross-section of the population of Michigan was studied in 1978, 5 years following the agriculture episode involving FireMaster BP-6, to determine the levels of PBBs in human tissues. Levels of PBBs were highest in the part of state in which the episode occurred (median: adipose tissue, 500 µg/kg (ng/g); serum, 1.7 µg/L) and were lowest in the upper peninsula (median: adipose tissue, 15 µg/kg (ng/g); serum, 0.2 µg/L), farthest from the source of contamination. Levels in the rest of the state were in between (median: adipose tissue, 240 µg/kg [ng/g]; serum, 0.9 µg/L) (Wolff et al. 1982). The estimated concentration ratio of PBBs in adipose tissue over serum was near 300 among 31 Michigan dairy farm residents (Wolff et al. 1979a). The ratio of adipose tissue to serum PBB concentration was 363 to 1 for the general population and 100 to 1 in lactating women (Brilliant et al. 1978). The kinetics of fat

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metabolism in lactating women seems to alter PBB partitioning. The ratios of adipose tissue to serum PBB concentration for nonpregnant females and male chemical workers, farm workers and other males, and pregnant females in 3,683 Michigan residents with varying degrees of exposure were 190–260 to 1, 325–329 to 1, and 107–119 to 1, respectively (Eyster et al. 1983). The PBB ratios for cord to serum and placenta to serum in pregnant females were 0.10–0.14 to 1 and 0.10–0.17 to 1, respectively (Eyster et al. 1983). The PBB ratios for feces to serum and bile to serum in farm and chemical workers were 0.53–0.71 to 1 and 0.45–0.63 to 1, respectively (Eyster et al. 1983). The detection of PBBs in bile and feces indicates transfer into the intestinal tract. However, the concentration of PBBs in feces represented a minor proportion of the total body burden, indicating a slow rate of excretion (Eyster et al. 1983). Serum PBBs were determined in a nested case-control study of 1,925 women enrolled in the Michigan Department of Public Health registry for persons exposed to PBBs (Henderson et al. 1995). Study participants had lived on or received food from a farm quarantined by the Michigan Department of Agriculture, were recruited from July 1976 to December 1977, and were followed up annually from 1978 through 1993. Median serum PBB concentrations were 2 ppb (n=290; range=0.5–419 ppb [ $\mu\text{g/L}$ ]).

The concentrations of PBBs in the breast milk of females from the lower peninsula of Michigan (exposed area) were generally higher than in breast milk of females from the upper peninsula (farthest from the sources) (Brilliant et al. 1978). PBB levels in breast milk of five females from the exposed farms were 0.21–92.7 mg/kg (Cordle et al. 1978; Humphrey and Hayner 1976). In a cohort of Michigan residents, the ratio of PBBs in breast milk to maternal serum was 107–122 to 1 and in adipose tissue to breast milk was 1.1–1.5 to 1 (Eyster et al. 1983; Landrigan et al. 1979). The concentrations of PBBs found in human tissues and body fluids are given in Table 8-5. Recent levels of PBBs in human breast milk (i.e., 1990 to present) were not located (WHO 1994b).

***Cow's Milk.*** In an attempt to determine the metabolites of PBBs, whole milk of lactating cows from contaminated areas of Michigan was analyzed for monohydroxy metabolites, but none were found (Gardner et al. 1976). In a later study, the feces of dogs fed FireMaster BP-6 in corn oil was found to contain a metabolite identified as 6-hydroxy-2,2',4,4',5,5'-hexabromobiphenyl (Gardner et al. 1979).

The effects of processing cow's milk containing PBBs also has been studied (Murata et al. 1977; Zabik et al. 1978). Spray-drying reduced PBB levels in whole and skim milk, whereas pasteurization, freeze-drying, aging of cheese, and condensation were not effective in reducing the level of PBBs in milk products. Pressure-cooking meat containing PBBs reduced the level of PBBs in the cooked meat (Zabik et al. 1978).

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**Table 8-5. Tissue Levels of PBBs in Michigan Residents**

Tissue	Subject(s)	Mean/median concentration <sup>a</sup>	Year	Reference	
Serum	Exposed farm workers	14 µg/L	1976	Stross et al. 1979	
	Chemical workers	48 µg/L	No data	Stross et al. 1981	
	Chemical workers	1.1–1,000 µg/L	1976	Anderson et al. 1978d	
	Exposed farm workers	BDL to 1,000 µg/L	1976	Anderson et al. 1978d	
	Residents from quarantined farms	26.9 µg/L	1976–1977	Landrigan et al. 1979	
	Residents from non-quarantined farms	3.5 µg/L	1976–1977	Landrigan et al. 1979	
	Farm product consumers	17.1 µg/L	1976–1977	Landrigan et al. 1979	
	Chemical workers and families	43.0 µg/L	1976–1977	Landrigan et al. 1979	
	Control group	3.5 µg/L	1976–1977	Landrigan et al. 1979	
	General population (lower peninsula)	1.9 µg/L	1978	Wolff et al. 1982	
	General population (upper peninsula)	0.2 µg/L	1978	Wolff et al. 1982	
	General population (remainder of state)	0.9 µg/L	1978	Wolff et al. 1982	
	Chemical workers	25.4 µg/L	No data	Eyster et al. 1983	
	Farm and other workers	5.4 µg/L	No data	Eyster et al. 1983	
	Mothers from lower peninsula	26.2 µg/L	1976–1977	Landrigan et al. 1979	
	Exposed mothers from farms	3.4 µg/L	No data	Eyster et al. 1983	
	Non-pregnant women from exposed farms	3.1 µg/L	No data	Eyster et al. 1983	
	Exposed women enrolled in the Michigan Department of Public Health registry	2 µg/L	1993	Henderson et al. 1995	
	Cord serum	Exposed mothers from lower peninsula	3.2 µg/L	1976–1977	Landrigan et al. 1979
		Mothers from lower peninsula	<1.0 µg/L	No data	Eyster et al. 1983
Blood plasma	Workers from quarantined farms	14 µg/L	1974	Humphrey and Hayner 1975	
	Children from quarantined farms	35 µg/L	1974	Humphrey and Hayner 1975	
	Adults from non-quarantined farms	3 µg/L	1974	Humphrey and Hayner 1975	
	Children from non-quarantined farms	6 µg/L	1974	Humphrey and Hayner 1975	
Placenta	Exposed mothers	<1 µg/L	No data	Eyster et al. 1983	
Breast milk	Exposed mothers	370 µg/kg (fat basis)	No data	Eyster et al. 1983	
	Exposed mothers from lower peninsula	3,614 µg/kg (fat basis)	1976–1977	Landrigan et al. 1979	
	Mothers from lower peninsula	68 µg/kg (fat basis)	1976	Brilliant et al. 1978	
	Mothers from upper peninsula	<44 µg/kg (fat basis)	1976	Brilliant et al. 1978	

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**Table 8-5. Tissue Levels of PBBs in Michigan Residents**

Tissue	Subject(s)	Mean/median concentration <sup>a</sup>	Year	Reference
Adipose tissue	Population of lower peninsula	500 µg/kg	1978	Wolff et al. 1982
	Population of upper peninsula	15 µg/kg	1978	Wolff et al. 1982
	Population of rest of the state	240 µg/kg	1978	Wolff et al. 1982
	Chemical workers	9,330 µg/kg	No data	Brown et al. 1981
	Farm residents from lower peninsula	3,940 µg/kg	No data	Brown et al. 1981
	Farm residents from lower peninsula	3,260 µg/kg	1976	Stross et al. 1979
	Chemical workers	12,820 µg/kg	No data	Stross et al. 1981
	Workers from quarantined dairy farms	12,500 µg/kg	1974	Humphrey and Hayner 1975
	Pregnant females from lower peninsula	400 µg/kg	No data	Eyster et al. 1983
	Chemical workers	5,290 µg/kg	No data	Eyster et al. 1983
	Farm and other workers from lower peninsula	1,650 µg/kg	No data	Eyster et al. 1983

<sup>a</sup>When both mean and median values are available, the former values have been used in the table. In some cases, when neither value is available, the range is given in the table.

BDL = below detection limit

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***Polybrominated Diphenyl Ethers.***

**Food.** Information about the concentrations of PBDEs in food-stuff is very limited. Huwe et al. (2002a) reported total PBDE levels in farm chickens raised in two different regions of the United States. The total PBDE level 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 and chickens bought in the grocery store were analyzed for total PBDEs (Environ 2003b). 2,2',4,4',5-Pentabromodiphenyl ether (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. Recently, Ohta et al. (2002a) 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 2,2',4,4'-tetrabromodiphenyl ether (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 2,2',4,4',5,5'-hexabromodiphenyl ether (BDE 153), spinach (representing a leafy vegetable) might be strongly influenced by PBDE contamination in air. The concentration 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. 2002a). 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 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, and 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 seafoods from the Inland Sea of Japan were determined for samples collected in 1998 (Hori et al. 2000). The congeners, 2,4,4'-tribromodiphenyl ether (BDE 28); BDE 47; 2,3',4,4'-tetrabromodiphenyl ether (BDE 66); BDE 99; 2,2',4,4',6-pentabromodiphenyl ether (BDE 100); BDE 153; and 2,2',4,4',5,6'-hexabromodiphenyl ether (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 vegetarian diet samples from the United Kingdom. Median concentrations of BDE 47

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(2,2',4,4'-tetraBDE), 99 (2,2',4,4',5-pentaBDE), 100 (2,2',4,4',6-pentaBDE), 153 (2,2',4,4',5,5'-hexaBDE), and 154 (2,2',4,4',5,6'-hexaBDE) 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 (2,2',4,4'-tetraBDE), 99 (2,2',4,4',5-pentaBDE), 100 (2,2',4,4',6-pentaBDE), 153 (2,2',4,4',5,5'-hexaBDE), and 154 (2,2',4,4',5,6'-hexaBDE) were 47.2, 56.7, 10.0, 20, and 20 pg/g dry weight, respectively. Concentrations of BDE 47, 99, and total PBDE were found to be statistically higher in omnivorous diet samples compared to vegetarian diet samples.

***Biosolids and Effluents.*** The concentrations of PBDEs in biosolids (sewage sludge) and effluents are summarized in Table 8-6. PBDEs were detected in biosolids from four different regions of the United States (Pardini et al. 2001). The total concentrations of pentaBDE in biosolids ranged from 1,100 to 2,290 ng/g dry weight; the levels of pentaBDE were high and consistent, regardless of the region of origin. 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 ng/g dry weight in the biosolid samples. Sewage sludges 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 waste water-treatment plants in Germany were sampled (Hagenmaier et al. 1992). The mean concentration of tri- to hepta-BDEs was 8.37 ng/g with tri-, tetra-, penta-, hexa-, and heptaBDE at concentrations of 0.65, 3.06, 3.02, 0.49, and 0.22 ng/g, respectively. Levels of penta- and hexaBDEs were highest in these samples. de Boer et al. (2000b) determined the concentration of PBDEs in sewage treatment-plant effluents (STP) from the Netherlands. The concentration of total PBDEs ranged from 11 to 35 ng/g dry weight, while the concentrations of 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and BDE 209 were 11–35 and 310–920 ng/g dry weight, respectively. Kohler et al. (2003) determined the levels 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%.

***World Trade Center Site.*** In 2001, PBDEs were detected in dust and smoke samples taken near the World Trade Center (WTC) disaster site (Liroy et al. 2002). The highest concentration was for decaBDE (i.e., BDE 209), which was present in thermoplastics (e.g., computers). Concentrations of PBDE

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**Table 8-6. Concentrations (ng/g Dry Weight) of Several PBDEs in Biosolids (Sewage Sludge) and Effluents**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <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	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 1999; Sellström and Jansson 1995
Sewage Sludge	Rimbo, Sweden	53	53	13	119	No data	Sellström 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> $\Sigma$ PBDEs is the sum of BDE 47, BDE 99, and BDE 100, but if more congeners are included, this is marked with an asterisk (\*).

Source: de Wit 2002; Hale et al. 2002



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congeners were 107–174 µg/kg dry weight basis for 2,2',4,4'-tetrabromodiphenyl ether (BDE 47), 51.1–74.1 µg/kg dry weight basis for 2,2',4,4',6-pentabromodiphenyl ether (BDE 100), 155–293 µg/kg dry weight basis for 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), 42.0–53.5 µg/kg dry weight basis for 2,2',4,4',5,5'-hexabromodiphenyl ether (BDE 153), 219–305 µg/kg dry wt basis for 2,2',4,4',5,6'-hexabromodiphenyl ether (BDE 154)/PBB-153, and 1,330–2,660 µg/kg dry wt basis for decaBDE (BDE 209). Levels of PBDEs were found to be similar to levels found in sewage sludge (Lioy et al. 2002). No further information on levels of PBDEs in environmental was located for the WTC site.

**Freshwater Fish.** Monitoring data indicated that the levels of PBDEs are increasing in freshwater organisms with higher concentrations near point sources. The congener profiles show the highest levels for 2,2',4,4'-tetrabromodiphenyl ether (BDE 47). The presence of PBDEs in freshwater aquatic organisms taken from remote regions suggests that diffuse sources of PBDEs are also important. The concentrations of PBDEs in freshwater fish samples in the United States are summarized in Table 8-7. 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. 2000a). Mean total PBDE concentrations (sum of BDE 47, 2,2',4,4',5-pentabromodiphenyl ether [BDE 99], 2,2',4,4',6-pentabromodiphenyl ether [BDE 100], 2,2',4,4',5,5'-hexabromodiphenyl ether [BDE 153], and 2,2',4,4',5,6'-hexabromodiphenyl ether [BDE 154]) were higher in crappie (*Poxomis 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 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 dominate 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, BDE 153, and BDE 154 was 3,000 ng/g lipid weight (Asplund et al. 1999b). Lake trout (*Salvelinus manaycush*) from Lakes Ontario, Huron, and Superior were also found to have di- to

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**Table 8-7. Concentrations (ng/g Lipid Weight, Except as Noted) of Several 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
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
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

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**Table 8-7. Concentrations (ng/g Lipid Weight, Except as Noted) of Several 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
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
					297 (fw) (whole)		
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
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 (\*).

Sources: Luross et al. 2002; Manchester-Neesvig et al. 2001; Rayne et al. 2003; Stapleton and Baker 2003

dw = dry weight; fw = fresh weight

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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 fresh water 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 levels 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 large mouth bass), the congener patterns were dominated by BDE 47; however, in the Des Plaines River carp, the dominant congeners were heptaBDE congeners (2,2',3,4,4',5,6-heptBDE [BDE 181] and 2,2',3,4,4',5',6-heptaBDE [BDE 183]), lesser amounts of 2,3,3',4,4',5,6-heptaBDE (BDE 190), and two hexaBDEs (BDE 154 and BDE 153). Possible sources for the heptaBDE congeners were not obvious since none of the commercial mixtures are known to contain these congeners. Three possible explanations were proposed to explain the presence of the heptaBDE congeners found in Des Plaines River carp: waste discharge from an industrial facility; publicly-owned treatment works (POTW) effluents; or formation *in situ* by decaBDE deposits.

The concentrations of PBDEs in freshwater fish samples from Europe are summarized in Table 8-8. Between 1986 and 1988, levels of 2,2',4,4'-tetrabromodiphenyl ether (BDE 47), 2,2',4,4',5-pentabromodiphenyl ether (BDE 99), and 2,2',4,4',6-pentabromodiphenyl ether (BDE 100) were measured in whitefish (*Coregonus spp.*) from a remote mountain lake in Northern Sweden (Lake Storvindeln), 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 levels (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 levels 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 levels

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**Table 8-8. Concentrations (ng/g Lipid Weight Except as Noted) of Several PBDEs in Freshwater Fish Samples from Europe**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
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	Lake Bolmen, Sweden	65	42	19	130	No data	Kierkegaard 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
Whitefish	Lake Thun, Switzerland	19	12	2.5	36*	No data	Zennegg et al. 2003

<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 (\*).

Sources: de Wit 2002; Zennegg et al. 2003

dw = dry weight; fw = fresh weight

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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 levels 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 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 levels 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 levels 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). Levels 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 (*Abramais 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. 2000). 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 (*Platichys flesus*), and dab (*Limanada limanada*) 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. Levels 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 to 1,200 ng/g lipid weight, and not detected to 1,200 ng/g lipid weight. The highest levels 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. 2000). 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.

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**Saltwater Fish.** No identifiable temporal trends were found for PBDE levels in marine aquatic species. Spatial trends show higher levels of lower brominated BDE congeners found near human populated areas. The congener profiles show the highest levels for 2,2',4,4'-tetrabromodiphenyl ether (BDE 47). The levels of several PBDEs in marine aquatic species are summarized in Table 8-9. 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. 2002b); 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 (2,2',4,4',6-pentabromodiphenyl ether [BDE 100]), and 9.5–46 ng/g lipid (2,2',4,4',5-pentabromodiphenyl ether [BDE 99]), respectively. The highest levels 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 level (690 and 2,600 ng/g wet weight) followed by BDE 99 and BDE 100; total BDE congener levels 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. Levels 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 levels of any PBDEs, while samples collected in industrial areas of southeast Asia had the highest. Fall-caught 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 dominate 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 levels and 5-year-old herring had the highest levels. 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

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**Table 8-9. Concentrations (ng/g Lipid Weight) of Several PBDEs in Marine Aquatic Species**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
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
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 8-9. Concentrations (ng/g Lipid Weight) of Several 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	0.6–20 (dw)	<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 (\*).

Source: Easton et al. 2002; de Wit 2002; Ikonomidou et al. 2002; Ueno et al. 2003

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primary congener with levels ranging from 7.6 to 24 ng/g lipid weight for 1–3-year-old sprat, 17–140 ng/g lipid weight for 3- to 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 levels 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 levels were 47, 7.2, and 6.3 ng/g lipid, respectively, in whole-body composites (Burreau et al. 1999). In another study, the levels 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 levels 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. 2000). Concentrations of BDE 153 and BDE 209 were below the detection limit. In 1996, de Boer et al. (2001) measured the levels 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 levels ranged from 0.1 and 17 ng/g fresh weight, with tetraBDE being the major congener. decaBDE was also detected in a mussel sample from Osaka Bay (at 1.4 µg/kg wet weight). Recently, Japanese market fish were analyzed for PBDEs. The highest combined PBDE levels (2,4,4'-triBDE [BDE 28], 2,2',4,4'-tetraBDE [BDE 47], 2,3',4,4'-tetraBDE [BDE 66], 2,2',4,4',5-pentaBDE [BDE 99], 2,2',4,4',6-pentaBDE [BDE 100], 2,2',4,4',5,5'-hexaBDE [BDE 153], and 2,2',4,4',5,6'-hexaBDE [BDE 154]) were in salmon, cultured yellowtail, and wild yellowtail muscle (46, 44, and 30.5 ng/g lipid weight, respectively) and lowest levels 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 levels ranged from 0.00136 to 2.1 ng/g

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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. Levels 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 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE (BDE 99), 2,2',4,4',5,5'-hexaBDE (BDE 153), and decaBDE (BDE 209) (de Boer et al. 2000). Concentrations of BDE 47 and BDE 99 ranged from 0.9 to 4.3 ng/g dry weight and from 0.3 to 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 levels of lower brominated BDE congeners with higher levels found near human-populated areas. In all marine animal studies, the congeners profile show the highest levels for BDE 47. The concentrations of several PBDEs in marine animals are summarized in Table 8-10. PBDEs have been detected in several species of seal from several different sites. In San Francisco Bay, California, the concentrations of PBDEs in harbor seals have increased dramatically over the past decade, with current levels among the highest reported for this species (She et al. 2002). The concentration of total PBDEs (sum of BDE 47, 2,2',4,4',5-pentaBDE [BDE 99], 2,2',4,4',6-pentaBDE [BDE 100], 2,2',4,4',5,5'-hexaBDE [BDE 153], and 2,2',4,4',5,6'-hexaBDE [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

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**Table 8-10. Concentrations (ng/g Lipid Weight) of Several 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	San Francisco Bay, California	46–6,682	17–303	No data	No data	No data	She et al. 2000
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
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
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 et al. 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
Galauous 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

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**Table 8-10. Concentrations (ng/g Lipid Weight) of Several PBDEs in Marine Animals**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Harbor porpoise	British Columbia, Canada	50–1,200	No data	No data	350–2,300*	No data	Ikononmou et al. 2000b
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 et al. 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. 1998
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. 1998
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
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	Ikononmou et al. 2000
Sperm whale	Netherlands	130–250	32–64	21–35	187–349	No data	de Boer et al. 1998
Whitebeaked dolphin	Netherlands	5,500	1,000	1,200	7,700	No data	de Boer et al. 1998

<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 (\*).

Sources: de Wit 2002; Norstrom et al. 2002; She et al. 2002

fw = fresh weight

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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 et al. 1987, 1993; Sellström et al. 1993). Higher levels 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 level 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 seal from the San Francisco Bay area (She et al. 2000). 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 seal 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 levels in female seals suggest that PBDEs are transferred to young through breast milk. On Holman Island, Northwest Territory, Canada (Arctic) in 1996, ringed seal had total PBDE concentrations of 2.4–4.9 ng/g lipid for males. The levels of PBDEs were found to increase with age (Ikonomou 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 levels. The concentration of PBDE in samples collected between 1981 and 1996 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 levels of PBDEs have recently 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 levels (sum of tri- to hepta-congeners) were 350–2,300 ng/g lipid weight; 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) was found at the highest levels in these samples (range, 50–1,200 ng/g lipid weight) (Ikonomou 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 levels 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. (1998) found PBDEs in blubber of one whitebeaked dolphin (*Lagenorhynchus albirostris*); the levels of 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE (BDE 99), and 2,2',4,4',6-pentaBDE (BDE 100) were 5,500, 1,000, and 1,200 ng/g lipid weight, respectively.

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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 levels, ranging from 3,000 to 3,160 ng/g lipid; lower levels 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 2,2',4,4',5-pentaBDE (BDE 99), accounting for 70% of the sum of 19 congeners. van Bavel et al. (1999) also studied the levels 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 levels (females, 230 ng/g lipid; males, 540 ng/g lipid). In Beluga whales sampled in 1997 from southeast Baffin (Cumberland Sound), the levels 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, 2,2',4,4',6-pentaBDE (BDE 100), and 2,2',4,4',5,6'-hexaBDE (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 (*Pyseter macrocephalus*) and one minke whale (*Balaenaoptera 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 concentrations of PPBEs 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.

**Marine Birds.** Increasing levels of PBDEs have been found in marine birds and eggs, with 2,2',4,4'-tetraBDE (BDE 47) found at the highest levels. 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 throughout 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, 2,4,4'-triBDE (BDE 28), BDE 47, 2,2',4,4',5-pentaBDE (BDE 99), 2,2',4,4',6-pentaBDE (BDE 100), 2,2',4,4',5,5'-hexaBDE (BDE 153), 2,2',4,4',5,6'-hexaBDE (BDE 154), and 2,2',3,4,4',6,6'-heptaBDE (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

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(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 et al. 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 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 1999; Sellström et al. 1993). Between 1992 and 1997, PBDE levels started to decrease statistically. In 1997, the PBDE level (sum of BDE 47, BDE 99, and BDE 100) was 190 ng/g lipid, with BDE 47 as the predominant congener.

**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 levels 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.

Tables 8-11, 8-12, and 8-13 summarize the concentrations of PBDEs found in blood (serum), adipose tissue, breast milk, and other body tissues or fluids, respectively. Levels of PBDEs in body tissues and fluids from individuals living in the United States have recently been determined. These studies indicate that levels 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. 2001). 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. 2001). DecaBDE was found at levels above the limit of quantification (1 pmol/g lipid) in 5 of 12 serum samples (Patterson et al. 2000). Serum samples were



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**Table 8-11. Concentrations (ng/g Lipid Weight) of Several PBDEs in Human Blood Samples**

Sample type	Location	BDE 47	BDE 99	BDE 100	ΣPBDEs <sup>a</sup>	BDE 209	Reference
Human blood	San Francisco, California	ND	No data	No data	No data	No data	Petreas et al. 2002
Human blood	San Francisco, California	95	No data	No data	No data	No data	Petreas et al. 2002
Human blood	San Francisco, California	16.5	No data	No data	No data	No data	Petreas et al. 2003
Human blood	United States (in 1988)	0.63 (median)	0.32 (median)	0.17 (median)	No data	<0.1	Sjödin et al. 2001
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
Human blood	Sweden	No data	No data	No data	2.1*	No data	Klasson Wehler et al. 1997
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 (median)	No data	No data	3.3–4.1*	<0.7 (median)	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 (\*).

Source: de Wit 2002; Sjödin et al. 2001

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**Table 8-12. Concentrations (ng/g Lipid Weight) of Several PBDEs in Human Adipose Tissue Samples**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Human adipose tissue	Northern California	7.0–28	3.1–7.3	No data	No data	No data	She et al. 2000
Human adipose tissue	San Francisco, California	16.5 (5.2–196)	No data	No data	No data	No data	Petreas et al. 2003
Human adipose tissue	United States	No data	No data	No data	No data	ND–0.7	Cramer et al. 1990; Stanley et al. 1991
Human adipose tissue	Sweden	8.8	1.1	1.8	11.7	No data	Haglung et al. 1997
Human adipose tissue	Sweden	3.8–16	No data	No data	No data	No data	Lindström et al. 1998
Human adipose tissue	Sweden	2.2	1.6	0.1	5*	No data	Meironyté Guvenius and Norén 1999
Human adipose tissue	Finland	7.3	2.3	No data	6.2–22*	No data	Strandman et al. 1999
Human adipose tissue	Finland	1.20	0.26	0.09	No data	No data	Smeds and Saukko 2003
Human adipose tissue	Spain	1.36	0.42	No data	No data	No data	Meneses et al. 1999
Human adipose tissue	Japan	459	118	250	1,288	No data	Choi 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 (\*).

Source: de Wit 2002; Petreas et al. 2003

ND = not detected

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**Table 8-13. Concentrations (ng/g Lipid Weight) of Several PBDEs in Human Breast Milk Samples**

Sample type	Location	BDE 47	BDE 99	BDE 100	$\Sigma$ PBDEs <sup>a</sup>	BDE 209	Reference
Human breast milk	Texas	18.4	5.7	2.9	34.0	8.24 (max)	Schechter et al. 2003b
Human breast milk	Germany	No data	No data	No data	0.6–11*	No data	Krüger 1988
Human breast milk	Uppsala County, Sweden	2.35	0.62	0.38	4.01	No data	
Human breast milk	Sweden	2.3	0.5	0.4	4*	No data	Norén and Meironyté 1998, 2000
Human breast milk	Sweden	2.5	0.7	0.5	4.4*	No data	Darnerud et al. 1998
Human breast milk	Finland	1.31	0.39	No data	No data	No data	Strandman et al. 2000
Human breast milk	Quebec and Ontario, Canada	3.4	1.2	0.44	5.8*	No data	Ryan and Patry 2000
Human breast milk	Maritimes, Canada	No data	No data	No data	19*	No data	Ryan and Patry 2000
Human breast milk	Quebec, Canada	No data	No data	No data	18.8*	No data	Ryan and Patry 2000
Human breast milk	Ontario, Canada	No data	No data	No data	2.8*	No data	Ryan and Patry 2000
Human breast milk	Prairies, Canada	No data	No data	No data	5.7*	No data	Ryan and Patry 2000
Human breast milk	Canada (wide area)	No data	No data	No data	16.2	No data	Ryan and Patry 2000
Human breast milk	Japan	0.18–0.57	0.09–0.13	0.07–0.18	0.65–1.48*	No data	Ohta et al. 2000

<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 (\*).

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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 level 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. Levels of BDE 47 in all archived samples were below the limit of detection. Recently, 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, 2,2',4,4',5-pentaBDE [BDE 99], 2,2',4,4',6-pentaBDE [BDE 100], BDE 153, 2,2',4,4',5,6'-hexaBDE [BDE 154], and BDE 183) and total PBDEs in maternal and fetal blood samples taken from subjects in Indianapolis, Indiana. Median levels 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 levels for a similar population of Swedish mothers and newborns, the levels for the Indiana population were 20- to 69-fold higher for maternal blood and 30- to 106-fold higher for fetal blood. In fact, the median blood levels 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 levels of lower brominated BDEs (i.e., BDE 47) than are 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).

Six PBDE congeners (2,4,4'-triBDE [BDE 28], 2,2',4,4'-tetraBDE [BDE 47], 2,3',4,4'-tetraBDE [BDE 66], 2,2',4,4',5-pentaBDE [BDE 99], 2,2',4,4',6-pentaBDE [BDE 100], and 2,2',4,4',5,5'-hexaBDE [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 PBDEs were  $2.1 \pm 1.4$  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, 2,2',3,4,4'-hexaBDE (BDE 85), BDE 99, BDE 100, BDE 153, and 2,2',4,4',5,6'-hexaBDE (BDE 154). An increasing temporal trend was also observed; the mean total PBDE concentration (sum of eight congeners) increased

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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).

2,2',4,4'-TetraBDE (BDE 47), 2,2',4,4',5,5'-hexaBDE (BDE 153), 2,2',4,4',5,6'-hexaBDE (BDE 154), 2,2',3,4,4',5',6-heptaBDE (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 levels. 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. 2000). 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 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 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE (BDE 99), and 2,2',4,4',5,5'-hexaBDE (BDE 153) (She et al. 2000). Mean

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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 levels (86 ng/g lipid) were the highest human levels reported to date. Recently, 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 (2,4,4'-triBDE [BDE 28], 2,2',4,4'-tetraBDE [BDE 47], 2,2',3,4,4'-pentaBDE [BDE 85], 2,2',4,4',5-pentaBDE [BDE 99], 2,2',4,4',6-pentaBDE [BDE 100], 2,2',4,4',5,5'-hexaBDE [BDE 153], and 2,2',4,4',5,6'-hexaBDE [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, 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 levels 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. Levels 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 PeBDE and HexaBDE were 0.93 and 1.83 ng/g lipid weight, respectively (Meneses et al. 1999).

**Human Milk.** Recently, Schechter et al. (2003b) reported the first findings on levels 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

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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 2,2',4-triBDE (BDE 17), 2,4,4'-triBDE (BDE 28), 2,3',4,4'-tetraBDE (BDE 66), 2,2',3,4,4'-pentaBDE (BDE 85), 2,2',3,4,4'-pentaBDE (BDE 99), 2,2',4,4',6-pentaBDE (BDE 100), 2,2',3,4,4',5'-hexaBDE (BDE 138), 2,2',4,4',5,5'-hexaBDE (BDE 153), 2,2',4,4',5,6'-hexaBDE (BDE 154), and 2,2',3,4,4',5',6-heptaBDE (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 levels in breast milk from this study were similar to levels found in U.S. blood and adipose tissue lipid from California and Indiana and are 10–100 times greater than human tissue levels in Europe (Schechter et al. 2003b).

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 and home environments. PBDE levels 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. Recently, Lind et al. (2003) reported levels of PBDEs in human breast milk sampled from Uppsala County, Sweden. Total PBDEs, 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE (BDE 99), and 2,2',4,4',6-pentaBDE (BDE 100) levels were 4.01, 2.35, 0.62, and 0.38 ng/g lipid, respectively. In human breast milk from 25 German mothers, the levels of PBDEs ranged from 0.6 to 11 ng/g lipid (de Wit 2002). In 1992, the mean concentration of total PBDEs (sum of 2,4,4-triBDE [BDE 28], -47, -99, -100, 2,2',4,4',5,5'-hexaBDE [BDE 153], and 2,2',3,4,4',5',6-heptaBDE [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

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al. 2002a). 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 levels were analogous to BDE 47 levels in some samples (Ohta et al. 2002a).

***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; Olsson et al. 2000) 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 have 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'-tetrabromodiphenyl ether in herring from five locations along the Swedish coast increased from south to north in the Baltic Sea. No correlation between the concentrations of BDE congeners and methoxy-brominated diphenyl ethers was observed, indicating sources other than PBDEs for these compounds.

## 8.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

***Polybrominated Biphenyls.*** PBBs are no longer produced or used in the United States. Thus, the general population exposure to PBBs will only be from historical releases. For people residing in the lower peninsula of Michigan, especially in the immediate vicinity of the PBB contaminated areas of this region, exposure to PBBs may still be occurring today. However, environmental levels have decreased since the 1970s and current exposure, if any, will be at low levels. For other regions of the United States, the levels of exposure will either be very low or none.

In the past, the general population may have been exposed to PBBs by inhaling contaminated air, ingesting contaminated water and food, and using consumer products containing PBBs. Other than in air in the vicinity of PBB production plants (see Section 8.1), no current or historical data exist that would indicate that PBBs might be present in ambient air. There are no current or historical data on the direct



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exposure of humans to PBBs from water. The general population may have been exposed to low levels of PBBs from the consumption of contaminated foods, but no estimate is available that quantitated this exposure. Historical monitoring and body burden data indicate that low level exposures to PBBs were limited to the population within the state of Michigan (see Section 8.4 and Table 8-5). The level of exposure to PBBs was slightly higher for the people residing in the lower peninsula of Michigan and highest among people residing in the immediate vicinity of the contaminated dairy farms, where people consumed contaminated meat, eggs, and dairy products (see Section 8.4 and Table 8-5). Consumer exposure in the past (plastics containing PBBs may not be in circulation anymore since PBB production ceased in the 1970s) from using PBB-containing plastics (e.g., typewriters, calculators, projector housings, and movie equipment cases) is expected to be very low since the PBBs were incorporated into the plastic and their mobilization could only have occurred under conditions such as combustion (Di Carlo et al. 1978).

Workers involved in the historical production of PBBs, PBB-containing plastics, and PBB-containing plastic products could have been exposed to PBBs via inhalation of dust and vapor and/or dermal contact. Both workplace environmental monitoring and body burden monitoring data of workers (see Table 8-5) (Hesse and Powers 1978; Humphrey and Hayner 1975; Wolff et al. 1979b) indicated that workers in PBB industries were exposed to higher concentrations of PBBs than the general population. Although no evidence has been reported, workers in facilities that combusted or incinerated PBB-containing plastics might have been exposed to higher levels of PBBs.

***Polybrominated Diphenyl Ethers.*** Body burden data indicate that there are low-level exposures to PBDEs for the general population. However, the current understanding of exactly how low levels of certain PBDE isomers/congeners (e.g., 2,2',4,4'-tetraBDE [BDE 47]; 2,2',4,4',5-pentaBDE [BDE 99]; 2,2',4,4',6-pentaBDE [BDE 100]) came to be present in human tissues is insufficient to reach definitive conclusions. Humans appear to be exposed to lower brominated BDEs by ingestion of contaminated foods (e.g., fish) and possibly inhalation of ambient or contaminated air. Dermal exposure to PBDEs could occur by contact with products containing PBDEs such as textiles or polymers. Inhalation exposure could occur from outgassing of PBDEs from electrical appliances and furniture into indoor atmospheres. However, little information is known about the potential exposure from these routes.

In the United States, exposure is evident by the levels of lower brominated PBDEs found in tissues from individuals (see Section 8.4.4). For example, breast adipose samples collected in northern California in the late 1990s contained quantifiable amounts of 2,2',4,4'-tetraBDE (BDE 47), 2,2',4,4',5-pentaBDE

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(BDE 99), and 2,2',4,4',5,5'-hexaBDE (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 8.4.4; Haglund et al. 1997; Meneses et al. 1999). In general, levels of decaBDE in human tissues and body fluids are negligible which indicates that direct exposure to decaBDE congener appears to be low. However, the level current information is insufficient to draw conclusions on whether decaBDE degrades in the environment and if possible degradation products of decaBDE may be important sources of human exposure.

The concentration of total PBDEs in outdoor air ranges from 2 (rural) to 77 (urban)  $\text{pg}/\text{m}^3$  in the United States (Dodder et al. 2000a). Typically, lower brominated congeners (e.g., 2,2',4,4'-tetraBDE [BDE 47]) are predominant which indicates low levels of exposure to these congeners by the general population. Levels of higher brominated congeners (e.g., decaBDE) tend to be below the limit of quantitation. In Sweden, indoor air concentrations of PBDEs in lecture halls, computerized indoor environments, and rooms with electronic devices (e.g., televisions) have low levels of PBDEs (Lindström 1999). Point sources may result in increased concentrations of PBDEs in indoor air. For example, octaBDE has been found in indoor areas that contain electronic products containing PBDEs (e.g., televisions and computers) (Bergman et al. 1997). The release of PBDEs from polymers is dependant on the migration ability of the PBDE molecule through the polymer matrix to the polymer surface where emission is possible (Danish EPA 1999). Because PBDEs are large molecules, migration is expected to occur slowly. No experimental studies were located on the emission rate of PBDEs from plastics. Based on worst case emission factors, the estimated emission of deca-, octa-, and pentaBDEs from plastics are 0.038, 0.054, and 0.39% per year, respectively (Danish EPA 1999).

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 polyurethane foam. Concentrations of tetra- and pentabrominated congeners (BDE 47, 99, and 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 ( $1,700 \text{ pg}/\text{m}^3$ ), 99 ( $852 \text{ pg}/\text{m}^3$ ), and 100 ( $217 \text{ pg}/\text{m}^3$ ), respectively. In this study, the median lower bound estimates of inhalation exposure (i.e., where a congener is below the detection limit, the concentration is assumed to be zero) for BDE 47 (2,2',4,4'-tetraBDE), 99 (2,2',4,4',5-pentaBDE), 100 (2,2',4,4',6-pentaBDE), 153 (2,2',4,4',5,5'-hexa-

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BDE), 154 (2,2',4,4',5,6'-hexaBDE), and total PBDEs were 4.5, 1.2, 0.41, 0.016, 0.040, and 6.9 ng/day, respectively (Harrad et al. 2004).

Consumption of food is expected to be the major route of exposure in humans (Lindström 1999).

Consumption of fish has been associated with elevated levels 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 vegetarian 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 (2,2',4,4'-tetraBDE), 99 (2,2',4,4',5-pentaBDE), 100 (2,2',4,4',6-pentaBDE), 153 (2,2',4,4',5,5'-hexaBDE), 154 (2,2',4,4',5,6'-hexaBDE), and total PBDEs were 46.4, 42.6, 0, 0, 0, and 90.5 ng/day, respectively (Harrad et al. 2004). 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 levels 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 6-6); 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 (Sjödin et al. 1999a, 2001a). The electronics dismantling plant had the highest concentrations of PBDEs, with mean concentrations of 2.5 pmol/m<sup>3</sup> (1.25 ng/m<sup>3</sup>) for 2,2',4,4'-tetraBDE (BDE 47), 4.6 pmol/m<sup>3</sup> (2.6 ng/m<sup>3</sup>) for 2,2',4,4',5-pentaBDE (BDE 99), 6.1 pmol/m<sup>3</sup> (3.93 ng/m<sup>3</sup>) for 2,2',4,4',5,5'-hexaBDE (BDE 153), 26 pmol/m<sup>3</sup> (18.8 ng/m<sup>3</sup>) for 2,2',3,4,4',5',6-heptaBDE (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–

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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 (Sjödín et al. 1999a, 2001a).

2,2',4,4'-TetraBDE (BDE 47), 2,2',4,4',5,5'-hexaBDE (BDE 153), 2,2',4,4',5,6'-hexaBDE (BDE 154), 2,2',3,4,4',5',6-heptaBDE (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) (Sjödín 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 levels. 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. 2000). 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 Sjödín 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.

## 8.6 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 5.7 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).

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***Polybrominated Biphenyls.*** Infants who consume breast milk may have had a higher exposure to PBBs than children who drink formula milk, especially children exposed during the Michigan episode (see Section 8.4.4). No additional information was found in the literature about the exposure of children to PBBs (WHO 1994b).

***Polybrominated Diphenyl Ethers.*** Infants who consume breast milk may have a higher exposure to lower brominated BDEs than children who drink formula milk (see Section 8.4.4). Exposure of neonates is evident due to the presence of lower brominated BDEs (e.g., 2,2',4,4'-tetraBDE [BDE 47]) in breast milk and placental tissue. The PBDEs detected in breast milk are the tri- to hexaBDEs, but not the hepta- to decaBDEs (LaKind and Berlin 2000), which are the same congeners found in bioaccumulation studies with fish and other mammals. Schechter et al. (2003b) reported the first findings on levels 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 levels of PBDEs in breast milk from this study were 10–100 times greater than human tissue levels in Europe (Schechter et al. 2003b). Levels of lower brominated BDEs in the breast milk of Swedish women shows an exponentially increasing trend in exposure since the 1970s, with concentrations of lower brominated BDEs in breast milk doubling every fifth year (Lindström 1999). The concentrations of lower brominated BDEs in breast milk are increasing exponentially from about 300 pg/g lipid in 1976 to about 4,000 pg/g lipid in 1997 in Swedish women (Norön and Meironyté 2000). The sum of four PBDE congeners (2,4,4'-triBDE [BDE 28], BDE 47, 2,2',4,4',5-pentaBDE [BDE 99], and decaBDE [BDE 209]) was between 0.88 and 5.89 ng/g lipid in breast milk and between 1.00 and 4.40 ng/g lipid in placental tissue of 11 Finnish women (Strandman et al. 2000). The four highest concentrations of total PBDEs were found in nulliparous women. In 2001, Mazdai et al. (2003) determined the concentration of six PBDE congeners (BDE 47, BDE 99, 2,2',4,4',6-pentaBDE [BDE 100], BDE 153, 2,2',4,4',5,6'-hexaBDE [BDE 154], and 2,2',3,4,4',5',6-heptaBDE [BDE 183]) and total PBDEs in maternal and fetal blood samples taken from subjects in the United States. Median levels 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. Concentrations of PBDEs in fetal and maternal serum were comparable. Thus, the authors concluded that measurement of maternal serum PBDE levels could be used to determine fetal exposure to PBDEs. Concentrations of PBDEs were determined in children from Norway (Thomsen et al. 2002b). Samples were collected between the period of 1975–2002. Children ages 0–4 years had congener levels of BDE 28, BDE 47, BDE 100, BDE 99, 2,2',4,4',5,5'-hexaBDE (BDE 153), and BDE 154 at 0.26, 6.2,

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1.7, 1.6, 1.5, and 0.45 ng/g lipid, respectively; while children 4–14 years old had levels of BDE 28, BDE 47, BDE 100, BDE 99, BDE 153, and BDE 154 at 0.20, 2.0, 0.66, 0.37, 0.86, and 0.39 ng/g lipid, respectively. No additional information was found in the literature about the exposure of children to PBDEs (WHO 1994a).

### 8.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

***Polybrominated Biphenyls.*** The production of PBBs ceased in 1979, and the usable life of the plastics containing PBBs has expired. Therefore, these plastics are probably no longer in circulation. At the present time and in the near future, populations potentially exposed to low levels of PBBs are those living near hazardous waste sites in which the PBB-containing plastics have been disposed and the residents in and around the contaminated farms in Michigan. The lifetime of PBBs in soil is on the order of years (Jacobs et al. 1978), and the levels of PBBs in fish caught in contaminated waters have declined slowly (Hesse and Powers 1978). Therefore, concentrations of residual PBBs in soil and streams in the vicinity of PBB-containing hazardous waste sites, PBB production facilities, and contaminated farm areas are expected to remain above background levels for many years. The sources of potential exposure to PBBs for residents in these areas are consumption of contaminated meat and dairy products obtained from herds grazing over contaminated soil and consumption of fish from nearby contaminated streams. PBB contamination has triggered the issuance of one human health advisory in the state of Michigan. As of September 30, 1993, recreational and subsistence fishermen who consume appreciably higher amounts of fish caught in the Pine River downstream from St. Louis in Gratiot and Midland Counties (RTI 1993) may be exposed to above-average levels of PBBs associated with dietary intake (EPA 1993). The body burden for PBBs in residents of contaminated areas has been higher than in the general population (Brilliant et al. 1978; Cordle et al. 1978; Eyster et al. 1983; Humphrey and Hayner 1975; Kimbrough 1987; Lambert et al. 1990; Landrigan et al. 1979; Wolff et al. 1982). Therefore, babies breast fed by exposed mothers in the contaminated areas may also be at higher risk (Jacobson et al. 1989).

***Polybrominated Diphenyl Ethers.*** Subsistence fishermen who consume PBDE-contaminated fish and Native Americans who reside in the Arctic region and consume whale and seal blubber may have a higher risk of exposure to lower brominated PBDEs (WHO 1994a). Other populations with high exposure levels to PBDEs involve occupational exposures (see Section 8.6). No other information was located that identified specific populations with higher exposure levels to PBDEs.

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**8.8 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 PBBs and PBDEs are 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 PBBs and 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 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.

**8.8.1 Identification of Data Needs****Physical and Chemical Properties.**

***Polybrominated Biphenyls.*** Many of the relevant physical and chemical properties of the PBBs are not available (see Table 6-3). More data on the physical and chemical properties of hexabromobiphenyl are available relative to octabromo- and decabromobiphenyl. Even in the case of hexabromobiphenyl, not all relevant data are available, and the quality of data is questionable because the properties of FireMaster BP-6 have been reported as the properties of hexabromobiphenyls. More importantly, very limited data are available on the physical and chemical properties for the individual congeners of hexabromo-, octabromo-, and decabromobiphenyl. The absence of such important data as  $K_{oc}$ , vapor pressure, and Henry's law constant, is a major impediment in the prediction of the environmental fate and transport of PBBs.

***Polybrominated Diphenyl Ethers.*** Many of the relevant physical and chemical properties of the PBDEs are available (see Table 6-4). Very limited data are available on the physical and chemical properties for the individual congeners (Braekvelt et al. 2003; Tittlemier et al. 2002). Important data, such as  $K_{oc}$ , vapor pressure, and Henry's law constant, are necessary for the prediction of the environmental fate and transport of PBDEs.

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**Production, Import/Export, Use, Release, and Disposal.**

***Polybrominated Biphenyls.*** The production of all PBBs in the United States stopped in 1979 (IARC 1986). Data on the past production, import/export, and use of PBBs are available (Neufeld et al. 1977). In the past, PBB-containing plastic was used in consumer products, but the useful life of these products may have ended (Di Carlo et al. 1978; Neufeld et al. 1977), and these products are probably no longer in circulation. In the workplace, the environmental media contaminated by PBBs were air, water, and soil (DeCarlo 1979). Outside of the workplace, soil is expected to be the medium with significant contamination due to disposal of solid waste from production plants and disposal of PBB-containing plastics in landfills (Neufeld et al. 1977). Although it is known that PBB-containing plastics may have been disposed in landfills (Di Carlo et al. 1978), the amount that may have been incinerated is not known. No data were located from studies that determined the efficiency of incineration as a method of disposal of PBBs present in the neat form in industrial wastes or in plastics. Environmental regulations regarding the manufacture and disposal of PBBs have been established (EPA 1988a). According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit chemical release and off-site transfer information to the EPA. The Toxics Release Inventory (TRI), which contains this information for 1999, became available in 2002. This database will be updated yearly and should provide a list of industrial production facilities and emissions.

***Polybrominated Diphenyl Ethers.*** Production and use data are available for PBDE commercial mixtures (BSEF 2003). PBDEs are used as additive flame retardants in thermoplastics at levels ranging from 5 to 30% by weight (EU 2001). The commercial pentaBDE product is used predominantly (95–98%) for flame-retardant purposes as an additive in consumer products manufactured by the furniture industry (ENVIRON 2003a). The commercial octaBDE is used by the plastics industry as an additive flame retardant for manufactured products. It is used almost exclusively as a flame retardant for acrylonitrile-butadiene-styrene (ABS) terpolymers used in computer casings and monitors (ENVIRON 2003b). The commercial decaBDE product is an additive flame retardant used in a variety of polymer applications. Industry information indicates that decaBDE is used at loadings of 10–15% weight in polymers and is always used in conjunction with antimony trioxide (EU 2002). The Great Lakes Chemical Corporation recently announced that it is voluntarily phasing out production of pentaDBEs and octaDBEs by the end of 2004 (Tullo 2003). In the United States, waste disposal of PBDE-containing consumer products is described as transfers to disposal (landfill), recycling, energy recovery (incineration), or publicly owned treatment works (POTWs) (Darnerud et al. 2001). More information for PBDEs is needed on import/export, release, and disposal. In particular, the mechanism by which PBDEs are leaving the products in which they are used and entering the environment is not understood. Possibilities include



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disintegration of plastic products in particulates contaminated with PBDEs or volatilization of PBDEs from the plastic itself (Hites 2004). In the United States, waste disposal of PBDE-containing consumer products is described as transfers to disposal (landfills), recycling, energy recovery (incineration), or POTWs (Darnerud et al. 2001). Soil should be a medium of significant contamination due to disposal of PBDE-containing plastics in landfills. The commercial decaBDE and octaBDE products are used in hard dense plastics from which migration would be very difficult (BFRIP 2002). Although it is known that PBDE-containing plastics are disposed in landfills, information on the recycling of PBDE-containing plastics and the amounts of PBDE-containing plastics that are incinerated is not known. Additional information of levels of PBDEs and PBDD/PBDF from incineration of PBDE-containing plastics would be useful in determining exposure to the general population.

**Environmental Fate.**

***Polybrominated Biphenyls.*** Information regarding the environmental fate of PBBs in air was not located in the literature. The data about the fate of PBBs in air are important for the prediction of transport characteristics of these compounds in air. Photolysis of the PBBs will produce debrominated products in proton-donating organic solvents (Ruzo and Zabik 1975; Ruzo et al. 1976), but there is less certainty about the importance of photolysis of PBBs in water (Norris et al. 1973; Ruzo et al. 1976). PBBs will partition from the aquatic phase to sediment and suspended solids in water (Hesse and Powers 1978). PBBs will bioconcentrate in aquatic organisms, but the BCF may decrease as the bromine substitution exceeds six (Gobas et al. 1989; Opperhuizen et al. 1985; Zitko 1979; Zitko and Hutzinger 1976). However, the difference in the reported BCF values for hexabromobiphenyl among different investigators is vast (Gobas et al. 1989; Hesse and Powers 1978; Opperhuizen et al. 1985; Veith et al. 1979). PBBs will remain strongly sorbed to soil (Filonow et al. 1976; Griffin and Chou 1981a, 1981b) and will persist in soil because of the lack of suitable degradation pathways (Jacobs et al. 1978). The translocation of PBBs from soil to upper parts in plants was not observed, and the transfer of PBBs from soil to carrot roots was found to be minor (Jacobs et al. 1976, 1978). A recent article by de Boer et al. (1998) found PBBs in deep ocean marine mammals, which suggests that PBBs may be transported globally. More monitoring data for PBBs in the environment are needed to verify the possible global transport of PBBs. Since the toxicity and the environmental fate of PBBs depends on specific PBBs congeners, development of more data regarding congener-specific fate and transport of PBBs in the environment are needed.

***Polybrominated Diphenyl Ethers.*** 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

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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, 2003). Based on a very limited number of studies, biodegradation does not appear to be significant for PBDEs commercial mixtures (EU 2002, 2003). 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, 2003). More studies are needed to determine conclusively if commercial PBDE mixtures, such as decaBDE, are degraded to lower brominated congeners (e.g., 2,2',4,4'-BDE [BDE 47]), which appear to bioaccumulate in fish, animals, and humans (see Sections 8.4). 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.**

***Polybrominated Biphenyls.*** Available information regarding the rate of absorption of PBBs following inhalation, oral, or dermal contact is discussed in the Toxicokinetics Section (Section 5.4). Although no data on the bioavailability of PBBs from inhalation of contaminated air, or ingestion of or dermal contact with water, or inhalation of or dermal contact with soil are available, the bioavailabilities from these routes of exposure are expected to be far less than 100% because these compounds strongly sorb to particulate matter and soil. The estimated bioavailability of higher brominated biphenyls is expected to be even lower than the less brominated biphenyls due to stronger sorption characteristics of the former compounds. The estimated bioavailability of PBBs by farm animals from ingestion of contaminated soil was 56–65% (Fries 1985a). Also, studies on many persistent halogenated aromatic compounds clearly show that they become progressively less bioavailable with time (Alexander 2000). Often, three-fourths or more of the concentration of such compounds is not bioavailable. Information on the possibility of the very low bioavailability of PBBs is needed.

***Polybrominated Diphenyl Ethers.*** The absorption and distribution of PBDEs as a result of inhalation, ingestion, and dermal exposure are discussed in Sections 5.3.1, 5.3.2, and 5.3.3. Studies that describe the bioavailability of PBDEs commercial mixtures and congeners from ambient air, surface water, and groundwater, or soil do not exist. Essentially no good data exist on the adsorption of PBDEs commercial mixtures and congeners with the exception of decaBDE (BFRIP 2002). Studies on many persistent halogenated aromatic compounds clearly show that they become progressively less bioavailable with time (Alexander 2000). Often, three-fourths or more of the concentration of such compounds is not

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bioavailable. Information on the possibility of the very low bioavailability of PBDEs is needed. Studies determining the effect of particle size and organic matter content on the bioavailability of PBDEs from soil and the role of microparticle-sorbed PBDEs on the bioavailability of PBDEs from drinking water are needed. Such studies would be useful in assessing the health effects of PBDEs on people living near hazardous waste sites.

**Food Chain Bioaccumulation.**

***Polybrominated Biphenyls.*** PBBs do not readily translocate from soil to plants via root uptake (Jacobs et al. 1976, 1978). Therefore, PBBs may not bioconcentrate in plants. However, plant uptake data are limited, and it will be helpful to develop additional plant uptake data. Brominated biphenyls with bromine substitution 6 or less will bioconcentrate in aquatic organisms (Gobas et al. 1989; Norris et al. 1973; Opperhuizen et al. 1985; Zitko 1979; Zitko and Hutzinger 1976). PBBs are preferentially stored in the adipose tissue of animals (Kimbrough 1987). Although PBBs have been detected in fish-eating birds and predatory animals from the consumption of contaminated food (Heinz et al. 1983, 1985; Hesse and Powers 1978), no systematic study was located that analyzed the biomagnification potential in predators resulting from consumption of contaminated food.

***Polybrominated Diphenyl Ethers.*** An abundance of monitoring data illustrate the uptake of lower brominated diphenyl ethers by aquatic organisms, which results in bioconcentration (see Section 8.4.4; Hardy 2002b). Congener components of the pentaBDE commercial product tend to bioconcentrate to different extents. DecaBDE and octaBDE commercial products appear to not bioconcentrate, bioaccumulate, or biomagnify (Hardy 2002b). The limited existing data indicate that lower brominated BDE congeners (e.g., pentaBDE commercial mixtures) bioaccumulate in aquatic and terrestrial food chains and biomagnify in predators due to consumption of contaminated prey. Bioaccumulation of PBDEs in the aquatic food web is inversely related to the degree of bromination (Bureau et al. 2000; Jansson et al. 1993). More information on bioaccumulation and biomagnification of PBDE and its congeners is needed in assessing human health risks.

**Exposure Levels in Environmental Media.**

***Polybrominated Biphenyls.*** Only limited data on the levels of PBBs in ambient air are available (DeCarlo 1979). Data are available on the levels of PBBs in effluent water from manufacturing plants, in river water, stream sediment, and soil in the vicinity of the plants, in sludge of a waste treatment plant, and in groundwater of a landfill site (Hesse and Powers 1978; Shah 1978). No data on the level of PBBs

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in drinking water from the contaminated sites were located. No estimate on the human intake of PBBs from any of the various environmental media was located in the literature.

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

***Polybrominated Diphenyl Ethers.*** Information on the relative importance of different routes of exposure to PBDEs is limited especially in the United States. Atmospheric concentrations of PBDEs tend to be dominated by lower brominated congeners, e.g., 2,2',4,4'-pentaBDE (BDE 47) (Dodder et al. 2000a; Strandberg et al. 2001). 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. Due to the hydrophobic nature of PBDEs, this class of compounds has not been detected in water to any significant extent. BDE 47 was detected at low concentrations in Lake Ontario surface waters (Luckey et al. 2001). Although levels are predicted to be low, monitoring data on PBDE concentrations in finished drinking water nationwide would be helpful. No information was located on the ambient environmental concentrations of PBDEs in soils in the United States or other parts of the world. Hale et al. (2002) reported the concentrations of PBDEs in soil samples collected in the vicinity of a polyurethane foam manufacturing facility, which are higher than expected in rural and potentially urban areas. 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); temporal trends suggest that concentrations of PBDEs in sediments are increasing. Information about the concentrations of PBDEs in food stuffs is very limited (Bocio et al. 2003; Huwe et al. 2002a; Ohta et al. 2002a), especially in the United States. Data on the concentrations of PBDEs in foods, collected using a market-basket approach, are needed to determine concentrations of PBDEs in foods consumed by the general population. Data on the PBDE concentrations in foods grown in contaminated areas, particularly in the vicinity of hazardous waste sites, are also needed. Data on congener-specific PBDE analysis of food, especially plant products, would be useful. Monitoring data indicated that the levels of PBDEs are increasing in aquatic organisms with higher concentrations near point sources (Alaee et al. 1999; Dodder et al. 2000a; Johnson and Olson 2001; Loganathan et al. 1995; Luross et al. 2000). Additional monitoring data on environmental levels 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.

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**Exposure Levels in Humans.**

***Polybrominated Biphenyls.*** Body burden data indicate that low-level exposures to PBBs have occurred for people in the state of Michigan. No recent information about average daily intake of PBBs was located. The levels of PBBs in human tissue and body fluids, such as blood, serum, adipose tissue, breast milk, feces, cord blood, biliary fluid, and placenta, of people in the state of Michigan have been extensively studied (Brilliant et al. 1978; Cordle et al. 1978; Eyster et al. 1983; Humphrey and Hayner 1975; Lambert et al. 1990; Landrigan et al. 1979; Wolff et al. 1982). However, no recent data are available. Data on the levels of PBBs in tissues and body fluids of residents in the vicinity of sites of industrial discharge of PBB wastes were not located. Updated information would be useful to understand current exposure levels of people in the state of Michigan to PBBs. This information is necessary for assessing the need to conduct health studies on these populations.

***Polybrominated Diphenyl Ethers.*** Body-burden data indicate that there are low-level exposures to lower brominated PBDEs for the general population. The absence of decaBDE (BDE 209) in the ambient population is likely the result of analytical bias since most studies of the ambient population did not include BDE 209 as one of the analytes of interest. Thus, future studies that include the analysis of BDE 209 in body tissues and fluids would be useful (Hites 2004). Information about the average daily intake of PBDEs is limited to populations living in Sweden (Bergman et al. 1999; Lindström 1999; WHO 1994a). PBDE levels are reported in the current literature for blood, breast milk, and adipose tissue of the general population and occupationally exposed individuals (WHO 1994a). Limited information on the levels of PBDEs in body tissues and fluids from individuals living in the United States have been located (Mazdai et al. 2003; Patterson et al. 2000; Petreas et al. 2002, 2003; Schechter et al. 2003b; Sjödin et al. 2001). These studies indicate that levels of lower brominated BDEs in body fluids are a factor of 10–100 higher for individuals living in the United States compared to individuals living in other regions of the world (e.g., Europe). Limited surveys have ever been conducted in the United States to evaluate the trend of PBDE concentrations in human tissues over the years. It would be helpful to develop a database of information on congener-specific PBDE levels in tissues of exposed and control cases for studying clinical and epidemiological outcomes. In particular, a comprehensive study that monitors congener-specific concentrations in fish species and relates them directly to congener levels in human tissue would be extremely useful. Monitoring data indicate 2,2',4,4'-tetraBDE (BDE 47) and 2,2',4,4',5,5'-hexaBDE (BDE 153) are relatively higher in human samples than in the commercial pentaBDE product and that 2,2',4,4',5-pentaBDE (BDE 99) is relatively lower. The cause of this difference is not known. It may be due to its relatively higher vapor pressure of BDE 99, or it may be due to the selective environmental elimination of BDE 99, a process that has been observed in some biota (Hites 2004). Additional studies

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that examine these trends would be useful. 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.**

***Polybrominated Biphenyls.*** Children may be exposed to PBBs by a variety of exposure pathways. Levels will be highest for children living in the vicinity of the area affected by the Michigan contamination episode. The most important pathway appears to be consumption of contaminated mother's milk (see Section 8.4.4). More data are needed on the levels of PBB exposure in nursing women from consumption of fish and from those of the general population. Exposure and body burden studies related to consumption of fish in the U.S. population are needed to determine exposure levels, particularly in children of recreational and subsistence fishers. Information related to the exposure of children living near hazardous waste sites is also needed. In particular, information is needed that is related to the potential for children to be exposed to PBBs bound to soil and dust particles through pica or unintentional hand-to-mouth activity within homes located in these areas. Quantitative information regarding the bioavailability and amount of PBBs that children are exposed to through contact with contaminated soils are unavailable. Therefore, any information concerning this subject would be useful in evaluating children's exposure. Additional information on weight-adjusted intakes would be helpful for determining the health risks for young children. Infants and young children consume a greater amount of food per kilogram of body weight and, therefore, may have a proportionately greater exposure to PBBs than adults.

***Polybrominated Diphenyl Ethers.*** Children may be exposed to PBDEs by a variety of exposure pathways. The most important pathway appears to be consumption of contaminated foods, particularly fish (Bergman et al. 1999; Lindström 1999; WHO 1994a). Children can also be exposed to PBDEs from mother's milk (LaKind and Berlin 2000; Lindström 1999; Norön and Meironyté 2000; Schecter et al. 2003b; Strandman et al. 2000; WHO 1994a). More data are needed on the levels of PBDEs exposure in nursing women, from occupational situations, from consumption of fish, and from those of the general population. Exposure and body-burden studies related to consumption of fish in the U.S. population are needed to determine exposure levels, particularly in children of recreational and subsistence fishers. Exposure and body-burden studies are also needed in Native American communities that consume high levels of game and marine mammals. Information related to the exposure of children living near hazardous waste sites is also needed. In particular, information is needed that is related to the potential for children to be exposed to PBDEs bound to soil and dust particles through pica or unintentional hand-

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to-mouth activity within homes located in these areas. Quantitative information regarding the bioavailability and amount of PBDEs that children are exposed to through contact with contaminated soils is unavailable. Therefore, any information concerning this subject would be useful in evaluating children's exposure. Additional information on weight-adjusted intakes would be helpful for determining the health risks for young children, particularly those in Native American populations. Infants and young children consume a greater amount of food per kilogram of body weight and, therefore, may have a proportionately greater exposure to PBDEs than adults.

Child health data needs relating to susceptibility are discussed in Section 5.12.2 Identification of Data Needs: Children's Susceptibility.

**Exposure Registries.**

***Polybrominated Biphenyls.*** The Michigan Department of Community Health (MDCH), together with the Centers for Disease Control and Prevention (CDC) and three other federal agencies, began a major study to assess the health effects of PBBs after the Michigan contamination episode. A health questionnaire and blood samples were collected from people affected by the feed-contamination incident. MDCH had the responsibility to analyze several thousand samples for PBB from 1975 to 1978. MDCH continues contact with this cohort, updates health questionnaires, and collects blood samples to be analyzed (MDCH 2002).

***Polybrominated Diphenyl Ethers.*** No exposure registries for PBDEs were located. This substance is not currently one of the compounds for which a subregistry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for subregistries 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.

**8.8.2 Ongoing Studies**

***Polybrominated Biphenyls.*** A search in Federal Research in Progress (FEDRIP 2002) did not identify ongoing research studies for PBBs.

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***Polybrominated Diphenyl Ethers.*** A search of FEDRIP (2003) did not identify ongoing research studies for PBDEs. However, EPA has funded work in the Science To Achieve Results (STAR) Research Grants program, as well as regional and intramural efforts (EPA 2004). In addition, the National Toxicology Program (NTP) has committed to performing studies on the commercial pentaBDE and octaBDE products and their congener components (NTP 2004).