PERSISTENT POLUTANTS IN SERUM POOLS FROM THE NATIONAL HEALTH AND NUTRITION EXAMINATION SURVEY (NHANES): 2003 – 2008

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Introduction

The National Health and Nutrition Examination Survey (NHANES) provides bi-annual assessments of the exposure to select environmental compounds and/or their metabolites among the civilian non-institutionalized population of the United States through biomonitoring. Historically, biomonitoring in NHANES has been based on individual measurements of target chemicals in body fluids from a representative sample of the population. For example, individual measurements for polybrominated diphenyl ethers (PBDEs), 2,2',4,4',5,5'-hexabromobiphenyl (PBB-153), polychlorinated biphenyls (PCBs), coplanar PCBs (cPCBs), polychlorinated dibenzo-p-dioxins and furans (PCDD/F) and persistent pesticides are available for 2003–04 NHANES participants.[1,2]

Pooling allows for larger sample volumes for analysis which can result in lowered limits of detection primarily for PCDD/F where sample volume is a limiting factor. Hence, the increased detection frequency achieved with pooling can allow researchers to study the central tendency for a given compound. By contrast, for individual samples only estimates of upper percentiles (e.g., 90^{th}, 95^{th}) would have been possible for compounds with low detection frequencies due to low sample volume (i.e., PCDD/F). Pooling samples before making analytical measurements also decreases the number of analytical measurements required, thereby reducing the costs of biomonitoring. We report here the serum concentration of select PBDEs, PBB-153, PCBs, cPCBs, PCDD/F, and persistent pesticides in pools constructed from serum collected from 2005–06 and 2007–08 NHANES participants (data for NHANES 2007/08 PCDD/F are not included because they are not publically available). We also compare these pooled results to the arithmetic mean concentration of the 2003–04 NHANES individual results. [1,2]

Materials and methods

Pooling strategy: NHANES is a complex multistage-area probability design survey that includes approximately 10,000 persons per each two-year survey period. NHANES uses sampling weights to adjust for unequal selection probability; these sampling weights are used to adjust the analytical data to create a statistically representative subsample of the general US population. Demographic, clinical and other data from NHANES participants are collected through household interviews and through standardized physical examinations conducted in mobile examination centers. On the basis of self-reported data, a composite race/ethnicity variable defines four major racial/ethnic groups, i.e., non-Hispanic Blacks, non-Hispanic Whites, Mexican Americans, and other race/ethnicities. Informed consent is obtained from all study participants. Serum samples representing a one-third subset (approx. 2000 persons per survey period) were used to prepare pools for analysis stratified by the race/ethnicity categories above, sex, and age groups (12-19, 20-39, 40-59 and ≥60 years). In order to incorporate sample weighting into the pooled-sample design it was necessary to use a different volume of serum from each sample contributing to a pool. For NHANES 2005–06 and 2007–08, the volume chosen for each sample in a pool was based on the ratio of its sampling weight to the sum of the sampling weights of all samples in the pool.

Arithmetic mean concentrations and 95% confidence intervals (CIs) were also calculated adjusting for sampling weights and survey design effects for the available NHANES 2003–04[1] individual sample measurements. The design effects calculated from the weighted NHANES 2003–04[1] individual measurements were used to calculate adjusted 95% CIs for the weighted NHANES 2005–06 and 2007–08 pooled-sample results.

The analytical methodology used to analyze NHANES pooled samples[3-5] has been published and is not described here.

Table 1. Number of serum pools created per survey period and demographic group.

Race/Ethnicity	Age (Years)	Number of serum pools			
		NHANES 2005/06		NHANES 2007/08	
		Females	Males	Females	Males
Mexican American	12-19	16	11	5 [1]	6
(MA)	20-39	9	9	8	9 [1]
	40-59	6	4	6 [2]	6
	≥60	3	4	5 [3]	5 [1]
Non-Hispanic Black	12-19	14	13	5 [4]	6 [4]
(NHB)	20-39	7	6	8 [5]	6
	40-59	7	5	8 [5]	6
	≥60	5	5	7 [1]	8
Non-Hispanic White	12-19	10	9	7 [4]	6
(NHW)	20-39	16	12	13 [2]	15
	40-59	13	12	17	16
	≥60	17	15	21	23
Other	All	9 [6]		10 [7]	

[1] One six sample pool. [2] One 3 sample pool. [3] One 7 sample pool. [4] One 5 sample pool. [5] One 4 sample pool. [6] One 6 sample pool and one 7 sample pool. [7] Two 5 sample pools and one 7 sample pool.

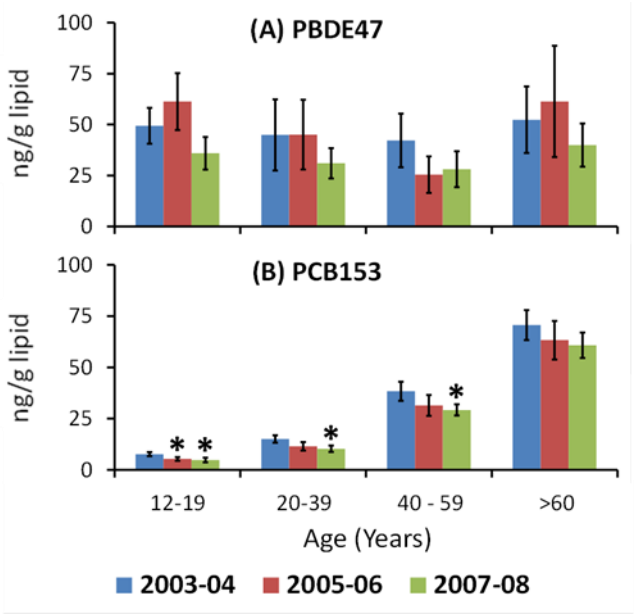
Results and discussion

Serum concentrations of 17 PCDD/F, 3 cPCBs, 35 PCBs, 9 PBDEs, PBB-153, and 8 persistent pesticides were measured in pools from two NHANES covering the time period 2005 through 2008 encompassing the voluntary withdrawal from the US market in 2004 of commercial Penta- and OctaBDE. In this abstract, we present only data for 2,2',4,4'-tetrabromodiphenyl ether (PBDE-47) and 2,2',4,4',5,5'-hexachlorinated biphenyl (PCB-153), 3,3',4,4'-tetraCB (PCB-77), 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TeCDD), 1,2,3,7,8-pentaCDD (1,2,3,7,8-PeCDD), 1,2,3,6,7,8-hexaCDD (1,2,3,6,7,8-HeCDD), and 2,3,4,7,8-pentachlorodibenzofuran (2,3,4,7,8-PeCDF) as representative markers of exposure.

The average concentration of BDE-47 and PCB-153 for the years 2003–04, 2005–06, and 2007–08 is given in Figure 1 for the age groups 12-19, 20-39, 40-59, and 60 years of age and older. For NHANES 2003–04, the weighted arithmetic mean concentrations and 95% CIs were calculated from publicly available individual measurements,[1,2] while for NHANES 2005–06 and 2007–08 the calculations were based on pooled serum results. Because survey design effect information was lost when samples were pooled during the 2005-06 and 2007-08 surveys, the survey design effects determined for NHANES 2003–04 were applied to the weighted results for NHANES 2005–06 and 2007–08 when calculating the 95% CIs. (It should be noted, however, that the design effects from one survey may not be applicable to another.) In the case of PBDE-47, the calculated mean concentrations were overall lower in NHANES 2007–08 than in NHANES 2003–04 suggesting a potential decrease in concentration following the discontinuation of commercial Penta- and OctaBDE in 2004, however, the 95% CIs for the NHANES 2003-04 overlap with those of the other survey periods. Data from future surveys are needed to confirm if this is the beginning of a decreasing concentration trend of PBDE-47 in serum from the general United States population.

The weighted arithmetic mean serum concentration of PCB-153 was significantly higher in 2003–04 than in NHANES 2005–06 for 12-19 year olds and in NHANES 2007–08 for 12-19, 20-39 and 40-59 year olds, suggesting a decreasing concentration trend for PCB-153. Decreasing serum concentration with time is expected for PCB-153 because PCBs were phased out in the mid-1970s.

Figure 1. Age distribution of weighted arithmetic mean concentrations (ng/g lipid) by survey period (NHANES 2003/04, individual samples; NHANES 2005–06 and 2007–08, pooled samples) for (A) 2,2',4,4'-tetrabromodiphenyl ether (BDE-47), (B) 2,2',4,4',5,5'-hexachlorobiphenyl (PCB-153. Error bars indicate 95% confidence intervals (95%CIs). Significantly different 95%CIs of the weighted arithmetic means compared with NHANES 2003–04 are indicated with an asterisk.



The concentration of the 4 PCDD/F and the cPCB that make up approximately 80% of the total toxicity equivalence (TEQ) is given in Table 2 for non-Hispanic white males in NHANES 2003–04 and 2005–06. We observe an increasing concentration trend with age for NHANES 2005–06 based on pooled measurements for all analytes (Table 2) with concentrations 3 to 4 times higher in subjects over the age of 60 vs. 12-19 year olds. A similar trend can be seen for the NHANES 2003–04 for the 90th and 95th percentile. However, the lower detection frequency in NHANES 2003–04 prevents the determination of an average concentration for 2,3,7,8-TeCDD except those over the age of 60 (when requiring an detection frequency over 50% for calculating an average concentration). Using the same requirement for calculating an average concentration we could not determine the average concentration for 1,2,3,7,8-PeCDD in 12-19 and 20-39 year olds and 2,3,4,7,8-PeCDF in 12-19 year olds in NHANES 2003/04. On the other hand in NHANES 2005–06 we were able to calculate an average concentration for all analytes except 2,3,7,8-TeCDD in the 12-19 year age group. Measurements for NHANES 2005–06 pooled samples were based on 30–35 mL of serum while the measurements conducted for NHANES 2003–04 were based on 6–7mL. Our results (Table 2) suggest that the pooled study design did enable the possibility for studying the central tendency for PCDD/F and cPCBs in the general population of the United States while also reducing the biomonitoring cost. Future NHANES for POPs will alternate between individual and pooled measurements to be able to provide data on both central tendency and upper percentiles of body burdens for these compounds.

Table 2. Concentration (pg/g serum lipids) of select co-planar polychlorinated biphenyl (cPCB) and polychlorinated dibenzo-p-dioxins and furans (PCDD/F)) in Non-Hispanic White Males in serum pools from NHANES 2005/06 and as a comparison NHANES 2003/04 individual measurements. [2] Mean concentrations were not determined (ND) when the detection frequency for an age group was less than 50%.

Congener (Abbreviation)	Age Group	NHANES 2003/04				NHANES 2005/06	
		Mean (SE)	90th Percentile (CI)	95th Percentile (CI)	% Detect	Mean (SE)	% Detect
2,3,7,8-Tetrachlorodibenzo-*p*-dioxin	12-19	ND	1.54 (1.25-1.96)	1.95 (1.51-2.14)	10 [b]	ND	23 [b]
(2378-TCDD)	20-39	ND	1.96 (1.54-2.61)	2.70 (1.87-3.30)	22 [b]	0.41 (0.09) [a,b]	57
	40-59	ND	3.81 (2.70-5.24)	5.10 (2.90-5.83)	46 [b]	1.21 (0.16)	87
	≥60	2.7 (0.2)	5.11 (4.45-5.40)	5.71 (4.85-6.84)	61	2.37 (0.19)	100
1,2,3,7,8-Pentachlorodibenzo-*p*-dioxin	12-19	ND	4.09 (1.88-4.62)	4.73 (3.73-6.85)	18 [b]	2.52 (0.14)	100
(1,2,3,7,8-PeCDD)	20-39	nd	4.82 (4.16-5.58)	5.59 (4.76-6.03)	44 [b]	2.47 (0.24)	92
	40-59	5.15 (0.51)	9.35 (7.00-10.92)	10.68 (8.63-13.46)	78	4.49 (0.55)	91
	≥60	7.63 (0.39)	12.63 (10.94-14.05)	15.04 (11.30-16.96)	86	7.25 (0.40)	100
1,2,3,6,7,8-Hexachlorodibenzo-*p*-dioxin	12-19	10.0 (0.6)	19.10 (15.29-21.54)	22.97 (18.14-30.45)	70	10.5 (0.8)	100
(1,2,3,6,7,8-HxCDD)	20-39	14.7 (0.8)	26.96 (22.40-28.83)	28.56 (26.63-30.37)	83	14.7 (1.4)	100
	40-59	33.1 (3.8)	56.81 (42.10-70.45)	70.29 (46.58-83.85)	92	31.8 (3.4)	100
	≥60	52.3 (2.9)	93.23 (70.60-111.50)	106.95 (89.84-129.50)	96	46.6 (3.4)	100
2,3,4,7,8-Pentachlorodibenzofuran	12-19	ND	4.28 (3.47-5.39)	6.78 (3.87-8.45)	30 [b]	2.25 (0.14)	100
(2,3,4,7,8-PeCDF)	20-39	3.9 (0.2)	6.91 (5.56-7.26)	7.23 (6.33-9.88)	68	3.46 (0.30)	100
	40-59	6.3 (0.4)	10.52 (8.19-15.15)	13.71 (9.96-18.56)	83	5.82 (0.26)	100
	≥60	8.2 (0.5)	12.89 (11.29-15.00)	15.56 (14.30-17.58)	94	8.03 (0.43)	100
3,3',4,4',5-Tetrachlorobiphenyl	12-19	10.2 (0.8)	15.88 (11.35-19.78)	19.18 (12.89-23.93)	86	6.23 (0.47)	100
(PCB-126)	20-39	13.7 (1.0)	21.39 (18.02-28.82)	28.18 (20.54-43.49)	88	8.13 (0.78)	100
	40-59	22.6 (3.2)	37.13 (26.11-49.52)	50.73 (30.86-82.13)	97	17.6 (2.4)	100
	≥60	30.8 (2.8)	55.33 (46.14-68.81)	75.44 (54.66-122.44)	97	26.8 (3.3)	100

Abbreviations: SE, Standard Error; CI, Confidence intervall; ND, not determined due to detection frequency less than 50%

[a] Standard error >30% of mean; [b] recovery <50%

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