Title
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Journal
Environmental Science and Technology, 48(1)

ISSN
0013-936X

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Publication Date
2014-01-07

DOI
10.1021/es403863m

Peer reviewed

Published in final edited form as:
Environ Sci Technol. 2014 January 7; 48(1): 157–164. doi:10.1021/es403863m.

Polychlorinated Biphenyls in Residential Dust: Sources of Variability

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Abstract

We characterized the variability in concentrations of polychlorinated biphenyls (PCBs) measured in residential dust. Vacuum cleaner samples were collected from 289 homes in the California Childhood Leukemia Study during two sampling rounds from 2001 to 2010 and 15 PCBs were measured by high resolution gas chromatography–mass spectrometry. Median concentrations of the most abundant PCBs (*i.e.*, PCBs 28, 52, 101, 105, 118, 138, 153, and 180) ranged from 1.0–5.8 ng per g of dust in the first sampling round and from 0.8–3.4 ng/g in the second sampling round. For each of these eight PCBs, we used a random-effects model to apportion total variation into regional variability (6–11%), intra-regional between-home variability (27–56%), within-home variability over time (18–52%), and within-sample variability (9–16%). In mixed-effects models, differences in PCB concentrations between homes were explained by home age, with older homes having higher PCB levels. Differences in PCB concentrations within homes were explained by decreasing time trends. Estimated half-lives ranged from 5–18 years, indicating that PCBs are removed very slowly from the indoor environment. Our findings suggest that it may be feasible to use residential dust for retrospective assessment of PCB exposures in studies of children's health.

Keywords

Environmental exposures; house dust; polychlorinated biphenyls

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ASSOCIATED CONTENT

Supporting Information. The supporting information features a map of the study area (Figure S1) and a graphical representation of changes in PCB concentrations within homes between sampling rounds (Figure S2) as well as details regarding analytical standards (Table S1), quality control (Table S2), questionnaires (Table S3), data imputation (Table S4), single variable (Table S5) and multivariable (Table S6) model fit, inter-round correlation coefficients by sampling interval (Table S7), and indoor PCB half-lives (Table S8). This material is available free of charge via the Internet at http://pubs.acs.org.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

INTRODUCTION

Polychlorinated biphenyls (PCBs) were used extensively in electrical, heat transfer, and hydraulic equipment.[1] The Toxic Substances Control Act ended U.S. production and distribution of PCBs in 1979,[2] but these contaminants persist in U.S. homes.[3]

Dust is an important route of exposure to PCBs. Knobeloch *et al.*[4] demonstrated that concentrations of PCBs in vacuum-cleaner dust were positively associated with concentrations of PCBs in human serum (p-value=0.07) in 26 sample pairs. Likewise, Rudel *et al.* found that residents of two homes with elevated dust-PCB levels also had high serum-PCB levels.[5] Unintentional ingestion of residential dust is a particularly important route of exposure to PCBs for young children.[6] Elevated levels of PCBs in residential dust have been associated with increased risks of childhood leukemia.[7] Moreover, prenatal and early childhood exposures to PCBs have been associated with adverse immunological[8] and neurological[9] effects, including diminished IQ.[10]

Several researchers have measured PCBs in dust, but few have sampled dust repeatedly in the same homes and characterized the variability of dust measurements over time.[3, 5, 11] When estimating the health effects related to a chemical exposure, the variance ratio (*i.e.*, ratio of within-subject variability to between-subject variability) is predictive of the underestimation of true risks due to exposure measurement error.[12] Variance ratios for PCBs in residential dust are relatively small when dust samples are collected at semiannual intervals.[11] Thus, one dust measurement can characterize average PCB levels found in a residence over a short period of time. Variance ratios for PCBs in residential dust have not been estimated over intervals of years or decades, but these estimates would be useful to investigators who plan to assess long-term exposures to PCBs and other persistent pollutants.

To characterize the long-term temporal variability of PCB concentrations in residential dust, we analyzed 15 PCB congeners in dust samples collected at intervals of 3–8 years. We also identified determinants of PCB levels and estimated long-term trends in PCB levels from 2001 to 2010.

MATERIALS AND METHODS

Study population

The California Childhood Leukemia Study (CCLS) is a case–control study of childhood leukemia conducted in the San Francisco Bay Area and California's Central Valley that aims to identify genetic and environmental risk factors for childhood leukemia. Cases aged 0–14 years were ascertained from pediatric clinical centers; controls, matched to cases on date of birth, sex, Hispanic ethnicity, and maternal race, were selected from the California birth registry. Dust samples were collected from homes of cases and controls from 2001 to 2007 (Round 1) if the children were 0-7 years-old at study enrollment and if the families had not moved since study enrollment. Subsequently, in 2010, households that participated in Round 1 were eligible for a second dust collection (Round 2) if the family was still living in the same home. Among 629 participants in Round 1, 225 were eligible for Round 2, and 204 participated. PCB analyses were performed on dust samples from 289 homes; 201 homes with one dust sample from each round, 87 homes with only a Round 1 dust sample (ineligible for Round 2), and one home with only a Round 2 dust sample (insufficient Round 1 dust remaining).

Dust collection

Dust was collected from the participants' vacuum cleaners, which were used for typical household cleaning. The median interval between rounds was 4.8 years (range: 2.6–8.6 years). During Round 1, a questionnaire was administered and vacuum-cleaner dust was obtained during an in-home visit. For Round 2, participants were interviewed via telephone and instructed to mail the contents of their vacuum cleaners to the study center in prepaid parcels. Dust samples were stored in the dark at ≤4 °C prior to analysis. Previously, we measured 6 PCBs in dust samples that were collected during Round 1 using either a high-volume small-surface sampler (HVS3) or household vacuum cleaners;[7, 13] however, for consistency within our analysis, vacuum-cleaner dust samples from Rounds 1 and 2 were extracted and analyzed concurrently, according to the protocol described below.

PCB analysis

Dust particles smaller than 150 μm were obtained with a 100-mesh sieve and 0.2-g portions were spiked with 15 ^{13}C-labeled internal standards (*i.e.*, 1 ng of each ^{13}C-labeled analog for each analyte), extracted via accelerated solvent extraction using 5 cycles of 95% hexane and 5% methylene chloride at 100 °C and 1500 psi (Dionex Corporation, Sunnyvale, CA), purified by silica-gel column chromatography and gel permeation chromatography, concentrated to 250 μL, solvent exchanged into tetradecane, and spiked with three ^{13}C-labeled recovery standards (*i.e.*, 5 ng each of ^{13}C-labeled PCBs 47, 128, and 178). A detailed description of the analytical standards can be found in the Supporting Information (Table S1). Finally, we analyzed 15 PCB congeners (PCBs 28, 52, 101, 105, 114, 118, 138, 153, 156, 157, 167, 180, 189, 194, and 209) by isotope-dilution/high resolution gas chromatography—mass spectrometry using a MAT95 high resolution mass spectrometer (Thermo-Fisher Scientific, Waltham, MA) operated in electron impact ionization-selective ion monitoring (EI-SIM) mode, dual 6890 GCs (Agilent Technologies, Santa Clara CA) equipped with a RTX-Dioxin2 column (60 m × 0.25-mm i.d., 0.25-μm film thickness; Restek, Bellefonte, PA), and a split/splitless injector operated in splitless mode (at 270 °C). The initial GC temperature was held at 140 °C for 1.0 min, then ramped to 235 °C at 19.8 °C/min, then to 283 °C at 1.5 °C/min, and finally to 300 °C at 4.25 °C/min with a 1.7 min hold. Helium was used as the carrier gas in the GC at a constant flow of 1.0 mL/min.

We analyzed samples in batches of 12, with each batch consisting of 8 single samples, 1 duplicate sample pair (*i.e.*, two 200-mg portions of dust taken from the same vacuum cleaner), 1 inter-batch quality control sample (*i.e.*, one of a series of 200-mg portions of dust taken from the same vacuum cleaner bag), and 1 method blank. We also analyzed 14 replicate samples of National Institute of Standards and Technology Standard Reference Material 2585 (NIST SRM2585). For 8 of the 15 PCBs analyzed in this study, we compared the NIST-certified concentrations to the average concentrations measured in 14 NIST replicates and found that the method was suitably accurate (*i.e.*, <35% error). Median internal standard recoveries in field samples ranged from 80 to 193% for the 15 ^{13}C-labeled internal standards, with 13 of the 15 medians above 100%. We calculated the coefficient of variation (CV) for replicate quality control samples (see Supporting Information, Table S2). For each PCB, the CV for 14 replicate NIST SRM 2585 dust samples was smallest (7-11%), followed by the CV for 55 pairs of duplicate samples (20-66%), and the CV for 40 replicate quality control samples (36-77%).

Household characteristics

In structured in-home interviews conducted during Round 1, parents provided information relevant to childhood leukemia, as well as socio-demographic information such as the annual household income and the parents' age, ethnicity, and race. Because demographic descriptors of mothers and fathers were mostly concordant, we used the more complete data

describing mothers. In a subsequent telephone questionnaire (see Supporting Information, Table S3), parents participating in Round 2 described sources of residential chemical exposures; residential characteristics, including the construction date, construction material, square footage, and type of residence (*e.g.*, single family home, apartment); vacuum characteristics, including the type of vacuum and the frequency of its use; and resident activities including remodeling, occupations, window and air conditioner use, and shoe removal habits. Finally, we used a global positioning device to locate each residence and classify it as belonging to one of six geographic regions as shown in the Supporting Information (Figure S1).

Data imputation

We set the method reporting limit (MRL) for each PCB equal to the standard deviation of the mass of each PCB in 56 method blanks, multiplied by three (*i.e.*, MRL = $3\times\sigma_{blanks}$). We assigned all values below the MRL (see Table 1) a concentration equal to MRL/$\sqrt{2}$.[14] Because some participants did not complete all parts of the questionnaires, missing responses were replaced by the population averages from non-missing households in regression analyses (*e.g.*, five respondents did not know their residence's construction date and we used the population average, *i.e.*, 1972; see Supporting Information, Table S4). Regression results were similar for the smaller number of observations with complete questionnaire responses (data not shown).

Random-effects models

To apportion the observed variance in PCB concentrations into four components describing regional variability, intra-regional between-home variability, within-home variability over time, and within-sample variability we used a hierarchical random-effects model of natural-log transformed PCB-dust concentrations,

$$Y_{hijk}=ln\left(X_{hijk}\right)=\mu_Y+b_h+b_{hi}+b_{hij}+e_{hijk} \quad \text{(Model 1)}$$

for h = 1, 2, …, 6 regions; i = 1, 2, …, 290 homes (*i.e.*, 289 CCLS homes and the inter-batch quality control home); j = Round 1 or 2; and k = 1, 2…, 40 replicate samples from the same vacuum cleaner bag, as previously described.[15, 16]

We assumed b_h, b_{hi}, b_{hij}, and e_{hijk} are independent and normally distributed random variables, with means of zero and variances of σ^2_{BR}, σ^2_{BH}, σ^2_{WH}, and σ^2_{WS}, representing the between-region variance, the intra-regional between-home variance, the within-home variance over time, and the within-sample variance, respectively. Using Proc Mixed (SAS v. 9.1, Cary, NC) we fit the random-effects model and estimated variance components $\hat{\sigma}^2_{BR}, \hat{\sigma}^2_{BH}, \hat{\sigma}^2_{WH}, \hat{\sigma}^2_{WS}, \hat{\sigma}^2_{Total}=\hat{\sigma}^2_{BR}+\hat{\sigma}^2_{BH}+\hat{\sigma}^2_{WH}+\hat{\sigma}^2_{WS}$ and variance ratios $\left(\lambda=\frac{\hat{\sigma}^2_{WH}+\hat{\sigma}^2_{WS}}{\hat{\sigma}^2_{BR}+\hat{\sigma}^2_{BH}}\right)$. For each PCB, we used the estimated variance ratio to predict the potential impact of measurement error on an odds ratio (OR_{True} = 2.0) for a hypothetical case–control study that employs a single dust sample to assess long-term average exposure to PCBs $\left(OR_{Expected}=\exp\left[\frac{ln\left(OR_{True}\right)}{1+\lambda}\right]\right)$, as previously described.[11]

Mixed-effects models

We estimated the proportion of variability in PCB concentrations explained by each factor considered for inclusion in the mixed-effects model (see Supporting Information, Table S5). We retained a group of factors that were each significantly (p-value<0.05) associated with concentrations of at least one PCB and which collectively minimized the unexplained

variance in PCB concentrations (see Supporting Information, Table S6). In addition to the random effects described above, the mixed-effects model (Model 2) included eight fixed effects, including residential construction date, residential square footage, mother's age at Round 1, vacuum type (*i.e.*, vacuum was equipped with a disposable bag), and the change in PCB levels between sampling rounds for four different groups of households (*i.e.*, for case households, for households that installed flooring between sampling rounds, for households that did construction between sampling rounds, and for all other households). Based on the regression coefficients for the four time trends included in Model 2, we estimated the indoor half-life of each PCB[17] [see Supporting Information].

We fit the mixed-effects models for 448 observations with covariate data (*i.e.*, 403 samples collected from 202 homes during two rounds and 45 duplicate samples) and excluded the 137 observations without covariate data (*i.e.*, 40 inter-batch quality control replicates and 87 samples with 10 duplicates collected from homes that participated in Round 1 only). For comparison, we re-ran the random-effects model (Model 1) using this set of 448 observations

RESULTS

Table 1 shows summary statistics for PCB measurements in 289 California homes. PCBs with five or fewer chlorines that were commonly used in commercial mixtures (*i.e.*, PCBs 28, 52, 101, 105, and 118) were detected in at least 90% of dust samples from both sampling rounds. Commonly used PCBs with more than five chlorines (*i.e.*, PCBs 138, 153, and 180), had higher MRLs and, consequently, lower detection frequencies (*e.g.*, 84%, 87%, and 67% for Round 2, respectively). In Round 1, eight PCBs had median concentrations of at least 1 ng/g (*i.e.*, PCBs 28, 52, 101, 105, 118, 138, 153, and 180; hereafter referred to as the "major PCBs"). Median PCB concentrations were consistently lower in Round 2 compared to Round 1. Spearman rank correlation coefficients for inter-round comparisons of dust concentrations of the major PCBs ranged from 0.54 to 0.69 (p-values < 0.0001). Inter-round correlations were equally strong for households with repeat samples collected 6-8 years apart compared to households with repeat samples collected 3-5 years apart (see Supporting Information, Table S7).

Table 2 summarizes the distribution of concentration ratios between Rounds 1 and 2 for the major PCBs and Figure S2 (Supporting Information) shows the change in PCB-153 concentrations between rounds for each home. Some homes had widely differing PCB concentrations in dust from the two sampling rounds. As shown in Table 2, PCB-153 concentrations decreased within one home by as much as 20-fold between sampling rounds (*i.e.*, minimum ratio between rounds of 0.05), whereas PCB-153 concentrations increased within another home by as much as 8.8-fold. Each median concentration ratio was less than one (range for 7 of the 8 major PCBs of 0.74-0.84; 0.44 for PCB-138), indicating that, in most homes, PCB concentrations decreased from Round 1 to Round 2. For example, PCB-138 concentrations decreased in 80% of homes and increased in 20% of homes.

Random-effects model

Table 3 shows estimated variance components from Model 1 with corresponding variance ratios for each major PCB. Between-region variability accounted for 6-11% of the total variability in PCB concentrations, intra-regional between-home variability accounted for 27-56%, within-home variability over time accounted for 18-52%, and within-sample analytical variability accounted for 9-16%. The variance ratio ranged from 0.5 to 1.9, with ratios for PCBs 52, 101, 105, 118, 153, and 180 estimated to be less than 1.0. For example, PCB-153 had a variance ratio of $\lambda = 0.5$, suggesting that an investigator using a single dust

sample to estimate exposures to PCB-153, would be expected to observe an OR_{Exp} = 1.6 compared to a true effect size of OR_{True} = 2.0.

Mixed-effects models

Table 4 shows the percent change in PCB concentrations associated with a unit increase in each of the fixed effects included in the mixed-effects model (Model 2) for each major PCB. Residential construction date was significantly associated with concentrations of each PCB using Model 2, with more recently constructed homes having lower concentrations (9–18% decrease in concentrations for each ten-year increment in construction date). PCB concentrations were positively associated with the mother's age and this effect was statistically significant for PCBs 28 and 52, with concentrations of these two PCBs increasing by 18 and 14% per 5-year increment in the mother's age, respectively. PCB concentrations were negatively associated with the residential square footage and this effect was statistically significant for PCBs 101, 105, 118, and 153 with concentrations decreasing by 10–11% per 500-sf increment. Concentrations of PCBs 101, 105, 118, 153, and 180 were 24-39% higher in dust samples collected from vacuum cleaners with a disposable bag (N=136, 116 in Rounds 1 and 2, respectively) compared to concentrations in dust samples collected from a bag-less vacuum cleaner (N=65, 86).

Concentrations of PCBs generally decreased from Round 1 to Round 2 and these trends were statistically significant for PCBs 28, 138, and 180, with concentrations of these three PCBs decreasing by 4–12% per year. PCB concentrations decreased more rapidly in case households and in households in which flooring was installed between sampling rounds (as much as an additional 6% decrease per year associated with case status and as much as an additional 7% decrease per year associated with floor installation). In contrast, in households where construction was done between sampling rounds, PCB concentrations decreased more slowly over time.

Based on the regression coefficients from Model 2, Table S8 (Supporting Information) shows the estimated indoor half-life of each major PCB for different groups of study homes. In general, indoor half-lives for each of the major PCBs were estimated to be between 11-18 years, with the exception of PCB-138, which had a shorter half-life of 5 years. In case households or in households that installed flooring between sampling rounds, PCBs were relatively short-lived.

Table S5 (Supporting Information) shows the proportion of variability in PCB concentrations accounted for by each explanatory factor considered for inclusion in the mixed-effects model. Residential construction date was the most influential factor, explaining 56-83% of the regional variability and 10-29% of the intra-regional between-home variability in PCB concentrations. As illustrated in Figure S1 (Supporting Information), we observed the highest PCB concentrations in homes from the metropolitan San Francisco Bay Area. Homes in this region were more likely to be constructed prior to 1980 (median construction date of 1960) compared to homes from other regions (median construction date of 1985). Table S6 (Supporting Information) shows the proportion of variability in PCB concentrations accounted for by all of the fixed effects included in Model 2 for each major PCB. Model 2 explained 61-88% of the regional variability, 0-25% of the intra-regional between-home variability, and 12-52% of the within-home variability in PCB concentrations.

DISCUSSION

Variability in repeat dust samples

Dodson *et al.*[3] previously reported temporal variability in concentrations of PCBs in repeat dust samples collected from 16 California homes in 2006 and 2011, with most homes experiencing decreases in PCB concentrations between sampling rounds. The authors reported that PCB-153 concentrations decreased within one home by as much as 6-fold between rounds; whereas other homes experienced as much as a 3-fold increase in PCB-153 concentrations. In comparison, over a slightly longer sampling interval and in a larger population of homes, we observed more dramatic changes in concentrations of PCB-153 (maximum decrease in a home of 20-fold; maximum increase in another home of 8.8-fold).

Despite observing temporal variability of PCBs in some homes, Dodson *et al.*[3] reported that concentrations of PCB-153 were significantly correlated between sampling rounds (r_s = 0.53). Likewise, we observed significant correlation between PCB concentrations from two sampling rounds (r_s of at least 0.6 for 7 of the 8 major PCBs). Our findings suggest that the rank order of PCB concentrations in homes remained consistent across our study population for up to 8 years. In comparison, we previously reported more modest inter-round correlation for concentrations of polycyclic aromatic hydrocarbons, PAHs[15] (*e.g.*, r_s = 0.50, 0.47, and 0.57 for chrysene, benzo[*a*]pyrene, and benzo[*g,h,i*]perylene, respectively) and polybrominated diphenyl ethers, PBDEs[16] (*e.g.*, r_s = 0.51, 0.56, and 0.18 for BDE-47, BDE-99, and BDE-209, respectively) measured in the same dust samples.

For PCBs 52, 101, 105, 118, 153, and 180 the ratio of within-home variance to between-home variance was relatively modest (*i.e.*, $\lambda \leq 0.8$). In comparison, we previously reported larger variance ratios for PAHs[15] (*e.g.*, λ = 1.2, 1.1, and 1.0 for chrysene, benzo[*a*]pyrene, and benzo[*g,h,i*]perylene, respectively) and PBDEs[16] (*e.g.*, λ = 1.1, 0.9, and 4.4 for BDE-47, BDE-99, and BDE-209, respectively) measured in the same dust samples. We deduce that, in contrast to other chemical sources, the sources of PCBs in the study homes remained consistent from 2001-2010.

Factors that explain changes in PCB concentrations within homes

Dodson *et al.*[3] found that when comparing dust-PCB concentrations from samples collected in 2011 to concentrations from samples collected in 2006, the median ratio between rounds for 16 homes was 0.77 for PCB-153, suggesting a decrease in most homes over time. When comparing PCB concentrations from two sampling rounds, we found a similar median ratio of 0.79. The principal factor that influenced the change in PCB concentrations was the time between sampling rounds, as homes that were sampled over a greater interval of time tended to experience larger reductions in PCB concentrations. Using models of chemical fate, Shin *et al.*[18] predicted the indoor half-lives of two PBDEs (BDEs 47 and 99) to be in the range of 7-11 years. Based on the observed decreases in PCB concentrations between two sampling rounds, we estimated a similar range of indoor half-lives for the major PCBs (5-18 years).

We identified three factors that modified the rate of change of PCB concentrations over time; namely, case-control status, floor installation, and residential construction. PCB concentrations decreased slightly faster in case households than control households. It is possible that case parents initiated more intense cleaning activities after their child was diagnosed with leukemia and that these new behaviors explain the faster decrease in PCB levels. We are limited in our ability to evaluate this hypothesis; however, frequency of vacuum use did not explain differences in PCB levels.

Households that reported installing floors between sampling rounds had more drastic decreases in PCB concentrations than other households. Most of these households (47 of 89)

also reported carpeting between sampling rounds. Investigators have demonstrated that carpet pads[19] and wood floor finishes[5] can be residential PCB sources. Moreover, it has been suggested that semivolatile organic compounds tend to partition to non-mobile household surfaces, such as carpet pads[18] and concentrations of PCBs in dust have been correlated with floor and carpet age.[20, 21] We hypothesize that PCB residues were removed in homes where wood floors (or possibly carpets/carpet pads) were replaced between sampling rounds, resulting in modest reductions of PCB concentrations.

In contrast, households that reported residential construction between sampling rounds experienced slower decreases in PCBs over time. Homes built before 1980 may contain PCB-contaminated construction materials, such as paint, ceiling tiles, insulation, caulk, and roofing.[1, 22] Construction activities may result in the release of PCB-contaminated dust particles from these materials.

Factors that explain differences in PCB concentrations between homes

Investigators have observed elevated PCB concentrations in dust from older residences.[4, 23] Likewise, we previously reported that home age was the strongest predictor of PCB loadings (*i.e.*, ng of PCB per m^2 of carpet) in dust samples collected using an HVS3 from an overlapping group of CCLS homes.[13] In this analysis we confirmed that home age was the factor that explained the greatest proportion of variability in PCB concentrations between homes. Since U.S. regulations ended the production and distribution of PCBs in 1979,[1] homes built before 1980 have a greater potential for historic PCB contamination via indoor sources than more recently constructed residences. Thus, older homes tend to have higher PCB dust concentrations.

In multivariable mixed-effects models, after adjusting for home age, we found that PCB concentrations were negatively associated with residential square footage. We suggest that home size may act as a surrogate for socioeconomic status. We found that household annual income and residential square footage were correlated in our study population; and, although, mixed-effects models including income explained less variability in PCB concentrations than those including residential square footage, the effect of both explanatory factors were similar when included in separate models (data not shown). Potentially, families of lower socioeconomic status are more likely to own PCB-contaminated consumer items, to live in homes that have PCB-contaminated construction materials, or to live near outdoor PCB sources. Alternatively, it may be that the number of PCB sources in a home is only weakly related to its square footage; whereas the mass of dust found in a home may be proportional to its size. If true, a dilution effect could explain the observed inverse relationship between PCB concentrations and home size.

We also found that mother's age was associated with concentrations of some PCBs in multivariable mixed-effects models after adjusting for home age. We speculate that older mothers (and fathers) are more likely to have owned PCB-contaminated consumer products manufactured before 1980, such as fluorescent lights, refrigerators, televisions, carpet pads, or air conditioners.[19, 24-26] While most of these pre-1980 consumer items were likely removed from homes prior to dust collection, these items could have contaminated carpet dust or household surfaces, creating persistent PCB residues.

Compared to a previous study that collected residential dust from homes in Fresno County, California from 2003–2005 and reported median concentrations of 3, 5, 4, and 3 ng/g for PCBs 118, 138, 153, and 180, respectively;[13] we report very similar median PCB concentrations for dust samples collected from 2001-2007 (*i.e.*, 3, 6, 4, and 2 ng/g, respectively). On the other hand, Dodson *et al.*[3] reported higher median concentrations of PCBs 153 and 180 (*i.e.*, 18 and 16 ng/g, respectively) for 16 homes in the San Francisco Bay

Area sampled in 2006. Differences in PCB levels between these studies may be explained in part by differences in the characteristics of the homes sampled (*e.g.*, construction date).

We previously reported that, in dust samples collected using an HVS3 from an overlapping population of CCLS homes; PCB loadings were associated with residential construction date, recent floor installation, surrounding population density, resident employment as an electrician, and the practice of wearing outdoor shoes in the home.[13] Only the first two of these explanatory factors influenced PCB concentrations in this analysis. Likewise, the determinants of PBDE concentrations that we previously identified using the same dust samples[16] were mostly unrelated to the factors that explained PCB concentrations in this analysis. For example, while concentrations of long-ago banned PCBs were strongly associated with home age, concentrations of their more modern counterparts, PBDEs, were marginally inversely associated with home age. However, in both cases, certain remodeling activities were associated with decreases in chemical concentrations, suggesting that floor and carpet replacement may reduce indoor half-lives of various classes of semivolatile organic compounds. Likewise, two possible surrogates for socioeconomic status, household annual income and residential square footage, were weakly and negatively associated with chemical concentrations of both PCBs and PBDEs, suggesting that low-income families may be disproportionately exposed to elevated levels of multiple chemical contaminants.

Limitations

Given that the focus of the CCLS is to identify risk factors for childhood leukemia, our questionnaire and sampling protocol were limited in scope. We lacked a detailed survey describing the age and quantity of household items and construction materials that may have contained PCBs. Moreover, we relied on dust samples from vacuum cleaners to characterize indoor PCB levels rather than collecting surface wipes or materials from putative PCB sources. Environmental sampling at a finer spatial scale, administration of more exhaustive questionnaires, measurement of gas-phase PCB concentrations, or characterization of organic matter content in dust samples might have revealed more information about residential PCB sources.

We obtained dust samples from vacuum cleaners, which, from home-to-home and from round-to-round, may have been used in a different combination of rooms and at different proximity to PCB sources. Differences in vacuum cleaning practices between and within homes as well as differences in the vacuum cleaners used to collect dust (*e.g.*, type, efficiency) could be responsible for some of the unexplained variability in PCB levels. Our use of vacuum-cleaner dust samples precluded our ability to evaluate the temporal variability of PCBs in a specific location or the spatial variability of PCBs from different locations in the same home.

Vacuum-dust PCB measurements in studies of children's health

We observed substantial variability in concentrations of PCBs measured in duplicate samples and inter-batch quality control replicates. The analytical variability observed in our study is attributable to the heterogeneous nature of vacuum dust and the heterogeneous distribution of chemicals in vacuum-dust samples.[27] Our dust preparation protocol relied on a mechanical sieve shaker to homogenize dust samples. The more thorough homogenization employed in the NIST SRM 2585 dust preparation protocol (*i.e.*, using a food processor, a compressed air jet, and a cone blender) improved analytical reproducibility. We recommend that future investigators homogenize dust samples using a commercial blender.

Given the moderate variance ratios and significant correlation between sampling rounds for concentrations of most major PCBs, we conclude that dust measurements would be useful in

case–control studies where past levels of PCB exposures are of interest and post-diagnosis sample collection is necessary. However, because we found that PCB concentrations decreased more rapidly between sampling rounds in case households compared to control households, we suggest that investigators who plan to use residential dust to estimate past levels of PCBs in case-control studies should start sampling as soon as possible after participant enrollment. If long-term average chemical exposures are of interest and prospective sample collection is feasible (*e.g.*, cohort studies), investigators can improve the precision of their exposure estimates and limit the attenuation of observed risk estimates by making repeated exposure measurements on each participant.[11]

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

This work was supported in part by the National Institute of Environmental Health Sciences (NIEHS, grant numbers R01ES009137, R01ES015899, P42ES0470518, and P01ES018172); by the Intramural Research Program of the National Cancer Institute (NCI), National Institute of Health (subcontracts 7590-S-04, 7590-S-01); by the NCI (contract N02-CP-11015); and by the Environmental Protection Agency (EPA, grant number RD83451101). We thank the families for their participation. We also thank the clinical investigators at the following collaborating hospitals for help in recruiting patients: University of California Davis Medical Center (Dr. Jonathan Ducore), University of California San Francisco (Drs. Mignon Loh and Katherine Matthay), Children's Hospital of Central California (Dr. Vonda Crouse), Lucile Packard Children's Hospital (Dr. Gary Dahl), Children's Hospital Oakland (Dr. James Feusner), Kaiser Permanente Oakland (Drs. Daniel Kronish and Stacy Month), Kaiser Permanente Roseville (Drs. Kent Jolly and Vincent Kiley), Kaiser Permanente Santa Clara (Drs. Carolyn Russo, Denah Taggart, and Alan Wong), and Kaiser Permanente San Francisco (Dr. Kenneth Leung). Finally, we acknowledge the entire California Childhood Leukemia Study staff for their effort and dedication.

ABBREVIATIONS

CCLS	California Childhood Leukemia Study
CV	coefficient of variation
HVS3	high-volume small-surface sampler
MRL	method reporting limit
OR	odds ratio
PCB	polychlorinated biphenyls

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Table 1

Summary statistics for PCB measurements in dust collected during two rounds from 289 residences in the California Childhood Leukemia Study, 2001–2007 and 2010

PCB	Number of chlorine atoms	MRL, ng/g	Round 1, 2001-2007, *N*=288		Round 2, 2010, *N*=202		Maximum Concentration, ng/g	Rank correlation between rounds	
			Values above MRL, *N* (%)	Median concentration, ng/g	Values above MRL, *N* (%)	Median concentration, ng/g		r_s	*p*-value
PCB-28	3	0.15	286(99)	1.5	199(99)	1.2	270	0.54	<0.0001
PCB-52	4	0.28	286(99)	2.9	200(99)	2.4	190	0.62	<0.0001
PCB-101	5	0.45	276(96)	3.8	182(90)	3.4	480	0.67	<0.0001
PCB-105	5	0.08	277(96)	1.0	191(95)	0.8	200	0.68	<0.0001
PCB-114	5	0.24	21(7)	<MRL	15(7)	<MRL	10	0.15	0.03
PCB-118	5	0.21	281(98)	2.5	186(92)	2.0	460	0.69	<0.0001
PCB-138	6	0.73	285(99)	5.8	169(84)	3.2	480	0.60	<0.0001
PCB-153	6	0.60	261(91)	3.6	176(87)	3.4	350	0.63	<0.0001
PCB-156	6	0.04	266(92)	0.3	144(71)	0.2	60	0.54	<0.0001
PCB-157	6	0.01	212(74)	0.05	61(30)	<MRL	20	0.35	<0.0001
PCB-167	6	0.04	223(77)	0.2	120(59)	0.10	20	0.45	<0.0001
PCB-180	7	0.72	224(78)	2.4	136(67)	1.8	180	0.62	<0.0001
PCB-189	7	0.01	169(62)	0.02	14(10)	<MRL	2	0.23	0.01
PCB-194	8	0.22	200(69)	0.7	102(50)	0.4	50	0.55	<0.0001
PCB-209	10	0.04	222(77)	0.2	63(31)	<MRL	50	0.49	<0.0001

MRL = Method reporting limit = 3× σ_{blanks}

Table 2

Distribution of concentration ratios between repeat samples (Round 2/Round 1)[a] for major PCBs, *N*=201 pairs

PCBs	Min	25th	Med	75th	Max	% <1
PCB-28	0.02	0.45	0.77	1.1	110	66
PCB-52	0.04	0.52	0.74	1.2	36	68
PCB-101	0.05	0.43	0.77	1.3	16	63
PCB-105	0.03	0.44	0.77	1.5	20	61
PCB-118	0.02	0.43	0.79	1.3	19	60
PCB-138	0.02	0.24	0.44	0.8	6.2	80
PCB-153	0.05	0.41	0.79	1.5	8.8	58
PCB-180	0.005	0.39	0.84	1.1	18	56

Min = minimum; Med = median; Max = maximum; 25^{th} = 25^{th} percentile; 75^{th} = 75^{th} percentile; % < 1 = Percent of homes with a ratio less than one

[a] Concentration ratio between repeat samples < 1 indicates decrease in PCB concentrations between sampling rounds

Table 3

Estimated variance components for major PCBs from the random-effects model (Model 1[a]) with corresponding variance ratios.

PCB	Variance Component Estimate (95% Confidence Interval)				λ [b]	OR_{Exp} [c]	Percent of Total Variance			
	Between-region, σ^2_{BR}	Intra-regional between-home, σ^2_{BH}	Within-home over time, σ^2_{WH}	Within-sample, σ^2_{WS}			$\frac{\sigma^2_{BR}}{\sigma^2_{Total}}$	$\frac{\sigma^2_{BH}}{\sigma^2_{Total}}$	$\frac{\sigma^2_{WH}}{\sigma^2_{Total}}$	$\frac{\sigma^2_{WS}}{\sigma^2_{Total}}$
PCB-28	0.04 (0.01, 0.49)	0.22 (0.16, 0.34)	0.33 (0.26, 0.42)	0.07 (0.05, 0.09)	1.5	1.3	6	34	50	10
PCB-52	0.07 (0.02, 0.77)	0.42 (0.33, 0.56)	0.27 (0.21, 0.36)	0.10 (0.08, 0.13)	0.8	1.5	8	49	32	11
PCB-101	0.16 (0.06, 1.68)	0.81 (0.66, 1.04)	0.31 (0.23, 0.43)	0.16 (0.12, 0.21)	0.5	1.6	11	56	21	11
PCB-105	0.15 (0.05, 1.93)	0.97 (0.78, 1.24)	0.31 (0.21, 0.51)	0.28 (0.22, 0.38)	0.5	1.6	9	56	18	16
PCB-118	0.18 (0.06, 1.96)	0.89 (0.71, 1.14)	0.41 (0.32, 0.55)	0.14 (0.11, 0.19)	0.5	1.6	11	55	25	9
PCB-138	0.08 (0.03, 1.23)	0.32 (0.21, 0.52)	0.60 (0.48, 0.78)	0.16 (0.13, 0.22)	1.9	1.3	7	27	52	14
PCB-153	0.14 (0.05, 1.55)	0.78 (0.62, 1.00)	0.34 (0.26, 0.48)	0.15 (0.12, 0.21)	0.5	1.6	10	55	24	11
PCB-180	0.13 (0.04, 1.94)	0.75 (0.59, 0.99)	0.35 (0.25, 0.52)	0.23 (0.18, 0.32)	0.7	1.5	9	52	24	16

[a] In this table, Model 1 was fit for 585 observations including 402 samples collected from 201 homes during repeat sample collections, 88 samples from homes that were sampled once, 55 duplicate samples, and 40 inter-batch quality control replicates.

[b] Estimated variance ratio, $\lambda = \frac{\hat{\sigma}^2_{WH} + \hat{\sigma}^2_{WS}}{\hat{\sigma}^2_{BR} + \hat{\sigma}^2_{BH}}$

[c] Expected odds ratio, $OR_{Expected} = \exp\left[\frac{ln\,(OR_{True})}{1+\lambda}\right]$

Table 4

Percent change in concentrations of major PCBs associated with a unit increase in each of the fixed effects included in the mixed-effects model (Model 2a).

Explanatory variables	PCB Congener							
	28	**52**	**101**	**105**	**118**	**138**	**153**	**180**
Residential construction date[b]	−9*	−9*	−15*	−18*	−17*	−14*	−15*	−15*
Mother's age[c]	18*	14*	13	11	12	7	11	12
Residential square footage[d]	−5	−6	−11*	−11*	−11*	−8	−10*	−9
Vacuum has disposable bag	4	8	24*	36*	39*	19	25*	33*
Time between rounds (reference)[e]	−4*	−3	−3	−0.4	−3	−12*	−1	−6*
Time between rounds (cases)[e]	−0.4	−4	−4	−6*	−4	−3	−3	−1
Time between rounds (construction)[e]	3	4*	5*	5	6*	7*	4	6*
Time between rounds (floors)[e]	−5*	−3	−6*	−6*	−6*	−6*	−7*	−4

* Regression coefficient for fixed effect in Model 2 is significantly different from 0, p-value<0.05.

[a] In this table, Model 2 was fit for 448 observations including 403 samples collected from 202 homes during repeat sample collections and 45 duplicate samples; excluding 139 observations without covariate data (40 inter-batch quality control replicates and 87 samples with 10 duplicates from homes that were sampled during Round 1 only).

[b] Percent change per 10 year increment in residential construction date

[c] Percent change per 5 years increment in mother's age

[d] Percent change per 500-sf increment in residential square footage

[e] Percent change per 1 year increment in time between rounds