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Exposure to Polychlorinated Biphenyls in Residential Indoor Air and Outdoor Air near a Superfund Site

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Polychlorinated biphenyl (PCB) concentrations were measured in indoor and outdoor air at 34 homes surrounding New Bedford Harbor during dredging of highly contaminated harbor sediments. PCB volatilization from harbor sediments and water leads to inhalation exposure for nearby residents. On each sampling day, 24 h indoor and outdoor air samples were collected simultaneously and analyzed for 65 PCB congeners to evaluate the relative importance of the harbor and indoor sources for human inhalation exposure. Outdoor air concentrations were higher in neighborhoods closest to the harbor (0.4–53 ng/m^3) relative to comparison neighborhoods (0.1–8.2 ng/m^3) and contained slightly greater proportions of volatile PCB congeners. Outdoor air concentrations near the harbor are some of the highest measured in recent years. Indoor air concentrations in homes near the most contaminated part of the harbor (7.9–61 ng/m^3) were slightly higher than concentrations in homes distant from this area (5.2–51 ng/m^3). In all neighborhoods, indoor concentrations exceed corresponding outdoor concentrations (mean ratio = 32), suggesting the importance of indoor PCB sources even near a highly contaminated waste site.

Introduction

PCBs were used in New Bedford, MA, in the production of capacitors from the 1940s until the late 1970s, and their disposal during this period resulted in contamination of harbor sediments and closure of the harbor to fishing. The harbor was designated a Superfund site in 1981, in part, because of the health risk to nearby residents given the potential estrogenic, carcinogenic, and immunotoxic effects of PCBs (*1–5*). In the most contaminated part of the harbor, referred to as the "hot spot", sediment PCB concentrations as high as 100 000 ppm have been measured (*6*). This study was conducted while hot spot sediments were dredged and piped to a confined disposal facility (CDF) along the western shoreline of the harbor about 1.5 km south of dredging activity.

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People living near New Bedford Harbor may be expos to PCBs emanating from contaminated harbor sediments a waters, whether or not dredging is occurring. Our aim w to assess indoor air and outdoor air PCB concentrations duri disturbance of these highly contaminated sediments. Becau PCBs are ubiquitous in the environment (*7–10*), we sampl concurrently in neighborhoods upwind of and at sor distance from the harbor to discern the portion of P contamination attributable to harbor proximity. While PC have been measured in outdoor air near waste disposal si (*11–14*), this study is among the first to include the indo environment.

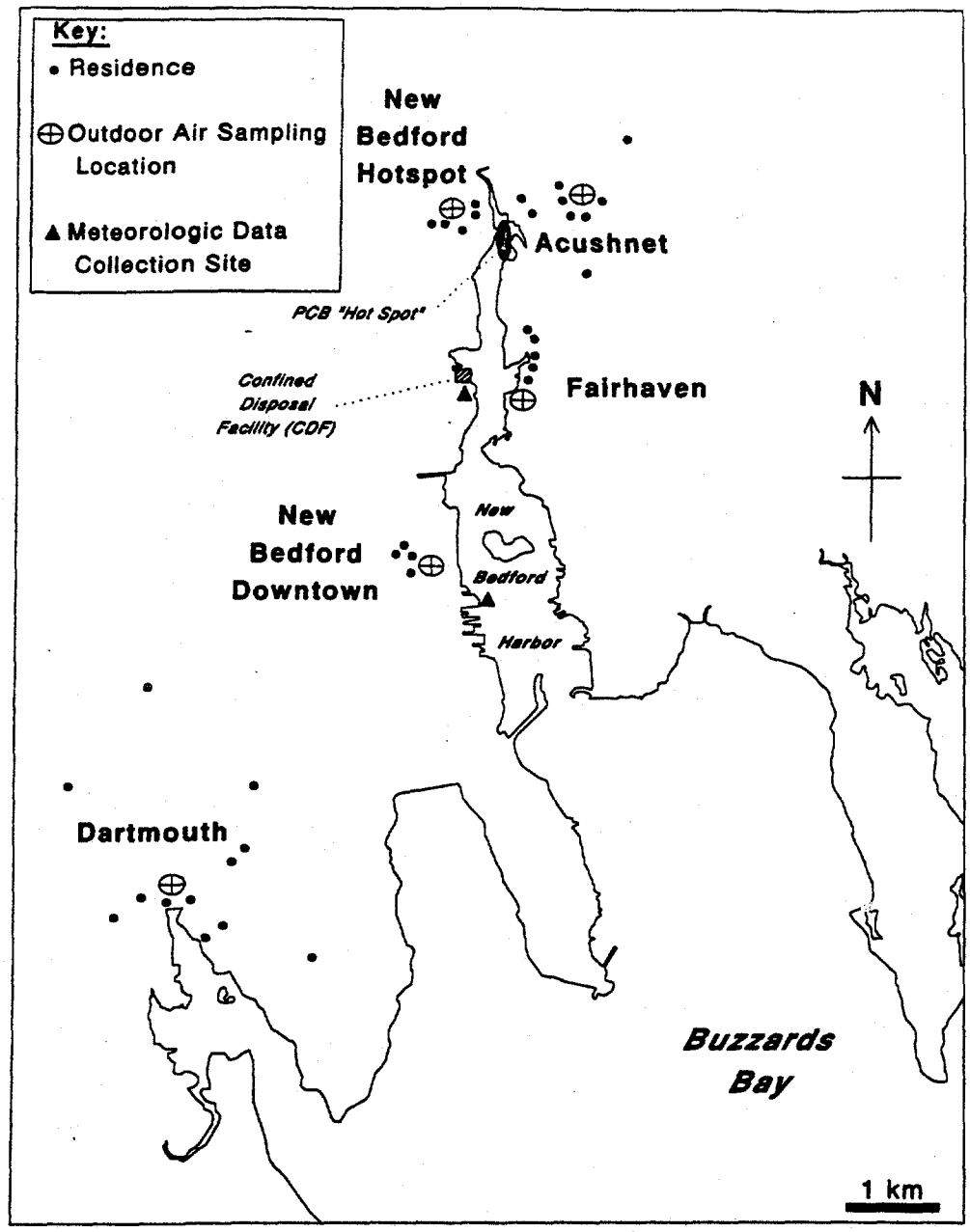
In addition to outdoor sources such as the harbor, the are smaller indoor sources of PCBs (*15–20*). Concurre indoor air and outdoor air sampling was conducted to explo the relative magnitude of indoor and outdoor source co tributions to human exposure. Despite the 20 year ban production of PCBs, many long-lasting household produ may contain PCBs, such as, wood product coatings, plastic ers in paints, sealants, flame retardants, plastics, fluoresce light ballasts, and small electrical capacitors in applianc (*21, 22*). Little research has been devoted to examini residential inhalation exposure to PCBs (*15*), even thou people spend the bulk of their time indoors (*23, 24*). Wh food is thought to be the most important human exposu pathway for PCBs, the Agency for Toxic Substances a Disease Registry (*25*) has suggested that inhalation inta may exceed ingestion intake due to a steady decrease in dieta PCB concentrations since 1978. This research is part o larger multimedia PCB exposure study that, in addition indoor air and outdoor air, estimates exposure to house du yard soil, tap water, and local produce collected near N Bedford Harbor (*26, 27*).

Experimental Methods

Indoor and outdoor air samples were collected for 34 hom between April 1994 and April 1995 on days when hart dredging was scheduled (Figure 1). These homes w recruited from five neighborhoods: three harbor neighb hoods (Acushnet, Fairhaven, and New Bedford Hot Sp immediately downwind of the hot spot and CDF (based prevailing winds during fair weather when warm temperatu induce the greatest amount of volatilization from sedime and water) and two comparison neighborhoods (Dartmou and New Bedford Downtown).

On each sampling day, two homes were sampled: o harbor neighborhood home and one comparison neighb hood home. Because higher PCB concentrations have be measured historically in urban areas than in rural areas rural Dartmouth homes were paired with rural Acushnet a Fairhaven homes and urban New Bedford Downtown hom were paired with urban New Bedford Hot Spot neighborho homes. Outdoor air samples were collected concurrently fr a central site in each neighborhood (Figure 1).

Outdoor air samples consisted of 200–300 m^3 of collected over 24 h using high-volume samplers (Grasel GMW, OH) operated at flow rates between 0.19 and 0.23 m min. Samplers were calibrated at the beginning and end the study, and after each move to a new location. T sampling module held a Whatman quartz fiber filter (10 cm diameter) followed by two polyurethane foam (PUF) pl (5.5 cm diameter, 7.6 cm length and 5.5 cm diameter, 2.5 length, respectively; density of 0.022 gm/cm^3). The secc PUF was used to check for any sample breakthrough. Filt were cleaned prior to sampling by heating overnight at °C. PUFs were cleaned by three consecutive 24 h Soxl extractions with hexane.



IGURE 1. Sampling locations for outdoor air, indoor air, and meteorological data.

Outdoor air samplers were located away from known PCB ources, air conditioners, vents, heaters, and walls. Field lanks were collected by assembling a sampling module and ransporting it to the field along with the other sampling iodules. Sampling modules were prepared and unloaded 1 the laboratory.

Indoor air samples were collected in the room used most ften by residents with the sampler inlet 1.5 m above the oor. Sampling modules identical to outdoor air sampler iodules and a vacuum pump were used to collect 24 h air amples at a flow rate of 5 or 10 L/min. Samplers were located t least 0.75 m away from walls or internal doors, vents, air onditioners, space heaters, and windows. Homeowners ecorded activities during the sampling period that could ffect air quality such as cleaning and opening of windows nd doors.

Analytical Methods. All samples were stored at less than -20 °C prior to extraction. PUFs were brought to room emperature, spiked with two surrogates, IUPAC no.103 and UPAC no.112, and extracted with hexane in a Soxhlet pparatus for 24 h. Sample extracts were reduced using Kuderna–Danish apparatus, then evaporated to 1 mL at room temperature under a gentle stream of ultrahigh purity grade nitrogen. They 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, which ranged from 200 μL to 3 mL.

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 DB5 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. Helium was used for the carrier gas at 1 mL/min. 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

TABLE 1. Summary Statistics for Indoor Air and Outdoor Air PCB Concentrations (ng/m³) in Harbor and Comparison Neighborhoods

		geometric mean	geometric standard deviation	minimum	maximum
harbor neighborhoods	indoor air (n = 18)	18	1.8	7.9	61
	outdoor air (n = 20)	4.9	4.6	0.4	53
comparison neighborhoods	indoor air (n = 16)	10	1.8	5.2	51
	outdoor air (n = 20)	0.6	3.3	0.1	8.2

peaks were quantified as individual 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 field blank standard deviation value for each congener by 3. Sample concentrations were corrected using corresponding field blanks on a congener-by-congener basis.

Reported PCB concentrations are based only on analysis of PUFs. Prefilters were analyzed for a subset of indoor and outdoor air samples collected over the range of ambient temperatures during the study. They contained, on average, 0.8 and 5.6% of the amount of PCBs collected on PUFs for outdoor air and indoor air samples, respectively.

Quality Control and Quality Assurance. Duplicate indoor air samples were collected in 60% of the homes, with relative percent differences for the sum of PCB congener concentrations ranging from 0.04 to 35%, with a median value of 8%. Average recoveries for individual target congeners across 16 spiked PUFs ranged from 83 to 98%. Average recoveries for surrogates 103 and 112 across all air samples and quality control samples were 93 ± 22 and 95 ± 11%, respectively. PCB concentrations were not corrected for surrogate recoveries. Average field blank congener mass was 12% (range, 0–45%) of indoor air sample mass and 6.5% (range, 0–48%) of outdoor air sample mass.

To determine if PCB concentration estimates would differ using the indoor or outdoor sampler, these samplers were collocated outdoors for two 24 h periods. Concentration estimates for the sum of PCB congeners from both samplers compared favorably, with relative percent differences of 2.2 and 6.1%.

Breakthrough checks resembled procedural blanks, except for three samples collected on the hottest sampling days (30 and 20% breakthrough of congeners 6 and 8, respectively). Overall, PCBs lost to breakthrough represent only 1 to 4% of the sum of PCB congener concentrations and are not included in the reported PCB concentration results.

Results

Indoor Air and Outdoor Air PCB Concentrations. Table 1 includes summary statistics for indoor and outdoor air PCB concentrations in harbor and comparison homes, based on the sum of individual congener concentrations. While samples were collected in all four seasons, the summary statistics do not represent annual averages because sampling events were not equally distributed over the one-year sampling period.

In presenting concentration results, the three neighborhoods near the hot spot and CDF are combined in the category harbor neighborhoods and the two comparison neighborhoods are combined. Because air samples were collected from one home and one outdoor location from each of these two neighborhood groups on each sampling day, variation in meteorological conditions does not affect comparison of these two groups.

Indoor air concentrations in harbor homes exceed those in comparison homes (two-sided Wilcoxon rank sum test, p = 0.002). Outdoor air concentrations are higher in harbor neighborhoods than in comparison neighborhoods in 15 out of 19 comparisons, with geometric means of 4.9 ng/m³ and 0.6 ng/m³, respectively (two-sided Wilcoxon rank sum test, p < 0.001). Little or no difference is seen between these neighborhoods in colder months when less PCB volatilization is expected from outdoor sources.

Figure 2 depicts daily indoor and corresponding outdoor air PCB concentrations for harbor and comparison homes. With few exceptions, indoor concentrations exceed outdoor concentrations, regardless of neighborhood. In winter, indoor air to outdoor air concentration ratios increase, probably because PCB volatilization from outdoor sources decreases with colder outdoor temperatures, while indoor temperatures remain relatively constant throughout the year. Indoor to outdoor air concentration ratios are smaller for harbor homes than for comparison homes. Indoor PCB concentrations were not found to be significantly correlated with outdoor PCB concentrations, regardless of neighborhood.

Influences on Indoor Air PCB Concentration. While indoor air PCB concentrations appear to be affected by neighborhood location, other factors may be influential. Correlation analyses were performed between indoor air PCB concentration and number of potential indoor sources, residents' occupational exposure to PCBs, house age, carpet age, and number of open windows during sampling.

No significant correlation was found between indoor air concentration and the number of potential indoor PCB sources (i.e., 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 1977 ban on PCB manufacture. Despite finding no correlation between PCB concentrations and potential indoor sources, we noted that the comparison home with the highest PCB concentration (52 ng/m³) reported 11 potential indoor sources.

Three residents with occupational exposure to PCBs lived in harbor neighborhoods. One was currently employed in a PCB-using industry, one had not worked for 13 years, and one had not worked for 30 years. After excluding these three homes, harbor and comparison indoor air concentrations were still significantly different (p value = 0.008).

Houses ranged from 8 to 164 years old. No correlation was found between indoor air PCB concentration and house age, even after excluding homes built prior to the year PCBs were first manufactured (1929) and after the year PCB manufacture was banned (1977). No correlation was evident between indoor air concentration and carpet age or number of windows left open during the sampling period.

To assess short-term variation in concentrations, 24 indoor air and outdoor air samples were collected on three consecutive days for one home in a harbor neighborhood. While no general conclusions can be derived from one short-term study of one home, PCB congener patterns and concentrations (59, 60, and 61 ng/m³) did not vary.

Six homes were sampled in two different seasons. However, little variation in congener pattern or concentration was apparent with season, although the sample size was small.

PCB Congener Patterns in Indoor Air and Outdoor Air. Average congener patterns in indoor air and outdoor air differ with a smaller proportion of low molecular weight congeners in indoor air compared to outdoor air (Figure 3). We plotted outdoor air congener patterns for harbor and comparison neighborhoods to observe any pattern differences due to neighborhood. Because only daily paired data were used

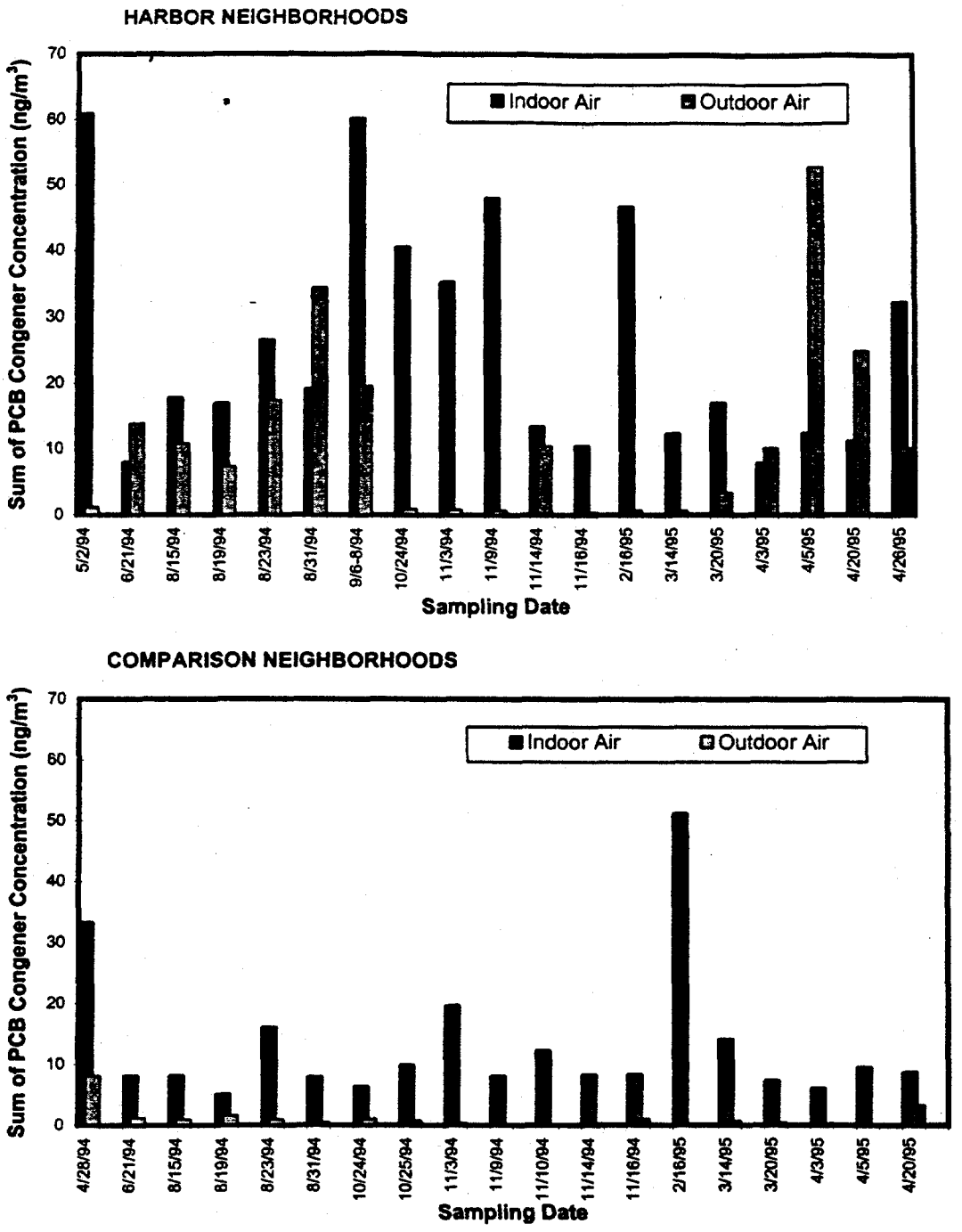


FIGURE 2. PCB concentrations in indoor air and outdoor air collected simultaneously for homes in harbor and comparison neighborhoods.

calculate average congener patterns, variations in dredging activity or weather conditions do not explain any pattern difference. Two harbor neighborhood (Fairhaven and Acushnet) patterns differ from the corresponding comparison neighborhood (Dartmouth) pattern, with a slightly higher proportion of the more volatile congeners. See Figure 4 for an example of this pattern difference. Outdoor air samples collected in one harbor neighborhood (New Bedford Hot Spot) did not differ from the corresponding comparison neighborhood (New Bedford Downtown).

The same plots of indoor air congener patterns suggest no differences among neighborhoods.

Discussion

The results of this study show that, during dredging of harbor sediments, PCB concentrations in harbor neighborhood outdoor air exceed comparison neighborhood concentrations by about a factor of 8. Harbor neighborhood indoor air concentrations are approximately 1.5 times the concentrations found in comparison neighborhood homes. Regardless of neighborhood, indoor air concentrations exceed outdoor air concentrations in all but 5 of 38 comparisons. What is less certain are the sources of PCBs and their relative contribution to indoor and outdoor air concentrations.

PCB Sources. In addition to PCBs entering homes during dredging, PCBs transported from the harbor and neighboring industries over time may have accumulated in indoor sinks that are now re-emitting PCBs. A large portion of indoor PCB concentrations probably originates from indoor sources given that indoor concentrations almost always exceed outdoor concentrations and given the lack of correlation between indoor air and outdoor air concentrations.

Indoor sources of PCBs have been characterized in only a few cases, including sealants and fluorescent light ballasts

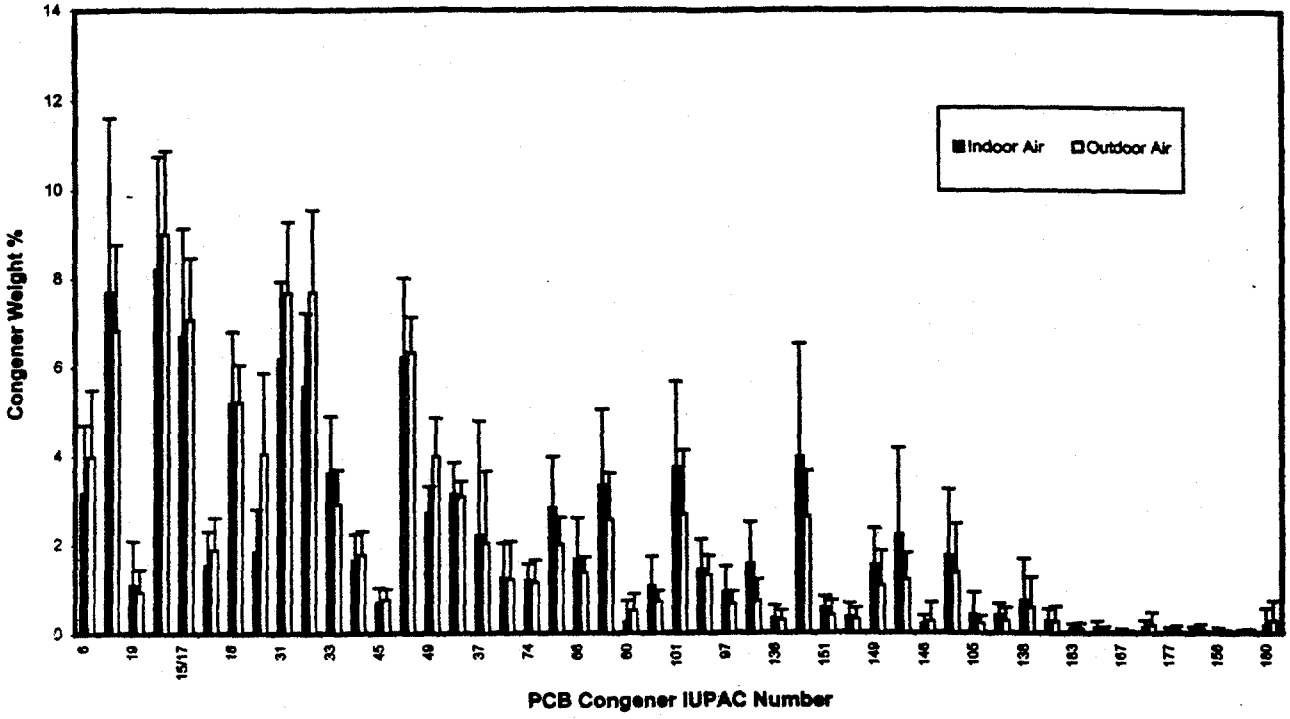


FIGURE 3. Average indoor air and outdoor air PCB congener patterns for all neighborhoods.

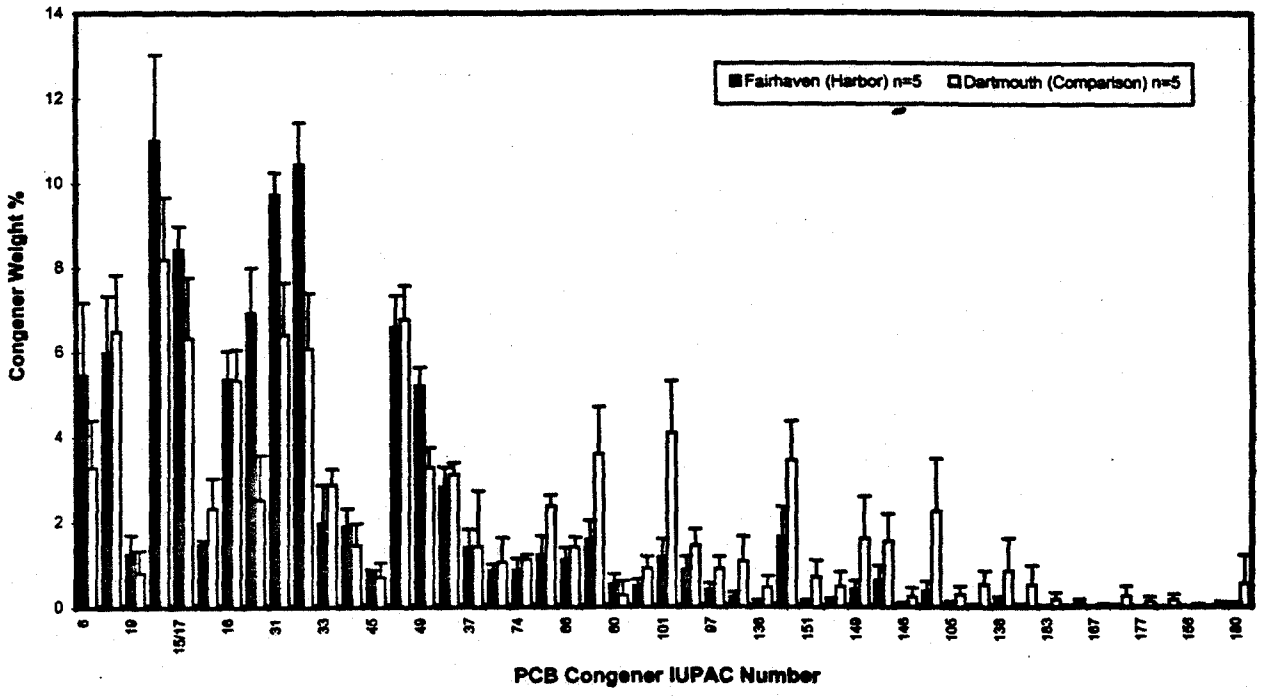


FIGURE 4. Comparison of average PCB congener patterns in outdoor air collected simultaneously in a harbor neighborhood (Fairhav and a comparison neighborhood (Dartmouth). Error bars represent one standard deviation.

(*17, 18*). New Bedford Harbor sediments, on the other hand, have been well characterized (*28, 29*). Source apportionment efforts are hindered by PCB partitioning and aging in the environment and the fact that commercial congener mixtures disposed of in the harbor also were used in the manufacture of products that might be found in residences (e.g., paints, sealants, small capacitors, and fluorescent light ballasts) (*30*).

Despite these barriers to apportioning PCB sources, the trend of "heavier" congener patterns (i.e., a higher proportion of high molecular weight PCB congeners) in indoor air than outdoor air suggests indoor sources, especially given higher indoor air concentrations than outdoor air concentrations. In fact, the two highest indoor air concentrations measured in this study also exhibit the heaviest congener pattern. O of these homes has a resident occupationally exposed to PC although that exposure occurred 30 years ago. The heav indoor air pattern is found in both harbor and comparis homes and in homes of heavy smokers and nonsmokers. P patterns on prefilters for these homes did not differ fr patterns for prefilters from other homes, so their exclus from congener pattern calculations does not explain pattern difference. No source or activity during sampl could be identified that could affect the congener patt

Potential Influence of Harbor Contamination on O door Air Congener Patterns. The congener pattern differe observed between outdoor air in harbor and compari

neighborhoods suggests some harbor influence. The higher proportion of lighter congeners in harbor neighborhood air may be the result of PCBs volatilizing from harbor sediment and water. Some investigators have shown that the rate of volatilization from fresh surface water to air is greater for the less chlorinated congeners (*31*, *32*). Interestingly, tomato samples collected in a harbor neighborhood during dredging show this same enrichment of the more volatile congeners (18, 28, 31, 49, and 70) relative to tomatoes grown in the same season in a comparison neighborhood (*26*). No such pattern difference was apparent in tomatoes collected from these neighborhoods prior to dredging. Outdoor air congener patterns in the New Bedford Hot Spot and the New Bedford Downtown neighborhoods were similar, perhaps because the latter neighborhood is closer to the harbor than the Dartmouth comparison neighborhood.

Comparison of PCB Concentrations with Recent Findings from Other Studies. MacLeod (*15*) measured indoor air PCB concentrations in nine North Carolina residences that, on average, were 10 times higher than outdoor air concentrations. While we detected a similar average ratio of indoor to outdoor air concentration, our highest indoor measurement was 61 ng/m^3 in contrast with 580 ng/m^3 measured by MacLeod (*15*). The use of different laboratory analytic and PCB quantification techniques over time complicates such comparisons. However, we simply may be observing a general decline over the past 15 years as the number of possible PCB sources in homes decreases.

There are still cases where indoor air levels may reach and exceed these 1981 levels. Ballfanz et al. (*18*) measured indoor air levels as high as 7500 ng/m^3 in buildings containing permanently elastic sealant composed of about 40% PCBs by weight. Wallace et al. (*19*) recently measured levels as high as 300 ng/m^3 in public buildings in Indiana. Oatman and Roy (*16*) found mean PCB concentrations of 460 ng/m^3 in office and school buildings containing transformers compared with 230 ng/m^3 in buildings not using these transformers. The homes in our study contained no transformers or other known significant source such as the sealant detected in Ballfanz et al. (*18*).

Gas-phase outdoor air PCB levels have been reported for many locations. Hermanson and Hites (*12*) measured gas phase PCB levels at three sites within 14 km of three landfills in the Bloomington, IN region that were known to be contaminated with PCBs. They measured average levels between 1.7 and 3.8 ng/m^3 in summer and 0.27 to 0.58 ng/m^3 in winter. A follow-up study conducted 6 years later (*33*) detected no concentration change. More recently, Leister and Baker (*34*) detected PCB concentrations ranging from 0.02 to 0.51 ng/m^3 over Chesapeake Bay, while Manchester-Neesvig and Andren (*35*) measured a maximum PCB concentration of 1.8 ng/m^3 in summer and a minimum of 0.14 ng/m^3 in winter over the Great Lakes watershed. Hoff et al. (*9*) found PCB levels ranging from 0.55 to 0.82 ng/m^3 over rural Ontario, Canada. Panshin and Hites (*36*) detected an average annual PCB concentration of 0.38 ng/m^3 in Burmuda. In comparing these studies, one must consider the influence of different analytic and quantification techniques as well as outdoor air temperature and wind direction relative to known sources during sample collection. Despite these uncertainties, summer New Bedford Harbor neighborhood outdoor air PCB concentrations are among the highest levels measured anywhere in recent years.

This study demonstrates that PCBs persist in residences despite the 20 year ban on PCB production, whether or not they are located near a significant outdoor PCB source. While measurements in homes near the harbor reflect exposure conditions during dredging, measurements inside comparison homes may be similar to background PCB exposure in other northeastern U.S. housing. For this larger group of residences, this study reinforces the importance of including the indoor environment in assessing exposure to environmental contaminants.

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