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Influence of Harbor Contamination on the Level and Composition of Polychlorinated Biphenyls in Produce in Greater New Bedford, Massachusetts

ALISON C. CULLEN,*,†,‡
DONNA J. VORHEES,† AND
LARISA M. ALTSHUL†

Department of Environmental Health, Harvard School of Public Health, Boston, Massachusetts 02115, and University of Washington, Seattle, Washington 98195-3055

Samples of produce (tomatoes, potatoes, carrots, and lettuce) from the area surrounding the New Bedford Harbor Superfund site were analyzed for the concentration and distribution of 47 polychlorinated biphenyl (PCB) congeners. Samples of produce from out of state also were analyzed for comparison purposes. New Bedford Harbor sediments are highly contaminated with PCBs, which may partition into the seawater and ultimately into air. During low tide, direct volatilization of PCBs is likely since the sediment is in contact with air. Also, sediment dredging, from the spring of 1994 until the fall of 1995, daily exposed fresh layers of contaminated sediment. Overall levels of PCBs in produce are within the range observed at local and out-of-state background sites, with the exception of greatly elevated levels in tomatoes grown during harbor dredging. Spatial and temporal differences in PCB concentrations and congener profiles indicate the effect of the harbor and other sources of environmental contamination. Our results are consistent with the view that atmospheric transport and gas-phase transfer play a pivotal role in influencing the concentration of PCBs in plant tissue. This work is an initial step toward gauging the significance of the consumption of local produce as a pathway of human exposure to PCBs in New Bedford before and during harbor dredging.

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Introduction

The use of polychlorinated biphenyls (PCBs) by industrial facilities in New Bedford, MA, and their disposal in the local harbor between World War II and the 1970s resulted in severe contamination, up to 15% PCBs by weight in sediment. Prior to 1977, a range of commercial Aroclors including 1016, 1242, and 1254 was used at these plants (*1*). Although PCBs have been banned in the United States, there is evidence that they persist in marine and terrestrial ecosystems (*2*). Remediation efforts place freshly dredged sediment in contact with air during low tides. Migration of PCBs from the contaminated sediments, through the water column into air, or directly into air at low tide, may lead to contamination of the surrounding environment. The focus of this work is the abundant local produce of Greater New Bedford, a potentially important source of PCB exposure to humans (*2, 3*).

?? why in very limited areas

There are dozens of small commercial farms and backyard gardens located within a 5-mi radius of New Bedford Harbor. They are popular sources of fresh fruit and vegetables to residents, of whom 36% identify themselves as Portuguese (*4*). In a survey administered as part of a related study, 26 out of 32 local homeowners and 43 out of 48 police officers and fire fighters surveyed reported eating locally grown produce at least twice a week in season (*5*). Furthermore, 25% of those surveyed reported that their household canned or stored produce for the winter. While these results by no means represent a random survey, they are consistent with the abundance of roadside produce stands. The present work considers potential exposure to PCBs by ingestion of local produce, before and during harbor remediation.

PCBs in the environment often cannot be described adequately as commercial Aroclor mixtures. The physical and chemical differences among congeners have implications for environmental fate and transport of the compounds as well as for toxicological impacts. The differential toxicity of the PCB congeners is well established, although there are many unanswered questions about health effects (*6*). Therefore, in our work, quantification was based on specific responses of 47 individual PCB congeners selected for their persistence, prevalence, and/or toxic potential (*7, 8*).

Sampling Strategy. Produce samples were collected from four sites located roughly downwind of the harbor's most contaminated areas, the "hot spots", and one upwind background site (Figure 1, Table 1). The prevailing wind during the growing season is from the south-southwest (*9, 10*). The downwind sites included two backyard gardens (sites A and B) and two commercial farms (sites C and D). A commercial farm (site E), upwind of the hot spots, was the source of local background samples. An additional set of samples (grown in New Jersey) was collected at site F, the supermarket. Also, PCB measurements for air and soil samples from the vicinity of the growing locations were collected to explore PCB transport pathways.

Sampling round I took place prior to the onset of dredging in September and October 1992. Three types of produce were collected: (i) carrots and potatoes representing root vegetables, (ii) tomatoes representing vine vegetables, and (iii) lettuce representing leafy vegetables. The full range of produce was not available at every site; however, all produce types were available from the local background site. Batches of soil and air samples were collected from as close to the produce sampling sites as was feasible.

* Corresponding author present address: 208 Parrington Hall, Box 353055, University of Washington, Seattle, WA 98195-3055; telephone: (206) 543-4900; fax: (206) 543-1096; e-mail address: alison@u.washington.edu.
† Harvard School of Public Health.
‡ University of Washington.

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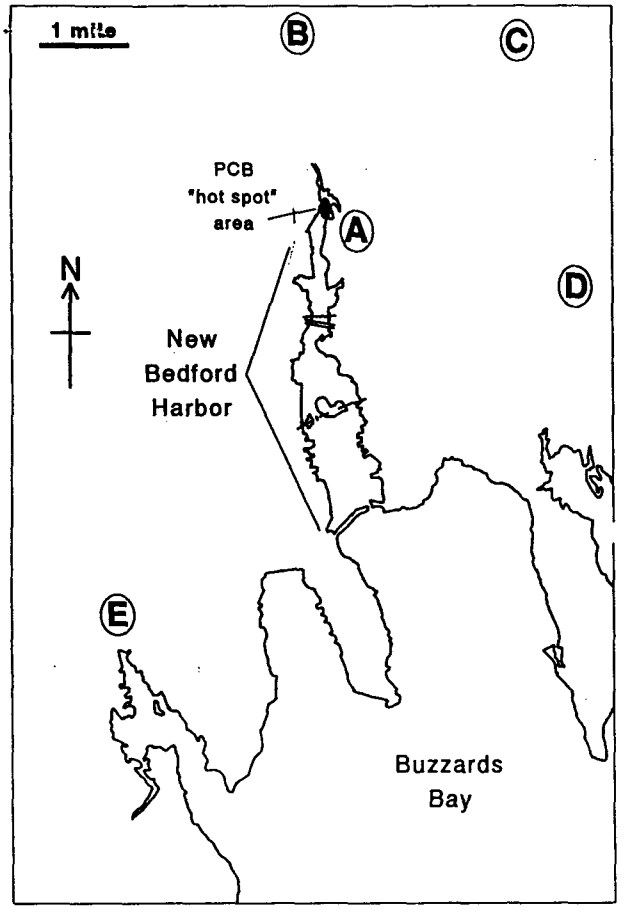

FIGURE 1. Produce sampling sites downwind of New Bedford Harbor are marked A–D, and the upwind background site is marked E.

only A&E sampled during dredging

Dredging of the harbor sediments began in the spring of 1994, prompting a second round of sampling. Batches of tomatoes again were collected from site A, the downwind site closest to the harbor, and site E, the background site, in early October 1994.

Experimental Methods

Produce Sampling. Produce samples (250–500 g) were placed in brown paper bags at the point of purchase and transported to the laboratory. In the laboratory, produce was transferred into precleaned, hexane-rinsed amber glass jars with Teflon-lined lids. All produce was washed with water to simulate normal household preparation of food, except where noted otherwise. Produce samples were stored at −15 to −20 °C until extraction.

Air Sampling. Ambient air sampling took place during late summer and fall of 1992 near sites A and E. Samples were collected with Graseby-GMW Model PS-1 high volume units, each outfitted with a 5.5 cm by 7.6 cm polyurethane foam (PUF) plug of density 0.022 g/cm^3 backing up a 10.16 cm diameter quartz fiber filter. All PUF plugs were cleaned by three consecutive 24-h extractions with hexane, dried in a vacuum desiccator, and placed in precleaned, hexane-rinsed amber glass jars with Teflon-lined screw caps sealed with Teflon tape until use. The samplers were calibrated after each relocation. The quartz fiber filters were baked at 400 °C overnight and stored in hexane-rinsed foil prior to use. Samples of volume 200–400 m^3 were collected over 24-h periods.

Soil Sampling. Soil samples were collected using hexane-rinsed stainless steel chisels. Three 50–100-g soil samples from sites A and E were scraped from the top 2 cm of exposed surface soil and transferred to precleaned, hexane-rinsed amber glass jars.

TABLE 1

Produce Samples Collected Prior to and during Dredging

site ID	location relative to hot spots	tomatoes	potatoes	carrots	lettuce
	Round I: Samples Grown prior to Dredging				
A	home garden, 0.3 mi E	6			
B	home garden, 3 mi NNE		9		
C	farm site, 3 mi NE		5		
D	farm site, 3.5 mi E	3		4	3
E	farm site, 5 mi SW	4	3	5	6
F	remote background, supermarket	7	7		
total		20	24	9	9
	Round II: Samples Grown during Dredging				
A	home garden, 0.3 mi E	6			
E	farm site, 5 mi SW	6			
total		12			

Chemical Analysis. *Preparation of Produce, Air, and Soil Samples.* Throughout this paper the IUPAC PCB nomenclature was used, which corresponds largely to the numbering system devised by Ballschmiter et al. (*11*). Prior to extraction, each field sample and quality control sample was spiked with two surrogate compounds, IUPAC No. 103 (2,2′,4,5′,6-pentachlorobiphenyl) and IUPAC No. 112 (2,3,3′,5,6-pentachlorobiphenyl). Internal standard IUPAC No. 166 (2,3,4,4′,5,6-hexachlorobiphenyl) was added to all samples to quantify target analytes.

Produce samples were cut and homogenized before subsampling for extraction and dry weight determination. Surrogate compounds, 100 g of sodium sulfate, and 100 mL of a hexane/acetone mixture (4:1) were added. The sample was homogenized at high speed for 2 min and then centrifuged for 3 min at ~1500 rpm. The extract was decanted through a powder funnel containing a glass wool plug and sodium sulfate. The extraction was repeated twice. Sodium sulfate (20–50 g) was added, and extracts were concentrated 10–15 min later. Air samples were extracted with 350 mL of hexane for 24 h using Soxhlet apparatus. Soil samples were well mixed prior to taking an approximately 30-g aliquot for extraction. Samples were shaken on a shaker table for 12 h with 100 mL of a 1:1 hexane/acetone mixture and 60 g of sodium sulfate and then centrifuged for 2 min. Soil sample extracts were decanted, and the extraction procedure was repeated two more times, for 4 and 1 h, respectively.

Air, soil, and produce extracts were concentrated by Kuderna–Danish apparatus to less than 10 mL. The K-D apparatus was disassembled, 2 drops of "keeper" solution was added to avoid analyte loss, and the extract volume was reduced to ~0.5 mL at room temperature under a gentle stream of nitrogen.

Sample extract was eluted through an 18 cm × 9 mm glass chromatographic column with a 50-mL reservoir, containing no glass frit, and packed as follows: (1) glass wool plug, (2) about 1-cm layer of anhydrous sodium sulfate,

(3) 3% deactivated silica gel, (4) 2% deactivated alumina, and (5) about 1-cm layer of anhydrous sodium sulfate. The column was washed with 30 mL of hexane before adding the concentrated extract. The sample tube was rinsed three times with 0.5 mL of hexane, and rinsates were added to the column. The column was eluted with hexane. Two drops of keeper solution were added to the eluate, and the volume was reduced to about 0.5 mL by nitrogen evaporation. An internal standard was added before instrumental analysis.

Reagents. Analytical standards of individual PCB congeners (purchased from Ultra-Scientific, North Kingston, RI, and AccuStandard, New Haven, CT) were at least 97% pure. All solvents were 'Resi-Analyzed' grade. Prior to their use, silica gel, sodium sulfate, and glass wool were extracted with dichloromethane and hexane in a sonication bath. Silica gel and aluminum oxide were baked at 400 °C prior to use. Milli-Q water used for sorbent deactivation was extracted three times with dichloromethane and three times with hexane. A keeper solution was prepared by adding 1 g of paraffin oil to 100 mL of hexane.

Instrumental Analysis. The extracts were analyzed by gas chromatography with electron capture detection (GC/ECD) using a Hewlett-Packard 5890 Series II GC. We used a capillary column (DB5, 30 m, 0.25 mm, 0.25 μm, from J&W Scientific, Folsom, CA) and the following instrumental conditions: injector at 280 °C, detector at 300 °C, initial oven temperature at 60 °C, hold for 1 min, heating to 140 °C at 15 °C/min, then to 220 °C at 1 °C/min, with final 40-min hold. For the soil samples, the final 40-min hold at 220 °C was changed to 15 min, and a third temperature ramp to 270 °C at 20 °C/min was added and held for 40 min to allow approximately 20 min of column bake-out time between analyses. The carrier gas was helium at about 1 mL/min, and the makeup gas was argon/methane (95:5) at about 60 mL/min.

Quantification. Samples were quantified by the internal standard method using pure standards of individual PCB congeners. GC data were quantified using Hewlett Packard EnviroQuant GC software. All target congeners, except three co-eluting pairs (110/77, 157/201, 196/203), were completely separated or were separated using the integrating system when eluted very closely. Several peaks were quantified as individual target congeners even though they may have had a small contribution from a co-eluting congener (138/158, 153/132, 170/190). Target analytes and method detection limits appear in Table 2. In the trace level PCB analysis of produce, all of the reported congener concentration values fall above the instrument detection limit (IDL), and a majority of them (>70%) fall above the method detection limit (MDL) based on the standard deviations of PCB congener measurements across 11 procedural blanks, all those analyzed with batches of produce samples. Rather than omit information of potential value, we report all PCB measurements falling above the IDL based on nine replicates of the standard containing the lowest concentration of individual PCB congeners. However, we present for comparison a pair of PCB congener profiles, one consisting of data that fall above the IDL and the other restricted to data falling above the MDL (Figure 2).

Quality Control. The extraction procedures for each produce type were validated by analyzing a set of replicate samples, fortified with target congeners and two surrogates. For produce, recoveries of all target analytes ranged from 67 to 120%. Surrogate recoveries for the [illegible]

TABLE 2

Method Detection Limits for PCB Congeners[a]

IUPAC No.	chlorobiphenyl structure	MDL produce, ng/g (wet weight)	MDL air, ng/m³	MDL soil, ng/g (dry weight)
6	2,3′	0.0025	0.0013	0.0059
8	2,4′	0.0335	0.0046	NA
18	2,2′,5	0.0090	0.0018	0.0090
16	2,2′,3	0.0025	0.0011	0.0038
31	2,4′,5	0.0025	0.0012	NA
28	2,4,4′	0.0040	0.0013	0.0272
33	2′,3,4	0.0035	0.0010	0.0019
22	2,3,4′	0.0035	0.0005	NA
52	2,2′,5,5′	0.0035	0.0013	0.0208
49	2,2′,4,5′	0.0015	0.0006	0.0057
44	2,2′,3,5′	0.0020	0.0007	0.0026
37	3,4,4′	0.0130	0.0005	0.0023
74	2,4,4′,5	0.0015	0.0003	NA
70	2,3′,4′,5	0.0030	0.0006	0.0018
66	2,3′,4,4′	0.0020	0.0003	0.0024
95	2,2′,3,5′,6	0.0090	0.0016	0.0051
84	2,2′,3,3′,6	0.0025	0.0004	NA
101	2,2′,4,5,5′	0.0050	0.0016	0.0060
99	2,2′,4,4′,5	0.0020	0.0003	0.0018
97	2,2′,3′,4,5	0.0015	0.0004	0.0159
87	2,2′,3,4,5′	0.0020	0.0011	0.0085
136	2,2′,3,3′,6,6′	0.0045	0.0008	0.0012
110/77+	2,3,3′,4′,6/3,3′,4,4′	0.0100	0.0017	0.0047
151	2,2′,3,5,5′,6	0.0030	0.0006	0.0030
135	2,2′,3,3′,5,6′	0.0095	0.0011	0.0020
149	2,2′,3,4′,5′,6	0.0110	0.0018	0.0070
118	2,3′,4,4′,5	0.0045	0.0011	0.0060
153	2,2′,4,4′,5,5′	0.0170	0.0029	0.0140
105	2,3,3′,4,4′	0.0035	0.0006	0.0013
141	2,2′,3,4,5,5′	0.0035	0.0005	0.0024
138	2,2′,3,4,4′,5′	0.0115	0.0035	0.0310
187	2,2′,3,4′,5,5′,6	0.0060	0.0008	NA
183	2,2′,3,4,4′,5′,6	0.0030	0.0004	0.0590
128	2,2′,3,3′,4,4′	0.0040	0.0003	0.0020
167	2,3′,4,4′,5,5′	0.0005	0.0001	NA
174	2,2′,3,3′,4,5,6′	0.0040	0.0008	0.0050
177	2,2′,3,3′,4′,5,6	0.0025	0.0004	0.0030
171	2,2′,3,3′,4,4′,6	0.0020	0.0003	0.0010
156	2,3,3′,4,4′,5	0.0005	0.0002	NA
157/201+	2,3,3′,4,4′,5′/ 2,2′,-3,3′,4,5′,6,6′	0.0045	0.0001	0.0014
180	2,2′,3,4,4′,5,5′	0.0090	0.0028	0.0060
170	2,2′,3,3′,4,4′,5	0.0125	0.0019	0.0060
199	2,2′,3,3′,4,5,5′,6′	0.0030	0.0003	0.0020
196/203+	2,2′,3,3′,4,4′,5′,6/ 2,2′,3,4,4′,5,5′,6	0.0035	0.0004	0.0020
189	2,3,3′,4,4′,5,5′	0.0005	0.0000	NA
195	2,2′,3,3′,4,4′,5,6	0.0015	0.0002	NA
206	2,2′,3,3′,4,4′,5,5′,6	0.0020	0.0001	NA

[a] MDLs are calculated from background contamination in procedural blanks. MDL = 3SD (ng/g in procedural blanks across all batches of produce and soil samples, ng/m³ in field blanks across all batches of air samples). NA, congener was detected in less than three procedural blanks.

were 78 ± 9.2 for congener no. 103 and 82 ± 11 for congener no. 112.

Samples were processed in batches of 15 with one procedural blank and two matrix spikes. The sum of PCB congeners in procedural blanks for produce batches ranged from 0.10 to 0.20 ng/g. Surrogate recoveries were 90 ± 12% for congener no. 103 and 94 ± 12% for congener no. 112. Average recoveries for the two matrix spikes fortified with all target analytes ranged from 71 to 114%. Relative percent differences for most of the congeners in the matrix spike duplicates were below 10%.

Data Analysis Methods

We present summary statistics of the sum of the concentrations of the 47 target PCB congeners in each produce

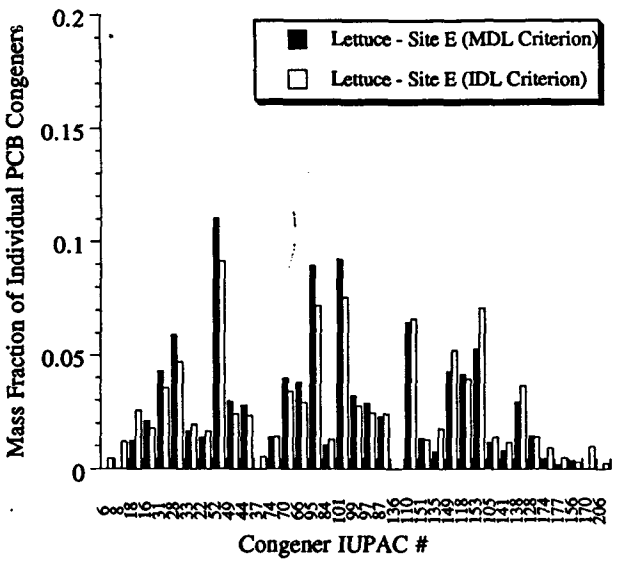

FIGURE 2. Profiles of the average mass fraction of individual PCB congeners in lettuce samples (sites D and E). Comparison of the congener profile resulting from application of the MDL (method detection limit) based on the procedural blanks versus the IDL (instrument detection limit) as acceptability criteria for PCB measurements.

type and at each location sampled. Furthermore, we construct biomagnification factors, ratios of the concentration of four individual congeners in the plant tissue to the concentration in the soil, to assess transfer between media. We also compare PCB concentrations in the inner and outer leaves of heads of lettuce and estimate reductions in the contaminant concentrations in whole potatoes with washing.

Principal components analysis (PCA) was used to compare congener profiles between and within environmental media and to generate hypotheses about mechanisms of PCB transport and uptake by plants. PCA has been used by other researchers to compare the PCB congener profiles (histograms of the mass fraction of individual congeners) of various types of environmental samples (*12, 13*). We present a selected set of PCB congener profiles summarizing the results of this analysis. In addition, the proportion of the total PCB mass consisting of congeners more volatile than no. 70, i.e., lighter than no. 70, is reported for comparison among sample types.

Results

Sampling Round I: Prior to Dredging Period. Among tomatoes grown prior to dredging, out-of-state samples contain the highest concentrations, followed by those from the downwind sites, and lastly those from the upwind site (Table 3). Among potatoes, samples grown downwind of the harbor have the highest concentration of PCBs, followed by those from the upwind site and lastly the out-of-state site. PCB concentration in carrots does not vary with location, while in lettuce the upwind samples exhibit higher levels than those from downwind. At sites D and E multiple types of produce were available for within site comparisons. At site D, PCB concentration (wet weight) is ordered as follows: carrots > lettuce > tomatoes, while at site E the order is as follows: lettuce > carrots > potatoes > tomatoes.

The congener profiles for local tomatoes and potatoes are characterized by a larger proportion of heavily chlorinated congeners, relative to those for out-of-state samples (Figure 3a,b).

In a comparison of washed and unwashed potatoes (site B), unwashed potatoes are found to contain about three times the concentration found in washed potatoes. Furthermore, the congener profiles for unwashed potatoes are characterized by a large proportion of highly chlorinated congeners relative to those for washed.

The partitioning of individual PCB congeners into plants may occur from soil through the roots of the plant or from air through the foliage. We explore the relationship of PCBs in air and soil relative to their presence in produce in several ways. First, congener profiles from samples of air and soil are compared to those for produce. For this comparison,

TABLE 3

PCB Levels in Produce from Local and Out-of-State Sites (ng/g)[a,b]

site	tomatoes	potatoes	carrots	lettuce
		Round I: Samples Grown prior to Dredging		
A	0.14 ww (0.06) 3.0 dw (1.3)			
B		0.15 ww (0.09) 0.78 dw (0.55)		
C		0.33 ww (0.21) 1.7 dw (1.1)		
D	0.09 ww (0.03) 2.1 dw (0.57)		0.17 ww (0.07) 1.9 dw (0.98)	0.16 ww (0.16) 5.0 dw (5.5)
E	0.07 ww (0.02) 1.4 dw (0.53)	0.12 ww (0.04) 0.62 dw (0.20)	0.18 ww (0.07) 2.0 dw (1.1)	0.25 ww (0.05) 6.3 dw (1.8)
F	0.20 ww (0.06) 4.4 dw (1.2)	0.08 ww (0.03) 0.46 dw (0.20)		
		Round II: Samples Grown during Dredging		
A	0.83 ww (0.29) 25 dw (23.5)			
E	0.16 ww (0.08) 3.8 dw (2.0)			

► other produce not looked at
-due to air v. soil uptake preferences??

[a] PCB levels represent the sum of 47 congeners in ng/g; ww, wet weight basis; dw, dry weight basis. [b] Table entries are averages within each

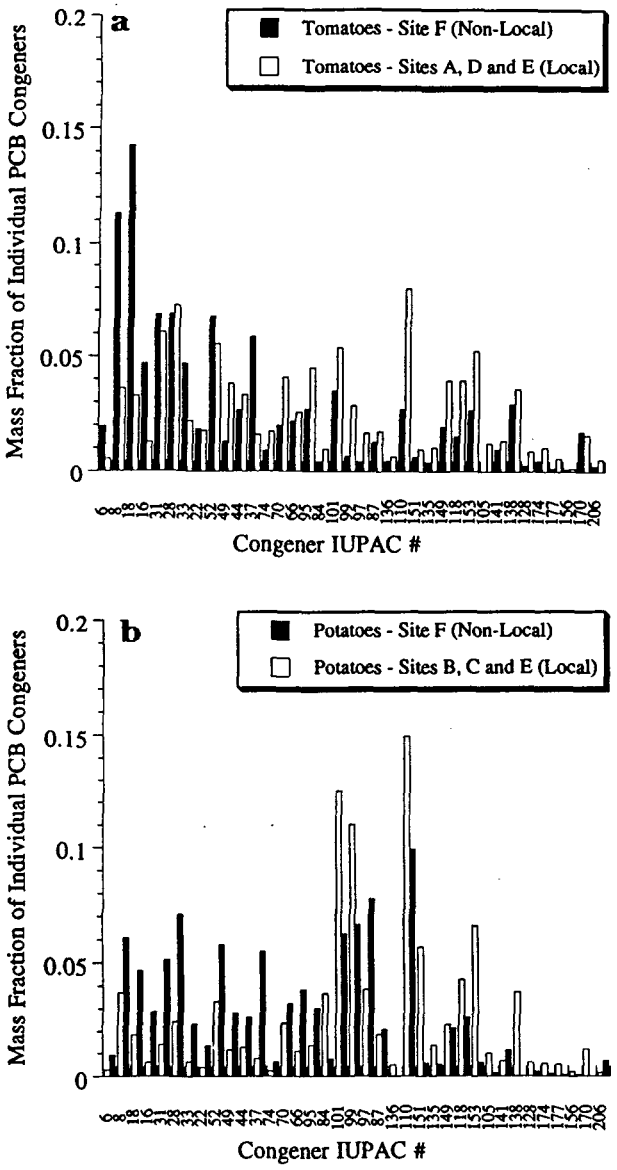

FIGURE 3. (a) Profiles of the average mass fraction of individual PCB congeners in local (sites A, D, and E) versus out-of-state (site F) tomato samples prior to dredging. (b) Profiles of the average mass fraction of individual PCB congeners in local (sites B, C, and E) versus out-of-state (site F) potato samples.

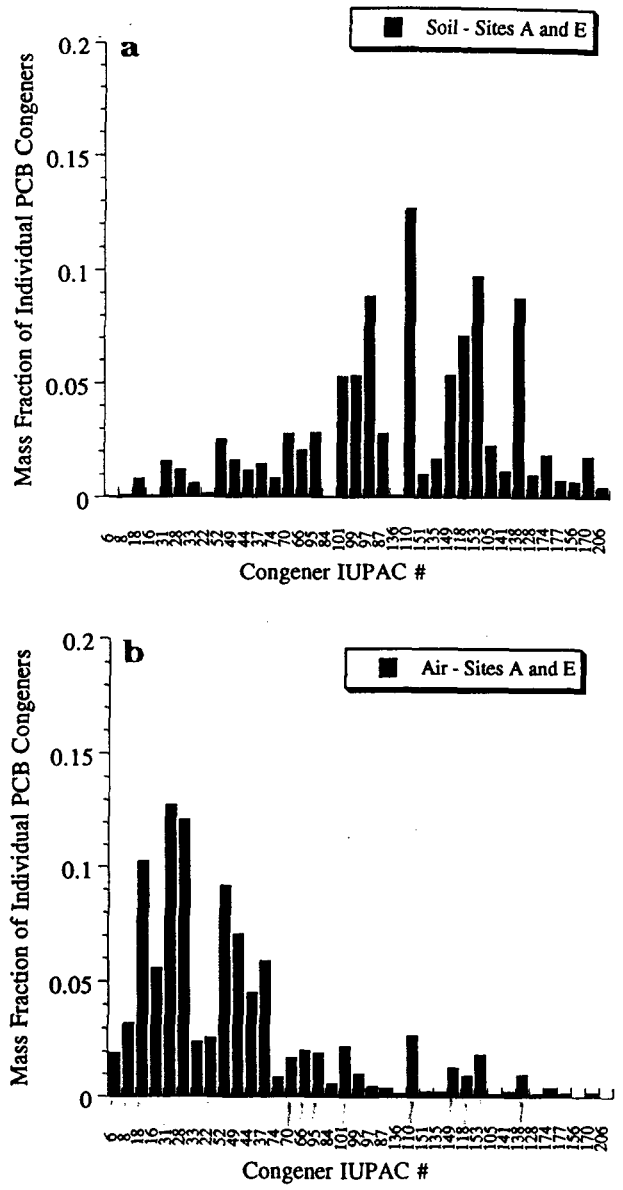

FIGURE 4. (a) Profile of the average mass fraction of individual PCB congeners in soil samples (sites A and E). (b) Profile of the average mass fraction of individual PCB congeners in air samples (in the vicinity of sites A and E).

we define the PCB congeners more volatile than no. 70 to be the "light" congeners. The soil profile is characterized by a large proportion of heavily chlorinated PCB congeners, only 15 ± 8% of the mass is comprised of congeners more volatile than no. 70. In contrast, in air 80 ± 7% of the PCB mass consists of light congeners (Figure 4a,b). From Figure 3a,b we observe that potato profiles resemble those of soil (16 ± 7% of the mass consists of light congeners), while tomato profiles resemble those of air (40 ± 5% of the mass consists of light congeners). Next, congener profiles of inner and outer leaves in heads of lettuce are compared. The profile for outer leaves contains a large proportion of heavier congeners (only 24 ± 3% light congeners) relative to the profile for the inner leaves (40 ± 2% light congeners) (Figure 5). Finally, we explore the potential uptake of PCBs by plants from soil using biomagnification factors (BFs) (Table 4). Overall, lettuce was associated with the highest biomagnification factors, followed by carrots, tomatoes and potatoes.

Sampling Round II: During Dredging Period. A significant increase in PCB concentrations was found in samples of tomatoes grown during a summer of harbor dredging compared to those from the pre-dredge period (Table 3). Downwind of the harbor (site A), PCB concentrations in tomatoes grown during dredging exceed the concentrations in pre-dredging samples by a factor of 6. Upwind (site E) the PCB concentration in tomatoes from the dredging period exceeds pre-dredging levels by a factor of about 2. In addition, the congener profiles change with dredging status. Congener profiles for tomatoes grown downwind during dredging are characterized by somewhat larger proportions of light PCBs (40 ± 5% light congeners, i.e., those more volatile than no. 70) relative to those grown prior to dredging (30 ± 2%) (Figure 6a). Furthermore, we observe that the profile for tomatoes grown downwind during dredging contains a larger proportion of light congeners (40 ± 5%) than the profile for those grown upwind (25 ± 6%) (Figure 6b).

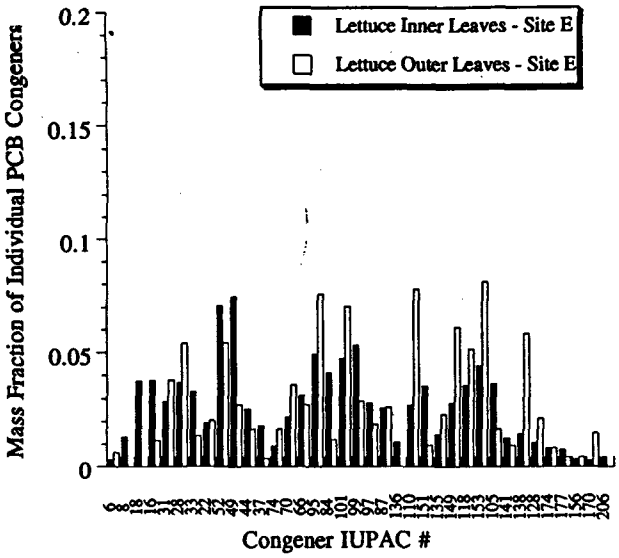

FIGURE 5. Profiles of the average mass fraction of individual PCB congeners in inner and outer leaves in heads of lettuce (site E).

TABLE 4

Biomagnification Factors (BFs) for Selected PCB Congeners[a–c]

food	no. 52	no. 101	no. 138	no. 153
	Results for New Bedford Site E			
potatoes	0.29 (0.07)	0.01 (0.02)	0.17 (0.04)	0.08 (0.01)
carrots	1.5 (1.2)	0.35 (0.38)	0.38 (0.24)	0.28 (0.17)
tomatoes	0.64 (0.29)	0.23 (0.11)	0.15 (0.10)	0.01 (0.02)
lettuce	6.0 (3.7)	1.5 (0.57)	1.1 (0.48)	0.74 (0.36)
	Results of Rippen and Wesp (*14*)[d]			
green kale	0.3–4.3	0.13–0.86	0.09–0.6	0.08–0.42

[a] Biomagnification factors are defined as the ratio of PCB congener concentration in produce to PCB congener concentration in soil. [b] New Bedford results are averages for site E, with standard deviations in parentheses. [c] All calculations were performed with dry weight concentrations. [d] Rippen and Wesp report ranges indicating variability across sampling sites.

Discussion

PCBs have been detected in all parts of the world in a range of environmental media including foods from field settings, the commercial food supply, and cultivated in laboratory environments (*14–20*). Surveys specifically targeting produce in the food supply are somewhat less common (*14–17, 20*). We found that PCB concentrations in pre-dredging produce samples from the New Bedford area are similar to those reported by other researchers, while tomatoes grown during dredging contain elevated levels. However, there are a number of issues that complicate direct comparison of our results with published data, thus a finding of similar concentrations may in fact indicate elevated levels in New Bedford produce. First, different PCB quantification methods may lead to differences in PCB measurements. Most published concentrations of PCBs in produce are based on previous analytical approaches, for example, the use of packed GC columns and quantification using direct comparison with Aroclor standards. Furthermore, our results may appear lower than those quantified by alternative methods, for example, the Deutsche Industrie Norm (DIN) (*20, 21*). The DIN approach, adopted in much of Europe and by the WHO, specifies that the sum of the concentra-

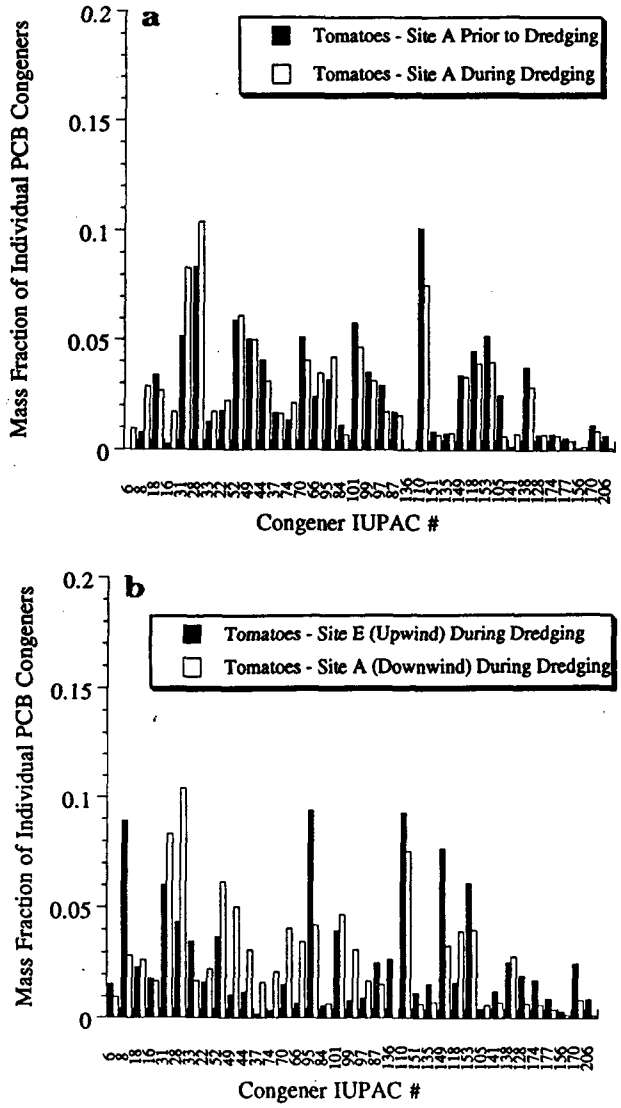

FIGURE 6. (a) Comparison of profiles of the average mass fraction of individual PCB congeners in tomatoes prior to and during dredging (site A). (b) Comparison of profiles of the average mass fraction of individual PCB congeners in tomatoes grown upwind (site E) versus downwind (site A) during dredging.

tions of six PCB congeners (IUPAC Nos. 28, 52, 101, 138, 153, and 180) multiplied by a factor of 5 approximates the total PCB concentration in a sample. Application of the DIN approach to our samples would result in concentrations between 80% and 210% of those we report, with an average increase of our values of 40%. Finally, it is not always clear whether measured values reported in the literature are based on the wet weight or dry weight of samples, which differ by up to a factor of 30 (see Table 3). Nevertheless, the background sites (E and F) included in this work allow internal comparison and provide context for the PCB measurements in downwind samples. In summary, except for potato samples, we found that the levels of PCBs in produce vary only slightly between upwind and downwind sites except during the harbor dredging. Notably, downwind tomato samples collected during harbor dredging contained higher levels of PCBs than those from upwind sites, the supermarket, or published surveys.

The WHO has reported PCB concentrations for food supplies in many countries (*16*). In a 1982 German survey, 16 potato samples were analyzed and found to be below

the limit of quantitation (LOQ), 2 ng/g wet weight (*18*). In Ontario, a survey of composite food samples found no reportable levels of PCBs in produce with an LOQ of 0.5 ng/g wet weight (*19*). In a 1980–1983 study by the Joint FAO/WHO Food Contamination Monitoring Programme, the concentration of PCBs in fresh fruits and vegetables ranged from 0.5 to 5.0 ng/g wet weight (*16*). Under its Total Diet Study (TDS), the U.S. Food and Drug Administration (FDA) reported the PCB content of two types of produce, raw celery and raisins (*17*). Only 1 out of 24 celery samples contained PCBs at levels above the LOQ, at 0.9 ng/g wet weight. Only 1 out of 24 raisin samples exceeded the LOQ, at 1.4 ng/g. The FDA has established PCB tolerances for a variety of foods, ranging from 2 ppm for fish and shellfish to 0.2 ppm for foods consumed by infants and juniors [*21 CFR*, Chapter 1, April 1, 1989, edition; Compliance Policy Guides (CPG) 7111.03]. None of the analyzed produce samples exceeded these limits.

The quantification of individual congeners is important not only because of the higher accuracy of measurements but also for combining with toxicological data and as a basis for identifying PCB sources. Information about the presence of more highly chlorinated congeners and coplanar, dioxin-like congeners is increasingly sought by risk assessors.

We find greatly elevated PCB concentrations in unwashed potatoes relative to washed, consistent with previous measurements of dioxin in root vegetables (*22, 23*). The inner and outer lettuce leaf samples we analyzed contained approximately equal PCB concentrations, consistent with earlier measurements of dioxin in lettuce (*24*). Furthermore, we find that unwashed potatoes (relative to washed) and the outer leaves in a head of lettuce (relative to inner) are enriched with more heavily chlorinated congeners. The increased proportion of heavier PCB congeners in outer lettuce leaves (even when washed) and unwashed potatoes is likely attributable to the presence of soil.

During harbor dredging, concentrations of PCBs rose substantially in tomatoes and exceeded levels reported in the literature. Also, slightly increased proportions of the more volatile PCB congeners are observed in tomatoes grown downwind during dredging relative to those upwind and those grown downwind prior to dredging. This finding is consistent with the exposure of freshly dredged PCB-contaminated sediment to the air during low tide. The hot spot sediment contains an abundance of light PCB congeners (those more volatile than no. 70), characteristic of Aroclors 1016, 1242, and 1254, which were disposed there (*1*). The volatilization, and thus depletion, of PCBs in the top layers of sediment is counteracted by the exposure of undepleted sediments during dredging.

Pal et al. (*25*) reviewed the literature on the fate of PCBs in soil–plant systems and summarized biomagnification factors (BFs) for total PCBs (mostly quantified as Aroclor 1254) from a range of field studies. The BFs we obtained are high relative to published values for produce grown in controlled field and laboratory settings; however, they are consistent with other published results for field samples. We found lettuce to biomagnify PCBs relative to soil more strongly than other produce analyzed due to its interaction with air. Lettuce is characterized by a large surface area exposed to the air and an extremely low percentage of dry matter by weight, both characteristics relevant to high biomagnification. In field samples of green kale, BFs comparable to our lettuce BFs are reported (*14*) and are consistent given the physiological similarities between the plants. In studies of edible tissue of carrots, radishes, and sugarbeets, BFs below 0.25 were reported, while in the carrot and sugarbeet greens, BFs as high as 1.0 were observed (*25*). The BFs for potatoes, carrots, and tomatoes from the New Bedford area are consistent with these results.

Within each type of produce, biomagnification of PCB congeners is inversely correlated with molecular weight and degree of chlorination, i.e., congeners with lower molecular weights exhibit higher BFs. Thus, BF also is correlated with vapor pressure. This result supports previous research findings that dry gaseous deposition onto plants is a key transport mechanism by which semi-volatile compounds enter edible tissue (*14, 15, 24, 26–28*). This premise is plausible regardless of whether the contaminant enters the air due to volatilization from soil or due to direct emission from a waste source. Alternative explanations include the degradation of heavily chlorinated congeners in soil and plants to those with lower chlorination and more mobility or the preferential uptake of less chlorinated PCBs by plants. As no evidence of metabolism of PCB congeners by plants has been reported in laboratory transport studies, this interpretation is not likely (*26, 27*).

Finally, we note that PCB levels in food may reflect the influence of multiple contaminant sources. In addition to New Bedford Harbor, sites of PCB disposal in the surrounding towns also have been identified. We deliberately avoided sampling at sites near other known PCB disposal areas; however, undiscovered sources are always possible, especially in an industrial setting.

Conclusions

Prior to harbor dredging, the presence of a major Superfund site, the PCB contaminated harbor, did not appear to result in elevated PCB concentrations in New Bedford area produce relative to local and remote background sites. The effect of growing location relative to the harbor is inconsistent between produce types. During harbor dredging there is a marked change in the PCB concentrations downwind with a lesser effect observed upwind. In particular, tomatoes grown downwind during harbor dredging exhibit elevated levels of PCBs and are especially enriched with lighter weight PCB congeners, but we acknowledge that with data from only two growing seasons we are unable to evaluate season to season variability.

In summary, accumulation of PCBs in food warrants attention because it presents opportunities for (i) assessing the efficiency of the harbor cleanup in reducing human exposure, (ii) predicting the environmental fate of PCBs during and after remediation, (iii) informing us about practical ways to reduce personal exposure.

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