Polybrominated Diphenyl Ethers in the Environment and in People: A Meta-Analysis of Concentrations

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Polybrominated diphenyl ethers (PBDEs) are used as flame retardants in many types of consumer products. Perhaps as a result of their widespread use and their lipophilicity, these compounds have become ubiquitous in the environment and in people. This review summarizes PBDE concentrations measured in several environmental media and analyzes these data in terms of relative concentrations, concentration trends, and congener profiles. In human blood, milk, and tissues, total PBDE levels have increased exponentially by a factor of ~100 during the last 30 yr; this is a doubling time of ~5 yr. The current PBDE concentrations in people from Europe are ~2 ng/g lipid, but the concentrations in people from the United States are much higher at ~35 ng/g lipid. Current PBDE concentrations in marine mammals from the Canadian Arctic are very low at ~5 ng/g lipid, but they have increased exponentially with a doubling time of ~7 yr. Marine mammals from the rest of the world have current PBDE levels of ~1000 ng/g lipid, and these concentrations have also increased exponentially with a doubling time of ~5 yr. Some birds' eggs from Sweden are also highly contaminated (at ~2000 ng/g lipid) and show PBDE doubling times of ~6 yr. Herring gull eggs from the Great Lakes region now have PBDE concentrations of ~7000 ng/g lipid, and these levels have doubled every ~3 yr. Fish from Europe have ~10 times lower PBDE concentrations than fish from North America. From these and other data, it is clear that the environment and people from North America are very much more contaminated with PBDEs as compared to Europe and that these PBDE levels have doubled every 4−6 yr. Analyses of the relative distributions of the most abundant PBDE congeners (using category averages and principal component analysis) indicated that these patterns cannot yet be used to assign sources to these pollutants.

Introduction. Polybrominated diphenyl ethers (PBDEs) save lives by serving as flame retardants in a wide variety of commercial and household products. For example, polyurethane foam, which is widely used in upholstered furniture, is flammable unless it is treated with suitable flame retardants such as PBDEs. In fact, some polyurethane foam is treated with 10−30 wt % of PBDEs to make this material safe for home use (*1*). Because many states and the federal government now have regulations requiring most household products, such as mattresses and electronics, to be flame resistant (*2*), PBDEs have become an important commercial substance. Not surprisingly, the use of PBDEs has increased over the years, and annual sales are now ~70 000 t (t = metric ton) (*3*).

PBDEs are commercially available as three products, two of which are mixtures of several congeners (*4*). The so-called penta-product contains 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′-tetrabromo-diphenyl ether (BDE-154), in a ratio of about 9:12:2:1:1 (*5, 6*). The octa-product contains several hexa- to nonabrominated congeners, and the deca-product is almost entirely composed of decabromodiphenyl ether (BDE-209) (*4*). (For convenience, the congeners are numbered from 1 to 209 using the same IUPAC scheme used for polychlorinated biphenyls.) Like most commercial chemical mixtures, the compositions of these products vary with manufacturer and with the year in which they were produced. Table 1 gives the market demand for these products in 1999 and in 2001. Notice that >95% of the penta-product is now used in the Americas.

Despite their societal benefits, PBDEs seem to be migrating from the products in which they are used and entering the environment and people. PBDEs are now ubiquitous; they can be found in air, water, fish, birds, marine mammals, and people, and in many cases, the concentrations of these compounds are increasing over time. Several reviews on the presence of PBDEs in the environment have been published over the past few years (*7*−*10*), including a complete issue of *Environment International* (*11*). Many of these reviews have been exhaustive in scope (including both chemistry and toxicology), but none of them have analyzed the published concentration data in detail. This paper is, therefore, a meta-analysis of the available PBDE concentration data. Recommendations for future research will also be presented.

Strategy of This Review. Most of the literature on PBDEs in the environment or in people, published up until early August of 2003, was acquired and classified. Papers on the toxicology or metabolism of PBDEs are not included here. Because PBDEs are present as mixtures of congeners, only papers that reported specific congener information are included. Unfortunately, the set of congeners reported in the various papers was not consistent. Some papers reported on as few as two or three congeners (BDE-47, -99, or -153), while some reported on 10 or more congeners. As a compromise between these two extremes, only congeners BDE-47, -99, -100, -153, -154, and -209 are included here because the concentrations of most of these congeners were reported most of the time. From this congener-specific database, the units were made consistent (for example, pmol/g wet wgt was converted to ng/g lipid, using reported lipid concentrations), geometric averages were calculated for some sets of samples, values for total PBDE (∑PBDE)

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TABLE 1. PBDE Market Demand (in t) in 1999 and 2001 (*3*)

	Americas	%[a]	Europe	%	Asia	%	rest of world	%	total
				1999					
deca-BDE	24 300	44	7 500	14	23 000	42			54 800
octa-BDE	1 375	36	450	12	2 000	52			3 825
penta-BDE	8 290	98	210	2					8 500
total	33 965	51	8 160	12	25 000	37			67 125
				2001					
deca-BDE	24 500	44	7 600	14	23 000	41	1 050	2	56 150
octa-BDE	1 500	40	610	16	1 500	40	180	5	3 790
penta-BDE	7 100	95	150	2	150	2	100	1	7 500
total	33 100	49	8 360	12	24 650	37	1 330	2	67 440

[a] Percent of total (last column).

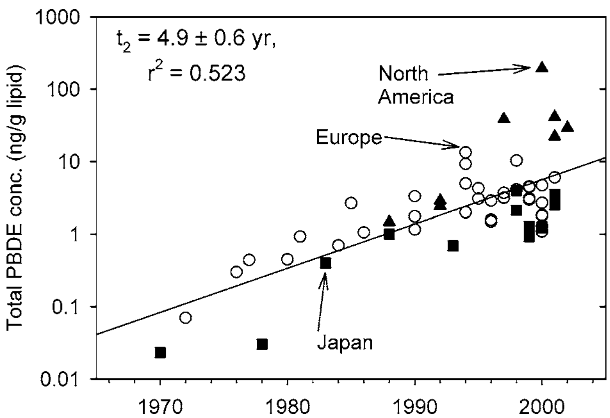

FIGURE 1. Total PBDE concentrations (∑PBDE) in human blood, milk, and tissue (in ng/g lipid) shown as a function of the year in which the samples were taken; see Table 2. The three symbol types indicate the location from which the samples were collected. The overall regression is shown.

concentrations were calculated, and congener patterns (the percent of the total due to each congener) were determined. In this case, geometric averaging was used because environmental concentration data are log-normally distributed. The year the samples were taken was also tabulated; in those cases (which were numerous) where this information was not given, it was assumed that the samples were taken 1.5 yr before the publication date of the paper.

Human Samples. A large number of samples from people have been analyzed for PBDEs. Table 2 gives these concentrations in ambient human tissue, blood (usually serum), and milk; these data are sorted by the year in which the sample was taken. In this case, "ambient" means samples from people who were not known to have been occupationally exposed. ∑PBDE concentrations range from <0.03 ng/g lipid for adipose tissue from Japan in 1970 to >190 ng/g lipid for milk from Austin and Denver in the United States in 2000. A plot of all these concentrations versus sampling year (see Figure 1) shows an exponential increase with a doubling time of ~5 yr. In general, the PBDE concentrations in people have increased by a factor of ~100 during the last 30 yr. The regression of these data as a function of year is good despite the disparate sample types (blood, milk, and tissue), the different continents of origin, and the various congeners measured. This analysis shows that the North American samples are always above the regression line (in recent years by a factor of >10) and that the Japanese samples are usually below the regression line (by a factor of ~5). This suggests that people in the United States are exposed to higher levels of these PBDE congeners than are Europeans and that the Japanese are exposed to less than the Europeans.

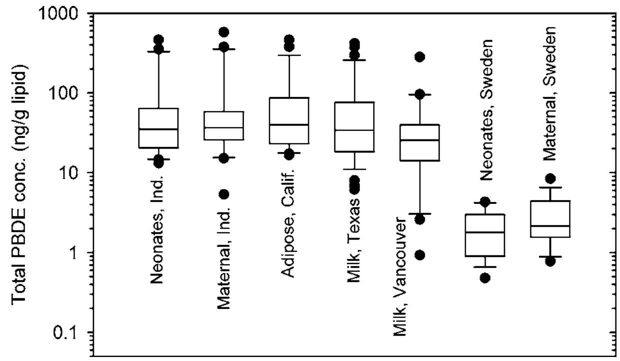

FIGURE 2. Distributions of the ∑PBDE concentrations in human blood, milk, and tissue from different locations. The horizontal lines indicate the 10th, 25th, 50th, 75th, and 90th percentiles of the distributions. Outliers are shown as dots. The data are taken from the following sources: neonatal and maternal blood from Indiana (*25*); adipose tissue from California (*38*); milk from Texas (*50*); milk from Vancouver, Canada (*49*); and neonatal and maternal blood from Sweden (*44*).

The variations in PBDE concentrations among populations can be examined in more detail using those published data that were given in sufficient detail to determine the statistical distribution of the concentrations. Figure 2 shows data from seven such data sets; this figure shows the 10th and 90th percentiles (the "whiskers") and the 25th and 75th percentiles (the boxes); the median and outliers are also shown. The median concentrations in the four United States samples (neonatal and maternal blood, tissue, and milk) are all about the same at ~35 ng/g lipid; the median concentration in the Vancouver (Canada) milk samples is somewhat lower than the United States value; and the Swedish neonatal and maternal blood concentrations are much lower at ~2 ng/g lipid. In other words, the concentrations of PBDEs are ~20 times higher in people from the United States as compared to people from Europe. It is also interesting to note that all samples show a few high outliers. In North America, these outlier concentrations usually exceed 300 ng/g lipid, which is ~10 times the average level. The reason for these outliers is not yet clear (*12*). In general, it is likely that the reason that the North American samples show such relatively high concentrations is the focused use of the penta-BDE product in North America. As shown in Table 1, the United States and Canada used 7100 t/yr of the penta-BDE product, which was 95% of the world's production (*3*).

The average congener distribution in the ambient human samples is shown in Figure 3; this information should not be taken too literally because, in some studies (see Table 2), only a few congeners were measured. The most notable missing congener was BDE-209, decabromodiphenyl ether, which was rarely measured in the ambient human samples. (It was measured more frequently in occupationally exposed people; see below.) Given this measurement bias, one can only use literature-based congener distribution patterns for approximate comparisons. In this case, the congener pattern from the ambient human samples can be compared with that of the penta-BDE product (*5*), the presumptive source of these compounds in people (see Figure 3). It is clear that BDE-47 and-153 are relatively higher in the human samples than in the penta-product and that BDE-99 is relatively lower. The cause of this difference is not known. It may be due to the enhanced environmental availability of BDE-47 due to its relatively higher vapor pressure, or it may be due to the selective environmental elimination of BDE-99, a process that has been observed in some biota (*13*).

Given that some PBDEs are used in plastics that end up in consumer electronics, one would expect that workers involved in assembling or disassembling these products

TABLE 2. PBDE Concentrations and Congener Distributions in Ambient Human Samples (in ng/g Lipid)

location	type	date	reps[a]	47	99	100	153	154	∑PBDE	% 47	% 99	% 100	% 153	% 154	ref
Japan	tissue, adipose	1970	10	0.017	0.004	0.002			0.023	73.9	17.0	9.1			(*26*)
Sweden	milk	1972	P[b]	0.06			0.01		0.07	85.7			14.3		(*27*)
Sweden	milk	1976	P	0.18	0.04	0.05	0.02	0.01	0.30	60.0	13.3	16.7	6.7	3.3	(*27*)
Norway	blood	1977	34	0.25	0.09		0.10		0.44	56.8	20.5		22.7		(*28*)
Japan	milk	1978	1	0.03					0.03	100.0					(*29*)
Sweden	milk	1980	P	0.28	0.09	0.04	0.03	0.01	0.45	62.2	20.0	8.9	6.7	2.2	(*27*)
Norway	blood	1981	17	0.32	0.13	0.079	0.18	0.22	0.93	34.4	14.0	8.5	19.4	23.7	(*28*)
Japan	milk	1983	1	0.26	0.04	0.02	0.07	0.01	0.40	65.0	10.0	5.0	17.5	2.5	(*29*)
Sweden	milk	1984	P	0.49	0.08	0.06	0.05	0.02	0.70	70.0	11.4	8.6	7.1	2.9	(*27*)
Germany	blood	1985	16	1.86	0.23	0.20	0.37		2.66	69.9	8.6	7.5	14.0		(*30*)
Norway	blood	1986	24	0.41	0.13	0.12	0.14	0.26	1.06	38.7	12.3	11.3	13.2	24.5	(*28*)
U.S.	blood	1988	12	0.63	0.32	0.17	0.35		1.47	43.1	21.6	11.6	23.7		(*31*)
Japan	milk	1988	1	0.67	0.08	0.05	0.18	0.02	1.00	67.0	8.0	5.0	18.0	2.0	(*29*)
Germany	blood	1990	19	2.32	0.29	0.25	0.46		3.32	69.9	8.6	7.5	14.0		(*30*)
Norway	blood	1990	20	0.89	0.24	0.13	0.27	0.23	1.76	50.6	13.6	7.4	15.3	13.1	(*28*)
Sweden	milk	1990	P	0.81	0.15	0.06	0.10	0.04	1.16	69.8	12.9	5.2	8.6	3.4	(*27*)
Canada	milk	1992	10	1.75	0.65	0.21	0.29		2.90	60.3	22.4	7.2	10.0		(*32*)
Canada	milk	1992	72	1.40	0.50	0.20	0.30	0.05	2.45	57.1	20.4	8.2	12.2	2.0	(*33*)
Japan	milk	1993	1	0.32	0.06	0.07	0.21	0.03	0.69	46.4	8.7	10.1	30.4	4.3	(*29*)
Sweden	milk	1994	P	1.48	0.26	0.09	0.15	0.02	2.00	74.0	13.0	4.5	7.5	1.0	(*27*)
Sweden	tissue, adipose	1994	5	2.37	1.29	0.29	0.96	0.06	4.97	47.7	25.9	5.8	19.3	1.2	(*34*)
Sweden	tissue, adipose	1994	1	8.80	1.10	1.80	1.70		13.4	65.7	8.2	13.4	12.7		(*35*)
Sweden	tissue, liver	1994	5	2.75	3.05	0.40	1.54	1.54	9.28	29.7	32.9	4.3	16.6	16.6	(*34*)
Germany	blood	1995	19	2.98	0.37	0.32	0.60		4.27	69.9	8.6	7.5	14.0		(*30*)
Norway	blood	1995	19	1.40	0.33	0.32	0.52	0.50	3.07	45.6	10.7	10.4	16.9	16.3	(*28*)
Sweden	milk	1996	P	2.08	0.41	0.15	0.24	0.01	2.89	72.0	14.2	5.2	8.3	0.3	(*27*)
Finland	milk	1996	11	0.85	0.35		0.29		1.49	57.0	23.5		19.5		(*36*)
Finland	tissue, placenta	1996	11	0.77	0.41		0.40		1.58	48.7	25.9		25.3		(*36*)
Sweden	milk	1997	39	1.83	0.44	0.34	0.48	0.06	3.15	58.1	14.0	10.8	15.2	1.9	(*37*)
Sweden	milk	1997	P	2.28	0.48	0.42	0.46	0.05	3.69	61.8	13.0	11.4	12.5	1.4	(*27*)
San Francisco	tissue, breast	1997	22	18.3	6.59	3.17	4.09	6.40	38.6	47.5	17.1	8.2	10.6	16.6	(*38*)
Japan	blood	1998	24	0.52	1.20		2.25		3.97	13.0	30.3		56.7		(*39*)
Japan	milk	1998	1	1.03	0.53	0.22	0.29	0.05	2.12	48.6	25.0	10.4	13.7	2.4	(*29*)
Spain	tissue, adipose	1998	13	1.36	0.42	0.51	1.83		4.12	33.0	10.2	12.4	44.4		(*40*)
Finland	tissue, adipose	1998	10	6.14	2.02		2.18		10.34	59.4	19.5		21.1		(*41*)
Germany	blood	1999	20	3.17	0.39	0.34	0.63		4.53	69.9	8.6	7.5	14.0		(*30*)
Norway	blood	1999	29	1.50	0.31	0.35	0.59	0.35	3.10	48.4	10.0	11.3	19.0	11.3	(*28*)
Sweden	milk	1999	39	2.52	0.72	0.48	0.65	0.07	4.43	56.9	16.2	10.7	14.6	1.6	(*42*)
Sweden	milk	1999	124	1.77	0.37	0.27	0.51	0.06	2.98	59.4	12.4	9.1	17.1	2.0	(*37*)
Japan	milk	1999	1	0.62	0.16	0.18	0.29	0.03	1.28	48.4	12.5	14.1	22.7	2.3	(*29*)
Japan	milk	1999	6	0.34	0.10	0.13	0.32	0.03	0.93	36.7	11.3	14.3	34.6	3.2	(*43*)
Sweden	blood, fetal	2000	15	0.98	0.07	0.07	0.17		1.29	76.0	5.4	5.4	13.2		(*44*)
Sweden	blood, maternal	2000	15	0.83	0.19	0.17	0.56	0.04	1.79	46.4	10.6	9.5	31.3	2.2	(*44*)
Sweden	milk	2000	15	1.15	0.21	0.14	0.32	0.02	1.84	62.5	11.4	7.6	17.4	1.1	(*44*)
Japan	milk	2000	1	0.53	0.15	0.17	0.34	0.03	1.22	43.4	12.3	13.9	27.9	2.5	(*29*)
U.S.	milk	2000	4	126	27.0	23.5	14.8	1.66	193	65.3	14.0	12.2	7.7	0.9	(*45*)
Japan	tissue, adipose	2000	10	0.46	0.12	0.25	0.38	0.06	1.27	36.2	9.3	19.7	30.1	4.7	(*26*)
Belgium	tissue, adipose	2000	20	1.45	0.28	0.48	2.49		4.70	30.9	6.0	10.2	53.0		(*46*)
Czech Republic	tissue, adipose	2000	14	0.40	0.12	0.13	0.41	0.03	1.09	36.8	10.8	12.1	38.0	2.4	(*47*)
Czech Republic	tissue, adipose	2000	10	1.18	0.34	0.59	0.52	0.06	2.69	43.9	12.7	22.0	19.4	2.0	(*47*)
Japan	bile	2001	10	0.70	0.14	0.20	1.42	0.07	2.54	27.7	5.6	7.8	56.1	2.8	(*48*)
Japan	blood	2001	10	1.63	0.26	0.29	1.25	0.09	3.52	46.3	7.3	8.1	35.6	2.7	(*48*)
Sweden	blood	2001	143	2.77	1.39		1.87		6.03	45.9	23.1		31.0		(*12*)
U.S.	blood, fetal	2001	12	25.0	7.10	4.10	4.40	0.70	41.3	60.5	17.2	9.9	10.7	1.7	(*25*)
U.S.	blood, maternal	2001	12	28.0	5.70	4.20	2.90	0.30	41.1	68.1	13.9	10.2	7.1	0.7	(*25*)
Canada	milk	2001	20	13.3	3.00	2.30	3.00	0.60	22.2	59.9	13.5	10.4	13.5	2.7	(*49*)
Japan	tissue, liver	2001	10	1.38	0.18	0.22	1.55	0.14	3.48	39.8	5.2	6.4	44.7	4.0	(*48*)
U.S.	milk	2002	47	18.4	5.70	2.90	2.00	0.22	29.2	63.0	19.5	9.9	6.8	0.8	(*50*)
	averages			5.21	1.36	1.02	1.12	0.36	8.73	54.9	14.4	9.7	20.2	5.0	
	standard errors			2.26	0.51	0.48	0.28	0.17	3.47	2.1	0.8	0.5	1.7	1.0	
	geometric means			1.21	0.35	0.24	0.47	0.08	2.32	52.4	13.1	9.1	17.2	2.9	

[a] "reps" represents number of replicate samples. [b] "P" represents several different samples pooled into one analytical sample.

would have an excess load of some PBDEs in their blood. Table 3 lists PBDE blood concentrations for people who had been exposed occupationally, primarily in the dismantling of electronics (such as computers) and for people who had not been so exposed (controls). The latter were usually people working in the same facility but not working with the electronic products. The data here are much less complete than for the ambient samples. For example, several studies did not measure congeners 99 and 100, but they did measure congener 209. Thus, using these data to compare PBDE levels between exposed people and controls must be done on a congener-specific basis. In this case, only BDE-47 and -153 have sufficient data to make this comparison. In these cases, the PBDE concentrations in the blood of the exposed workers were about twice that in the blood of the controls. This difference is statistically significant; $t = 2.49$ for BDE-47 and 3.09 for BDE-153 as compared to the critical value of 2.23 for $p = 0.05$. There may be more BDE-99, -100, and -209 in the exposed workers than in the controls, but there are too few data for these differences to be statistically significant.

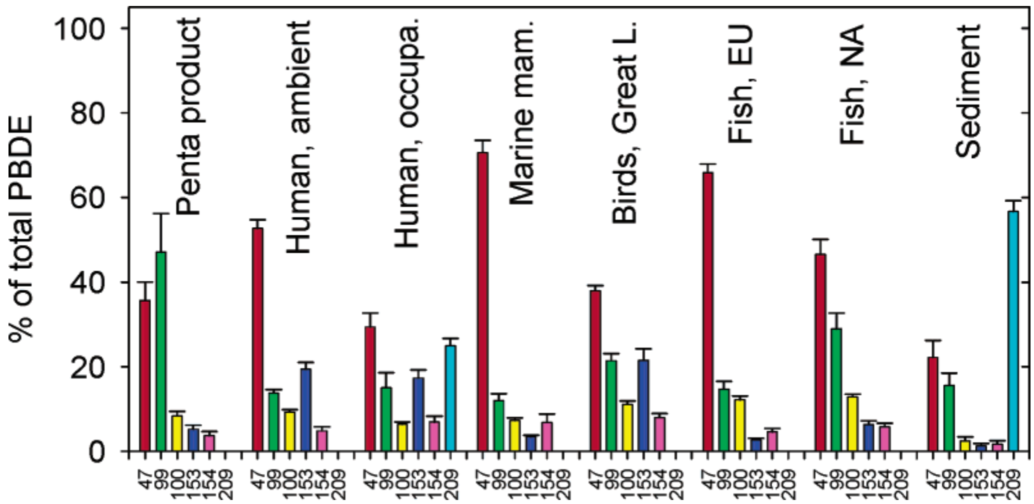

FIGURE 3. Congener distributions, given as the percent of ∑PBDE concentration, for the commercially available penta-product (*5*, *6*) and averaged for samples of ambient and occupationally exposed human blood, milk, and tissue; marine mammals; birds from the Great Lakes region; fish from Europe and North America; and sediment. The error bars indicate standard errors.

TABLE 3. PBDE Concentrations and Congener Distributions in Human Blood Taken from Occupational Settings (in ng/g Lipid)

type	date	reps	47	99	100	153	154	209	∑PBDE	% 47	% 99	% 100	% 153	% 154	% 209	ref
controls, Sweden	1997	20	1.56			0.57	0.38		2.51	62.0			22.9	15.1		(*52*)
controls, Norway	2000	5	1.50	0.40	0.23	0.54			2.67	56.2	15.0	8.6	20.2			(*51*)
controls, Sweden	1997	20	1.46			0.84	0.51		2.80	52.0			29.9	18.1		(*52*)
controls, Sweden	1999	2	1.44			1.02		2.37	4.83	41.8			23.2		34.9	(*53*)
averages			1.49	0.40	0.23	0.74	0.44	2.37	3.20	53.0	15.0	8.6	24.0	16.6	34.9	
standard errors			0.03			0.11	0.06		0.55	4.2			2.1	1.5		
exposed, Norway	2000	5	1.60	0.32	0.38	0.95	0.21		3.46	46.2	9.2	11.0	27.5	6.1		(*51*)
exposed, Sweden	1999	19	1.30			2.53	0.57	1.52	5.93	21.9			42.7	9.7	25.6	(*54*)
exposed, Norway	2000	5	4.00	0.97	0.71	1.70	0.44		7.82	51.2	12.4	9.1	21.7	5.6		(*51*)
exposed, Sweden	1999	6	1.29			1.50		6.89	9.68	35.0			24.7		40.2	(*53*)
exposed, Sweden	2001	21	4.20	3.70		1.90			9.80	42.9	37.8		19.4			(*12*)
exposed, Sweden	2001	3	4.11	3.69	0.76	1.82	0.40		10.8	38.2	34.3	7.0	16.9	3.7		(*55*)
exposed, Sweden	1997	19	2.87			4.51	1.22	4.80	13.4	21.4			33.6	9.1	35.8	(*52*)
averages			2.77	2.17	0.62	2.13	0.57	4.40	8.69	36.7	23.4	9.0	26.6	6.8	33.9	
standard errors			0.51	0.89	0.12	0.43	0.17	1.56	1.24	4.3	7.3	1.1	3.4	1.1	4.3	
***t*-values**			**2.49**			**3.09**										

Nevertheless, these data certainly suggest that some people can accumulate higher than ambient levels of PBDEs through their work.

The congener distribution for these occupationally exposed blood samples is shown in Figure 3. While there are some similarities of this distribution to that for the ambient human samples, the presence of BDE-209 in some of these occupationally exposed people suggests that this compound can accumulate into people and presumably into other animals as well. The "absence" of BDE-209 in the ambient population is likely a result of analytical bias; that is, most studies of the ambient population did not include BDE-209 as one of the analytes of interest. This compound should not be ignored in future studies.

Air. There have been a few measurements of PBDEs in air (see Table 4); many of these measurements have been of indoor dust collected to assess occupational exposure. Outdoor levels are on the order of 5−300 pg/m^3, with the higher values observed in cities. In one study of air around the Great Lakes, the atmospheric concentrations of PBDEs were strongly correlated with those of PCBs (*14*). Indoor PBDE air concentrations can be high, ranging up to ∼1800 pg/m^3. In some occupational settings, the concentrations can range up to 67 000 pg/m^3, a value observed in an electronics shredding plant. In these occupationally exposed cases, BDE-209 is usually the dominant congener, no doubt reflecting the extensive use of this congener for flame retarding the high impact plastics used in consumer electronics.

Marine Mammals. There have been several studies of PBDE concentrations in the tissues of marine mammals (mostly seals and porpoises). These concentrations are given in order of increasing ∑PBDE concentration in Table 5. While there are natural variations due to different species, sexes, and ages of the animals, it is appropriate to treat these data as two separate time series. The first series is the samples from the Canadian Arctic (*15*), all of which have ∑PBDE concentrations below 5 ng/g lipid, and the second series is the other samples, all of which are from industrially well-developed parts of the world. The ∑PBDE concentrations as a function of time for these two series are shown in Figure 4. There is a strong correlation for the Arctic samples with time, showing a doubling time of ∼7 yr, which is a bit slower than observed in the human samples discussed above. The samples from the rest of the world show more scatter, but the correlation is still statistically significant ($p < 0.01$). For these samples, the doubling time is ∼5 yr, which is statistically indistinguishable from the doubling times observed in people and in marine mammals from the Canadian Arctic. This observation suggests that PBDE levels in the fish-based diet of marine mammals are increasing at about the same rate

TABLE 4. PBDE Concentrations and Congener Distributions in Ambient (Outdoor) and Indoor Air Collected near and in Occupational Settings (in pg/m³)[a]

location	type[b]	reps	47	99	100	153	154	209	∑PBDE	% 47	% 99	% 100	% 153	% 154	% 209	ref
U.K.	out	129	2.46	1.96	0.44	0.23	0.18		5.27	46.7	37.2	8.3	4.3	3.4		(*56*)
Great Lakes	out	36	4.52	3.15	0.45	0.18	0.11		8.42	53.7	37.4	5.3	2.2	1.3		(*14*)
Japan	out	5	1.72	1.21		0.36		13.9	17.2	10.0	7.0		2.1		80.8	(*57*)
Great Lakes	out	12	33.0	16.0	2.00	0.53	0.41		51.9	63.5	30.8	3.9	1.0	0.8		(*14*)
Canada	out	32	150	110	20	13	8		301	49.8	36.5	6.6	4.3	2.7		(*58*)
averages			38.3	26.5	5.72	2.86	2.18	13.9	76.8	44.8	29.8	6.0	2.8	2.1	80.8	
standard errors			28.5	21.1	4.77	2.54	1.94		56.7	9.1	5.8	0.9	0.7	0.6		
Sweden	in	4						82.6	82.6						100	(*52*)
Sweden	in	4						83.0	83.0						100	(*59*)
Sweden	in	2	759	379	55.9	22	12.0	170	1400	54.3	27.1	4.0	1.6	0.9	12.2	(*59*)
U.K.	in	9	1 350	302	113	8	8		1780	75.7	17.0	6.3	0.4	0.4		(*60*)
averages			1 050	341	84.3	15.0	9.93	112	835	65.0	22.1	5.2	1.0	0.7	70.7	
standard errors			293	38.3	28.4	7.02	2.07	29.1	441	10.7	5.0	1.2	0.6	0.2	29.3	
Sweden	occ	2						93.0	93.0						100	(*59*)
Sweden	occ	6	350	150	41	19	6.9	220	787	44.5	19.1	5.2	2.4	0.9	28.0	(*59*)
Sweden	occ	12	1 210	2 580		408		36 100	44 000	2.7	5.9		9.3		82.1	(*52*)
Sweden	occ	12	1 200	2 600	250	3 900	570	36 000	45 000	2.7	5.8	0.6	8.8	1.3	80.9	(*59*)
Sweden	occ	2	1 500	1 900	320	5 000	1 300	57 500	67 000	2.2	2.8	0.5	7.4	1.9	85.2	(*59*)
averages			1 063	1 807	203.7	3 248	609.4	25 980	31 360	13.0	8.4	2.1	7.0	1.3	75.2	
standard errors			247.6	575.5	83.8	1 103	359.8	11 250	13 320	10.5	3.6	1.6	1.6	0.3	12.3	

[a] All samples were collected between 1998 and 2001 inclusive. [b] "out", outdoor air; "in", indoor air from homes; "occ", air from occupational settings.

TABLE 5. PBDE Concentrations and Congener Distributions in Marine Mammals (in ng/g Lipid)

location	type	date	reps	47	99	100	153	154	∑PBDE	% 47	% 99	% 100	% 153	% 154	ref
Canadian Arctic	seals, 0−15 yr	1981	7	0.38	0.012	0.023	0.004	0.002	0.42	90.1	2.9	5.5	1.0	0.5	(*15*)
Canadian Arctic	seals, 0−15 yr	1991	5	1.43	0.076	0.10	0.006	0.012	1.63	87.9	4.7	6.4	0.4	0.7	(*15*)
Canadian Arctic	seals, 16−35	2000	5	1.27	0.16	0.17	0.055	0.041	1.69	74.9	9.4	10.1	3.3	2.4	(*15*)
Canadian Arctic	seals, 0−15 yr	1996	9	2.04	0.17	0.17	0.042	0.023	2.44	83.6	6.8	7.0	1.7	0.9	(*15*)
Canadian Arctic	seals, 0−15 yr	1996	8	2.71	0.17	0.21	0.050	0.025	3.17	85.5	5.4	6.7	1.6	0.8	(*15*)
Canadian Arctic	seals, 16−35 yr	2000	8	3.41	0.19	0.28	0.098	0.050	4.03	84.7	4.7	6.9	2.4	1.2	(*15*)
Canadian Arctic	seals, 0−15 yr	2000	7	3.75	0.19	0.25	0.069	0.036	4.30	87.2	4.4	5.9	1.6	0.8	(*15*)
Sweden	ringed seal, blubber	1981	7	47.0	1.70	2.30			51.0	92.2	3.3	4.5			(*17*)
San Francisco	harbor seals	1989	1	45.6	16.2	4.16	4.06	17.7	87.7	52.0	18.5	4.7	4.6	20.2	(*38*)
Baltic Sea	two seal species	1984	4	80.3	9.13	9.48	6.42	8.20	114	70.7	8.0	8.3	5.6	7.2	(*35*)
Faroe Islands	pilot whales	1997	6	117	22.6	12.2	2.90	10.3	165	71.0	13.7	7.4	1.8	6.2	(*61*)
San Francisco	harbor seals	1991	2	164	59.3	14.0	16.8	93.5	348	47.2	17.0	4.0	4.8	26.9	(*38*)
Netherlands	several species	1997	13	465	71.2	38.3			574	80.9	12.4	6.7			(*62*)
Canada, BC	porpoise	1992	5	369	116	94.8	11.5	21.0	612	60.3	18.9	15.5	1.9	3.4	(*63*)
San Francisco	harbor seals	1993	3	455	79.5	44.2	28.8	49.9	658	69.2	12.1	6.7	4.4	7.6	(*38*)
U.K.	porpoise blubber	1999	9	492	107	47	20		666	73.9	16.1	7.1	3.0		(*64*)
Sweden	grey seal, blubber	1982	8	650	40	38			728	89.3	5.5	5.2			(*17*)
San Francisco	harbor seals	1992	1	350	151	26.4	35.4	220	783	44.7	19.3	3.4	4.5	28.1	(*38*)
U.K.	porpoise blubber	1996	4	582	408	117	66		1170	49.6	34.8	10.0	5.6		(*64*)
Faroe Islands	pilot whale blubber	1996	5	896	321	162	52.8	124	1560	57.6	20.6	10.4	3.4	8.0	(*65*)
U.K.	porpoise blubber	1997	27	1140	275	201	55		1670	68.2	16.4	12.0	3.3		(*64*)
U.K.	porpoise blubber	1998	20	1080	341	206	51		1670	64.3	20.4	12.3	3.0		(*64*)
San Francisco	harbor seals	1997	1	1630	61	121	74	55	1940	84.0	3.1	6.2	3.8	2.8	(*38*)
North Sea	two species	1997	12	1170	478	174	156	85.9	2070	56.7	23.1	8.4	7.6	4.2	(*66*)
San Francisco	harbor seals	1998	2	3960	228	266	322	174	4950	80.0	4.6	5.4	6.5	3.5	(*38*)
	averages			548	111	63.2	41.1	47.8	793	72.2	12.2	7.5	3.4	7.0	
	standard errors			169	28.9	16.1	15.6	15.7	220	2.9	1.6	0.6	0.4	2.1	
	geometric means			89.8	12.3	8.9	3.3	2.3	127	70.7	9.7	7.0	2.9	3.4	

as the levels in the various media to which people are exposed.

The ∑PBDE concentrations in the non-Arctic marine mammals are ~100-fold higher than in people (compare Figures 1 and 4). This is not surprising given that these animals are high-level predators and that people are generally eating food from comparatively lower trophic levels. The congener distribution for these samples is shown in Figure 3. This distribution resembles that of the human samples.

Birds. There have been two noteworthy studies of PBDEs in birds' eggs, one for herring gull (*Larus argentatus*) eggs collected in the Great Lakes region from 1981 to 2000 (*16*) and the other for guillemot (*Uria aalge*) eggs collected in Sweden from 1970 to 1989 (*17*). There have been five other studies of PBDEs in bird tissue. These studies are all summarized in Table 6.

It is clear from these data that PBDE concentrations have been increasing rapidly in both the gull and guillemot eggs. Both of these concentration sets are plotted as a function of time in Figure 5. It appears that the Swedish guillemot samples have been increasing at a somewhat slower rate

TABLE 6. PBDE Concentrations and Congener Distributions in Birds (Mostly Their Eggs; in ng/g Lipid)

type	date	reps	47	99	100	153	154	∑PBDE	% 47	% 99	% 100	% 153	% 154	ref
guillemot eggs[a]	1970	1	130	24	4.2			158	82.2	15.2	2.7			(*17*)
guillemot eggs	1974	10	170	48	8.5			227	75.1	21.2	3.8			(*17*)
guillemot eggs	1975	1	130	33	4.6			168	77.6	19.7	2.7			(*17*)
guillemot eggs	1976	10	600	130	32			762	78.7	17.1	4.2			(*17*)
guillemot eggs	1978	8	260	70	22			352	73.9	19.9	6.3			(*17*)
guillemot eggs	1979	1	640	130	37			807	79.3	16.1	4.6			(*17*)
guillemot eggs	1982	10	820	200	44			1060	77.1	18.8	4.1			(*17*)
guillemot eggs	1983	1	880	210	49			1140	77.3	18.4	4.3			(*17*)
guillemot eggs	1986	1	1200	260	48			1510	79.6	17.2	3.2			(*17*)
guillemot eggs	1987	10	650	160	40			850	76.5	18.8	4.7			(*17*)
guillemot eggs	1989	1	1500	330	79			1910	78.6	17.3	4.1			(*17*)
guillemot eggs	1989	10	910	240	61			1210	75.1	19.8	5.0			(*17*)
guillemot averages			658	153	35.8			846	77.6	18.3	4.1			
standard errors			126	28.3	6.6			161	0.7	0.5	0.3			
geometric means			496	117	25.8			639	77.5	18.2	4.0			
gull eggs[b]	1981	3	41.4	12.3	7.9	42.3	20.0	124	33.4	9.9	6.4	34.1	16.1	(*16*)
gull eggs	1983	3	49.4	19.4	11.3	58.6	19.1	158	31.3	12.3	7.2	37.1	12.1	(*16*)
gull eggs	1987	3	280	126	70.3	285	69.6	831	33.7	15.2	8.5	34.3	8.4	(*16*)
gull eggs	1988	3	453	241	120	319	92.9	1230	37.0	19.7	9.8	26.0	7.6	(*16*)
gull eggs	1989	3	430	236	111	321	143	1240	34.6	19.0	8.9	25.9	11.5	(*16*)
gull eggs	1990	3	456	240	115	352	103	1270	36.0	19.0	9.1	27.8	8.1	(*16*)
gull eggs	1992	3	752	410	212	271	131	1780	42.3	23.1	11.9	15.3	7.4	(*16*)
gull eggs	1993	3	1330	882	399	650	233	3490	38.1	25.2	11.4	18.6	6.7	(*16*)
gull eggs	1996	3	1720	1110	619	584	255	4280	40.1	25.8	14.5	13.6	6.0	(*16*)
gull eggs	1998	3	2260	1740	851	861	330	6040	37.4	28.8	14.1	14.3	5.5	(*16*)
gull eggs	1999	3	2570	1930	938	804	353	6600	38.9	29.3	14.2	12.2	5.4	(*16*)
gull eggs	2000	15	2490	1450	766	597	232	5540	45.0	26.2	13.8	10.8	4.2	(*16*)
gull eggs	2000	3	3450	1840	1090	745	380	7510	45.9	24.5	14.6	9.9	5.1	(*16*)
gull averages			1250	787	409	453	182	3080	38.0	21.4	11.1	21.5	8.0	
standard errors			314	205	109	75.2	34.4	727	1.2	1.7	0.8	2.7	0.9	
geometric means			658	356	187	340	130	1740	37.8	20.4	10.7	19.5	7.4	
starling, Sweden	1988	4	4.2	3.1	0.9			8.2	51.5	38.1	10.4			(*17*)
chickens, U.S.	1997	13	3.2	4.3	0.7	0.9	0.3	9.4	34.0	46.2	7.0	10.0	2.8	(*67*)
cormorant, U.K.	1998	47	453	148	217	68		886	51.1	16.7	24.5	7.7		(*64*)
cormorant, U.K.	1999	20	554	176	249	88.5	89.3	1160	47.9	15.2	21.5	7.7	7.7	(*68*)
osprey, Sweden	1984	35	1800	140	200			2140	84.1	6.5	9.3			(*17*)
averages			563	94.3	134	52.5	44.8	840	53.7	24.5	14.6	8.4	5.3	
standard errors			329	37.5	54.8	26.4	44.5	398	8.2	7.5	3.5	0.8	2.5	
geometric means			90.5	34.5	22.7	17.8	4.9	176	51.5	19.6	12.9	8.4	4.7	

[a] From Sweden. [b] From the Great Lakes region.

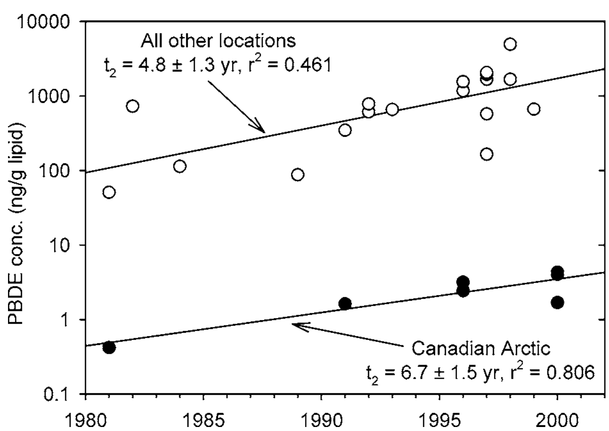

FIGURE 4. ∑PBDE concentrations in marine mammals (in ng/g lipid) shown as a function of the year in which the samples were collected; see Table 5. The bottom line with filled symbols represents samples from the Canadian Arctic (*15*), and the top line with open symbols is for all other samples. The regressions for the two data sets are shown separately; the doubling times of the types of samples are not significantly different.

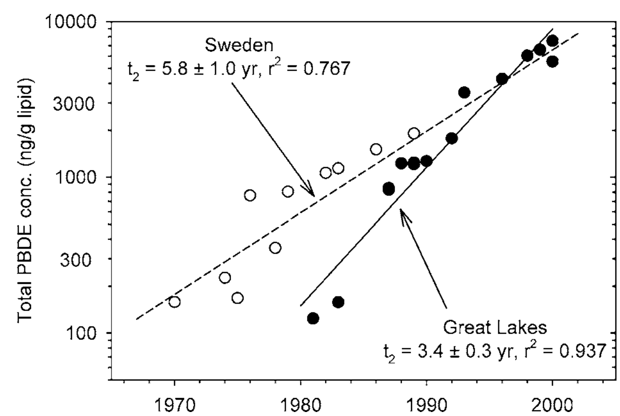

FIGURE 5. ∑PBDE concentrations in birds' eggs (in ng/g lipid) shown as a function of the year in which the samples were collected; see Table 6. The bottom line with filled symbols represents samples of herring gull eggs from the U.S. and Canadian Great Lakes (*16*), and the top line with open symbols is for guillemot eggs from Sweden (*17*). The regressions for the two data sets are shown separately; the doubling times of the two types of samples are significantly different.

than the North American gull samples (doubling times of 5.8 vs 3.4 yr, respectively, $t = 2.31$). It is interesting to note that the extrapolated guillemot egg concentrations are about the same as the measured gull egg concentrations in the year 2000. On the other hand, it should be pointed out that the guillemot egg concentration data are not strictly compatible

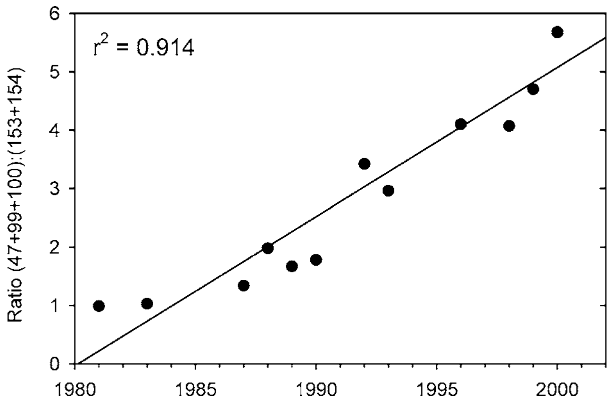

FIGURE 6. Ratio of the sum of the concentrations of BDE-47 plus -99 plus -100 divided by the sum of the concentrations of BDE-153 plus -154 (see eq 1) in herring gull eggs from the Great Lakes (*16*) shown as a function of the year in which the samples were collected.

with the gull egg concentration data because BDE-153 and -154 were not measured in the former study, but these congeners were measured in the latter study. In general, both of these doubling times are about the same as those observed for people (see Figure 1) and for marine mammals (see Figure 4). The ∑PBDE concentrations in these bird eggs are about the same as in the non-Arctic marine mammals (compare Figures 4 and 5), which is expected given the similarity of their food supplies.

Because of the lack of BDE-153 and -154 data (*17*), there is not much to say about the congener distribution of the guillemot egg samples other than that BDE-47 is by far the most abundant congener. In the Great Lakes samples, the gull egg congener distribution changes systematically with the year in which the sample was taken. In the 1981 samples, BDE-47 is ~33% of the total, but in 2000, it has increased to ~45% of the total; at the same time BDE-154 has decreased from ~16% in 1981 to ~5% in 2000. These changes can be demonstrated by plotting the ratio:

$$\mathscr{R} = \frac{[47] + [99] + [100]}{[153] + [154]} \quad (1)$$

(where [*xx*] is the concentration of congener *xx* in any units) as a function of sampling year; see Figure 6. The regression is excellent, but the meaning of these changes is not clear—perhaps the use or the composition of the various commercial PBDE products has changed systematically over the years. The average congener distribution for the Great Lakes gull egg samples is shown in Figure 3; this distribution is similar to that of the human samples, probably indicating that the gulls and the people are receiving PBDEs from similar sources.

The five other bird samples are tissue samples (rather than eggs) and show a wide range of concentrations, ranging from <10 ng/g lipid in chickens and starlings to >1000 ng/g lipid in cormorants and ospreys. The higher PBDE levels in the latter may be related to their higher trophic level as compared to the former.

Fish. The concentrations of PBDEs measured in several fishes at several locations are given in Table 7. The concentrations do not vary systematically with the sampling date either in Europe, in North America, or for both combined. In fact, the concentrations are highly variable depending on the type of fish and the location from which they were collected. For example, whitefish from the Columbia River have ∑PBDE concentrations ranging from 12 to 1060 ng/g lipid, depending on where they were caught. These variations are likely due to the proximity of the fishes' feeding grounds to PBDE sources. In general, the concentrations of PBDEs in fishes in Europe are significantly lower than in fishes from North America; the arithmetic and geometric averages for ∑PBDE are 120 and 49 ng/g lipid for the European fishes, respectively, and 1050 and 310 ng/g for the North American fishes, respectively. Student's *t*-test values average ~2.4 (see Table 7), indicating significant differences with 95% confidence. The ratio of about 7:1 for North American versus European fish ∑PBDE concentrations is in the same direction but somewhat smaller than the ratio observed for PBDE concentrations in people (see Figure 2), where the ratio was about 20:1.

The congener distributions for the European and the North American fishes are shown in Figure 3. In both cases, the distributions favor BDE-47, but they are different from one another. For the North American fishes, the ratio of BDE-47 to -99 is ~2:1, which is significantly lower than this ratio in the European fishes (at ~5:1). This difference may indicate a difference in the congener mixtures to which European and North American fishes have been exposed. The European fish distribution is similar to the marine mammal distribution, perhaps indicating that the marine mammals are feeding on these fishes.

Sediment. Several sediment samples have been collected from rivers and lakes from Europe, and PBDEs have been measured in these samples (see Table 8). Sediment collects unevenly in rivers (more sediment deposits where the current is slow and less where the current is rapid); therefore, the PBDE concentrations in rivers is highly variable, ranging from undetectable in some places to almost 7200 ng/g dry weight in other places. Thus, the PBDE concentrations in river sediment cannot be compared from river to river, but these values can give indications where the sources are on a given river.

From the limited data in the literature, it is clear that BDE-209 (decabromodiphenyl ether) is often present in sediment (see Table 8), even though it was not abundant in other samples, including biota. Presumably, the very high K_{ow} of this compound causes it to partition onto the particles in water, which in turn sink to form the sediment. Unfortunately, not all studies measured BDE-209 in the sediment, and this makes the congener distributions, as given in Table 8 and in Figure 3, particularly unreliable. Nevertheless, the congener distribution for the sediment samples is somewhat similar to that of the occupationally exposed human samples. These two types of samples are linked by the presence of particles laden with PBDEs (especially BDE-209), which are inhaled by people in occupational settings and which are deposited to water, where they sink and become incorporated into the sediment in aquatic settings.

Two sediment studies have measured PBDEs as a function of depth in dated sediment cores; this allows one to track the rate of concentration increase in sediment in much the same way as one can track trends using archived samples. The first such study is by Nylund et al. (*18*), who sampled and measured a sediment core from the Baltic Sea. The depth in the core was calibrated by counting laminae; thus, it was possible to know the PBDE concentrations in the sediment at this location as a function of time (see Figure 7). The data show a doubling time of 3−4 yr, which is similar to that observed for humans, marine mammals, and bird eggs.

The other sediment core study collected samples from Drammenfjord near Oslo, Norway (*19*). In this case, the relationship between depth in the core and date was established using the radioactive isotopes, ^{137}Cs and ^{210}Pb. The concentrations of PBDEs are also shown in Figure 7. It is interesting to note that the absolute concentrations here are ~10-fold more than in the Baltic Sea, presumably because the Baltic site has significantly less nearby human activity than the Norwegian site, which is adjacent to the city of Oslo. At this location, the PBDE concentrations increased with a doubling time of ~3 yr until the mid-1980s,

TABLE 7. PBDE Concentrations and Congener Distributions in Fishes (in ng/g Lipid)

location	type	date	reps	47	99	100	153	154	∑PBDE	% 47	% 99	% 100	% 153	% 154	ref
Germany	eel, river	2001	5	4.50	0.14	0.98	0.21	0.48	6.31	71.3	2.2	15.5	3.3	7.6	(*69*)
Japan	several species	1998	25	5.14	0.41	0.57	0.18	0.52	6.82	75.4	6.0	8.4	2.6	7.6	(*70*)
Netherlands	mackerel	1997	1	5.40	1.90	1.80			9.10	59.3	20.9	19.8			(*62*)
Baltic Sea	herring	1985	4	10.3	1.70	1.57			13.6	75.9	12.5	11.6			(*35*)
Baltic Sea	three species	1998	22	10.8	1.47	1.60	0.95	0.48	15.3	70.4	9.7	10.5	6.2	3.2	(*71*)
Switzerland	rainbow trout	2002	4	11.5	2.27	1.70	0.27	0.36	16.1	71.5	14.0	10.5	1.7	2.3	(*74*)
Baltic Sea	herring	1998	3	12.4	4.14		0.75		17.3	71.7	24.0		4.3		(*41*)
Greenland	three species	2000	36	15.6	0.69	1.28			17.6	88.8	3.9	7.3			(*72*)
Scotland & Belgium	salmon	2001	13	10.9	2.87	3.56	1.01	0.81	19.2	57.0	15.0	18.6	5.3	4.2	(*73*)
Sweden	whitefish	1986	35	15.0	7.20	3.90			26.1	57.5	27.6	14.9			(*17*)
Sweden	herring	1987	50	24.1	9.33	4.01			37.4	64.4	24.9	10.7			(*17*)
Baltic Sea	sprat	1998	9	49.4	6.34		1.03		56.7	87.0	11.2		1.8		(*41*)
Switzerland	whitefish	2002	8	44.3	24.0	4.63	1.21	1.52	75.6	58.6	31.7	6.1	1.6	2.0	(*74*)
North Sea	several species	1999	28	47.6	11.2	13.5	1.17	3.36	76.8	61.9	14.6	17.6	1.5	4.4	(*66*)
Sweden	herring	1987	260	130	23.0	13.0			166	78.3	13.9	7.8			(*17*)
Germany	bream, river	2001	22	127	0.49	31.8	4.80	18.2	182	69.7	0.3	17.4	2.6	10.0	(*69*)
Baltic Sea	salmon	1995	8	132	35.0	37.0	3.20	6.00	213	61.9	16.4	17.4	1.5	2.8	(*75*)
Baltic Sea	salmon fillet	1991	1	167	52.0	44.0	4.20	11.0	278	60.0	18.7	15.8	1.5	4.0	(*35*)
Sweden	several species	1987	12	269	41.8	34.5			345	77.9	12.1	10.0			(*76*)
Sweden	pike, rivers	1995	14	226	128	53.9			408	55.4	31.4	13.2			(*77*)
Sweden	Arctic char	1987	15	400	64.0	51.0			515	77.7	12.4	9.9			(*17*)
European averages				81.8	19.9	16.0	1.58	4.27	119	69.1	15.4	12.8	2.8	4.8	
standard errors				23.4	6.77	4.38	0.46	1.89	32.9	2.1	1.9	1.0	0.5	0.8	
geometric means				33.6	5.54	6.23	0.97	1.71	49.1	68.5	11.3	12.1	2.5	4.2	
Slocan River, U.S.	whitefish	1996	3	4.20	4.70	1.50	0.93	0.76	12.1	34.7	38.9	12.4	7.7	6.3	(*78*)
Canada, BC	sole	1992	26	14.7	7.36	4.45	6.16	1.51	34.2	43.0	21.5	13.0	18.0	4.4	(*63*)
Columbia River, U.S.	whitefish	1992	4	16.8	22.8	5.20	3.00	2.00	49.8	33.7	45.8	10.4	6.0	4.0	(*78*)
Columbia River, U.S.	whitefish	1992	2	20.0	27.7	8.20	4.10	2.90	62.9	31.8	44.0	13.0	6.5	4.6	(*78*)
Michigan & Illinois	two species	1999	36	34.0	7.28	6.83	8.96	11.4	68.4	49.7	10.6	10.0	13.1	16.6	(*79*)
Canada, BC	sole	2000	60	48.5	16.8	15.4	6.21	4.93	91.8	52.9	18.3	16.7	6.8	5.4	(*63*)
Columbia River, U.S.	whitefish	2000	9	63.4	83.4	22.3	12.9	7.40	189	33.5	44.0	11.8	6.8	3.9	(*78*)
Great Lakes	lake trout	2000	40	151	37.0	19.9	9.96		217	69.3	17.0	9.2	4.6		(*80*)
Kootenay Lake, U.S.	whitefish	1998	5	125	135	38.2	17.0	13.8	330	38.0	41.1	11.6	5.2	4.2	(*78*)
Great Lakes	several species	1999	20	208	59.0	45.5	14.7	40.4	368	56.6	16.0	12.4	4.0	11.0	(*81*)
Columbia River, U.S.	whitefish	1996	1	132	184	43.5	23.8	14.8	398	33.2	46.2	10.9	6.0	3.7	(*78*)
Columbia River, U.S.	whitefish	2000	12	179	227	68.8	32.9	20.0	527	33.9	43.0	13.0	6.2	3.8	(*78*)
Columbia River, U.S.	whitefish	1994	1	185	263	71.6	40.6	30.2	590	31.3	44.5	12.1	6.9	5.1	(*78*)
Columbia River, U.S.	whitefish	1995	4	325	479	148	63.7	44.0	1060	30.7	45.2	13.9	6.0	4.2	(*78*)
Lake Michigan	salmonids	1996	21	1340	239	249	30.3	116	1970	67.9	12.1	12.6	1.5	5.9	(*82*)
Kootenay River, U.S.	suckers	2000	6	2110	6.60	461	24.4	168	2770	76.2	0.2	16.6	0.9	6.1	(*78*)
Lake Michigan	trout	1996	6	1700	600	360	110	200	2970	57.2	20.2	12.1	3.7	6.7	(*83*)
Virginia, eastern	three species	1998	25	4540	783	1410	235	235	7200	63.0	10.9	19.6	3.3	3.3	(*84*)
North American averages				622	177	165	35.8	53.7	1050	46.5	28.9	12.9	6.3	5.8	
standard errors				275	53.7	79.5	13.3	18.5	423	3.6	3.8	0.6	0.9	0.8	
geometric means				136	63.1	38.9	16.0	16.6	308	44.3	20.5	12.6	5.2	5.3	
***t*-values**				**1.96**	**2.90**	**1.88**	**2.57**	**2.65**	**2.20**						

at which time the concentrations seem to have leveled off. This is surprising given that European restrictions on the use of these compounds were not promulgated until the late 1990s.

Other. Table 9 shows PBDE concentrations in a variety of other types of samples, such as invertebrates and sewage treatment plant sludge. As expected, the concentrations are highly variable, but the ∑PBDE concentrations in the invertebrates are less than those in fish from similar locations, presumably because the invertebrates are operating at a much lower trophic level. The ∑PBDE concentration in sewage sludge from North America is much higher than in sludge from The Netherlands; this observation mirrors the relative concentrations found in people and fishes from North America versus Europe. Sewage sludge has a relatively high proportion of BDE-209, probably because of the effective partitioning of this compound to the solids.

Principal Component Analysis of Congener Distributions. The comparison of the average congener distributions shown in Figure 3 is rather crude; principal components analysis (PCA) is a better way to make these comparisons. PCA is a statistical tool that can project multidimensional data onto two dimensions, which are easier to view. PCA uses the full data sets and does not require averaging by categories. In this case, only biotic samples were included, and only samples with measurements of all five of the main congeners (BDE-47, -99, -100, -153, and -154) were used. This gave 98 useable data sets, which were analyzed by the PRINCOMP procedure of the Statistical Analysis System (SAS Institute, Cary, NC). The first two principal components explained 63% of the variability of the data set. For principal component 1, the loadings (or eigenvectors) for the five main PBDE congeners were as follows: BDE-47, −0.730; -99, 0.443; -100, 0.101; -153, 0.411; and -154, 0.303. For principal component 2, the loadings the five main congeners were as follows: BDE-47, −0.034; -99, 0.524; -100, 0.580; -153, −0.479; and -154, −0.399. Principal component 1 is plotted against principal component 2 in Figure 8 with the four types of samples indicated by different color symbols.

In these plots, one is usually looking for clustering of the data; for example, the human samples might all cluster together and be separated from, say, the bird samples, which

TABLE 8. PBDE Concentrations and Congener Distributions in Sediment (in ng/g Dry Weight)

location	type	date	reps	47	99	100	153	154	209	∑PBDE	% 47	% 99	% 100	% 153	% 154	% 209	ref
Baltic Sea	core	1939	1	0.009						0.009	100						(*18*)
Baltic Sea	core	1946	1	0.017						0.017	100						(*18*)
Baltic Sea	core	1953	1	0.021	0.010					0.031	67.7	32.3					(*18*)
Baltic Sea	core	1961	1	0.029						0.029	100						(*18*)
Baltic Sea	core	1967	1		0.005					0.005		100					(*18*)
Baltic Sea	core	1971	1	0.020	0.009					0.029	69.0	31.0					(*18*)
Baltic Sea	core	1974	1		0.007					0.007		100					(*18*)
Baltic Sea	core	1976	1	0.038	0.011					0.049	77.6	22.4					(*18*)
Baltic Sea	core	1978	1	0.042	0.015					0.057	73.7	26.3					(*18*)
Baltic Sea	core	1980	1	0.053	0.014					0.067	79.1	20.9					(*18*)
Baltic Sea	core	1982	1	0.075	0.050					0.125	60.0	40.0					(*18*)
Baltic Sea	core	1984	1	0.102	0.054					0.156	65.4	34.6					(*18*)
Baltic Sea	core	1986	1	0.122	0.032	0.011				0.165	73.9	19.4	6.7				(*18*)
Baltic Sea	core	1987	1	0.288	0.176	0.056				0.520	55.4	33.8	10.8				(*18*)
Norway	core	1975	1	0.041	0.065	0.007			0.044	0.16	26.1	41.4	4.5			28.0	(*19*)
Norway	core	1978	1	0.040	0.071	0.006			0.372	0.49	8.2	14.5	1.2			76.1	(*19*)
Norway	core	1981	1	0.038	0.065	0.014	0.003		0.702	0.82	4.6	7.9	1.7	0.4		85.4	(*19*)
Norway	core	1983	1	0.052	0.081	0.016	0.010	0.016	1.79	1.97	2.6	4.1	0.8	0.5	0.8	91.1	(*19*)
Norway	core	1985	1	0.042	0.062	0.016	0.010		1.82	1.95	2.2	3.2	0.8	0.5		93.3	(*19*)
Norway	core	1986	1	0.076	0.113	0.032	0.016	0.022	2.32	2.58	2.9	4.4	1.2	0.6	0.9	90.0	(*19*)
Norway	core	1988	1	0.081	0.138	0.036	0.018	0.023	2.57	2.87	2.8	4.8	1.3	0.6	0.8	89.7	(*19*)
Norway	core	1993	1	0.140	0.179	0.049	0.031	0.036	2.29	2.72	5.1	6.6	1.8	1.1	1.3	84.0	(*19*)
Norway	core	1994	1	0.083	0.127	0.039	0.029	0.031	3.35	3.66	2.3	3.5	1.1	0.8	0.8	91.6	(*19*)
Norway	core	1996	1	0.111	0.141	0.050	0.029	0.036	2.86	3.22	3.4	4.4	1.6	0.9	1.1	88.6	(*19*)
Norway	core	1998	1	0.114	0.186	0.058	0.032	0.036	2.90	3.33	3.4	5.6	1.7	1.0	1.1	87.2	(*19*)
Norway	core	1999	1	0.145	0.208	0.070	0.040	0.048	2.63	3.14	4.6	6.6	2.2	1.3	1.5	83.7	(*19*)
U.K.	estuary	2000	23	4.80	6.50				146	157	3.1	4.1				92.8	(*89*)
U.S.	lake	1999	4	1.37	3.70	0.63	1.76	1.60	27.9	37.0	3.7	10.0	1.7	4.8	4.3	75.5	(*81*)
Korea	marine	2000	80	1.14	1.33		0.39	0.41		3.27	34.9	40.8		11.8	12.4		(*85*)
Denmark	marine & fresh	2000	16	0.16	0.23	0.10	0.04		3.67	4.19	3.7	5.5	2.4	1.0		87.5	(*86*)
Netherlands	particles	2000	44	2.20	2.40				71.0	75.6	2.9	3.2				93.9	(*87*)
Sweden	rivers	1995	7	56.6	14.9	13.7			7100	7190	0.8	0.2	0.2			98.8	(*77*)
U.K.	rivers	1996	27	8.47	14.9					23.3	36.3	63.7					(*88*)
Netherlands	rivers	2000	22	1.10	0.60				22.0	23.7	4.6	2.5				92.8	(*87*)
U.K.	rivers	2000	7	3.80	7.10				16.0	26.9	14.1	26.4				59.5	(*89*)
U.K.	rivers	2000	15	7.20	9.60				119	136	5.3	7.1				87.6	(*89*)
U.K.	rivers	2000	6	0.24	0.22				0.57	1.03	23.3	21.4				55.3	(*89*)
Portugal	rivers	2001	32	0.39	0.40	0.24				1.02	37.9	38.9	23.2				(*90*)
averages				2.48	1.82	0.84	0.19	0.23	359	203	32.2	22.6	3.6	1.9	2.5	82.5	
standard errors				1.58	0.67	0.76	0.13	0.16	337	189	5.8	4.2	1.3	0.9	1.1	3.6	
geometric means				0.18	0.18	0.05	0.03	0.06	5.67	1.02	13.4	12.1	1.9	1.1	1.5	80.1	

TABLE 9. PBDE Concentrations and Congener Distributions in Other Miscellaneous Samples (in ng/g Wet, Lipid, or Dry Weight as Indicated)

location	sample	date	reps	units	47	99	100	153	154	209	∑PBDE	% 47	% 99	% 100	% 153	% 154	% 209	ref
Belgium	crab	2001	1	wet	17.1	4.60	3.10	0.60	1.10		26.5	64.7	17.4	11.7	2.3	4.2		(*91*)
Canada, B. C.	crab	1994	23	lipid	84.2	15.3	10.2	2.67	4.09		116	72.3	13.2	8.7	2.3	3.5		(*63*)
Sweden	frog liver	1999	7	wet	0.06	0.06					0.12	48.0	52.0					(*92*)
North Sea	invertebrates	1999	40	lipid	23.6	8.25	10.0	4.09	6.16		52.1	45.3	15.8	19.2	7.8	11.8		(*66*)
Sweden	moose muscle	1985	13	lipid	0.82	0.64	0.24				1.70	48.2	37.6	14.1				(*17*)
Netherlands	mussels	2000	8	dry	1.20	0.50					1.70	70.6	29.4					(*87*)
Netherlands	mussels	2000	8	dry	1.80	1.40					3.20	56.3	43.8					(*87*)
Greenland	mussels	2000	20	lipid	5.00	1.00					6.00	83.3	16.7					(*72*)
U.K.	mussels	1996	1	wet	3.50	3.90					7.40	47.3	52.7					(*88*)
Denmark	mussels	2000	15	lipid	8.47	3.44	0.81	0.81			13.5	62.6	25.4	6.0	6.0			(*86*)
Sweden	reindeer fat	1986	31	lipid	0.17	0.26	0.04				0.47	36.2	55.3	8.5				(*17*)
Sweden	sewage sludge	2000	105	wet	7.00	10.0	1.70	0.86	0.72	11.0	31.3	22.4	32.0	5.4	2.7	2.3	35.2	(*93*)
Netherlands	sewage sludge	2000	6	dry	2.30	5.20				24.0	31.5	7.3	16.5				76.2	(*87*)
Netherlands	sewage sludge	2000	3	dry	0.40	6.60				45.0	52.0	0.8	12.7				86.5	(*87*)
Netherlands	sewage sludge	2000	6	dry	22.0					350	372	5.9					94.1	(*87*)
U.S.	sewage sludge	2000	11	dry	556	636	137	86.6	88.4	554	2058	27.0	30.9	6.7	4.2	4.3	26.9	(*5*)
Netherlands	water	1999	6	pg/L	1.00	0.50		0.10		0.40	2.00	50.0	25.0		5.0		20.0	(*94*)

might cluster together. Figure 8 does not show such strict clustering. In the middle of the figure (cluster A), the human (red), marine mammal (green), and fish (blue) data all overlap, which may indicate that these three types of samples have similar PBDE sources. The bird data (yellow) cluster together (cluster B) and are somewhat separated from cluster A, which may indicate that the birds have different PBDE sources than humans, marine mammals, or fish. However, it is wise to remember that most of these bird data are for herring gull eggs from the Great Lakes and that the human, marine

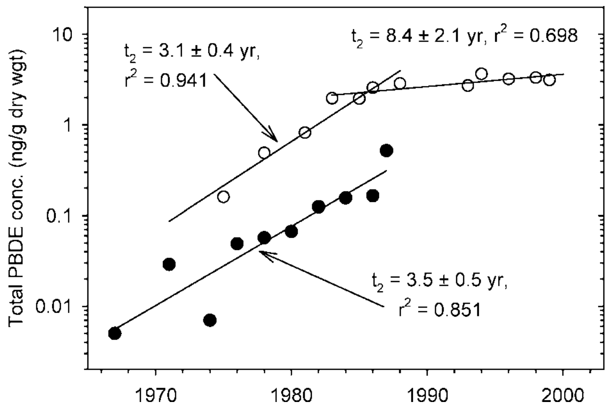

FIGURE 7. ∑PBDE concentrations in two sediment cores (in ng/g dry weight) shown as a function of depth in the core expressed as the year in which the sediment was deposited; see Table 8. The bottom line with filled symbols represents samples from the Baltic Sea (*18*), and the top line with open symbols represents data from Drammenfjord, Norway (*19*). Regressions for the two data sets are shown separately; the doubling times of the pre-1985 data sets are not significantly different.

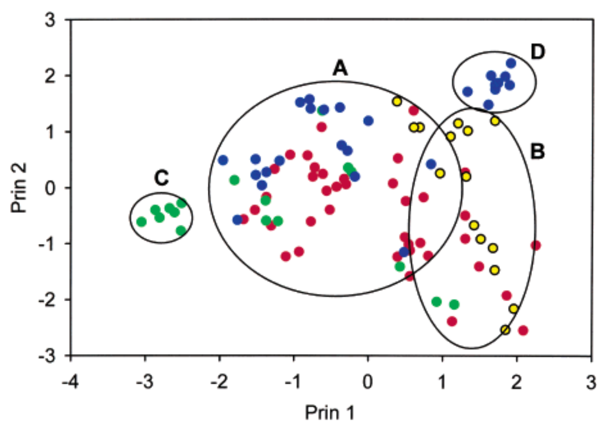

FIGURE 8. Principal component 1 vs principal component 2 for the PBDE congener distribution profiles. Only data with all five of the "main" congeners were used to find the principal components, and only these data are plotted. The green symbols are for marine mammals, the blue are for fish, the red are for people, and the yellow are for birds (primarily herring gull eggs). The clusters A–D are discussed in the text.

mammal, and fish data are mostly from samples collected in Europe. Thus, Figure 8 may simply be telling us that European samples have different PBDE sources than Great Lakes samples. The two most distinct clusters (C on the left and D on the top right) represent self-consistent data sets: C is the Canadian Arctic seal data, and D is the Columbia River whitefish data. The distance between these clusters may indicate very different PBDE sources for these two types of organisms. In general, it is too early to say that one or two BDE congeners or ratios of congeners can be used as "markers" of a particular PBDE source. Data of consistent quality from many more locations around the world are needed before one can approach this problem with even modest certainty.

Research Recommendations. By now it is clear that PBDEs are ubiquitous environmental pollutants and that their concentrations in most environmental compartments are exponentially increasing with doubling times of about 4–6 yr. The mechanisms by which these compounds are leaving the products in which they are used and entering the environment are much less clear. Possibilities include inhalation and ingestion of particles from polyurethane foam or other plastics, volatilization from the plastic itself, consumption of contaminated food, or in the case of human exposure, direct dermal exposure to the flame-resistant product. The latter may be important for human exposure to the penta-product used in polyurethane-foam-filled furniture cushions or mattresses. Research on all of these mechanisms, particularly the latter, is needed.

In most of the human sample data sets, there are a few people with very high levels as compared to the average (see the high outliers in Figure 2). Attention needs to be paid to these data—why are these few people so highly contaminated? Is it related to their occupation or to their home setting? Understanding these outliers may contribute to understanding the mechanisms by which people have become contaminated.

In the United States, regulations banning products containing penta- and octa-BDE will take effect in California in the year 2008 (*20*), and Great Lakes Chemical, one of the major manufacturers, will voluntarily phase-out the production of these two PBDE products by the end of 2004 (*21*). In Europe, the use of the penta-product is already being phased-out (*22*). It will be important to evaluate the effectiveness of these regulations by careful measurements of human blood or milk PBDE levels as a function of time. Presumably, these concentrations will no longer increase and will start to decrease, but it is important to actually track these changes. In fact, there are already indications that PBDE levels in Swedish human milk have decreased over the last few year as a result of regulations that are now being implemented in Europe (*23*, *24*).

As mentioned above, more good-quality, congener-specific data are needed if there is to be any hope of using these data to elucidate sources. It is obviously important to measure all of the industrially significant congeners in all samples. Considering that analytical standards are available for most of the common congeners (and many of the uncommon ones too), such measurements are not difficult. One analytical difficulty is for the decabromo congener (BDE-209), but even in this case, thermal decomposition can be avoided by using a short GC column (10 m will do) and a thermally inert GC injection port. The conventional wisdom that BDE-209 does not accumulate in biota may simply be an analytical artifact; analysts need to ensure that this congener is properly measured by their analytical method by running spiked, positive quality-control samples.

Although not mentioned here, 2,2′,4,4′,5,5′-hexabromobiphenyl (PBB-153) is often present in these samples (*25*). This compound is left over from an industrial accident in Michigan, which contaminated dairy cow feed and thus the milk supply of the entire state in the early-1970s. This compound usually coelutes from most gas chromatographic columns along with BDE-154; thus, analysts need to ensure the separation of these two compounds by using a sufficiently long GC column (60 m will do). Incidentally, the concentrations of PBB-153 did not correlate with those of ∑PBDEs in the Indianapolis, IN, human samples (*25*). Clearly, separating PBB-153 from BDE-154 (with a long GC column) and measuring BDE-209 (with a short column) requires two different GC measurements, but this is a small price to pay for having a complete and accurate data set for every sample.

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