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Polychlorinated Biphenyls in House Dust and Yard Soil near a Superfund Site

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Polychlorinated biphenyls (PCBs) were measured in house dust and yard soil at 34 homes surrounding New Bedford Harbor during dredging of highly contaminated harbor sediments. PCBs can volatilize from sediments and seawater and subsequently deposit on surrounding soil, resulting in potential exposures for nearby residents. House dust was collected from carpet, while yard soil was collected from the main entryway to evaluate whether PCBs might be tracked indoors. All samples were analyzed for 65 PCB congeners to evaluate the relative importance of the harbor and indoor sources for human exposure. PCB concentrations (260−23 000 ng/g) in house dust were about 10 times higher than yard soil concentrations (15−1800 ng/g), although similar congener patterns were detected in these two media. Yard soil concentrations in neighborhoods closest to the harbor were significantly higher than those in comparison neighborhoods (23−1800 ng/g and 15−290 ng/g, respectively), while house dust concentrations did not differ significantly between these two locales (320−23 000 ng/g and 260−3600 ng/g, respectively). PCB concentrations in house dust were correlated with those in indoor air, but house dust and yard soil concentrations were not correlated, suggesting that track-in may not be the only source of PCBs in house dust.

Introduction

Polychlorinated biphenyls (PCBs) were used in New Bedford, MA, in the production of capacitors from the 1940s until the late 1970s. Their disposal during this period resulted in contamination of harbor sediments and closure of the harbor to fishing. People living near New Bedford Harbor may be exposed to PCBs that volatilize from contaminated harbor sediments and waters, deposit on soil, and are tracked into homes. We measured house dust and yard soil concentrations during dredging of these highly contaminated sediments to understand the harbor's role in residential PCB exposure. Because PCBs are ubiquitous in the environment, we sampled concurrently in distant, upwind neighborhoods relative to the harbor to discern the portion of PCB contamination attributable to harbor proximity.

For small children, house dust appears to be a primary route of exposure for pesticides, lead, and allergens (*1, 2*). Polycyclic aromatic hydrocarbons (PAHs) and PCBs also have been detected in house dust (*3, 4*). Lewis et al. (*5, 6*) concluded that pesticides persist longer in dust deep in carpets than in soil because they are more protected from sunlight, moisture, temperature extremes, wind and rain dispersal, and microbial activity. PCBs in house dust may likewise be protected from degradation (*7*). House dust may provide an important indicator of chronic exposure because PCBs persist in dust and because people, especially children, spend more time indoors than outdoors (*8*).

Many long-lasting household products may contain PCBs, such as wood product coatings, plasticizers in paints, sealants, flame retardants, plastics, fluorescent light ballasts, and small electrical capacitors in appliances (*9, 10*). Yard soil and house dust data were used to explore the relative contribution of indoor sources and harbor sediments to human exposure. Correlations between entryway soil and house dust pollutant concentrations have been calculated elsewhere (*4*, *11*). In this study, house dust and yard soil concentrations and congener profiles are compared to determine whether PCBs in dust may originate from yard soil.

This research is part of a multimedia PCB exposure study that included measurement of PCB concentrations in air, local produce, and tap water from the New Bedford area (*12*−*14*). Using data from this larger study, we compared PCB concentrations and congener patterns in soil, dust, residential indoor air, outdoor air, and sediment to investigate possible sources of PCBs found inside New Bedford area homes.

Experimental Methods

Yard soil and house dust samples were collected for 34 homes between April 1994 and April 1995 on days when harbor dredging was scheduled (Figure 1). These homes were recruited from five neighborhoods: three harbor neighborhoods (Acushnet, Fairhaven, and New Bedford Hot Spot) immediately downwind of the hot spot and Confined Disposal Facility for dredged sediments (based on prevailing winds during fair weather when warm temperatures induce the greatest amount of volatilization from sediments and water) and two comparison neighborhoods (Dartmouth and New Bedford Downtown) distant from the harbor.

On each sampling day, two homes were sampled: one in a harbor neighborhood and one in a comparison neighborhood. Because higher PCB concentrations have been measured in urban areas than in rural areas (*15*), rural Dartmouth homes were paired with rural Acushnet and Fairhaven homes and urban New Bedford Downtown homes were paired with urban New Bedford Hot Spot neighborhood homes (Figure 1).

In each home, one composite house dust sample was collected from the main foot traffic area on carpet or area rugs in the room used most often by residents. Dust samples were collected with the HVS3 high volume small surface sampler (HVS3) (CS3, Inc., Bend, OR) according to ASTM Standard D-5438-94 (*16*) and the manufacturer's instructions (*17*). In the laboratory, house dust samples were sieved into coarse and fine ($<150\ \mu$m) fractions.

Soil samples were collected from approximately the top 2 cm of soil at the main entryway or as close to the main

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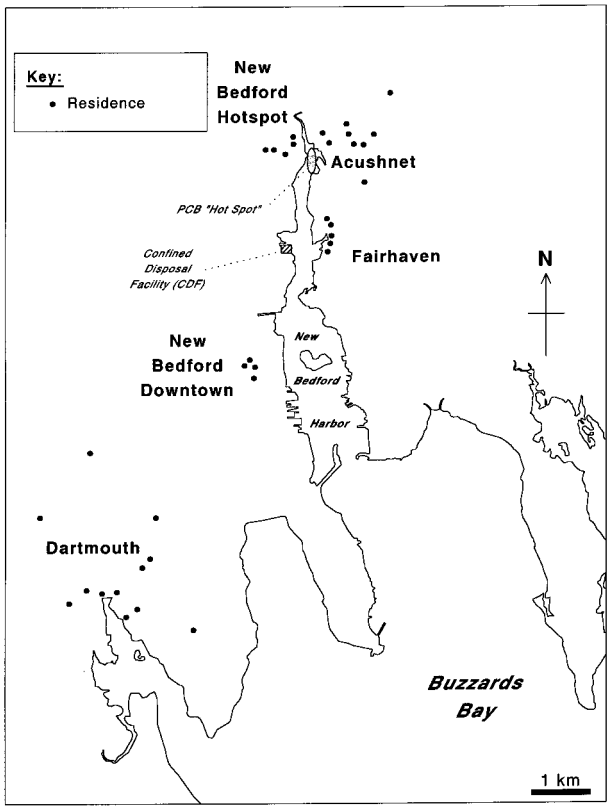

FIGURE 1. Residential sampling locations for house dust and yard soil.

entryway as possible using a hexane-rinsed stainless steel spatula and amber glass jars with PTFE-lined lids.

Homeowners responded to questionnaires with questions about general household information, the occupational histories of residents, and possible indoor sources of PCBs.

Organic Carbon Analysis. A Perkin-Elmer PE2400 Series II CHN/O analyzer was used to determine the amount of organic carbon in house dust and yard soil samples, which were dried overnight and crushed with a plastic pestle prior to analysis. Soil and dust replicates had average coefficients of variation equal to 6.2% and 1.7%, respectively. Results for a standard (BCSS-1 marine sediment, National Research Council of Canada) fell within the known range of 2.19 ± 0.09%.

Analytical Methods. All samples were stored at less than −20 °C prior to extraction. Samples were brought to room temperature and spiked with two surrogates, IUPAC no. 103 and IUPAC no. 112. Yard soil was extracted three times with 100 mL of hexane:acetone containing 60 g of sodium sulfate for 12 h, 2 h, and 30 min, respectively, on a mechanical shaker (250 rpm) overnight. Between extractions, samples were centrifuged for 2 min and the extract was decanted into a 250 mL Erlenmeyer flask through glass wool. House dust was sonicated for 5 min with 2 mL of methanol and sonicated three times for 30 min each with 25 mL of 10% diethyl ether in hexane. Sample extracts were reduced using a Kuderna−Danish apparatus and evaporated to 1 mL under a gentle stream of ultrahigh purity grade nitrogen. Extracts were cleaned by elution through a chromatographic column packed with anhydrous sodium sulfate, 3% deactivated silica gel (Scientific Adsorbents Inc., Atlanta, GA), and 2% deactivated aluminum oxide (J. T. Baker). Internal standard IUPAC no. 166 was added to the final volume of about 500 μL.

Extracts were analyzed on a Hewlett-Packard 5890 Series II gas chromatograph equipped with a ^{63}Ni electron capture detector and a 30 m, 0.25 mm i.d., 0.25 μm film thickness DB-5 capillary column (J&W Scientific, Folsom, CA). The following analytical conditions were used: splitless injection (2 μL), injector temperature, 280 °C; detector temperature, 320 °C; initial oven temperature, 60 °C; held 1 min, heated to 140 °C at 15 °C/min, then to 220 °C at 1 °C/min with a 40 min hold. The make-up gas was argon/methane (95:5) at 60 mL/min.

Calibration standards contained 65 target PCB congeners, surrogates, and the internal standard (AccuStandard, New Haven, CT, and Ultra-Scientific, North Kingstown, RI). Several peaks were quantified as individual target congeners but may contain a small contribution from coeluting congeners (i.e., IUPAC 138, 153, and 170). Method detection limits (MDLs) were calculated for each congener by multiplying the procedural blank standard deviation value for each congener by 3.

Quality Assurance and Quality Control. Average recoveries for surrogates 103 and 112 across all soil samples were 91 ± 11% and 87 ± 12%, respectively, and 99 ± 21% and 94 ± 10%, respectively, for dust samples. PCB concentrations were not corrected for surrogate recoveries. Six soil samples were split and analyzed separately to assess the laboratory error rate. Relative percent differences for six duplicates were 3.1, 4.8, 6.2, 6.8, 10.8, and 33. Split dust samples had relative percent differences of 3.0, 3.4, and 8.2. Target congener average recoveries were 88 ± 7% across 8 soil matrix spikes and 101 ± 8% across six dust matrix spikes. Confirmatory analysis of 7 dust samples was conducted using gas chromatography/mass spectrometry in selective ion monitoring mode using a Hewlett-Packard 5890 Series II gas chromatograph (relative percent differences ranged from 2 to 19%).

TABLE 1. Summary Statistics for House Dust and Yard Soil PCB Concentrations (ng/g, dry weight) in Harbor and Comparison Neighborhoods[a]

	geometric mean	geometric std dev	min	median	max
Harbor Neighborhoods					
house dust	1400	3.1	320	880	23000
(n = 19)	*8800*	*3.3*	*1700*	*7300*	*86000*
yard soil	200	3	23	200	1800
(n = 17)	*6900*	*2.7*	*1300*	*6400*	*33000*
Comparison Neighborhoods					
house dust	690	2	260	710	3600
(n = 15)	*3700*	*2.3*	*920*	*3200*	*23000*
yard soil	60	2.3	15	62	290
(n = 16)	*2000*	*2.2*	*370*	*1900*	*6900*

[a] Values in italics are concentrations normalized to the amount of organic carbon present in soil and dust (ng/g organic carbon).

Results

House Dust and Yard Soil Concentrations. Table 1 includes summary statistics for house dust and yard soil PCB concentrations, based on the sum of individual congener concentrations. Concentrations in yard soil from homes in harbor neighborhoods are significantly higher than in comparison neighborhoods (two-sided $t = -2.16$, $p = 0.014$). In contrast, concentrations in house dust did not differ significantly between these two neighborhood groups (two-sided $t = 1.50$, $p = 0.14$).

In all neighborhoods, concentrations in house dust exceed those in yard soil by approximately 1 order of magnitude (two-sided $t = 2.03$, $p = 0.013$). However, concentrations in dust were only slightly higher than those in soil after being normalized to organic carbon content (3 ± 1% and 4 ± 2% in harbor and comparison neighborhood soil, respectively; 19.5 ± 6.3% and 19.3 ± 6.0% in harbor and comparison neighborhood dust, respectively) (Table 1). Furthermore, the

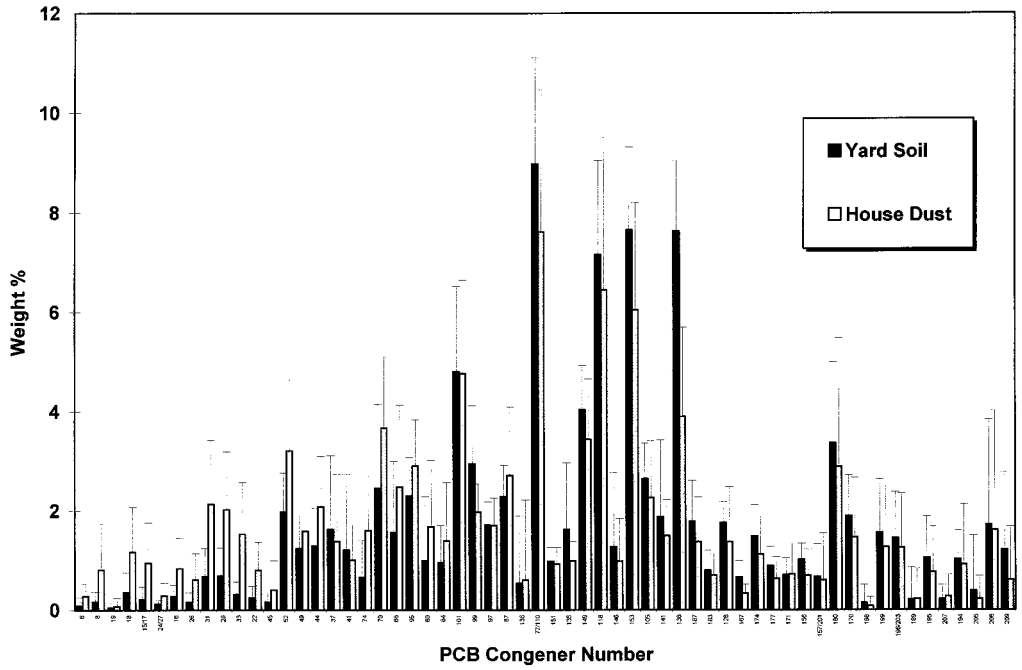

FIGURE 2. Average house dust and yard soil PCB congener patterns for all neighborhoods.

difference in dust and soil concentrations might be explained, in part, by sieving of dust samples prior to analysis. With a higher surface area to mass ratio than larger particles, these smaller particles might contain higher concentrations of PCBs. Soil samples were not sieved because PCBs likely would have been lost while drying soil samples in preparation for sieving.

For all but three dust samples, only the fine fraction (<150 μm) was analyzed because of its relevance to human exposure (*18*). PCB concentrations in fine dust fractions exceeded those in coarse dust fractions in two of three comparisons (12 000 vs 9300; 530 vs 550; and 260 vs 130 ng/g). The coarse dust fractions (>150 μm) comprised, on average, 20% of total sample weight and contained sand, paint chips, pebbles, plant matter, hair, carpet fiber, and foam padding.

Influences on Dust PCB Concentration. Four residents reported occupational exposure to PCBs, but all exposures ended at least 13 years before sample collection. All four residents lived in harbor neighborhoods. Although the highest concentrations in dust were measured in two of these homes, concentrations in dust and soil in homes with occupationally exposed residents were slightly different from those in other homes based on a two-sided Wilcoxon rank sum test (p = 0.052).

In a study of pesticides, old carpets tended to have more pesticides than newer carpets (*6*). In this study, the highest concentration in house dust, 23 ppm, was collected from a 30-year old carpet, which was the oldest carpet sampled. This concentration exceeds the next highest dust concentration measured in this study by about a factor of 3 (23 ppm vs 7.9 ppm). The correlation coefficient for house dust PCB concentration and carpet age is significant (Spearman's rank correlation coefficient = 0.35, p = 0.05). However, after excluding the home with 23 ppm PCBs in house dust, dust PCB concentrations were not correlated with carpet age (Spearman's rank correlation coefficient = 0.28, p = 0.27 for harbor neighborhoods; Spearman's rank correlation coefficient = 0.12, p = 0.66 for comparison neighborhoods).

No significant correlation was calculated between concentrations in house dust and the number of potential indoor PCB sources across all neighborhoods (Spearman's rank correlation coefficient = −0.18, p = 0.29). Indoor sources were defined as electrical appliances and fluorescent lights more than 10 years old. This time period assumes that such PCB-containing products were available for sale several years following the decline in PCB manufacturing during the 1970s.

Yard Soil and House Dust PCB Congener Patterns. Average PCB congener patterns in soil and dust are similar, regardless of neighborhood or occupational exposure, although dust appears to contain a slightly higher proportion of more volatile congeners (Figure 2). We used principal component analysis (PCA) to examine congener pattern differences among neighborhoods. No difference was observed for house dust. However, PCA results suggest a slight pattern difference between yard soil in harbor and comparison neighborhoods, with 55% of the variance in the underlying data explained (Figure 3a). Standardized congener weight percent values were used in the PCA. Removal of the two points with large negative loadings on PC2 did not otherwise change interpretation of the PCA analysis.

Most harbor and comparison neighborhood yard soil samples are divided along PC1. Harbor samples have low PC1 loadings, and so do several relatively volatile congeners (IUPAC numbers 49, 70, 66, 95, 101, 99, 77/110, 118, and 105) (Figure 3b). Along PC2, harbor and comparison samples are divided, again with a higher proportion of more volatile congeners associated with harbor neighborhoods (Figure 3c). Because only daily paired data were entered into the PCA, variations in dredging activity or weather conditions do not explain this pattern difference.

Discussion

Carpet dust is not the only kind of residential dust, but it may represent the largest repository integrating exposure over a long period. If dust collected with the HVS3 represents what people are exposed to during normal activities around the home, this study shows the importance of considering exposure to dust in addition to soil, which is routinely evaluated in human health risk assessments.

Comparison to Other Dust PCB Concentration Data. PCBs have been measured in house dust in nine Seattle, WA, homes (240 to 760 ng/g) and eight Columbus, OH, homes (210 to 1900 ng/g) (*3*, *19*). These homes were not reported to be located near known PCB sources as were the homes in this study, perhaps explaining why concentrations in these

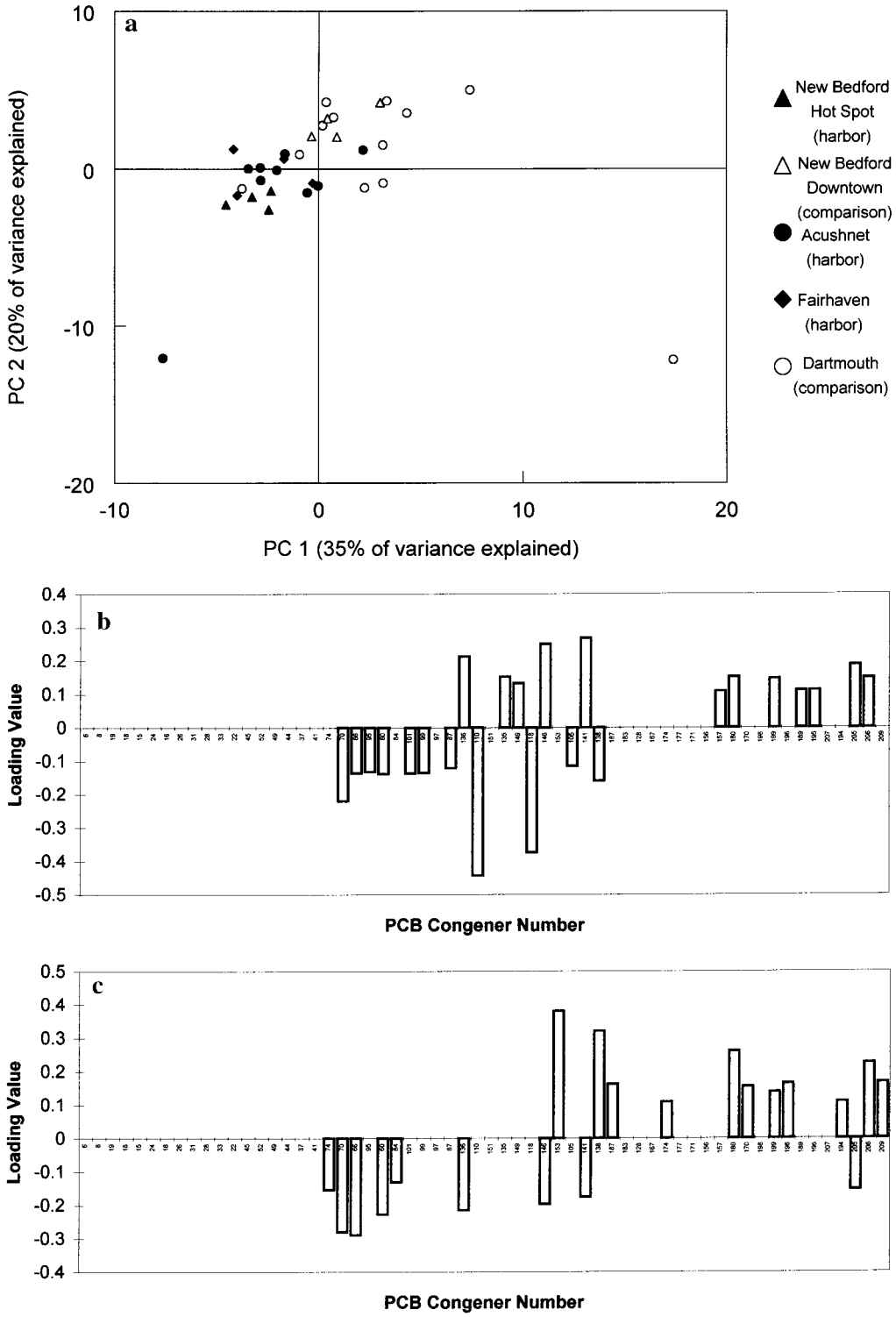

FIGURE 3. (a) Principal components analysis (PCA) score plot for yard soil. Standardized congener weight percent values were used. The percent of variance in the underlying data that is explained by each principal component is indicated on the axes. (b) Loadings for principal component 1. (c) Loadings for principal component 2.

homes are generally lower than those measured in this study (260 to 23 000 ng/g). Yard soil samples also were collected from the Seattle homes (58 to 240 ng/g). The difference between soil and dust concentrations measured in these homes is similar to the difference observed in this study.

Dermal Bioavailability. To estimate risk resulting from dermal exposure to PCBs in soil and dust, one must consider more than PCB concentration. Several factors can influence how much soil or dust adheres to skin, including moisture content, particle size, and perhaps organic carbon content (*18*, *20*−*26*). PCB absorption through the skin might decrease with increasing organic carbon content of soil and dust (*20*). Even if this is true, we detected higher PCB concentrations in dust than in soil after adjusting for organic carbon content, although this difference might not be as great if we were able to collect the $<150\,\mu$m fraction of soil samples as we did with dust samples.

Track-In of PCBs from Yard Soil. Track-in of soil to the indoor environment may occur when outdoor contaminants attach to shoes or clothing worn indoors. Chuang et al.

TABLE 2. Spearman's Rank Correlation Coefficients for the Sum of PCB Congener Concentrations in Residential Media

comparison	neighborhood	Spearman's rank correlation coeff, *r*	probability
house dust vs yard soil	all	0.28	0.1186
	harbor	0.24	0.3391
	comparison	0.21	0.4219
house dust vs indoor air	all	0.62	0.0003[a]
	harbor	0.56	0.0202[a]
	comparison	0.67	0.0092[a]
yard soil vs outdoor air	all	0.46	0.0118[a]
	harbor	0.5	0.0623
	comparison	0.07	0.7802

[a] Correlation analysis is significant at the $\alpha = 0.05$ level.

(*4*, *19*) detected evidence for PAH and PCB track-in, with significant correlation coefficients between house dust and entryway soil concentrations. Soil and dust concentrations in harbor and comparison neighborhoods are not correlated (Table 2). In the PAH study, Chuang et al. (*4*) provided homeowners with doormats from which samples were collected. We collected soil near the entryway, but we elected not to alter the home environment by providing subjects with doormats. Instead, our soil samples are more like "pathway soil" described in Chuang et al. (*4*, *19*) that was not correlated with house dust. The lack of correlation may reflect the fact that individuals could track contaminants indoors from many locations other than their yards. Alternatively, the PCB concentration gradient moving indoors to outdoors might indicate that indoor sources are important, perhaps overshadowing any PCB track-in. Indoor sources could include PCB-containing products or indoor sinks containing and re-emitting PCBs tracked into the home over many years.

Comparison to Indoor Air and Outdoor Air PCB Concentrations. Unlike soil and dust concentrations, indoor air and dust concentrations are significantly correlated in all neighborhoods (Table 2). Indoor air and outdoor air samples were collected at the same time soil and dust samples were collected, and these data are described in detail elsewhere (*13*). The correlation between indoor air and house dust is logical given the exchange that occurs between these media via deposition and re-suspension processes. Weak correlations exist for yard soil and outdoor air.

PCB Congener Patterns and Potential Sources. This study suggests a difference in congener pattern between soil from harbor and comparison neighborhoods, but not dust. Harbor neighborhood soils contain a slightly higher proportion of more volatile congeners and slightly higher overall PCB concentrations. In a previous study of these homes, a similar difference in both congener pattern and concentration was reported for outdoor air in harbor and comparison neighborhoods, but not indoor air (*13*). These results are consistent with volatilization of lighter congeners from harbor sediments and waters, impacting outdoor air and soil to a greater degree than indoor air and dust.

To reduce residential exposure to PCBs, it would be useful to identify PCB sources. Pattern and concentration differences suggest some harbor influence, but other sources may exist. Unfortunately, source apportionment of PCBs in the residential environment is made difficult by a number of factors. First, the principal Aroclor mixtures disposed of in New Bedford Harbor were likely Aroclor 1242 and Aroclor 1254 and perhaps some Aroclor 1016 (*27*, *28*). These same Aroclors were used in the manufacture of adhesives, small capacitors, fluorescent light ballasts, and other products that might be found inside homes (*9*, *10*). Second, congener patterns in dust, soil, and air do not necessarily match these Aroclor mixtures, because the physical and chemical properties that

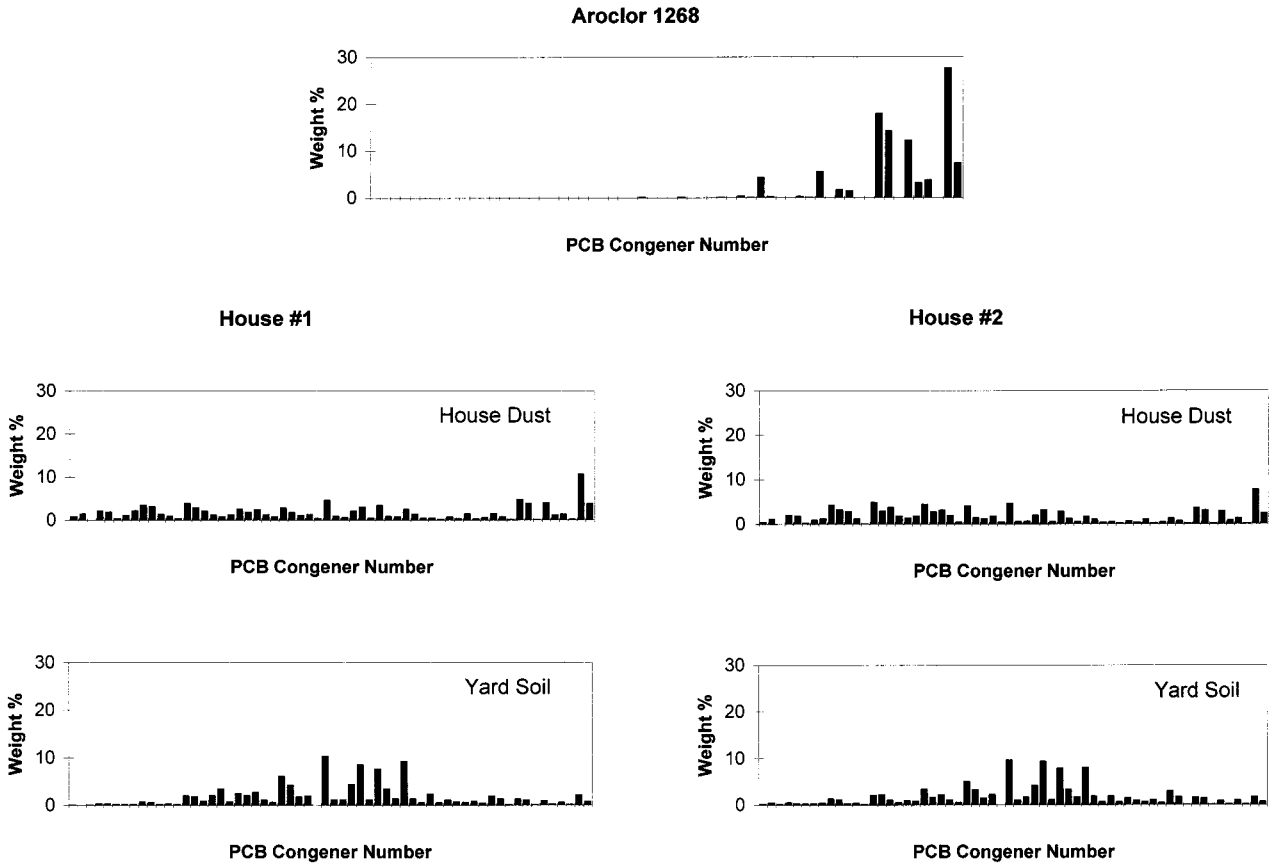

FIGURE 4. Aroclor 1268 congener pattern compared with congener patterns in house dust and yard soil from two residences.

affect partitioning, persistence, and bioaccumulation differ among congeners (*29*, *30*). Third, PCB degradation processes may differ among environmental media.

Despite these limitations, two dust sample congener patterns provide evidence that some PCBs might originate from a source other than the harbor. Figure 4 compares the congener pattern of Aroclor 1268 to patterns in dust and soil samples collected from two neighboring homes, built at the same time. A portion of the dust pattern matches the Aroclor 1268 pattern (Figure 4). This pattern was not observed in other dust samples or in any soil samples. Aroclor 1268 was not generally used in the production of capacitors (*9*, *10*); therefore, it probably was not used by the capacitor manufacturing companies in New Bedford. However, Aroclor 1268 may be present in building materials, such as sealants and ceiling tiles (*9*, *10*). The soil samples do not reflect this pattern, although weathering processes in soil likely differ from such processes in house dust. If this Aroclor was present in harbor sediments at some point, it may have been dechlorinated to produce the current sediment congener pattern (*31*, *32*). However, using New Bedford Harbor sediment congener-specific concentration data, Brown et al. (*28*) deduced that historic contamination of the sediments most likely includes Aroclors 1242 and 1254. Therefore, this pattern in dust probably originated from a source other than harbor sediments.

This study highlights the importance of considering residential PCB concentrations, whether or not one lives near a Superfund site. Decisions about harbor cleanup should consider the contribution of alternative remediation techniques to reducing total human exposure to PCBs because, regardless of harbor cleanup efforts, indoor sources of PCBs may persist.

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