Environ. Sci. Technol. **2008,** *42,* 1377–1384

Serum Concentrations of Polybrominated Diphenyl Ethers (PBDEs) and Polybrominated Biphenyl (PBB) in the United States Population: 2003–2004

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Received October 2, 2007. Revised manuscript received November 7, 2007. Accepted November 14, 2007.

Polybrominated diphenyl ethers (PBDEs) and 2,2′,4,4′,5,5′-hexabromobiphenyl (BB-153) are chemicals known as brominated flame retardants. We have assessed the exposure status of the United States population to PBDEs and BB-153 and explored associations with demographic information, including participants' age, sex, and race/ethnicity. A total of 2,062 serum samples, from participants in the National Health and Nutrition Examination Survey (NHANES) 2003–2004 aged 12 years and older, were analyzed for PBDEs and BB-153; stratified and regression analyses were used to examine levels among demographic groups. The congener with the highest serum concentration was 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) [geometric mean 20.5 ng/g lipid]; followed by 2,2′,4,4′,5,5′-hexaBDE (BDE-153) [5.7 ng/g lipid]; 2,2′,4,4′,5-pentaBDE (BDE-99) [5.0 ng/g lipid; a value equal to the highest limit of detection for an individual sample]; 2,2′,4,4′,6-pentaBDE (BDE-100) [3.9 ng/g lipid]; BB-153 [2.3 ng/g lipid]; and 2,4,4′-triBDE (BDE-28) [1.2 ng/g lipid]. For BDE-47, we observed no significant difference in the least-squares geometric mean (LSGM) by sex, but with age we found both a linear decrease ($p = 0.01$) and a positive quadratic trend ($p = 0.01$). Its LSGM, 27.9 ng/lipid, in the 12–19 year olds decreased to 17.2 ng/g lipid in the 40–49 year group, and then curved upward to 20.4 ng/g lipid in the ≥60 years olds. Mexican Americans had the highest LSGM of BDE-47 (24.5 ng/g lipid), which was significantly higher than that of non-Hispanic whites (19.7 ng/g lipid, $p = 0.01$). Adults 60 years and older were twice as likely as adults 20–59 years old to have a serum BDE-47 concentration above the 95th percentile ($p = 0.02$).These data provide needed exposure assessment data for public health decisions.

Introduction

The National Health and Nutrition Examination Survey (NHANES) provides an ongoing exposure assessment of the civilian, non-institutionalized United States population to a wide range of chemicals through biomonitoring. The survey results are reported in CDC's *National Report on Human Exposure to Environmental Chemicals* (*1*). We report here individual serum concentrations of polybrominated diphenyl ethers (PBDEs) and 2,2′,4,4′,5,5′-hexabromobiphenyl (BB-153), based on NHANES sampling conducted during the years 2003 and 2004.

PBDEs and BB-153 are chemicals commonly known as brominated flame retardants (BFRs), of which PBDEs are the most well-known class (*2*). BB-153, on the other hand, is an older use BFR; its production was discontinued in the mid-1970s in the United States after its accidental contamination into dairy feed (*3*) in Michigan. This incident led to widespread exposure to cattle and poultry, to humans who consumed these food items, and to breast feeding infants of exposed mothers (*3*).

PBDEs are produced in three different technical preparations, commonly named according to their average bromine content; i.e., PentaBDE, OctaBDE, and DecaBDE (*4*). Technical PentaBDE is a mixture of congeners containing approximately 37% by weight tetraBDEs, 43% pentaBDEs, 6.8% hexaBDEs, and trace levels of triBDEs and heptaBDE (*5*). Technical OctaBDE is a mixture of congeners with 6−8 bromine atoms on the diphenyl ether backbone, while the DecaBDE mixture almost exclusively contains BDE-209 (*6*). PBDE congeners containing 7−10 bromine atoms have relatively short biological half-lives in humans; e.g., 2,2′,3,4,4′,5′,6-heptabromophenyl ether (BDE-183) has a half-life of 94 days (95% CI 68–120 days) (*7*). These highly brominated congeners are not likely to biomagnify through the food chain, and thus direct exposures in the indoor environment and occupational exposures are most likely the dominant pathways of human exposure. The half-lives of PBDE congeners with fewer than 7 bromine substituents have not been determined in humans, although, the 2,2′,4,4′,5,5′-hexabromodiphenyl ether (BDE-153) half-life has been estimated to be in the range of 2 years (*8*).

The PentaBDE formulation has been used primarily in the United States. Of the 7,500 metric tonnes of technical PentaBDE produced in 2001, ∼95% was consumed in North America (*9*). The PentaBDE product is used in most modern homes in the United States as a flame retardant in polyurethane, which is used in furniture applications and in the foam padding under wall-to-wall carpets (*4, 10*). The PBDEs originating from the PentaBDE mixture have been reported at much higher concentrations in residential dust from the United States than from Germany, England, and Australia (*11*) and in serum samples worldwide (*12–17*). Technical PentaBDE and OctaBDE were voluntarily phased out in the United States beginning at the end of 2004. However, because of the widespread distribution in U.S. homes, as measured in the indoor environment (*11, 18–20*), and high environmental levels, as measured and documented in, for example, herring gull eggs from the Great Lakes (*21*), it is likely that human body burdens in the United States will remain higher than European levels in the near future.

The OctaBDE and DecaBDE preparations are used in hard plastics, such as those found in the casings of electronic equipment. The 2001 industrial consumptions of OctaBDE and DecaBDE in the United States were estimated to be 1,500 and 24,500 metric tones per year (*9*), which represent 40% and 44% of the total world market, respectively.

From a toxicological standpoint, PBDEs have been shown to cause neurodevelopmental effects, as measured by altered habituation patterns in pups born from mice dosed during

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10.1021/es702451p
Published on Web 01/08/2008

TABLE 1. Polybrominated Diphenyl Ethers and Brominated Biphenyl Measured and Their Abbreviations, Total Number of Valid Measurements, Percentage of Measurements above the Limit of Detection (LOD), and the Maximum LOD Applied

analyte	abbreviation	no. measured	% >LOD	maximum LOD
Polybrominated Diphenyl Ethers (PBDEs)				
2,2′,4-tribromodiphenyl ether	BDE-17	1992	5	1.0
2,4,4′-tribromodiphenyl ether	BDE-28	1987	79	0.8
2,2′,4,4′-hexabromodiphenyl ether	BDE-47	2016	97	4.2
2,3′,4,4′-tetrabromodiphenyl ether	BDE-66	1999	21	1.0
2,2′,3,4,4′-pentabromodiphenyl ether	BDE-85	2000	23	2.4
2,2′,4,4′,5-pentabromodiphenyl ether	BDE-99	1985	65	5.0
2,2′,4,4′,6-pentabromodiphenyl ether	BDE-100	2040	93	1.4
2,2′,4,4′,5,5′-hexabromodiphenyl ether	BDE-153	2039	93	2.2
2,2′,4,4′,5,6′-hexabromodiphenyl ether	BDE-154	2014	51	0.8
2,2′,3,4,4′,5′,6-heptabromophenyl ether	BDE-183	1993	15	1.7
Polybrominated Biphenyl (PBB)				
2,2′,4,4′,5,5′-hexabromobiphenyl	BB-153	2032	83	0.8

a critical window of the developing mice fetus (*22*). These experiments also indicated that 2,2′,4,4′,5-pentabromodiphenyl ether (BDE-99) is more potent than 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) (*22*). Potential toxicological end points of concern for PBDEs also include thyroid hormone disruption and cancer (*23*).

We report here serum concentrations of ten PBDE congeners (tri to heptaBDEs) and BB-153 (Table 1) in a representative sampling of U.S. residents in 2003 and 2004; we assess the variation of concentrations by age, sex, and race/ethnicity. Further, the influences of lifestyle factors, such as the age of the primary residence and country of origin, have been investigated. DecaBDE was not included in this survey due to difficulties with analytical measurement.

Experimental Section

Survey Overview and Sampling. The National Center for Health Statistics (NCHS) and the National Center for Environmental Health (NCEH), both of the Centers for Disease Control and Prevention (CDC), administered NHANES and performed the analytical analyses, respectively. The goal of the NHANES is to obtain a representative sample of the noninstitutionalized population residing in the United States and thus by assessing the health and nutrition status of this subsample of the population, estimate the status of the population as a whole. A statistically representative sample of the population is collected by dividing the country into geographic areas known as primary sampling units (PSUs). The PSUs are then combined to form strata, and each strata is then divided into a series of neighborhoods. Households are thereafter chosen at random from these neighborhoods, and residents are interviewed to determine eligibility for participation in the survey. Over-sampling of some demographic groups, such as Mexican Americans and the socioeconomically underserved, ensures reliable sample size for these demographic groups. This over-sampling is adjusted during data analyses by sampling weights. Each year approximately 5,000 subjects are recruited from 15 locations across the United States. However, the NHANES is not designed to assess geographical differences. A random one-third subset of eligible participants ($N = 2{,}305$) age 12 years and above of the NHANES 2003/2004 survey was chosen for measurement of BFRs. Of these, 2,062 serum samples were available for analysis. The population consisted of 48.4% male subjects. The race/ethnicity distribution of the sample was 71.5% non-Hispanic white (NHW), 12.0% non-Hispanic black (NHB), and 8.2% Mexican American (MA); other race/ethnicity groups corresponded to 8.3%. The percentages of participants below the age of 20 years and above the age of 60 years were 12.2 and 17.0%, respectively.

Analytical Methods. All sample preparation work was performed in a clean room; its supply air is passed through Ultra-Low Penetration Air (ULPA) filters, which have a 99.999% efficiency for removal of particles of 0.12 μm diameter or larger. Inside the clean room, biological safety cabinets are used as a second barrier against particle contamination during all phases of sample handling. The analytical methodology employed for the measurement of BFRs in serum has been reported previously (*24*); detailed information on the determination of the limit of detection (LOD) is given in SI-1 the Supporting Information.

Statistical Methods. SAS software (version 9.1.3, SAS Institute, Cary, NC) and SUDAAN software (version 9.0.1, Research Triangle Institute, Research Triangle Park, NC) were used to produce estimates, regression coefficients, and related standard errors. We used sample weights to account for the unequal probability of selection and log transformed data to compute all the estimates. Geometric means are calculated for analytes detected in ≥60% of the samples. For a concentration below the maximum LOD, a value equal to the LOD divided by the square root of 2 was used. Weighted Pearson correlation coefficients were calculated in SAS.

For race/ethnicity group, we included all samples in the calculation for total estimates, but we only used MA, NHB, and NHW for other descriptive analyses and for regression analyses. We stratified age by 12–19, 20–39, 40–59 years old, and 60 years and older for the geometric mean and the various percentiles.

We compared the brominated chemical concentrations by the participants' country of birth (inside or outside of U.S.; variable DMDBORN) and by the construction year of the residents' housing (before 1959, 1960−1977, and 1978 and later). Because only 74.5% of the participants responded to construction year of the residents' housing question and because the number of the foreign born was very small in the NHW ($n = 50$) and the NHB ($n = 52$), we did not include these two variables in the multiple regression.

We used analysis of covariance to examine the influence of demographic variation of the log-transformed serum concentrations for the analytes with ≥60% detection frequency; i.e., BB-153, BDE-28, BDE-47, BDE-99, BDE-100, and BDE-153. The least-squares geometric mean (LSGM) was calculated from the regression model. For multiple regression analyses, the variables included in the initial model are age (continuous), sex (male or female), and race (MA, NHB, and NHW). We also included an age-squared term in the model for BB-153, BDE-28, BDE-47, BDE-99, BDE-100, and BDE-153 because the unadjusted geometric mean by age groups indicated a curvilinear relationship between age and the PBDE lipid-adjusted serum concentration. When both age and age-squared were included in the model, we found that age and age-squared terms had the least correlation at age 46 years old, so to avoid multicollinearity, we subtracted 46

from each participant's age (*25*). We assessed all possible two-way interaction terms in the model. To evaluate the relation between the log-transformed concentration of all analytes and age, we changed the continuous age to be a categorical age-decades variable in the model in order to generate a bar chart of LSGM concentrations by age decades.

To reach the final model, we used backward elimination with a threshold of $p < 0.05$ for retaining the variable in the model, using Satterwaite-adjusted F statistics. We evaluated for potential confounding by adding each of the excluded variables back into the final model one by one and examining changes in the β coefficients of the statistically significant main effects. If addition of one of these excluded variables caused a change in a β coefficient by ≥10%, the variable was added back into the model.

We conducted weighted univariate and multiple logistic regression to examine the association of BDE-47 or BDE-153 concentrations above the 95th percentile with sex (male, female), age (12–19, 20–59, and 60 years and older), and race/ethnicity. These congeners were selected based on their having the highest serum concentrations.

Results

The serum concentrations of ten PBDE congeners with 3−7 bromine substituents and BB-153 were measured in 2,062 serum samples from the NHANES 2003/2004; 99% were analyzed successfully giving a total of 2,040 samples with data available for statistical analysis. BDE-47 was measured above its LOD in 2016 samples and also the concentration of this congener was the highest in all stratifications of the population and the entire population. Its geometric mean concentration in all participants was 20.5 ng/g lipid; followed by BDE-153 (5.7 ng/g lipid), BDE-99 (5.0 ng/g lipid; a value equal to the highest limit of detection for an individual sample), BDE-100 (3.9 ng/g lipid), BB-153 (2.3 ng/g lipid), and BDE-28 (1.2 ng/g lipid) (Tables 2 and 3). Geometric means could not be calculated for BDE-17, BDE-66, BDE-85, BDE-154, and BDE-183 because these congeners had a detection frequency of less than 60% (Table 1). The geometric mean concentration and various percentiles stratified by age groups (age 12–19, 20–39, 40–59, and ≥60 years), race/ethnicity (MA, NHB, NHW), and sex are given in Tables 2 and 3 and Figure 1 for congeners with a detection frequency above 60%, while congeners with a lower detection frequency are given in Table S1 in the Supporting Information.

When examining these data by multiple regression, the final model for BB-153 included sex, age, age-square ($p < 0.01$), race ($p < 0.01$), and an interaction term between age and sex ($p = 0.02$); for BDE-28, age ($p = 0.18$) and age-squared ($p = 0.04$); for BDE- 47, race ($p = 0.06$), age ($p = 0.01$), and age-squared ($p = 0.01$); for BDE-99, race ($p = 0.02$), age ($p = 0.01$), and age-squared ($p < 0.01$); for BDE-100, age ($p < 0.01$), and age-squared ($p = 0.045$); and for BDE-153, sex ($p < 0.01$), age ($p < 0.01$), and age-squared ($p = 0.02$).

We observed a curvilinear relationship between the log-transformed lipid-adjusted BDE-47 concentration and age (Tables S2 and S3, and Figure S1 in the Supporting Information). We found both a linear decrease and positive quadratic trend with age (linear $p = 0.01$, quadratic $p = 0.01$), cf. Table S3. The LSGM of BDE-47 had the highest level (27.9 ng/g lipid) in the 12–19 years old and decreased through the 40–49 years age group (17.2 ng/g lipid) and then curved upward to reach 20.4 ng/g lipid in the ≥60 year olds. MA participants had the highest LSGM of BDE-47 (24.5 ng/g lipid), which was significantly higher than that for NHW (19.7 ng/g lipid, $p = 0.01$). MA and NHB had similar LSGMs for BDE-47 concentrations. Similarly, no significant difference of LSGMs for BDE-47 concentrations was observed between males and females.

For log-transformed lipid-adjusted BDE-153, we also observed a similar curvilinear trend with age—both a linear decrease ($p < 0.01$) and a positive quadratic ($p = 0.02$) trend (Tables S2 and S3). The LSGM of BDE-153 had the highest level in the 12–19 years old sample (8.0 ng/g lipid). Female participants had significantly lower BDE-153 lipid adjusted concentrations then males ($p < 0.01$). No significant difference of the LSGMs for BDE-153 concentrations was observed among race/ethnicity groups.

For BDE-99, we also observed a curvilinear relationship with age (Tables S2 and S3). The LSGM of lipid-adjusted BDE-99 concentrations was the highest in the 12–19 years old samples (6.8 ng/g lipid). We observed a linear decreasing trend ($p < 0.01$) as well as a positive quadratic trend with age ($p = 0.01$). For BDE-99: NHB participants had the highest LSGM (6.1 ng/g lipid), which was significantly higher than the LSGM for NHW (4.8 ng/g lipid, $p = 0.02$). The LSGM of NHWwas significantly lower than the LSGM for MA (5.7 ng/g lipid, $p = 0.046$). There was no significant difference in the LSGMs for BDE-99 between NHB and MA, nor between males and females.

Similarly, we observed the curvilinear relationship between age and the log-transformed lipid-adjusted BDE-100 concentration. The highest LSGM of BDE-100 was in the 12–19 years old group with both a linear decreasing trend ($p < 0.01$) and a quadratic trend ($p = 0.045$). There was no significant difference in the LSGMs of BDE-100 among race/ethnicity groups or between the sexes.

Although there was a curvilinear relationship between age and the log-transformed lipid-adjusted BDE-28 concentration, only the positive quadratic curve was significant ($p = 0.04$). No significant difference for LSGM of BDE-28 was observed among race/ethnicity groups or between sexes.

For the log-transformed concentration of BB-153, there was a relationship with age (Tables S2 and S3, and Figure S1) characterized by a positive linear trend ($p < 0.01$) and a small negative quadratic trend ($p < 0.01$). We also observed that both males and females had an increasing LSGM with age ($p < 0.01$ for both male and female); however, concentrations in males increased more sharply with age than in females ($\beta = 0.002$, $p = 0.02$). The LSGM of BB-153 in males is significantly higher than in female regardless of age ($p < 0.001$). The LSGM of both NHB and NHW were higher than that of MA ($p = 0.001$ and $p < 0.001$ for NHB and NHW, respectively). NHB and NHW had similar LSGM concentrations of BB-153.

Relating to potential pathways of exposure, we note that all PBDE congeners with a detection frequency over 60% were significantly correlated with each other (Table S4). BB-153 was significantly correlated ($p = 0.04$) only with BDE-47 (Table S4). BDE-153 was found at a higher concentration than BDE-47 in 10.5% of the participants. The median ratio of BDE-153 to BDE-47 was 0.54 (quartile range 0.38–0.75).

BB-153 was found at 18% higher geometric mean concentration in participants living in housing constructed between 1960 and 1977 as compared to participants living in housing constructed after 1978 ($p = 0.03$), cf. Table 4. The geometric means of BDE-47 and BDE-153 did not vary significantly with the age of the participant's primary residence (Table 4). The geometric mean of BDE-153 was found to be 31% lower ($p < 0.01$) in foreign-born subjects than in participants born in the United States, while for BDE-47 no significant difference was observed between these groups (Table 4). The geometric mean for BB-153 was 62% lower in foreign-born participants as compared to participants born in the United States ($p < 0.01$).

In examining those with the highest serum concentrations (above the 95th percentile) of BDE 47, age, but not sex and race/ethnicity, was significantly associated. Senior adults (60 years and older) were two times more likely to be above the

TABLE 2. Geometric Mean (GM) Concentrations (ng/g lipid), Selected Percentiles, and Their 95% Confidence Intervals (95% CI) for 2,4,4′-Tribromodiphenyl Ether (BDE-28), 2,2′,4,4′-Hexabromodiphenyl Ether (BDE-47), and 2,2′,4,4′,5-Pentabromodiphenyl Ether (BDE-99) for All Participants in the NHANES 2003/2004 Stratified by Age, Race/Ethnicity, and Sex[a]

demographic group	geomean GM (95% CI)	percentiles P10 (95% CI)	P25 (95% CI)	P50 (95% CI)	P75 (95% CI)	P90 (95% CI)	P95 (95% CI)
2,4,4′-tribromodiphenyl ether (BDE-28)							
all participants	1.2 (1.0–1.4)	<LOD	<LOD	1.1 (0.9–1.2)	2.1 (1.9–2.5)	4.8 (3.3–6.7)	7.9 (5.3–10.9)
age groups							
12–19 years	1.3 (1.2–1.5)	<LOD	<LOD (<LOD−0.8)	1.2 (1.0–1.3)	2.3 (2.0–2.6)	3.9 (2.8–4.9)	6.0 (4.0–9.4)
20–39 years	1.1 (1.0–1.3)	<LOD	<LOD	1.1 (0.8–1.3)	1.9 (1.5–2.3)	3.4 (2.4–5.1)	6.1 (3.7–9.3)
40–59 years	1.1 (0.9–1.3)	<LOD	<LOD	1.0 (0.8–1.3)	2.1 (1.7–2.7)	4.7 (2.8–8.3)	8.2 (4.6–11.8)
>60 years	1.4 (1.1–1.7)	<LOD	<LOD	1.1 (0.9–1.4)	3.2 (2.1–4.8)	6.6 (5.6–7.7)	9.6 (7.2–18.1)
race/ethnicity							
MA	1.4 (1.3–1.6)	<LOD	<LOD (<LOD−0.8)	1.4 (1.2–1.5)	2.3 (1.9–2.8)	4.7 (3.7–5.5)	7.3 (5.6–8.3)
NHB	1.2 (1.0–1.4)	<LOD	<LOD	1.0 (0.8–1.2)	2.1 (1.8–2.4)	5.2 (3.5–6.7)	8.4 (5.4–12.3)
NHW	1.2 (1.0–1.4)	<LOD	<LOD	1.1 (0.8–1.3)	2.2 (1.8–2.7)	4.8 (3.0–8.0)	8.0 (4.5–13.6)
sex							
female	1.2 (1.0–1.4)	<LOD	<LOD	1.0 (0.8–1.3)	2.1 (1.7–2.5)	4.6 (3.0–6.7)	7.7 (4.7–11.8)
male	1.2 (1.1–1.4)	<LOD	<LOD	1.0 (0.9–1.2)	2.2 (2.0–2.7)	5.1 (3.5–7.3)	8.0 (5.7–11.3)
2,2′,4,4′-hexabromodiphenyl ether (BDE-47)							
all participants	20.5 (17.6–23.9)	5.0 (4.3–6.1)	9.3 (8.2–10.9)	19.2 (15.7–22.3)	40.9 (35.6–49.2)	84.9 (66.8–127.0)	157.0 (108.0–240.0)
age groups							
12–19 years	28.2 (24.6–32.3)	7.8 (5.2–10.6)	14.1 (12.0–16.6)	27.2 (22.1–33.6)	53.6 (44.9–63.6)	104.0 (82.4–145.0)	174.0 (115.0–211.0)
20–39 years	21.5 (18.3–25.4)	6.5 (4.8–7.5)	10.3 (9.2–12.0)	21.6 (17.0–25.1)	38.3 (34.8–44.2)	72.0 (53.0–102.0)	114.0 (73.9–216.0)
40–59 years	17.7 (14.4–21.7)	4.6 (<LOD-5.5)	7.7 (6.3–10.0)	14.7 (12.3–19.2)	35.8 (27.1–50.8)	84.9 (54.9–142.0)	142.0 (94.3–240.0)
>60 years	19.7 (15.8–24.5)	<LOD (<LOD−5.0)	7.7 (6.1–9.1)	16.9 (13.2–21.7)	48.9 (31.2–70.7)	108.0 (81.4–173.0)	235.0 (171.0–283.0)
race/ethnicity							
MA	25.5 (23.0–28.1)	8.0 (7.2–9.1)	12.0 (10.9–14.4)	23.4 (21.2–25.5)	47.1 (38.2–56.5)	85.8 (72.0–105.0)	151.0 (104.0–188.0)
NHB	24.3 (20.9–28.2)	6.0 (4.8–7.4)	10.3 (9.3–11.2)	21.3 (18.2–25.4)	47.5 (40.7–53.2)	116.0 (81.8–149.0)	242.0 (136.0–481.0)
NHW	19.5 (16.1–23.7)	4.7 (<LOD−5.6)	8.6 (7.5–10.1)	17.3 (14.4–22.2)	40.2 (33.1–51.9)	85.1 (60.3–142.0)	157.0 (90.2–283.0)
sex							
female	19.6 (16.4–23.5)	4.8 (<LOD−6.1)	9.2 (8.0–10.7)	19.1 (13.9–23.0)	38.3 (31.5–44.7)	79.2 (60.7–121.0)	155.0 (102.0–235.0)
male	21.4 (18.1–25.3)	5.1 (<LOD−6.6)	9.5 (7.8–11.3)	19.2 (15.7–24.0)	45.2 (37.3–54.9)	94.3 (66.8–148.0)	168.0 (112.0–382.0)
2,2′,4,4′,5-pentabromodiphenyl ether (BDE-99)							
all participants	5.0 (4.4–5.6)	<LOD	<LOD	<LOD	9.2 (7.5–10.9)	21.7 (17.0–29.1)	42.2 (33.3–54.8)
age groups							
12–19 years	6.9 (6.1–7.7)	<LOD	<LOD	5.6 (<LOD−7.6)	12.8 (11.4–15.7)	27.9 (19.6–37.9)	45.2 (35.9–56.8)
20–39 years	5.2 (4.4–6.1)	<LOD	<LOD	<LOD (<LOD−5.6)	8.3 (7.1–10.6)	18.0 (13.2–22.6)	28.8 (20.4–52.7)
40–59 years	[b]	<LOD	<LOD	<LOD	8.7 (5.5–10.9)	19.7 (14.4–31.9)	41.2 (25.1–80.7)
>60 years	[b]	<LOD	<LOD	<LOD	9.4 (6.5–12.8)	29.3 (15.3–47.9)	52.3 (33.5–74.0)
race/ethnicity							
MA	5.9 (5.5–6.4)	<LOD	<LOD	5.4 (<LOD−5.7)	10.6 (9.3–12.6)	20.0 (17.0–23.5)	30.8 (24.5–41.7)
NHB	6.2 (5.4–7.1)	<LOD	<LOD	5.0 (<LOD−5.7)	11.3 (9.5–12.8)	30.2 (21.5–42.2)	74.7 (30.2–155.0)
NHW	[b]	<LOD	<LOD	<LOD	8.8 (6.8–11.3)	21.5 (15.3–34.0)	43.6 (30.7–71.4)
sex							
female	[b]	<LOD	<LOD	<LOD	8.6 (6.6–10.6)	18.3 (14.7–28.8)	41.2 (22.9–60.3)
male	5.3 (4.5–6.1)	<LOD	<LOD	<LOD (<LOD−5.1)	10.0 (8.4–11.5)	23.8 (18.0–37.3)	44.0 (33.8–57.3)

[a] The limits of detection are given in Table 1. All data adjusted for design sampling weights. Abbreviations: MA, Mexican American; NHB, non-Hispanic black; NHW, non-Hispanic white. [b] Estimate is below the highest limit of detection for individual samples.

TABLE 3. Geometric Mean (GM) Concentrations (ng/g lipid), Selected Percentiles, and Their 95% Confidence Intervals (95% CI) for 2,2′,4,4′,6-Pentabromodiphenyl Ether (BDE-100), 2,2′,4,4′,5,5′-Hexabromodiphenyl Ether (BDE-153), and 2,2′,4,4′,5,5′-Hexabromobiphenyl (BB-153) for All Participants in the NHANES 2003/04 Stratified by Age, Race/Ethnicity, and Sex[a]

demographic group	geomean GM (95% CI)	percentiles P10 (95% CI)	P25 (95% CI)	P50 (95% CI)	P75 (95% CI)	P90 (95% CI)	P95 (95% CI)
2,2′,4,4′,6-pentabromodiphenyl ether (BDE-100)							
all participants	3.9 (3.4–4.5)	<LOD	1.6 (1.5–2.0)	3.6 (3.1–4.1)	7.8 (6.8–9.0)	18.3 (15.4–22.0)	36.5 (24.6–54.0)
age groups							
12–19 years	5.2 (4.5–6.0)	<LOD (<LOD−1.8)	2.6 (2.2–3.1)	4.8 (3.8–6.1)	9.4 (7.9–12.8)	19.3 (14.4–26.2)	34.3 (25.0–45.0)
20–39 years	4.4 (3.7–5.2)	<LOD	2.1 (1.7–2.6)	4.1 (3.3–5.0)	8.0 (6.6–10.0)	17.1 (13.2–21.1)	23.6 (19.7–47.1)
40–59 years	3.3 (2.8–4.1)	<LOD	<LOD (<LOD−1.6)	2.8 (2.1–3.4)	6.2 (5.3–7.5)	16.3 (10.5–29.2)	40.5 (19.5–91.6)
>60 years	3.6 (3.0–4.4)	<LOD	<LOD (<LOD−1.7)	2.9 (2.5–3.4)	8.1 (6.9–11.1)	21.5 (15.4–46.0)	56.6 (26.0–74.8)
race/ethnicity							
MA	4.6 (4.0–5.2)	<LOD (<LOD−1.6)	2.4 (2.2–2.6)	4.2 (3.8–5.2)	8.1 (6.5–9.0)	14.5 (11.4–20.3)	26.2 (20.3–36.2)
NHB	4.7 (4.0–5.6)	<LOD	2.0 (1.6–2.2)	4.2 (3.3–5.2)	9.4 (7.6–10.9)	23.7 (16.6–34.3)	39.2 (26.0–79.2)
NHW	3.8 (3.2−4.5)	<LOD	1.5 (<LOD−1.8)	3.3 (2.8−4.1)	7.6 (6.0–9.5)	18.5 (14.5–23.3)	38.8 (22.6–59.2)
sex							
female	3.7 (3.2–4.4)	<LOD	1.6 (<LOD−1.8)	3.2 (2.8–4.1)	7.0 (6.0–8.0)	18.4 (14.4–23.1)	32.8 (23.3–46.0)
male	4.2 (3.6–4.9)	<LOD	1.6 (1.5–1.9)	3.8 (3.1–4.4)	8.4 (7.5–9.9)	17.9 (14.9–25.1)	44.1 (21.9–61.5)
2,2′,4,4′,5,5′-hexabromodiphenyl ether (BDE-153)							
all participants	5.7 (5.1–6.3)	<LOD	2.4 (<LOD−2.6)	4.8 (4.1–5.3)	11.3 (9.8–12.7)	32.6 (27.9–40.3)	65.7 (54.9–88.4)
age groups							
12–19 years	8.1 (6.7–9.7)	2.2 (<LOD−2.7)	3.6 (3.0–5.2)	7.4 (6.0–9.5)	15.3 (11.7–19.4)	30.8 (22.9–44.2)	52.9 (35.9–68.5)
20–39 years	6.6 (5.6–7.9)	<LOD	2.8 (2.3–3.3)	5.4 (4.7–6.3)	13.7 (10.9–18.0)	34.6 (22.5–59.6)	63.5 (50.4–88.4)
40–59 years	4.6 (3.9–5.4)	<LOD	<LOD (<LOD−2.2)	3.6 (3.1–4.4)	7.3 (5.8–10.0)	28.5 (16.1–42.7)	62.8 (31.6–115.0)
>60 years	5.1 (4.3–6.1)	<LOD	<LOD (<LOD−2.3)	3.9 (3.3–5.1)	10.6 (8.4−13.6)	45.3 (20.9–58.9)	91.5 (58.9–134.0)
race/ethnicity							
MA	5.1 (4.3–6.1)	<LOD	2.7 (<LOD−3.1)	4.7 (4.2–5.4)	8.6 (6.5–11.0)	17.2 (13.0–25.2)	34.0 (18.3–55.1)
NHB	6.1 (5.4–6.8)	<LOD	2.4 (2.2–2.8)	5.5 (4.7–6.1)	12.9 (9.8–16.5)	30.0 (21.0–42.7)	53.0 (36.8–63.3)
NHW	5.9 (5.0–6.8)	<LOD	2.4 (<LOD−2.7)	4.9 (3.9–5.5)	11.9 (9.8–14.7)	39.0 (28.5–54.9)	75.9 (58.0–93.2)
sex							
female	4.8 (4.2–5.4)	<LOD	<LOD (<LOD−2.4)	4.0 (3.4–5.0)	9.7 (7.4–11.7)	25.9 (20.2–31.6)	54.5 (34.6–62.9)
male	6.9 (6.0–7.8)	<LOD	2.9 (2.4–3.2)	5.5 (4.7–6.5)	12.6 (11.2–16.1)	47.2 (31.0–62.9)	88.4 (63.4–115.0)
2,2′,4,4′,5,5′-hexabromobiphenyl (BB-153)							
all participants	2.3 (1.8–2.9)	<LOD	0.9 (<LOD−1.3)	2.1 (1.9–2.4)	4.4 (3.4–6.3)	12.8 (6.6–25.5)	26.1 (11.7–60.5)
age groups							
12–19 years	0.7 (0.6–0.8)	<LOD	<LOD	<LOD	1.0 (0.8–1.4)	2.7 (2.0–3.0)	3.7 (2.9–4.7)
20–39 years	1.6 (1.3–2.1)	<LOD	<LOD (<LOD−0.9)	1.6 (1.2–1.8)	3.0 (2.3–3.9)	6.6 (3.9–12.2)	12.2 (7.0–36.7)
40–59 years	3.4 (2.6–4.4)	0.8 (<LOD−1.5)	1.8 (1.6–2.1)	2.9 (2.4–3.2)	5.9 (3.7–11.9)	15.9 (7.9–38.9)	36.9 (13.7–70.3)
>60 years	4.5 (3.2–6.2)	1.4 (1.2–1.6)	2.0 (1.8–2.5)	3.4 (2.8–4.5)	8.1 (4.7–15.8)	25.0 (8.8–69.8)	52.8 (19.2–114.0)
race/ethnicity							
MA	1.1 (0.9–1.3)	<LOD	<LOD	1.1 (<LOD−1.3)	2.5 (2.0–2.9)	6.0 (3.5–8.6)	10.0 (6.5–15.2)
NHB	2.4 (1.6–3.4)	<LOD	0.8 (<LOD−1.4)	2.1 (1.6–2.8)	4.8 (3.0–12.1)	13.9 (7.0–38.2)	29.7 (12.1–70.2)
NHW	2.7 (2.1–3.4)	<LOD	1.2 (0.9–1.6)	2.4 (2.2–2.8)	4.9 (3.4–7.3)	13.5 (6.6–35.4)	34.1 (11.6–70.3)
sex							
female	1.9 (1.5–2.5)	<LOD	<LOD (<LOD−1.1)	2.0 (1.6–2.2)	3.6 (2.8–4.9)	9.7 (4.6–27.5)	23.9 (7.4–56.6)
male	2.8 (2.2–3.5)	<LOD	1.1 (0.8–1.6)	2.6 (2.2–3.1)	5.3 (3.8–8.4)	15.8 (9.3–27.2)	34.9 (13.5–70.3)

[a] The limits of detection are given in Table 1. All data adjusted for design sampling weights. Abbreviations: MA, Mexican Americans; NHB, non-Hispanic black; NHW, non-Hispanic white.

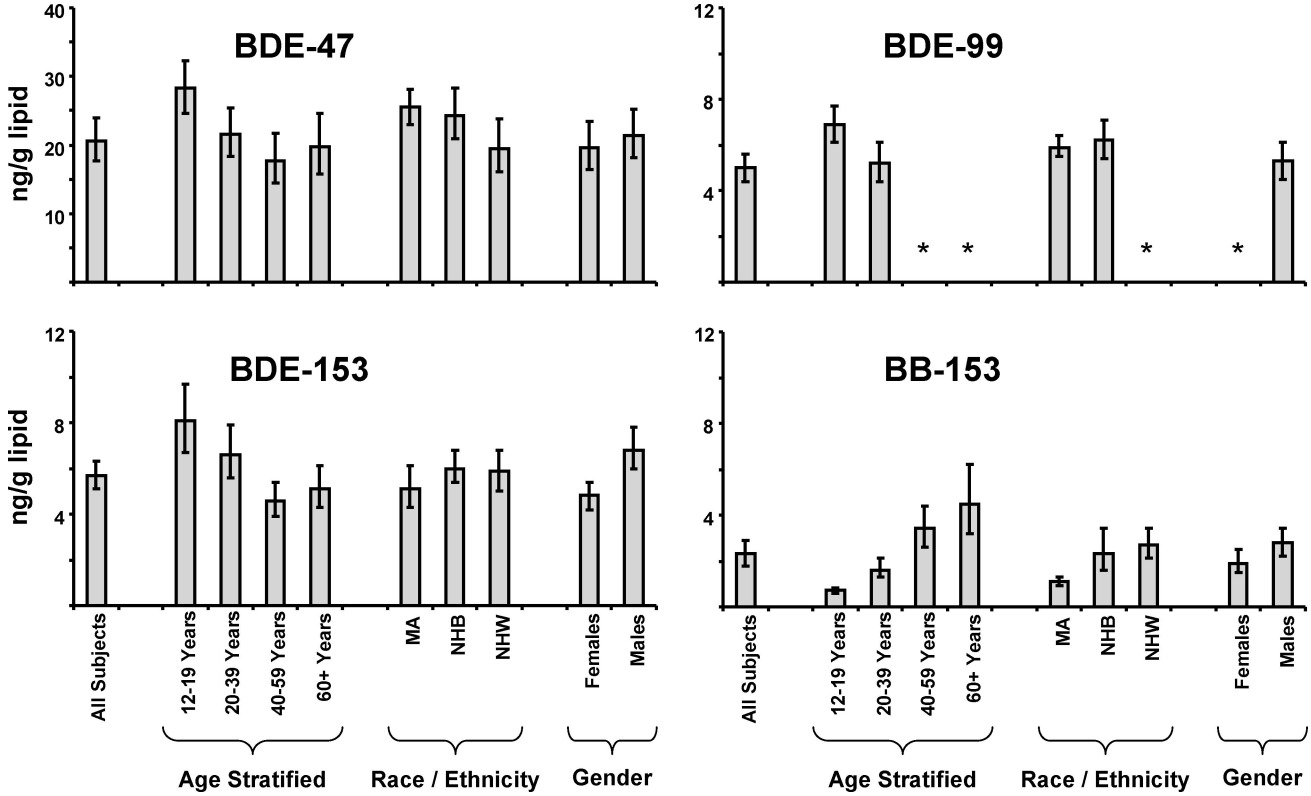

FIGURE 1. Geometric mean concentrations (ng/g lipid) and 95% confidence intervals adjusted for design sampling weights. Data stratified by age, race/ethnicity (MA, Mexican-Americans; NHB, non-Hispanic black; and NHW, non-Hispanic white), and sex. Congener abbreviations are given in Table 1. * Estimate is below the highest limit of detection for individual samples.

TABLE 4. Geometric Mean (GM) Concentrations (ng/g lipid) of 2,2′,4,4′,5,5′-Hexabromobiphenyl (BB-153), 2,2′,4,4′-Tetrabromodiphenyl Ether (BDE-47), and 2,2′,4,4′,5,5′-HexaBDE (BDE-153) in Participants Having a Primary Residence Constructed before 1959, between 1960 and 1977, and after 1978, and in Foreign-Born Participants and Participants Born in the United States, Adjusted for Design Sampling Weights[a]

	serum level (ng/g lipid)							pair-wise comparisons		
	BB-153		BDE-47		BDE-153					
strata	GM (95%CI)	N	GM (95%CI)	N	GM (95%CI)	N	group comparison	BB-153 *p*-value	BDE-47 *p*-value	BDE-153 *p*-value
construction year of primary residence										
<1959	2.7(1.9–3.9)	574	19.8(16.6–23.6)	564	5.7(4.8–6.8)	575	>1978 vs 1960–77	**0.03**	0.45	0.14
1960–77	2.6(2.0–3.4)	430	19.3(14.8–25.3)	428	5.0(3.9–6.4)	430	>1978 vs <1959	0.10	0.78	0.49
>1978	2.2(1.8–2.7)	534	20.4(17.1–24.4)	536	6.2(5.1–7.5)	536	1960–77 vs <1959	0.56	0.86	0.36
country of origin										
United States	2.6(2.0–3.3)	1658	20.7(17.4–24.6)	1647	6.0(5.4–6.7)	1664	U.S. vs Other	<**0.01**	0.46	<**0.01**
other	1.0(0.84–1.3)	373	19.4(17.1–22.0)	368	4.1(3.3–5.0)	374				

[a] All data adjusted for design sampling weights. *p*-Values for all pair wise comparisons are given.

95th percentile than adults 20–59 years old ($p = 0.02$) (odds ratio (OR) (95% CI): 2.0 (1.1, 3.6)); and 54% more likely than adolescents to be above the 95th percentile ($p = 0.067$) (OR (95% CI): 1.5 (1.0, 2.5). There was no difference in the likelihood of being above the 95th percentile between the 12–19 and 20–59 year old age groups.

For BDE-153, sex and race, but not age, were significantly associated with being above the 95th percentile univariately ($p = 0.018$, 0.03 for sex and race, respectively). In the final multiple logistics regression, sex ($p = 0.04$) and race ($p = 0.03$) were significant. We found that males were 2.1 times more likely than females to be above the 95th percentile (adjusted OR (95% CI): 2.1 (1.0, 4.3)). MA were 62% less likely than NHW to be above the 95th percentile ($p = 0.03$) (adjusted OR (95% CI): 0.38 (0.15, 0.91)). There was no significant difference between NHB and MA.

Discussion

These NHANES data from a representative civilian, noninstitutionalized U.S. population assess the internal dose concentrations of PBDE congeners and of BB-153, the congener of highest concentration in commercial PBBs. Among the PBDE congeners, BDE-47 was detected in serum from almost all of the participants and at the highest concentrations. Compared to median data from smaller nonrepresentative European studies, the median NHANES BDE-47 serum concentration of 19.2 ng/g lipids is 7.1–35 times higher (*13–15*). Among the PBDE congeners, in general BDE-47 serum concentrations were the highest, but 10.5% of persons had higher BDE-153 concentrations, which is present in both the PentaBDE and OctaBDE formulations (*6*); these formulations have different commercial applica-

tions. Because of the short serum half-life of BDE-183, which is found only in the OctaBDE formulation, BDE-153 concentrations or the ratio of concentrations of BDE-153/BDE-47 could potentially reflect exposure to the OctaBDE formulation, although the variance in this ratio could potentially also be explained by differences in pharmacokinetics. The presence of BDE-153 in the two formulations is also indicated by the lower correlation coefficients between its concentrations and those of the other prevalent PBDE congeners, which are found only in the PentaBDE formulation (Table S4).

In multiple regression models, we observed both a significant linear decrease and a positive quadratic trend with age for PBDE congeners detectable in more than 60% of the participants (Tables S2 and S3), in contrast to the traditional chlorinated persistent organic pollutants which tend to increase as age increases (*26*). The higher concentrations in the youngest age groups examined (Tables S2 and S3) are probably due to their lifestyle and activity differences, which lead to higher exposures, although potential differences in the pharmacokinetics of these chemicals in their bodies must be considered. The slight increase in the oldest participants could potentially be explained by bioaccumulation and/or by a slower metabolism and elimination of PBDEs at an older age.

For public health decisions, scientists are especially interested in persons with concentrations above the 95th percentile. For BDE-47 concentrations, participants aged 60 years and older were twice as likely to be above the 95th percentile as those aged 20–59 years. With regards to race/ethnicity, the BDE-47 LSGM of MA was significantly higher than for NHW; however, although this was not statistically significant, the likelihood for being above the 95th percentile was greater for NHW than for MA. For BDE-153, we did see differences with sex and race/ethnicity as males were twice as likely as females to have a concentration greater than the 95th percentile. This difference in concentrations between the sexes is consistent with the LSGM data for BDE-153. Also, NHW were more likely than MA to have a BDE-153 concentration above the 95th percentile, which was not indicated with the LSGM data from multiple regression analyses. The finding that MA participants are less likely to have the higher serum concentrations of BDE-153 as well as BB-153 is consistent with a higher proportion of immigrants who likely experienced a lower level of exposure in their country of origin, cf. Table 4.

In multiple regression models, the LSGMs of BB-153 increased linearly with age with a relatively small significantly negative quadratic term and a sex interaction term. An increasing concentration of BB-153 with age (Tables S2 and S3) is consistent with other traditional chlorinated persistent organic pollutants (POPs) (*26*) that were phased out in the 1970s. This increasing trend is because older participants have been exposed to BB-153 for a longer period of time than younger participants and that the older participants also were exposed to these chemicals when the environmental levels were higher than today. For BB-153, higher LSGM concentrations were found in males in all age and race/ethnicity groups (Figure S1 and Tables S2 and S3); this differs somewhat from data for traditional POPs which were found in higher concentrations in older females compared to older males (*26*). The observed sex difference potentially may be explained by depuration during nursing.

In addition to lower BB-153 concentrations being found in MA, the geometric mean for BB-153 was 62% lower in foreign-born participants compared to participants born in the United States ($p < 0.001$), cf. Table 4. We also found a significantly higher geometric mean BB-153 concentration in participants living in housing constructed between 1960 and 1977 (2.6 ng/g lipid; 95% CI 2.0–3.4) compared to participants living in housing constructed after 1978 (2.2 ng/g lipid; 95% CI 1.8–2.7, $p = 0.03$). This observed difference in geometric mean concentration by the construction year of primary residence could be explained by the fact that BB-153 was phased out in the United States in the mid-1970s and hence would be less likely to be found in homes constructed after 1978. Also, BDE-153 was found at a lower geometric mean concentration in foreign-born participants (4.1 ng/g lipid; 95% CI 3.3–5.0) than in participants born in the United States (6.0 ng/g lipid; 95% CI 5.4–6.7, $p = 0.01$). This difference is most likely explained by a higher exposure level to BDE-153 in the United States than abroad.

The 95th percentile for the sum of PBDEs was 291 ng/g lipid (95% CI, 224–540). The highest sum of PBDE levels observed in the NHANES 2003–2004 participants was 3680 ng/g lipid, which included BDE-47 at 2350 ng/g lipid and concentrations of BDE-99, BDE-100, and BDE-153 at 692, 339, and 152 ng/g lipid, respectively. In this individual, the level of 2,2′,4,4′,5,5′-hexachlorobiphenyl (CB-153) was only 13 ng/g lipid, indicating low exposure to PCBs (*1*). The concentration of BDE-47 was 180 times higher than that of CB-153. Children less than 12 years of age and some PBDE congeners, such as BDE-209, were not included in NHANES 2003–2004. Based on a NHANES 2001–2002 serum pool study the mean concentration of BDE-209 is about 2 ng/g lipids in those 12 years of age and older (unpublished data); therefore, BDE-209 is unlikely to contribute significantly to the total PBDE concentration. We are currently analyzing similar serum pools in children aged 3−11 years.

Although there have been no human health effects directly attributed to exposure to PBDEs and animal data are limited, public health concern has been expressed regarding the potential exposure to these persistent environmental chemicals (*9*). A recent time trend study of U.S. residents, although nonrepresentative, showed increasing serum PBDE concentrations from the mid-1980s to 2002 (*27*). In future studies, it will be of interest to compare changes in concentrations over time, particularly in light of the discontinuation of the PentaBDE and OctaBDE production in the United States. It is also important to conduct studies that will target children and workers in certain occupational settings and to focus on lifestyle factors that result in exposures that produce the higher serum concentrations of PBDEs.

Acknowledgments

We acknowledge the hard work of those who contributed to this work but are not listed as coauthors: Jeff Vancura for his expertise in high-resolution mass spectrometry, Cheryl McClure for database handling, Troy Cash for logistical support, and Sarah Niehüser for proofreading this manuscript.

Supporting Information Available

Tables S1−S4, Figure S1, and Supporting Information (SI-1). This material is available free of charge via the Internet at http://pubs.acs.org.

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ES702451P