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Estimating the half-lives of PCB congeners in former capacitor workers measured over a 28-year interval

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To date, most estimates of the half-life of polychlorinated biphenyls (PCBs) in humans have been based on relatively short follow-up periods. To address this issue, we determined the half-lives of PCB congeners of occupational origin in the serum of former capacitor workers as part of a study conducted in 2003–2006 — approximately 28 years after their last occupational exposure. A total of 241 persons from a source population of 6798 former capacitor workers were interviewed and asked to donate a blood sample for serum PCB congener analysis. A subgroup of 45 participants also had serum archived from 1976 and reanalyzed for the same 27 PCB congeners by the same laboratory. Our estimates of the half-lives of the congeners among these 45 persons were longer than those reported by Wolff et al. (1992), due primarily to the much longer interval between exposure and determination of serum PCB concentrations. Half-lives were significantly greater for the heavy *versus* light occupational congeners, for women *versus* men and for those with low *versus* high initial exposure. Current serum total PCB concentrations, expressed as the geometric mean of wet weight data, averaged 6.7 ng/g for the entire 241-person cohort, which represents a 10-fold decrease from values reported in the late 1970s, but is still nearly twice the average for persons of similar age residing in the same area, but without occupational exposure. In addition, current serum PCB concentrations remained significantly and positively associated with earlier occupational exposure, but were not associated with fresh water fish consumption. In general, the results support a consistent and long-duration trend of increased PCB body burden in this cohort of former capacitor workers compared with non-occupationally exposed individuals. The results may aid in further understanding the toxicological/epidemiological consequences of exposure to PCBs in humans.
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Introduction

Polychlorinated biphenyls (PCBs) are halogenated aromatic hydrocarbons with unique physical and chemical properties including thermal stability, resistance to acids, oxidation, hydrolysis, and low vapor pressure (ATSDR, 2000). They were produced by the Monsanto Company (St. Louis, MO, USA) under the Aroclor trade name and were used by the General Electric Company (GE) in two facilities in Fort Edward and Hudson Falls, New York in the manufacture of electrical capacitors beginning in 1946 until 1977 (National Research Council, 1979). Aroclor 1254 was used initially; however, beginning in 1953, Aroclor 1242 was used for all manufacturing with the exception of some specialty capacitors. In 1971, Aroclor 1242 was replaced with Aroclor 1016, while a small amount of Aroclor 1221 was used in the later years, that is, through 1977 (Fischbein et al., 1979). Air concentrations of PCBs in direct exposure job areas in 1977, just prior to the end of PCB use, were approximately 310 $\mu g/m^3$, whereas levels of PCBs in indirect exposure job areas were 10-fold lower (27 $\mu g/m^3$) (Taylor et al., 1991). Areas surrounding the plant had an

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average air PCB concentration of 6.2 $\mu g/m^3$ (Taylor et al., 1991), whereas ambient air concentrations in urban areas distant from the plants averaged 0.1 $\mu g/m^3$ (Kutz and Yang, 1975).

Fischbein et al. (1979) analyzed sera from 310 workers in these facilities in 1976 and found that they were, on average, 10- to 20-fold higher than in the general population (Wolff et al., 1982; Lawton et al., 1985). A subgroup of our cohort also had sera archived from 1976, allowing us to compare PCB concentrations and estimate their half-lives over a 28-year period. Although half-lives for PCBs have been estimated previously in these workers (Wolff et al., 1992), they were estimated after less than 5 years of follow-up — in fact, to our knowledge no other study has evaluated PCB half-lives over nearly 30 years. In addition, we also assessed variations in current PCB concentrations among all persons in the recreated cohort according to sociodemographic factors, past occupational exposure, and fish consumption. The results presented here are from a larger project designed to evaluate the neurobehavioral effects of PCBs. The rationale was that, although PCBs have been associated with deficits in behavior and cognitive performance in infants and children (Jacobson and Jacobson, 2003; Gray et al., 2005; Stewart et al., 2005), recent epidemiological findings from Schantz et al. (1996, 2001) and Fitzgerald et al. (2008) also suggest changes in the behavior of older adults.

Methods

Study Population

The study population was recruited between 2003 and 2006 and consisted of former capacitor workers who had been employed by GE for at least 3 months between 1946 and 1977 at either of two capacitor factories. This population was identified based on information generated in the 1970s by the New York State Department of Health (NYSDOH), in cooperation with GE and the National Institute of Occupational Safety and Health. Information included names, dates of birth, gender, social security number, job codes, and start and end dates for most jobs held by the 6798 former workers (3784 men and 3014 women). This cohort is believed to reflect virtually all persons who worked at the facilities in the time frame of interest.

Human Subjects Protection

Prior to initiation of the study, the design and instruments, including those associated with tracing, screening, recruitment, and data collection, were reviewed and approved by Institutional Review Boards at each of the study's collaborating institutions (NYSDOH, Albany Medical Center, University at Albany, Institute for Neurodegenerative Disorders and the U.S. Army).

Inclusion Criteria

To be considered for inclusion in the study, former workers had to: (i) currently live within a 100-mile radius of Albany, NY, USA; (ii) have worked at GE for at least 3 consecutive months between 1946 and 1977; and (iii) be at least 50 years of age. We oversampled certain categories of individuals, including those who had been previously tested by researchers from the Mount Sinai School of Medicine in the 1970s (Fischbein et al., 1979), and thus had sera archived from that period ($N = 310$), as well as workers who had held job(s) considered to place them at risk for high-level exposure to PCBs ($N = 589$). Finally, in the last 2 years of the study, we made efforts to oversample women to ensure that adequate numbers were available for statistical analyses stratified by gender.

Interview Assessment

Study participants were interviewed in person. Demographic information included education, race, ethnicity, marital status, gender, income, and residential history. A dietary history was also collected that included information on consumption of sport-caught fish, dairy products, and caffeinated beverages. In addition, the number and type of jobs held both at GE and elsewhere, as well as selected non-occupational activities and hobbies (e.g., stained glass making, painting), likely to result in exposure to neurotoxicants and lifestyle characteristics (smoking, alcohol consumption, and physical activity), were also collected. Finally, a comprehensive medical history, including use of over-the-counter and prescription drugs, as well as (if applicable) female reproductive histories (the number of pregnancies, breastfeeding history, and menopausal status) was collected.

Serum PCB Analysis

All samples, including current and archived sera, were analyzed for PCBs using the current methodologies of Gammon et al. (2002) based on the methods described by Brock et al. (1996). Quality control was monitored using three plasma pools fortified with PCB congeners corresponding to 0, 5, or 10 ng/ml total PCB. Detection limits (0.07 $\mu g/l$ per congener (Gammon et al., 2002)) were based on three times the SD of results from blanks and from the unfortified pool, which contained approximately 1 ng/ml each of DDE and of total PCBs. The higher detection limit compared with NHANES (CDC, 2005) is due to the analytic method, that is, the use of GC-ECD instead of GC-MS. Values less than the detection limit were set to the lowest positive value for a given analyte. This decision was based on the fact that many chemists and statisticians believe that a reported result, even if it is below the "criterion for detection," is the best available estimate of the true value, and is preferable to assigning a zero or an arbitrary constant, such as one-half the detection limit (Wolff et al., 2008; Fitzgerald et al., 2007b). Coefficients of variation (SD/mean) of DDE and PCBs

from the fortified pools were 10% to 15%. To ensure comparability between the current and historic serum PCB measurements for half-life estimation, archived sera from 1976 was reanalyzed using the same analytic methods described above for the current samples.

Total serum PCB concentration was defined as the sum of the 27 individual congeners that were analyzed. We also determined serum concentrations of ''light'' PCBs, that is, those congeners eluting before DDE; ''heavy'' PCBs, that is, those congeners eluting after DDE; and ''occupational'' PCB congeners, as previously defined by Wolff et al. (1982). Wolff M.S. (personal communication) identified PCB congeners 28, 74, 118, 105, and 156 as unique markers of occupational exposure based on demonstration that levels of these congeners are low in the general population and are derived almost exclusively from the Aroclor mixtures used in the capacitor plants.

We determined total serum lipid concentrations for samples collected in the period from 2003 to 2006 using the algorithm of Phillips et al. (1989a) to calculate total lipids from serum cholesterol and triglycerides. Results are expressed on both a wet weight and lipid basis for the current serum PCB concentrations to allow comparison of PCB levels with other published studies. Because of issues surrounding the accuracy of the measurement of lipid levels in the reanalyzed archived sera and the goal of comparing our half-life calculations with the published data of Wolff et al. (1992) which were based on wet weight data, we present all PCB data from the archived cohort subjects on a wet weight basis. This decision is supported by the fact that we are comparing paired data from the same individual at two time periods.

Fresh Water Fish Consumption

The exposure assessment for fish consumption focused on the Hudson River and other bodies of fresh water in New York State known to be contaminated with PCBs. Surveys conducted by the New York State Department of Environmental Conservation indicate that, in addition to the Hudson River, sport fish caught from Lakes Ontario and Champlain are also heavily contaminated with PCBs. These findings have resulted in bans or advisories against consumption of many fish species from those bodies of water being repeatedly issued by the NYSDOH (http://www.health.state.ny.us/nysdoh/environ/fish/htm).

Relatively few persons in our study consumed fish from the Hudson River. Hence, these data were combined with consumption data from Lakes Ontario and Champlain, the two most common sources of sport fish consumption for study participants, and other New York bodies of fresh water to provide a summary measure of fish consumption. This measure was computed by multiplying the yearly consumption rate of fresh water fish by the number of years fish were consumed for each of three time periods (before 1994, 1994 to the year before the interview, and the last year) and summed to estimate total cumulative exposure to PCBs from fresh water fish consumption. Persons who consumed fresh water fish from these bodies of water were then divided into groups above and below the median consumption value and compared with those who did not consume such fish. These consumption data were collapsed across species because species-specific fish PCB levels were not available for all the time periods of interest. This metric did not provide for individual estimates of PCB intake through fish consumption, but did allow for a semiquantitative comparison of serum PCB concentrations according to relative fish consumption.

Occupational Exposure to PCBs

Potential occupational PCB exposure was assessed by two industrial hygienists (IHs) who independently rated the probability of exposure for all GE jobs held for 3 or more months and all non-GE jobs held for more than 1 year. These assessments were based on the name(s) and location of the company, type of industry, and a brief description of work duties reported during the interview assessment. The IHs assigned one of four qualitative ratings to each self-reported job: (1) definitely not exposed, (2) possibly exposed, (3) probably exposed, and (4) definitely exposed. These ratings were then assigned weights of 0, 0.25, 0.5, and 1.0, respectively. When IH ratings differed, the IHs jointly reviewed the data and came to agreement on a rating. The weights for each job then were multiplied by how long the participant worked in that job, and the results were summed over all jobs to estimate total cumulative occupational exposure to PCBs for each participant. Occupational exposure was also partitioned according to the years when specific Aroclors were used by GE, that is, 1946–1953 for Aroclor 1254, 1953–1971 for Aroclor 1242, and 1971–1977 for Aroclor 1016 (with minor Aroclor 1221 usage) (Lawton et al., 1985).

Half-life Estimation

In order to estimate the half-lives (hl) of the PCB congeners for the 45 participants for whom we had both current serum as well as serum archived from 1976, we assumed an exponential decay model ($C_{(t)} = C_{(0)}e^{-kt}$) (Friedman, 1979) to describe the metabolism/excretion of PCBs across the 28-year span using methods previously used by Taylor and Lawrence (1992); Phillips et al. (1989b); and Shirai and Kissel (1996). In this model, $C_{(t)}$ represents the 2004 serum PCB measures, $C_{(0)}$ represents the 1976 serum PCB measures, t is the 28-year time interval, and k represents the decay constant. Since the value of $(\ln(C_{(0)}) - \ln(C_{(t)}) = kt)$ is known and with $k = \ln(2)/hl$, then it follows that the half-life of the PCBs can be estimated by $hl = \ln(2)*t/\ln(C_{(0)}) - \ln(C_{(t)})$. Different authors recommend different estimates of the C_0 and C_t, including either the median or

the geometric mean of the PCB quantities across the sample. As these two estimators are often very close in value, we chose the geometric mean to estimate the value of C_0 and C_t in this research. We did not have specific determinations of PCB intake from fish consumption or other foodstuffs to incorporate into our half-life estimates, so the half-life computations were restricted to those congeners that were occupational in origin, that is, PCB-28, 74, 118, 105, and 156.

Statistical Analysis

Rank transformation analysis of variance tests of simple main effect differences were used to compare calculated half-lives between men and women and exposure levels for the above described PCB congener groups. Multiple linear regression analysis was also used to test for associations between current serum PCB levels and demographic and background variables (age, gender, education, body mass index (BMI), marital status, cigarette smoking, alcohol consumption, and prescription drug use, some of which have been related to serum or plasma PCB levels in other studies (Rylander et al., 1997; Moysich et al., 2002). Variables found to be significant in a bivariate analysis were then regressed on the log-transformed serum total PCB concentration using stepwise procedures to add or remove the background variables one at a time. Cumulative exposure to PCBs from occupation and fresh water fish consumption were then added to the regression models to estimate their associations with current serum total PCB concentrations after adjustment for all remaining background variables.

Results

Recruitment and Participation

The population of former capacitor workers on computer tapes made available to us consisted of 6798 individuals; 6398 of these were retained as the potential pool of individuals to trace on the basis of a documented employment history at GE between 1946 and 1977. A total of 2844 (44%) were selected for tracing according to our previously discussed criteria. Of this group, 844 (30%) were deceased; 256 (9%) either lived beyond a 100-mile radius of Albany or had worked at either of the capacitor facilities for a period of less than 3 months during the specified time period; and 577 (20%) could not be located. In addition, 43 (2%) of those selected for tracing were not ultimately traced because they were selected for tracing just before the completion of the study.

The remaining 1124 persons (40%) were contacted by telephone and screened for eligibility. Forty-two individuals (4%) refused to participate in the screening interview; 110 (10%) agreed to answer the screening interview but refused to be contacted for recruitment into the study; and 80 (7%) never returned our screening telephone calls, despite numerous attempts. In addition, 348 of those screened (31%) were deemed ineligible for the medical reasons. Since the larger project focused on nervous system function, these persons included those diagnosed with or treated for conditions such as multiple sclerosis, stroke, head injury, epilepsy, or psychiatric disorders. Fifty individuals (5%) were considered ineligible because of their inability to travel to the study sites or complete the study measures, and 10 (1%) were entered into screening just before the conclusion of the study and were thus not screened.

The other 484 (43%) were eligible for participation in the study. Approximately half of these refused, while the remaining half agreed to participate in the study yielding a final study population of 241 persons. Compared with the 6157 former workers not enrolled in the study, the participants were significantly younger (38 years old as of 1 January 1978 *versus* 45 years old for the non-participants, $P \leq 0.001$) and more likely to be highly exposed to PCBs according to the job histories available from the computer tapes (25% *versus* 10% for the non-participants, $P \leq 0.001$; data not shown). There were no differences by gender.

Of the 241 former workers in this study, 33 men and 12 women also participated in the 1976 study conducted by Fischbein et al. (1979). Table 1 presents the demographic and background characteristics for both the entire cohort ($N = 241$) and for that subset with both current and archived serum ($N = 45$). The subgroup with archived sera was generally similar to the full cohort with the exception of the age of the women at the time that they were interviewed for this study (greater in the archived cohort) and the number of children (again greater in the archived cohort). BMI was lower in the women in the archived subgroup compared with the larger sample.

Current Serum PCB Concentrations

Current serum geometric mean PCB concentrations, for men and women separately, expressed on both a wet weight basis (ng/g) and a lipid adjusted basis (μg/g) are presented in Table 2. Total current serum PCB concentration for men was 7.47 ng/g or 1.19 μg/g. For women the corresponding values were 5.81 ng/g or 0.86 μg/g. For men $\sim$61% of the total PCB residue in serum consisted of heavily chlorinated PCBs (those eluting after DDE), most notably PCB congeners 153, 180, and 138. For women $\sim$55% of the total PCB residue consisted of heavily chlorinated PCBs. Men (Table 2a) had significantly higher serum concentrations of both heavy congeners ($P \leq 0.01$) and total PCBs ($P \leq 0.01$) than did women (Table 2b).

Predictors of Current Serum PCB Concentrations

Age was the demographic variable most strongly associated with log current serum total PCB concentrations ($\beta = 0.015$, $P \leq 0.001$, data not shown). Log current serum total PCB concentrations were also higher among men than women

Table 1. Demographic and background characteristics of all study participants ($N = 241$) and the cohort of subjects with archived sera ($N = 45$).

Characteristic	All study participants		Archived sera cohort	
	% or Mean (SD)	N[a]	% or Mean (SD)	N[a]
Gender				
Male	53.6	129	73.3	33
Female	46.5	112	26.7	12
Income				
<15,000	9.0	20	2.2	1
15,000–30,000	22.4	50	37.8	17
30,000–45,000	26.5	59	13.3	7
45,000–60,000	18.8	42	15.6	7
60,000–75,000	13.0	29	15.6	7
>75,000	10.3	23	0	0
Marital status				
Married or live with partner	70.5	165	73.2	30
Divorced, never married, separated, or widowed	29.5	69	26.8	11
Lost weight in past year				
No	80.8	189	80.5	33
Yes	19.2	45	19.5	8
Had hepatitis or cirrhosis of the liver				
No	97.0	225	97.5	39
Yes	3.0	7	2.5	1
Age (years)				
Male	64.1 (8.1)	129	64.0 (7.8)	33
Female	64.7 (9.3)	112	70.5 (8.6)*	12
Education (school years completed)				
Male	13.1 (2.2)++	122	12.7 (1.2)+	29
Female	12.4 (1.7)	112	12.2 (0.9)	12
BMI (kg/m^2)				
Male	29.1 (4.5)	122	28.6 (3.9)+	29
Female	29.9 (6.1)	112	25.9 (3.5)**	12
Number of cigarette packs in the previous year				
Male	38.4 (111.4)	122	44.1 (116.1)	29
Female	46.6 (122.8)	112	121.8 (179.9)	12
Number of cigarette packs in the last 10 years				
Male	630 (1404)	122	557 (1191)+	29
Female	867 (1946)	112	2100 (3169)	12
Total number of drinks/week in the last year				
Male	6.84 (9.16)+++	122	8.24 (9.9)+	29
Female	1.47 (3.46)	112	1.86 (4.0)	12
Total number of drinks/week in the last 10 years				
Male	7.01 (9.38)+++	122	8.50 (9.35)++	29
Female	1.14 (2.62)	112	1.05 (1.37)	12
Number of births (females only)	2.71 (1.63)	112	3.75 (2.14)*	12
Total weeks lifetime breastfeeding (females only)	7.18 (22.73)	112	6.92 (20.59)	12

[a]Number of observations varies across characteristics due to missing values.
* The t-test or χ^2 is significant at $P \leq 0.05$ for all study participants vs archived sera cohort.
** The t-test or χ^2 is significant at $P \leq 0.01$ for all study participants vs archived sera cohort.
+ The t-test or χ^2 is significant at $P \leq 0.05$ for male vs female.
++ The t-test or χ^2 is significant at $P \leq 0.01$ for male vs female.
+++ The t-test or χ^2 test is significant at $P \leq 0.001$ for male vs female.

Table 2a. Current serum PCB concentrations in men who participated in the study ($N = 129$).

IUPAC number	IUPAC structure	% of non-detectable or zero values	Wet weight (ng/g)		Lipid basis (μg/g)	
			Geometric mean	SD	Geometric mean	SD
Light PCBs[a]						
PCB-28[b]	2,4,4′	25.6	0.07	0.52	0.01	0.09
PCB-74[b]	2,4,4′,5	1.6	1.01	5.18	0.16	0.80
PCB-66	2,3′,4,4′	17.8	0.14	0.33	0.02	0.05
PCB-56	2,3,3′,4′	13.2	0.11	0.20	0.02	0.04
PCB-101	2,2′,4,5,5′	3.1	0.29	0.50	0.05	0.09
PCB-99	2,2′,4,4′,5	5.4	0.15	0.40	0.02	0.07
Total			2.84	5.83	0.45	0.92
Heavy PCBs[c]						
PCB-118[b]	2,3′,4,4′,5	8.5	0.16	0.73	0.03	0.14
PCB-146	2,2′,3,4′,5,5′	1.6	0.09	0.22	0.01	0.04
PCB-153	2,2′,4,4′,5,5′	0.0	0.90	1.70	0.14	0.29
PCB-105[b]	2,3,3′,4,4′	17.1	0.04	0.17	0.01	0.03
PCB-138	2,2′,3,4,4′,5′	0.8	0.73	1.37	0.12	0.23
PCB-178	2,2′,3,3′,5,5′,6	9.3	0.04	0.10	0.01	0.02
PCB-187	2,2′,3,4′,5,5′,6	3.1	0.14	0.21	0.02	0.04
PCB-183	2,2′,3,4,4′,5′,6	4.7	0.07	0.06	0.01	0.01
PCB-167	2,3′,4,4′,5,5′	16.3	0.03	0.08	0.004	0.02
PCB-174	2,2′,3,3′,4,5,6′	7.8	0.05	0.05	0.01	0.01
PCB-177	2,2′,3,3′,4,5′,6′	11.6	0.04	0.07	0.01	0.01
PCB-156[b]	2,3,3′,4,4′,5	0.8	0.19	0.59	0.03	0.09
PCB-172	2,2′,3,3′,4,5,5′	7.8	0.07	0.10	0.01	0.02
PCB-180	2,2′,3,4,4′,5,5′	0.8	0.51	1.05	0.08	0.18
PCB-170	2,2′,3,3′,4,4′,5	2.3	0.23	0.48	0.04	0.08
PCB-199	2,2′,3,3′,4,5,5′,6′	0.0	0.11	0.15	0.02	0.03
PCB-203	2,2′,3,4,4′,5,5′,6	0.8	0.11	0.12	0.02	0.02
Total			4.09	6.48	0.65	1.12
Total PCBs			7.47	10.71	1.19	1.75

[a]Elute before DDE.
[b]Markers for occupational exposure.
[c]Elute after DDE.

Table 2b. Current serum PCB concentrations in women who participated in the study ($N = 112$).

IUPAC number	IUPAC structure	% of non-detectable or zero values	Wet weight (ng/g)		Lipid basis (μg/g)	
			Geometric mean	SD	Geometric mean	SD
Light PCBs[a]						
PCB-28[b]	2,4,4′	15.2	0.11	1.53	0.02	0.27
PCB-74[b]	2,4,4′,5	6.3	0.55	6.25	0.08	1.24
PCB-66	2,3′,4,4′	10.7	0.21	0.34	0.03	0.06
PCB-56	2,3,3′,4′	15.2	0.09	0.26	0.01	0.05
PCB-101	2,2′,4,5,5′	6.3	0.26	0.30	0.04	0.05
PCB-99	2,2′,4,4′,5	5.4	0.15	0.46	0.02	0.09
Total			2.29	7.99	0.34	1.57
Heavy PCBs[c]						
PCB-118[b]	2,3′,4,4′,5	4.5	0.23	1.08	0.03	0.21
PCB-146	2,2′,3,4′,5,5′	6.3	0.05	0.33	0.01	0.07
PCB-153	2,2′,4,4′,5,5′	1.8	0.71	1.94	0.10	0.39
PCB-105[b]	2,3,3′,4,4′	8.9	0.05	0.21	0.01	0.04
PCB-138	2,2′,3,4,4′,5′	1.8	0.54	2.40	0.08	0.49
PCB-178	2,2′,3,3′,5,5′,6	9.8	0.03	0.08	0.004	0.01
PCB-187	2,2′,3,4′,5,5′,6	1.8	0.12	0.21	0.02	0.04
PCB-183	2,2′,3,4,4′,5′,6	2.7	0.05	0.06	0.01	0.01
PCB-167	2,3′,4,4′,5,5′	15.2	0.03	0.14	0.004	0.03
PCB-174	2,2′,3,3′,4,5,6′	2.7	0.06	0.04	0.01	0.01
PCB-177	2,2′,3,3′,4,5′,6′	9.8	0.04	0.08	0.01	0.02
PCB-156[b]	2,3,3′,4,4′,5	9.8	0.11	0.89	0.02	0.17
PCB-172	2,2′,3,3′,4,5,5′	11.6	0.05	0.09	0.01	0.02
PCB-180	2,2′,3,4,4′,5,5′	0.9	0.38	0.84	0.06	0.16
PCB-170	2,2′,3,3′,4,4′,5	0.9	0.16	0.51	0.02	0.10
PCB-199	2,2′,3,3′,4,5,5′,6′	0.0	0.08	0.12	0.01	0.02
PCB-203	2,2′,3,4,4′,5,5′,6	0.9	0.07	0.08	0.01	0.02
Total			3.21	8.58	0.47	1.72
Total PCBs			5.81	15.63	0.86	3.11

[a]Elute before DDE.
[b]Markers for occupational exposure.
[c]Elute after DDE.

($\beta = 0.176$, $P \leq 0.001$) and among persons with less education ($\beta = -0.022$, $P = 0.041$). BMI was positively associated with log current serum PCB concentrations, but this association was statistically significant only for the light congeners ($\beta = 0.012$, $P = 0.032$).

Table 3 demonstrates that reported total cumulative occupational exposure to PCBs was significantly and positively associated with log 2004 serum total PCB concentration ($\beta = 0.056$, $P \leq 0.001$) after adjustment for age, gender, education, and BMI. This association was strongest for the occupational light congeners, especially PCB-74 ($R^2 = 0.16$), although some heavy congeners, such as PCB-105 and 156, were also statistically significant. In general, the associations for heavy congeners present in the Aroclor mixtures used by workers were weaker in magnitude than those for the light congeners, and less difference was observed between the occupational and non-occupational heavy congeners compared with the lighter congeners.

Cumulative exposure during the years that Aroclor 1016 was used was most strongly related to the occupational light congeners, particularly PCB-74 ($\beta = 0.249$, $P \leq 0.001$, $R^2 = 0.17$), although two heavy occupational congeners (PCB-105 and 118) were also significant. In contrast, the strength of the association between log serum PCB levels and cumulative occupational exposure during the years that Aroclor 1242 was used was similar for both the occupational light ($\beta = 0.056$, $P \leq 0.001$, $R^2 = 0.09$) and occupational heavy congeners ($\beta = 0.032$, $P \leq 0.001$, $R^2 = 0.07$). Occupational exposure to Aroclor 1254 was significantly associated with only one congener (PCB-156).

Only seven participants (3%) ever ate fish from the Hudson River, and only two persons (1%) ate fish from the Hudson River after 1994 (data not shown). Thirty

Table 3. Multiple regression analysis of current serum PCB concentration (log adjusted, lipid basis) on Aroclor mixture use during years when participants were occupationally exposed[a] (log adjusted), by congener ($N = 233$).

Congener	Aroclor mixture in use during years when occupationally exposed							
	Aroclor 1254 (years of use: 1946–1953)		Aroclor 1242 (years of use: 1954–1972)		Aroclor 1016 (years of use: 1973–1977)		Total (years of use: 1946–1977)	
	β^b	R^2	β^b	R^2	β^b	R^2	β^b	R^2
Light PCBs[c]	−0.014	0.03	0.044***	0.10	0.041***	0.07	0.082***	0.09
Occupational light PCBs	−0.016	0.00	0.056***	0.09	0.078***	0.15	0.144***	0.15
PCB-28	0.005	0.00	0.083	0.01	0.063	0.01	0.104	0.00
PCB-74	−0.053	0.00	0.146***	0.07	0.249***	0.17	0.432***	0.16
Non-Occupational light PCBs	0.015	0.00	0.054*	0.03	0.002	0.00	0.040	0.00
Heavy PCBs[d]	0.018	0.00	0.022***	0.06	0.012	0.02	0.037**	0.05
Occupational heavy PCBs	0.021	0.00	0.032***	0.07	0.027**	0.04	0.060***	0.06
PCB-105	−0.093	0.00	0.151**	0.05	0.152**	0.04	0.278**	0.04
PCB-118	−0.122	0.00	0.100**	0.04	0.085*	0.02	0.112	0.01
PCB-156	0.177*	0.02	0.048	0.02	0.019	0.00	0.126*	0.03
Non-occupational heavy PCBs	0.045	0.00	0.043***	0.05	0.019	0.01	0.071**	0.03
Total PCBs	0.009	0.00	0.030***	0.08	0.023**	0.04	0.056***	0.08

[a]Cumulative occupational exposure based on industrial hygienist assessment and years on job; partitioned according to years when each Aroclor mixture was used.
[b]Adjusted for age, gender, education, and BMI.
[c]Elute before DDE: PCB-28, PCB-74, PCB-66, PCB-56, PCB-101, PCB-99.
[d]Elute after DDE: PCB-118, PCB-146, PCB-153, PCB-105, PCB-138, PCB-178, PCB-187, PCB-183, PCB-167, PCB-174, PCB-177, PCB-156, PCB-172, PCB-180, PCB-170, PCB-201, PCB-203.
*$P \leq 0.05$; **$P \leq 0.01$; ***$P \leq 0.001$.

individuals (13%) reported consuming fish from Lakes Ontario or Champlain, and 85 persons (36%) ate fish from other fresh bodies of water in New York. Combining these data yielded a total of 92 persons who ate a median of 47 fresh water fish meals in their lifetime. In contrast to occupational exposure, there were no significant differences in serum concentrations by total cumulative fresh water fish consumption for total PCB, light or heavy PCBs, or for individual congeners (data not shown).

Current versus *Archived Serum PCB Concentrations*

The geometric means of the archived and current serum PCB values, expressed on a wet weight basis, for the five occupational congeners, their light, heavy, and total sums, and the light, heavy, and total PCB values are shown in Table 4, separately for men and women and combined by gender for the 45 study participants with both measurements. Serum total PCB concentrations for men and women combined decreased significantly from a geometric mean of 37.8 ng/g in 1976 to 9.8 ng/g currently ($P \leq 0.001$). The relative decline was greater for the occupational light congeners (geometric mean of 21.3 ng/g to 2.8 ng/g) than for the occupational heavy congeners (geometric mean of 2.7 ng/g to 0.9 ng/g). Current serum PCB concentrations were significantly higher in women than men for congeners of both occupational origin and light, heavy, and total PCBs. Graphs of the relationships of serum PCB concentrations (log transformed) between archived and current samples for light, heavy, and total PCBs are presented in Figures 1a, 1b and 1c, respectively, separated into men and women after the removal of one extreme value for women. These results continue to indicate a strong association between values for the two time points, and that this association was greater for light congeners and for total PCB among women than men.

Half-Life Estimates

The half-lives of the occupational PCB congeners are shown in Table 5. They ranged from 4.6 years for PCB-28 to 41.0 years for PCB-156 when the data for men and women were combined. The half-life for the heavy congeners (17.8 years) was approximately twice that for the light PCBs (9.6 years). There were also gender differences with women exhibiting half-lives ranging from 1.5 to 10 times longer than those seen in men.

Table 6 shows the half-lives of the occupational PCB congeners according to whether the 1976 concentrations were above or below (''high'' or ''low'', respectively) the median serum PCB values for each group. These data indicate that PCB half-lives are greater among the low exposure group than in the high exposure group, with half-lives varying by a factor of 2 to 10. In addition, this inverse association between initial exposure levels and half-life estimates tended to be stronger for the heavy compared with light congeners.

Discussion

Of the 241 persons in the recreated cohort, a subgroup of 45 had serum PCB levels determined from both 1976 and

Table 4. Geometric means of current PCB concentrations (wet weight, ng/g) in sera from archived cohort ($N = 45$) for men, women, and all (men and women combined) capacitor workers[a].

PCB Congener or summed score	1976 Men and women combined	2004 Men and women combined	1976 Men	2004 Men	1976 Women	2004 Women
Occupational PCBs[b]						
PCB-28	11.27	0.17**	12.13	0.11*	9.23	0.49*
PCB-74	7.75	2.29***	8.67	1.74***	5.71	4.89 +
PCB-105	0.58	0.14**	0.68	0.12***	0.36	0.24
PCB-118	1.71	0.42***	1.69	0.32***	1.77	0.91 +
PCB-156	0.21	0.23	0.24	0.21**	0.15	0.30
Occupational summed PCBs						
Occupational light[b]	21.27	2.80***	23.20	2.15**	16.77	5.79* +
Occupational heavy[b]	2.74	0.92**	2.78	0.76***	2.62	1.55 +
Occupational total[b]	24.56	3.86***	26.56	3.05***	19.80	7.44* +
Summed PCBs[c]						
Light PCBs	26.41	4.28***	28.82	3.58**	20.76	6.98* +
Heavy PCBs	9.08	5.05**	9.01	4.42***	9.27	7.29 +
Total PCBs	37.82	9.80***	40.37	8.38***	31.62	15.05* +

[a]Total $N = 45$ (33 men and 12 women) in 1976 and in 2004.
[b]Occupational light = PCB28 + PCB74. Occupational heavy = PCB105 + PCB118 + PCB156. Occupational total = occupational light + occupational heavy.
[c]Summed PCBs are the sum of all occupational (PCB congener numbers 28, 74, 105, 118, and 156) and non-occupational (PCB congener numbers 66, 56, 101, 99, 146, 138, 178, 187, 183, 167, 174, 177, 153, 172, 180, 170, 201, and 203) PCB congeners.
*$P \leq 0.05$, paired t-test significant comparing 1976 and 2004 PCB concentrations.
**$P \leq 0.01$, paired t-test significant comparing 1976 and 2004 PCB concentrations.
***$P \leq 0.001$, paired t-test significant comparing 1976 and 2004 PCB concentrations.
+ $P \leq 0.05$, t-test significant comparing 2004 PCB concentrations for male vs female.

2004. A 75% decline in geometric mean serum total PCB concentration was observed in this subgroup over the 28-year period. This decrease was proportionately greater for the congeners thought to be of occupational origin than for all congeners combined and for the light congeners compared with the heavy congeners. Interestingly, the current serum concentrations for women were approximately twice those of the men, although their 1976 levels were lower than those for men.

Our serum PCB half-life estimates reflect these findings. For example, the half-life for all the occupational congeners combined was approximately 10 years, yielding about three half-lives over the study period. Similarly, shorter half-lives were found for the occupational light congeners, whereas the half-life for serum PCB concentrations in men was only 50% that of women. This difference in half-lives by sex may also explain why the association between the 1976 and 2004 serum concentrations was greater among women compared with men, at least for the light congeners. We also found an inverse relationship between initial serum levels and half-lives, with those workers whose 1976 concentrations were below the median having a half-life approximately twice that of those whose 1976 concentrations were above the median. As men had higher serum PCB levels in 1976 than did women, this difference in half-lives, according to initial serum PCB concentrations, may explain the greater half-lives for all categories of PCB congeners in women compared with men. Differences in BMI by gender probably are not involved, given the fact that BMI was not associated with half-life.

Similar associations were reported by Wolff et al. (1992), although their half-life estimates were shorter for the light, compared with heavy congeners, and for those individuals with high compared with those individuals with lower initial exposure. For the most part, however, the individual half-lives of the PCB congeners determined in this study are significantly longer than those reported by Wolff et al. (1992) with the exception of PCB-28. These differences in estimated half-lives between the two studies most likely reflect the longer time period over which we were able to estimate half-lives. Indeed, it has been shown that serum PCB concentrations decay in a non-linear, at least two-component, pharmacokinetic manner with the greatest decreases occurring shortly after exposure (Yakushiji et al., 1984; Lawton et al., 1985; Phillips et al., 1989b). Thus, for the PCB congeners with long half-lives, the half-lives estimated over the shorter interval (3.8 years) used by Wolff and co-workers would have emphasized this earlier and more rapid decline in serum PCBs, and therefore may be responsible for their shorter estimates of half-lives. Furthermore, due to the relatively short interval between the initial and follow-up

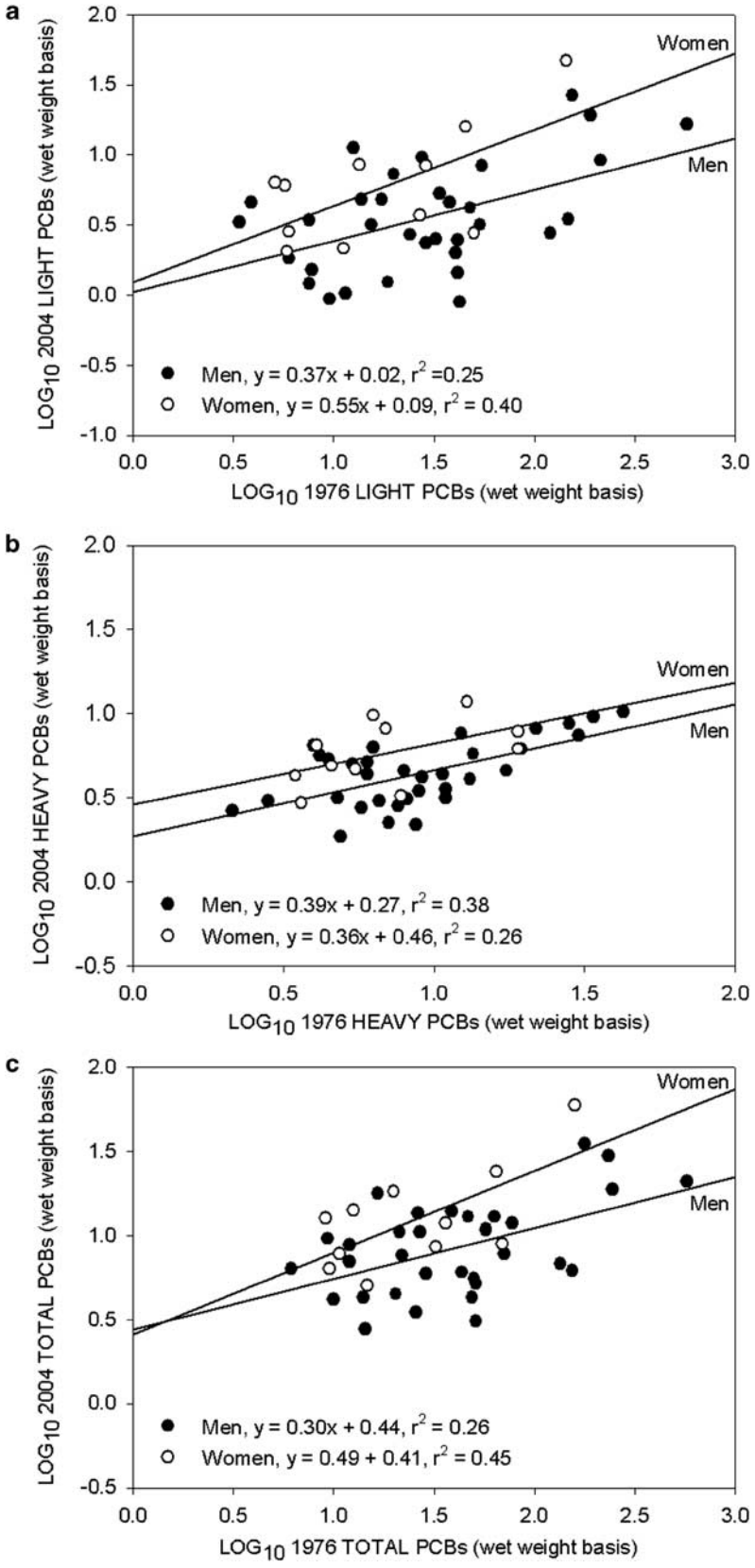

Figure 1. Graphs of the 1976–2004 relationships for log-transformed light (**a**), heavy (**b**), and total (**c**) PCBs expressed on a wet weight basis (ng/g) in men and women from the archived cohort ($N = 45$).

serum PCB measures available to Wolff and co-workers, two of the congeners reported to have an infinite half-life (PCB-74, 156) have now been estimated to have half-lives of 15.9 and 41 years, respectively.

Regarding the serum concentrations of the 241-person cohort as a whole, their geometric mean 2004 level for total serum PCB concentration of 6.65 ng/g (wet weight) or 1.02 μg/g (lipid weight) is approximately twofold higher than that for similarly aged individuals who resided in the same towns at that time, but did not work at GE (Fitzgerald et al., 2007a). This finding, that PCB levels remain elevated more than 30 years after the last direct occupational exposure to PCBs, confirms not only the high levels of occupational exposure but also our estimates of long half-lives for many congeners. Lightly chlorinated congeners, especially those thought to be occupational in origin (PCB-28, 74), were more dominant in the serum of capacitor workers than in the serum of individuals from the general population in the same towns (Fitzgerald et al., 2007a). This finding most likely reflects the impact of occupational exposure to Aroclor 1016 and 1242 in the later years during which PCBs were used.

The continuing impact of previous occupation also was apparent when regression analyses revealed that the 2004 serum PCB concentrations were positively and significantly associated with cumulative years of occupational exposure. In addition, these associations tended to be stronger for the occupational congeners (PCB-74, 105, 118, and 156) than for the non-occupational congeners. Furthermore, the associations with the occupational light congeners were strongest for exposures during the years when Aroclor 1016 was used. Associations between serum concentrations of light and heavy occupational congeners and exposure to Aroclor 1242 were similar. These findings reflect the fact that Aroclor 1016 is composed of lightly chlorinated congeners, whereas Aroclor 1242 is a mixture of both light and heavy congeners (Erickson, 1997). The magnitude of the association, however, was relatively modest, with a maximum R^2 of 17% for

Table 5. PCB half-lives in years (calculated using the geometric means of data expressed on a wet weight basis) for capacitor workers from archived cohort ($N = 45$)[a].

PCB Congener or summed score	Half-life men and women combined	Half-life men	Half-life women
Occupational PCBs			
PCB-28	4.6	4.2	6.6**
PCB-74	15.9	12.1	124.9*
PCB-105	13.7	10.9	46.5
PCB-118	13.8	11.6	29.2**
PCB-156	41.0	33.3	90.1
Occupational summed PCBs			
Occupational light[b]	9.6	8.2	18.2**
Occupational heavy[b]	17.8	14.9	37.2*
Occupational total[b]	10.5	9.0	19.8**

[a]Total $N = 45$ (33 men and 12 women) in 1976 and in 2004.
[b]Occupational light = PCB28 + PCB74. Occupational heavy = PCB105 + PCB118 + PCB156. Occupational Total = occupational light + occupational heavy.
*$P \leq 0.05$, significant rank transformation analysis of variance test between men and women.
**$P \leq 0.01$, significant rank transformation analysis of variance test between men and women.

Table 6. Geometric means of archived and current concentrations (ng/g wet weight) and calculated half-lives for high and low 1976 total PCB exposure groups from archived cohort ($N = 45$)[a].

PCB congener or summed score	Geometric mean			Geometric mean		
	1976 Low	2004 Low	Half-life low	1976 High	2004 High	Half-life high
Occupational PCBs						
PCB-28	4.62	0.20	6.13	28.65	0.14	3.67***
PCB-74	2.94	1.47	28.10	21.36	3.63	10.95*
PCB-105	0.23	0.09	19.92	1.49	0.23	10.35***
PCB-118	0.76	0.30	21.28	4.00	0.59	10.10***
PCB-156	1.03	0.94	214.80	2.65	1.17	23.71
Occupational summed PCBs						
Occupational light[b]	8.13	1.94	13.56	58.16	4.11	7.32**
Occupational heavy[b]	1.26	0.70	32.65	6.14	1.22	12.01***
Occupational total[b]	9.47	2.71	15.52	66.49	5.59	7.84***

[a]Total $N = 45$ (33 men and 12 women) in 1976 and in 2004; $N = 23$, low exposure (1976 TPCBs ≤ 36.534 (median)) and $N = 22$ and high exposure (1976 TPCBs > 36.534 (median)).
[b]Occupational light = PCB28 + PCB74. Occupational heavy = PCB105 + PCB118 + PCB156. Occupational total = occupational light + occupational heavy.
*$P \leq 0.05$, significant rank transformation analysis of variance test between high and low exposure groups for half-lives.
**$P \leq 0.01$, significant rank transformation analysis of variance test between high and low exposure groups for half-lives.
***$P \leq 0.001$, significant rank transformation analysis of variance test between high and low exposure groups for half-lives.

PCB-74 and occupational exposure during the years that Aroclor 1016 was used, probably due to the fact that nearly 30 years has elapsed from the time of last exposure. Only one congener was significantly associated with Aroclor 1254, which is likely related to the observation that only 10% of the study population was employed during the years that Aroclor 1254 was used. Employment during the years that Aroclor 1254 was used was moderately correlated with Aroclor 1242 ($r = 0.56$), but not associated with Aroclor 1016 ($r = -0.01$). In contrast, serum PCB concentrations were not associated with reported consumption of fish from PCB-contaminated bodies of fresh water in New York State, suggesting that the major source of PCB exposure in this cohort indeed was occupational.

Examination of the relationships between 2004 serum PCB concentrations and demographic variables provides additional insights, as well as confirming that the results reported here are similar to those found in previous studies. Current serum PCB concentrations were significantly and positively associated with the age of the individual, a finding that has been previously reported (Wolff et al., 2005). There were also significant gender differences in current serum lipid-adjusted PCB concentrations, with men in our cohort having higher serum PCB concentrations than women. This finding was not unexpected as men were more likely than women to have been occupationally exposed to PCBs, and thus had higher initial serum PCB levels. In addition, there were significant negative associations between both occupational light and total PCB concentrations and the number of years of education reported by the former capacitor plant workers. These associations most likely reflect the types of jobs held by the individuals, as $\sim$85% of those with a high school education or less were occupationally exposed compared with $\sim$74% of those who had some college education. Finally, BMI was positively associated with an increase in serum concentrations of lightly chlorinated PCBs. The findings of other studies, with regard to BMI are, however, mixed with some investigators reporting positive correlations (Falk et al., 1999; Fitzgerald et al., 2007a), whereas others have found either inverse associations (Wolff et al., 2000) or none (Hanrahan et al., 1999). For a review, the issues surrounding associations with BMI, see Wolff et al. (2005, 2007).

The major strength of this study is the ability to compare serum PCB concentrations in the same group of highly exposed persons over nearly three decades. With this information, we were able to more accurately and completely estimate half-lives for many congeners than was possible with shorter periods of follow-up. The fact that the PCB determinations were made by the same laboratory at the same time using the same analytical methods facilitated this comparison.

As in all observational studies, however, our findings should be considered in the context of possible limitations. For example, one possible problem is selection bias that might alter the type of persons participating in the study and their health status. Our results showed that, while the gender composition of the final study population ($N = 241$) did not differ from that of the source population, this group did differ in terms of both age and probable occupational PCB exposure level. The younger mean age of the participants probably reflects the fact that older persons were more likely to have died before the tracing began. The finding that the study participants were more likely to have been heavily exposed according to the original job codes reflected the fact that exposure levels were among the criteria used to select participants for recruitment in this study, that is, persons in the high exposure group were oversampled. As current serum PCB concentrations were measured prospectively, however, systematic bias is unlikely. In general, the 45-person subgroup that had both current and historical serum PCB data was similar to the larger cohort of 241 individuals. The only exceptions were among women, with those in the 45 person subgroup being older, more likely to have had children, and of lower BMI. These factors, however, should not bias the half-life data, as they were based on intra-person comparisons, and age, parity, and BMI were not associated with our estimates of half-lives.

An additional limitation that restricted our ability to better define the PCB half-lives was the relatively small sample size of those individuals for whom we had archived sera values, the 45-person subgroup, especially as it included only 12 women. A larger sample size would have allowed us to stratify both men and women into ''high'' and ''low'' exposures in order to determine whether the recognized inverse relationship between initial serum PCB concentrations and half-lives differed between men and women. Another limitation is the relatively crude nature of the exposure assessment for fish consumption, which did not take into account the PCB concentration(s) of any fish consumed. Hence, it is possible that some non-differential misclassification may have been introduced, making it more difficult to detect an association between serum PCB concentrations and fish consumption. We also did not have individual determinations of PCB intake from fish or other foodstuffs, restricting our half-life estimates to those five congeners that were primarily of occupational origin.

Conclusion

In summary, serum PCB concentrations in this cohort of former capacitor plant workers, determined nearly 30 years after direct occupational exposures ceased, remained substantially elevated relative to serum PCB levels seen in individuals residing in the same communities who did not work at the GE capacitor factories. By comparing serum concentrations in a subgroup of workers from 1976 to 2004, we demonstrated that some of the congeners that were

occupational in origin have longer half-lives than previously estimated. In general, the half-lives for light occupational congeners were shorter than those for heavy congeners. We have also shown that half-lives for the occupational congeners were inversely proportional to initial body burdens that may aid in explaining the significantly longer half-lives we observed in women. The revised half-lives should further aid in understanding the toxicological/epidemiological consequences of exposure to PCBs, as well as identifying earlier vectors of exposure to PCBs. Finally, earlier occupational exposure remained a significant predictor of current serum PCB concentrations, further supporting a consistent and long-duration trend of increased PCB body burden in this cohort of former capacitor workers.

Conflict of interest

The authors declare no conflict of interest.

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