

## AIR-VEGETATION PARTITIONING OF POLYCHLORINATED BIPHENYLS NEAR A POINT SOURCE

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**Abstract**—We investigated polychlorinated biphenyl (PCB) emissions to the environment from a waste treatment and transfer facility over the course of three years. We show that the facility, which undertakes PCB waste consolidation and maintains a low-yield incinerator for products such as light ballasts, acted as a point source for the spatial distribution of PCBs in vegetation. Concurrent air and vegetation sampling was performed to study the relationship between air-vegetation partitioning and the octanol-air partition coefficient ( $K_{OA}$ ). We show evidence of equilibrium partitioning for lower-chlorinated congeners ( $\log K_{OA}$  between 7 and 8.5), kinetically limited deposition on plants for intermediate congeners ( $\log K_{OA}$  between 8.5 and 11), and particle-bound deposition for congeners with high  $\log K_{OA}$  values ( $>11$ ), consistent with the McLachlan partitioning model. From spring to autumn, heavier congeners become much more concentrated in samples farther away from the facility, possibly because of higher temperatures, which enhance dispersal of these congeners. Multivariate principal components analysis showed that PCB composition in vegetation near the emission source most closely resembled the Aroclor mixtures processed by the treatment facility.

**Keywords**—Polychlorinated biphenyl Bioaccumulation Incinerator Partitioning Vegetation

## INTRODUCTION

Although North American production of polychlorinated biphenyls (PCBs) was prohibited in 1977, PCBs may still be produced as an industrial by-product. Polychlorinated biphenyls also currently are used in closed systems, such as fluorescent light ballasts and electrical transformers that were developed before the ban. As they are gradually removed from service, their PCB content usually is either stored in containers or destroyed almost completely by incineration [1]. Several media, including snowpack [2–5], sediments [5–8], and vegetation [5,9], have been used for studying the accumulation of PCBs in the environment.

Vegetation also acts as an important mediator in the exchange of PCBs between air and soil (see, e.g., [10]) because of its high storage capacity for PCBs. By examining the partitioning behavior between air and vegetation, the dominant uptake process for PCB can be determined. A theoretical model for the uptake of semivolatile organic compounds in plants was proposed by McLachlan [11]. The model predicts that the three major processes for plant uptake of semivolatile organic compounds from the atmosphere are equilibrium partitioning, kinetically limited deposition, and particle-bound deposition. Equilibrium partitioning results when the vegetation-gas partition coefficient ( $K_{vg}$ ) is relatively small ( $\log K_{vg} < 8$ ), because vegetation needs to extract a relatively small quantity of PCBs from air to reach equilibrium. As  $K_{vg}$  increases ( $8 < \log K_{vg} < 9$ ), the quantity of PCBs needed in vegetation to reach equilibrium with air will increase. Eventually, a point will be

reached at which the partition coefficient is so great that equilibrium will not be reached within the lifetime of the plant, at which point deposition is said to be transport limited. At very high  $K_{vg}$  values ( $\log K_{vg} > 9$ ), partitioning to particles in the air is so large that the dominant uptake process by vegetation will be particle-bound deposition. Each uptake process is represented by a linear relationship with unique slope on a graph of the log of PCB concentration in vegetation to air [ $\log (C_v/C_g)$ ] versus  $\log K_{OA}$ . Kinetically limited gaseous deposition appears as a plateau, and the overall graph is sigmoidal. In the McLachlan model,  $K_{OA}$  is used instead of  $K_{vg}$ , because it allows the description of particle-air and vegetation-air partitioning with a single parameter.

In the present study, we analyzed PCBs in air and vegetation around a PCB transfer and treatment facility near Cornwall, Ontario, Canada, to study their dispersal and air-surface partitioning in the field. We hypothesized that  $K_{OA}$  determines air-vegetation partitioning of PCBs, that concentrations in vegetation decline with distance from the treatment facility, that the slope of concentration versus distance is more pronounced for PCBs with higher  $K_{OA}$ , and that PCB congener composition in vegetation near the source would resemble most closely the PCBs emitted by the treatment facility.

## MATERIALS AND METHODS

## Study site

The PCB treatment facility (site A) is located at 45°3'21"N, 74°41'16"W, within a small industrial park northeast of Cornwall. Most of the surrounding area within a 1-km radius is largely rural but in land-use transition. A small number of recently constructed light industries (e.g., warehouse and distribution centers) are located within this area, but activities at these adjacent sites would not be expected to influ-

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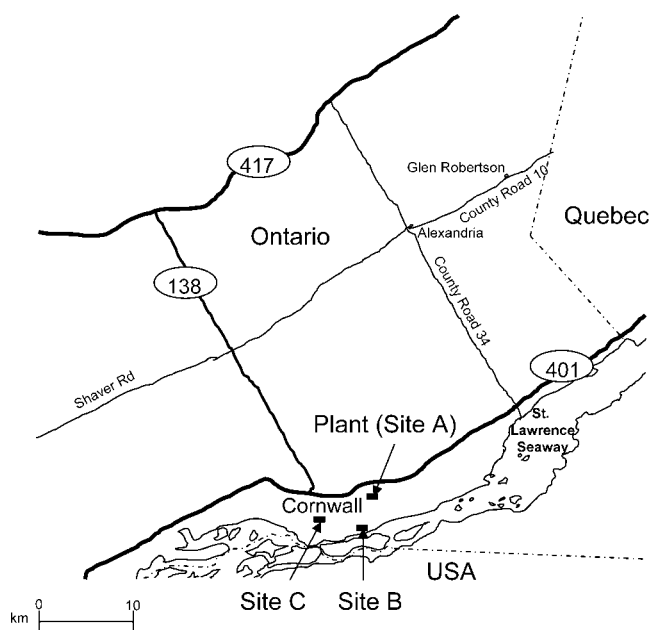


Fig. 1. Map of region of Cornwall (ON, Canada). Site A was the polychlorinated biphenyl treatment center; sites B and C were control sites.

ence the present results. Prevailing winds in this area are from the southwest. Sites B and C were used as reference sites; these sites are located 4.0 and 4.4 km, respectively, southwest of the treatment facility (Fig. 1).

#### Sampling, preparation, and extraction

Californian maple leaves (*Acer macrophyllum*) were collected along a transect extending northeast of the facility on three occasions: October 2, 1999 ( $n = 15$ ); September 22, 2000 ( $n = 10$ ); and May 14, 2001 ( $n = 6$ ). The precise location of each sample was determined by a global positioning system. The sampled trees were all mature (height,  $>8$  m). Leaves were sampled at a height of approximately 1.5 m on the side closest to the facility. Trees were resampled on each of the three sampling occasions. Approximately 20 g wet weight of each sample were collected and stored in a freezer at  $-20^{\circ}\text{C}$ .

Subsamples of 5 g of fresh foliage were cut with solvent-cleaned scissors; spiked with surrogate containing 1,3,5-tribromobenzene, 1,2,4,5-tetrabromobenzene,  $\delta$ -hexachlorocyclohexane, PCB 30, and PCB 204 in iso-octane; and immersed with 30 ml of 20:80 (v/v) acetone:hexane (Omnisolv Grade; EM Science, Gibbstown, NJ, USA). This mixture was sonicated for 30 min, and the extract was transferred to a separatory funnel with 20 ml of high-performance liquid chromatography (HPLC)-grade water. The aqueous layer was separated and re-extracted similarly twice more with hexane. Approximately 5 g of sodium sulfate were added to the combined hexane extract to remove any water in the hexane layer. The sample was transferred to the boiling flask with 1 ml of iso-octane (Optima Grade; Fisher, Fair Lawn, NJ, USA), followed by rotary evaporation of the sample down to approximately 3 to 5 ml at  $30^{\circ}\text{C}$ . The evaporated sample was then transferred to a clean, graduated centrifuge tube and topped up to 12 ml with hexane. A subsample of extract (10% by volume) was placed in a pre-weighed pan and allowed to dry to constant mass in a desiccator. The sample remaining in the tube was evaporated down to 2 ml at  $30^{\circ}\text{C}$  under a gentle stream of ultrahigh-purity (UHP)

Table 1. Total polychlorinated biphenyl (PCB) concentration in air collected during the years 1999 and 2000 at six different locations at a treatment facility near Cornwall, ON, Canada

Sample location	Collection date	Total PCB concn. (ng/m <sup>3</sup> )
Ballast splitting room	November 9, 1999	12,100
Furnace room	February 2, 2000	703
Warehouse	November 5, 1999	321
	February 3, 1999	550
Outside the facility	November 5, 1999	0.360
	November 9, 1999	0.277
	February 2, 2000	0.622
	February 3, 2000	0.196
	September 22, 2000	0.187
Site B	February 3, 2000	0.045
Site C	November 5, 1999	0.141
	November 9, 1999	0.104

nitrogen from a Reacti-Therm heating module (Pierce, Rockford, IL, USA).

Columns were prepared with glass wool (Soxhlet extracted in dichloromethane overnight, dried with UHP nitrogen, and stored at  $130^{\circ}\text{C}$ ), 8.0 g of 1.2% deactivated Florisil (7.9 g of Florisil [Suppelco, Bellefonte, PA, USA], baked at  $600^{\circ}\text{C}$  overnight, stored at  $130^{\circ}\text{C}$ , and spun with 96  $\mu\text{l}$  of HPLC-grade water for 2 h to ensure homogeneity), and 1 g of American Chemical Society Grade granular sodium sulfate (also baked at  $600^{\circ}\text{C}$  overnight and stored at  $130^{\circ}\text{C}$ ). The extract, containing 10.3 ng of 1,3-dibromobenzene and 10.2 ng of endrin ketone in iso-octane surrogate, was placed on the column (pre-rinsed with 50 ml of hexane). Three fractions were obtained with 37 ml of hexane for fraction 1, 38 ml of 15:85 (v/v) dichloromethane:hexane for fraction 2, and 52 ml of dichloromethane for fraction 3. All PCBs eluted into fraction 1. One milliliter of iso-octane was added to each fraction, and the mixture was evaporated down to between 3 and 5 ml by rotary evaporation and then to approximately 900  $\mu\text{l}$  under a gentle stream of UHP nitrogen in the Reacti-Therm heating module. Next, 106 ng of Mirex internal standard (ULTRA Scientific, North Kingston, RI, USA) were added, and the sample was topped up to 1 ml with iso-octane.

#### PCB concentrations in air

Air samples were obtained at 260 m northeast (i.e., downwind) of the facility on five occasions by drawing air from the atmosphere over a glass-fiber filter (estimated particle cut-off diameter, 0.4  $\mu\text{m}$ ), followed by two polyurethane foam filters ( $8 \times 6.5$  cm) with a high-volume air sampler for 16 h at a rate of 300 L/min. The sampling head geometry is similar to a General Metal Works PS-1<sup>®</sup> (CleanAir Engineering, Palatine, IL, USA) sampler that is widely used for air-quality monitoring. Four additional air samples were obtained inside the treatment facility (site A), and three samples were collected at two other sites in Cornwall (sites B and C) (Fig. 1 and Table 1). Filters were stored in a solvent-rinsed jar.

Each polyurethane foam filter was placed in 500-ml Soxhlet extraction vessels, to which 300  $\mu\text{l}$  of surrogate containing PCB 30, PCB 204, 1,3,5-tribromobenzene, 1,2,4,5-tetrabromobenzene, and  $\delta$ -hexachlorocyclohexane in iso-octane were added. In the boiling flask of each Soxhlet, 300 ml of Omnisolv Grade dichloromethane and two Teflon<sup>®</sup> boiling chips were added. Samples were Soxhlet extracted overnight. Approximately 25 g of granular sodium sulfate were added to the

boiling flask to remove water. The extract was pipetted out and then rotary evaporated down to between 0.5 and 1.0 ml after 1 ml of hexane was added. Florisil cleanup was the same as that for vegetation, except that only 35 ml of hexane were used to collect fraction 1. The fractions were evaporated down to less than 500  $\mu$ l. Next, 53 ng of Mirex internal standard were added, and the fractions were topped up to 0.5 ml with iso-octane. Both polyurethane foam filters in each sample were extracted separately to assess breakthrough of PCBs through the filters. No evidence of PCB breakthrough was found.

#### Gas chromatographic analysis

Samples were analyzed by gas chromatography (GC; model 6890; Hewlett-Packard, Wilmington, DE, USA) with a  $^{63}\text{Ni}$  microelectron-capture detector ( $\mu$ ECD) (Hewlett-Packard, Wilmington, DE, USA) and a DB-5 capillary column (J & W Scientific, Folsom, CA, USA) (length, 30 m; film thickness, 0.25  $\mu$ m; inner diameter, 0.25 mm; cross-linked 5% phenyl-methylsiloxane) with UHP helium as the carrier gas at 1.3 ml/min and UHP nitrogen as the detector makeup gas at 60 ml/min. The GC parameters were as follows: injector temperature, 250°C; splitless; oven temperature program, hold at 80°C for 2 min, ramp at 10°C/min to 110°C, and then ramp at 3°C/min to 280°C; total runtime, 61.67 min; and  $\mu$ ECD detector temperature, 350°C. The retention times of the congeners were compared to the reference retention times as determined with a known mixture of PCBs. The area of each peak was then converted to the concentration of the congener represented by that peak through a five-point calibration curve.

Results reported for PCBs are presented for each homologue group, from mono- to decachlorobiphenyl. In the case of two coeluting congeners that spanned more than one homologue group, the concentration was added to the lower homologue. In the case of three coeluting congeners, they were added to the majority or middle homologue group. The data were neither recovery nor blank corrected. The average recoveries for PCBs 30 and 204 were high ( $82\% \pm 8\%$  [SD] and  $92\% \pm 9\%$ , respectively), and the total PCB concentration in the blanks was low ( $0.15 \pm 0.03$  ng/g, assuming a typical sample weight of 5 g).

#### Quality control

The limit of detection in the method for the vegetation matrix was determined as described by the American Public Health Association [12] and was approximately 2 ng/g (assuming 5 g of sample extracted) of  $\Sigma\text{PCB}$ . The limits of detection for the homologue groups ranged from 0.039 to 1.605 ng/g.

To monitor potential laboratory contamination, procedural blanks were processed after every 10 vegetation extractions.

#### Multivariate analysis

To highlight the similarities in the PCB signature of air and vegetation samples as well as the technical Aroclor mixtures, we performed multivariate ordination analyses using CANOCO, Version 4.5 [13]. The PCB congener concentration data from each of the samples were first expressed as relative abundances (%) and then screened to only include congeners that were present in at least half the samples. The subset of 43 congeners or congener groups used in the analyses, however, represented an average of 81.5% of the total abundance of PCBs in each sample. For all analyses, the data were log ( $x$

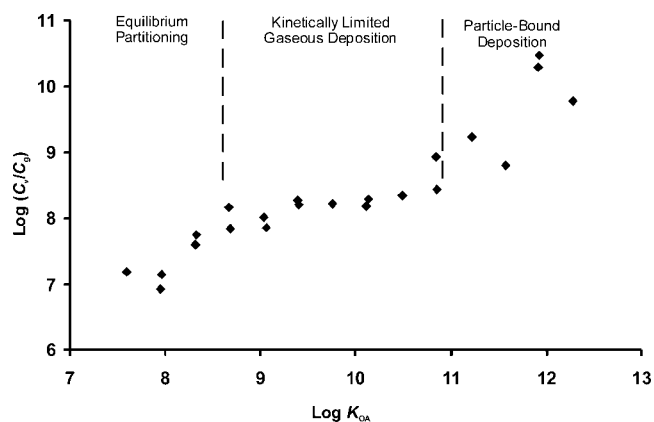


Fig. 2. Plot of log of concentration in vegetation to air (gas phase;  $\log C_v/C_g$ ) of all polychlorinated biphenyl congeners in the sample collected at 263 m (year 2000) versus log octanol-air partition coefficient ( $K_{OA}$ ). The  $K_{OA}$  values were estimated according to the quantitative structure-property relationship method and were corrected to 8°C [18]. Values of  $\log (C_v/C_a)$ , where  $C_a$  is the concentration in air (vapor phase), were averaged for congeners having the same predicted  $K_{OA}$ .

+ 1) transformed, where  $x$  refers to the relative abundance of each congener or congener group.

A preliminary, detrended correspondence analysis was first conducted to determine the gradient length of the first ordination axis, which often is used as a guide to determine whether the data are best fit with a linear ordination approach (i.e., a principal components analysis [PCA]) or a unimodal ordination approach (i.e., a correspondence analysis [14]). Using this approach, we found the data to be linear, so we applied a PCA analysis to the vegetation and air samples. In our final PCA analysis, we plotted passively the relative abundance data for commercial Aroclor mixtures [15], because these mixtures were being processed at the PCB transfer and treatment facility. The passive analysis of Aroclors means that these data were not included in the construction of the ordination analysis but were included afterward to assess the relationship among Aroclors, vegetation, and air samples. In summary, the PCA allowed us to identify which congeners were present in greater relative abundance in each of the sample types (i.e., air for 1999 and 2000, vegetation for 1999, or vegetation for 2000 and 2001) by comparing the congener scores with the sample scores.

## RESULTS AND DISCUSSION

To understand the spatial and temporal trends in PCB congener distributions, it was necessary to identify the main modes of their transport from air to vegetation. Plotting the log of the ratio of the concentration of PCBs in vegetation ( $C_v$ ) to that in the gas phase ( $C_g$ ) as a function of  $\log K_{OA}$  for our results from September 2000, a sigmoidal curve is observed similar to the one predicted and observed by McLachlan [11] and by Bohme et al. [16], confirming that all three deposition mechanisms are significant for PCBs (Fig. 2). The changes in uptake occur at  $\log K_{OA} = 8.5$  and  $10.5$ , very close to those predicted by the McLachlan model. The figure shows that lighter, more volatile PCBs likely are involved in equilibrium partitioning, whereas heavier PCBs are more likely to accumulate in vegetation as a result of deposition via aerosol particles.

A pronounced gradient in PCB concentrations and congener composition was apparent across our four different air-sam-

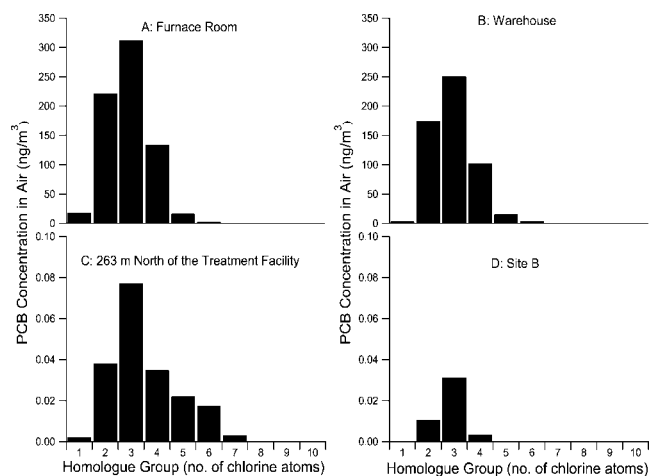


Fig. 3. Histograms of total polychlorinated biphenyl (PCB) concentration in air for each PCB homologue group at four locations. (A) Furnace room (on February 2, 2000). (B) Warehouse (on February 3, 2000). (C) 263 m north of the treatment facility (on September 22, 2000). (D) Site B (on February 3, 2000) near Cornwall, Ontario, Canada.

pling locations (Fig. 3 and Table 1). Air samples collected inside the furnace room and the warehouse showed an almost identical congener pattern, but PCBs there were more than 1,000-fold more concentrated than in the air taken outside the treatment facility. Polychlorinated biphenyl concentrations in air also were observed to decrease with distance by comparing the sample collected outside the treatment facility building with that collected at site B.

The total PCB concentration in vegetation samples generally decreased with their distance from the treatment facility (Fig. 4 and Table 2), and this decrease was most pronounced for heavier congeners (Fig. 5 and Table 3), indicating that the

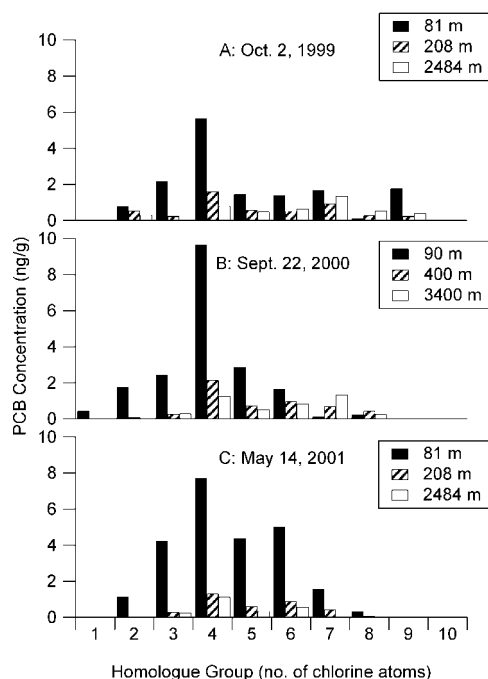


Fig. 4. Histograms of total polychlorinated biphenyl (PCB) concentration in each PCB homologue group at different distances for vegetation samples collected on (A) October 2, 1999; (B) September 22, 2000; and (C) May 14, 2001.

Table 2. Sum of polychlorinated biphenyl concentrations in vegetation collected in the years 1999, 2000, and 2001 at different distance from the treatment facility near Cornwall, Ontario, Canada

October 2, 1999		September 22, 2000		May 14, 2001	
Distance (m)	Concn. (ng/g)	Distance (m)	Concn. (ng/g)	Distance (m)	Concn. (ng/g)
81	15	90	19	55	22
83	9	125	10	81	24
208	5	263	49	83	8
293	19	276	26	208	4
1,060	10	342	22	1,060	3
2,484	5	400	5	2,484	2
3,758	2	1,100	2		
4,487	3	1,700	3		
4,745	1	3,400	5		
5,633	5	3,900	6		
6,922	4				
7,962	9				
12,406	2				
12,593	3				
20,651	1				

treatment facility was, indeed, a source of PCBs during this time period. Phytotoxicological monitoring of PCBs by the Ontario Ministry of the Environment (OMOE) at this site was done during the same time frame (from 1999 through 2002). The detection limits of the OMOE are less sensitive than ours, which are designed to look at the fate and dispersal of low concentrations; thus, all PCB congeners in their tree foliage samples were reported as being less than their limits of detection. They did, however, conduct coplanar PCB congener analyses at pg/g levels using high-resolution GC-mass spectroscopic techniques [17]. Concentrations of PCB in foliage near the building were several-fold greater than background levels for coplanar PCBs, leading the OMOE to conclude that during the period from 1999 to 2002, the facility was a marginal and localized, low-level source of coplanar PCBs.

We observed different spatial distributions for PCB homologues between late spring (May) and autumn (September/October). In May 2001 (Fig. 4C), heavier PCBs were absent from sites far from the facility. However, these heavier PCBs did appear at these sites in the samples from October 1999 (Fig. 4A) and September 2000 (Fig. 4B). Those PCBs that accumulate by either kinetically limited gaseous deposition or

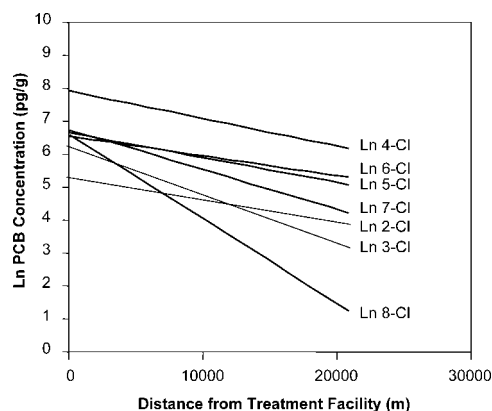


Fig. 5. Distribution of homologue concentration against distance from the treatment center (material resources recovery) on October 2, 1999. The equations of these lines are given in Table 3. In general, the heavier homologues declined more rapidly with distance from the treatment facility. PCB = polychlorinated biphenyls.

Table 3. Equations describing the ln concentration in vegetation (ng/g fresh wt) as a function of distance from the treatment facility

Concentration (ng/g fresh wt) <sup>a</sup>	Equation <sup>b</sup>	r <sup>2c</sup>
Ln dichloro-PCB	$(-2 \times 10^{-5})x + 5.05$	0.42
Ln trichloro-PCB	$(-1 \times 10^{-4})x + 6.19$	0.40
Ln tetrachloro-PCB	$(-8 \times 10^{-5})x + 7.93$	0.36
Ln pentachloro-PCB	$(-8 \times 10^{-5})x + 6.65$	0.35
Ln hexachloro-PCB	$(-6 \times 10^{-5})x + 6.56$	0.29
Ln heptachloro-PCB	$(-1 \times 10^{-4})x + 6.75$	0.25
Ln octachloro-PCB	$(-3 \times 10^{-4})x + 6.62$	0.52

<sup>a</sup> Mono-, nona-, and decapolychlorinated biphenyls (PCBs) are not included, because these homologues were undetected in more than half the samples.

<sup>b</sup> Where  $x$  is the distance (m) from the treatment facility.

<sup>c</sup> All values in this column are statistically significant (\* $p < 0.05$ ).

particle-bound deposition are more likely to be retained by vegetation and, therefore, will tend to deposit close to the treatment facility. With the higher mean temperature during the summer, these PCBs likely are transported greater distances, resulting in higher concentrations farther away from the source (Fig. 4A and B). Lighter PCBs, on the other hand, are accumulated by equilibrium partitioning. Therefore, their concentrations in vegetation do not vary as much with time, because they are near equilibrium with air.

Our multivariate analysis also showed a gradient where vegetation samples taken closest to the source are most similar to the air samples taken both in and around the treatment facility (Figs. 6A and 7A). The lower-chlorinated, more volatile congeners, such as PCBs 8+5, 15+17, 16+32, 18, and 33+20+53, had high relative abundance in air near the facility, as shown in the ordination analysis (Fig. 6B). These congeners also make up a substantial proportion of the PCBs in Aroclors 1016, 1232, and 1242 [15], which commonly are used in light ballasts, which in turn comprise a major portion of the materials processed at the facility. Interestingly, the air sample collected at site B (one of our reference sites) had a substantially different PCB signature compared to other air samples. This site is at the edge of Cornwall and, thus, represents background environmental conditions.

Vegetation samples closest to the treatment facility tended to partition near the tri- and/or tetrachlorinated biphenyls, such as PCBs 12+17, 18, and 33+20+53, in ordination space (Figs. 6 and 7B). This finding with multivariate analysis reiterates the pattern shown with the homologue histograms for vegetation (Fig. 4), but it also highlights which congeners are driving this trend.

Another striking feature of the PCA plot is the separation of vegetation samples among years, in which the 1999 samples differ substantially from the 2000 and 2001 samples (Fig. 6). This difference among sampling years also is suggested by Figure 4, in which the 1999 samples exhibit overall concentrations of PCBs lower than those in other sampling years. The multivariate analysis, however, clearly identifies which congeners differ most strongly among the years. For example, PCBs 46, 51, and 180 were more prevalent in the 1999 samples, whereas PCBs 70+76+98, 74, and 153+132+105 were more prevalent in the 2000 and 2001 samples. This outcome may have resulted because between our samplings in 1999 and 2000, the PCB treatment and transfer facility improved the ventilation of their building and their handling procedures to minimize exposure to workers, which should have in-

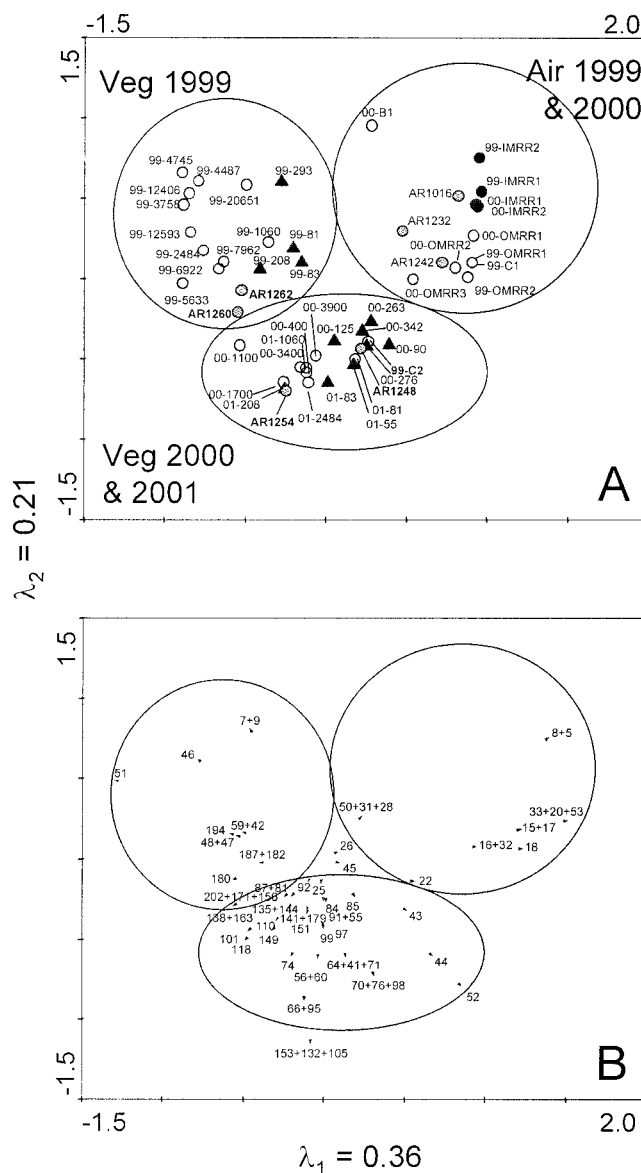


Fig. 6. Principal components analysis biplot of polychlorinated biphenyl (PCB) congeners analyzed in vegetation and air samples collected around the PCB transfer and treatment facility between the years 1999 and 2001. The biplots (A) and (B) show the relative distributions of samples and congeners. All samples are shown as white dots, with the exception of indoor air samples, Aroclor data, and vegetation samples. Indoor air samples are highlighted as black dots, and vegetation samples collected within 500 m of the transfer and treatment facility are shown as black triangles. The vegetation data points are labeled by the sampling year, followed by the distance (m) from the facility. Aroclor data are plotted passively (hatched dots) and denoted with an AR prefix.

creased the amount and dispersal of PCBs into the surrounding environment in 2000. Our results show that many of the sum and homologue PCB concentrations at sampling locations near the site were elevated in 2000 and 2001 relative to those in 1999 (Fig. 4 and Table 2). Changes in ventilation also would alter the relative contributions of regional atmospheric PCB deposition versus a localized source of atmospheric PCB deposition (i.e., from the incinerator) at sampling sites, and we suggest that this mechanism is largely responsible for the different congener fingerprint between 1999 and 2000/2001.

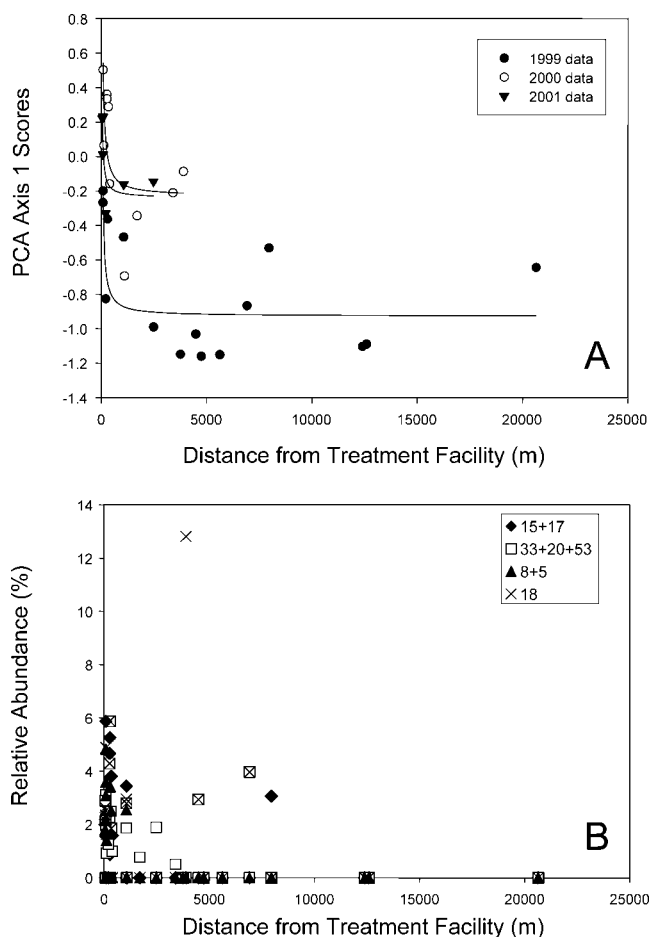


Fig. 7. (A) Plot showing the significant decrease ( $p < 0.05$ ) between principal components analysis (PCA) axis-one loading of each vegetation sample (from Fig. 6A) and distance from the incineration plant for each of the three sampling years. The  $r^2$  values for 1999, 2000, and 2001 were 0.51, 0.44, and 0.67, respectively. (B) Plot showing the relative abundances of incinerator congeners (i.e., congeners having  $>10\%$  relative abundances in air samples from inside the plant) as measured in vegetation samples versus distance from incinerator.

### CONCLUSION

In summary, the decrease in total PCB concentration with distance confirms that the treatment facility was a point source for PCBs during the period from 1999 to 2001. Air samples collected at different areas of the treatment facility reflect large differences between indoor and outdoor PCB concentration in air but also similarities in congener composition, suggesting that fugitive emissions rather than stack emissions were the major sources. In addition, extensive OMOE-regulated stack testing indicated high incinerator efficiency (99.999%). A study of air-vegetation partitioning showed agreement with the theoretical partitioning proposed by McLachlan [11], demonstrating that the three dominant uptake processes are active (equilibrium partitioning, kinetically limited gaseous deposition, and particle-bound deposition).

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