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# Redistribution of Polychlorinated Biphenyls from a Local Point Source: Terrestrial Soil, Freshwater Sediment, and Vascular Plants as Indicators of the Halo Effect

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## Abstract

Polychlorinated biphenyl (PCB) concentrations and congener profiles in terrestrial soils, vascular plants, and freshwater lake sediments were compared with respect to their relative utilities for assessing the redistribution of PCBs from a local contaminant source into the surrounding arctic environment.

Plants (n = 62), soils (n = 58), and surficial freshwater lake sediments (n = 16) were collected at varying distances up to 27 km from the source at Saglek, Labrador. Total PCB concentrations in these media exhibited similar negative log-linear relationships with increasing distance from the contaminant source. Analysis of congener compositions indicated that plants are more reflective of recent PCB exposure than either soils or sediments. Vascular plants reflect current ambient contaminant concentrations, whereas terrestrial soils and freshwater sediments represent much longer periods of contaminant deposition and are further influenced by such factors as their organic carbon content, particle size distribution, lake watershed size and productivity (in the case of sediments), and proximity to a drainage course (in the case of soils). Collectively, the data indicate that short-range transport of PCBs at Saglek has resulted in a halo of contamination that is up to 50 km in diameter.

## Introduction

Freshwater lake sediments have been widely used as indicators of atmospherically transported chemicals (Eisenreich et al., 1989; Lockhart et al., 1992; Charles and Smol, 1994; Camarero et al., 1995; Schindler et al., 1995; Allen-Gil et al., 1997; Braune et al., 1999). Lake sediments have been considered particularly useful in monitoring background concentrations of environmental contamination due to their ability to “focus” contamination from an area encompassing the lake’s entire watershed (Schindler et al., 1995; Muir et al., 1996; Semkin, 1996). This approach has been extensively applied in the study of spatial and temporal trends pertaining to low-level contamination of the Canadian Arctic by persistent organic contaminants (reviewed by Braune et al., 1999). Accumulated sediments provide an invaluable historical record of long-term contaminant chronologies, and analysis of biological remains through paleolimnological methods can be used to infer the impact of contaminants on freshwater food webs (Landers et al., 1995).

There has been comparatively less use of vascular plants in studies of organic contamination of arctic ecosystems (Dushenko et al., 1996; France et al., 1997; Kelly and Gobas, 2001), probably as a consequence of a perception among the scientific community of the inability of vegetation to absorb organic contaminants representative of ambient environmental contaminant loads. Plants have been used, however, in studies concerned with monitoring inorganic and/or radioactive contaminants in this remote region of the globe (Brakenhielm and Liu, 1995; Lippo et al., 1995; Steinnes, 1995), and it is increasingly recognized that concentrations of organic contaminants in plants do reflect conditions in the surrounding environment (Simonich and Hites, 1995; Ockenden et al., 1998). As primary producers, plants link abiotic and biotic ecosystem components, making them an ideal medium for monitoring ecosystem contamination. Hence, in our work

of assessing the impact of the radar sites of the Canadian Distant Early Warning (DEW) Line on surrounding arctic environments, we have used vascular plants to determine cleanup criteria for both inorganic and organic contaminants (ESG, 1993).

The relative significance of local versus more distant sources of contamination in the Arctic has been the source of some debate in the literature (Thomas et al., 1992; Norstrom and Muir, 1994; Stern et al., 1997; Braune et al., 1999). While long-range transport of organic contaminants is well known and understood, local source redistribution is less well documented and accepted. Previous studies conducted by our group around DEW Line radar stations have provided evidence that contaminant concentrations in the environment up to 20 km from the source of contamination were impacted by short-range transport of contaminants from the local source (Bright et al., 1995; Dushenko et al., 1996), creating a “halo” of contamination.

The discovery of large volumes of soil contaminated with PCBs (Table 1) at a remote military station in Saglek, Labrador (Fig. 1), presented a unique opportunity to study the fate and effects of a local source of PCB contamination on the surrounding environment (ESG, 1999). As a result of poor handling and disposal of waste transformer oil during its operation between 1953 and 1971 and subsequent abandonment, soil in three separate areas at Saglek was contaminated with PCBs at concentrations as high as 250  $\mu\text{g g}^{-1}$ . The Saglek Polevault Station had no other industrial activity and is isolated from external sources of industrial contaminants. Nain, the community closest to Saglek, is located 200 km to the south and has a population of approximately 1500.

The source of PCBs at Saglek is the commercial Aroclor 1260 mixture, the congener composition of which is well characterized (Schulz et al., 1989). Aroclor 1260 is dominated by higher-chlorinated congeners, such as 138, 149, 153, 180, and 170, and therefore its influence on the local environment can be distinguished from the

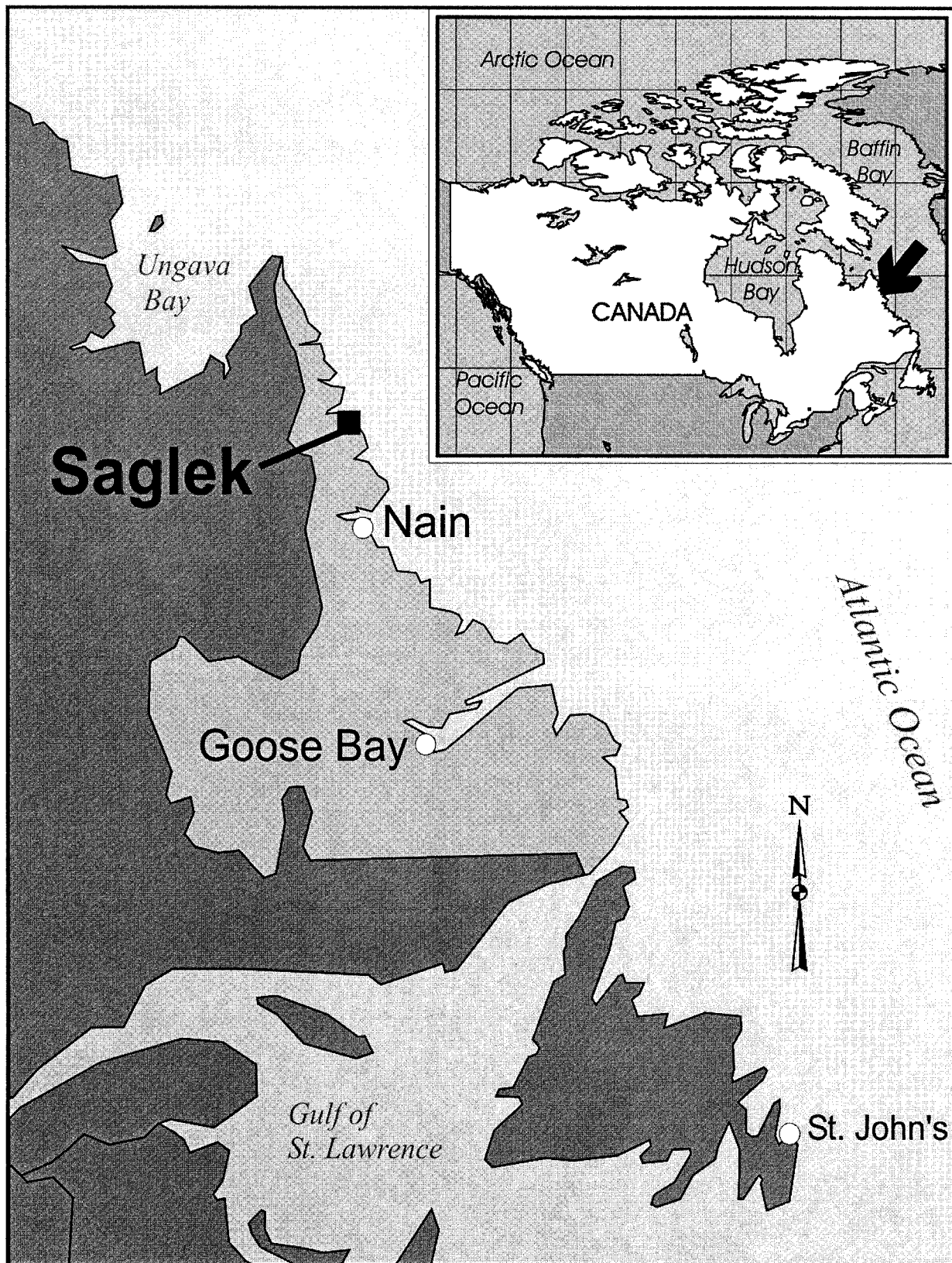


FIGURE 1. Location of the North Warning System radar site at Saglek, Northern Labrador.

TABLE 1

Volumes of soil and estimated concentration and mass of PCBs excavated from each of three areas at Saglek, Labrador, between 1997 and 1999 (from ESG, 1999)

Excavation Area	Volume (m <sup>3</sup> )	Average PCB Concentration (µg g <sup>-1</sup> )	Mass of PCB <sup>a</sup> (kg)
Antenna Hill	3300	250	1660
Beach	12,200	100	2440
	16,300	27.5	890
Site Summit	4000	75	590
Total >50 µg.g <sup>-1</sup>	19,500	120	4690
Total 5–50 µg.g <sup>-1</sup>	16,300	27.5	890

<sup>a</sup> Rough estimate based on total mass (kg) = volume (m<sup>3</sup>) × 2000 kg m<sup>-3</sup> × [PCB] (kg.kg<sup>-1</sup>), where 2000 kg m<sup>-3</sup> = estimated density of soil

background congener signature resulting from long-range atmospheric transport of less chlorinated, more volatile congeners (Fig. 2).

In this study, we compared total PCB concentrations and congener profiles in terrestrial soils, vascular plants, and freshwater lake sediments for their ability to reflect the distribution of PCBs in an arctic terrestrial ecosystem within 27 km of the local contaminant source. This study is unique in its concurrent examination of all three media and because it examines the redistribution of PCBs from a local source. This and related studies conducted at Saglek (Kuzyk, 2000) provide some of the first information regarding concentrations of persistent organic contaminants in coastal northern Labrador.

## Materials and Methods

### STUDY SITE CHARACTERISTICS

Saglek is located on the northeast coast of Labrador (58°23'N, 62°35'W), within the Torngat Mountains, approximately 600 km north of Goose Bay and Happy Valley (Fig. 1). From 1953 until 1971, a Polevault Line radar station was operated at Saglek by the U.S. Air Force. In 1971, the radar facility was abandoned, and in 1978 it was destroyed by fire. Saglek was reopened as one of the modern North Warning System long-range radar facilities operated by the Canadian Department of National Defence in 1988, and is still in operation today (ESG, 1999).

Saglek is approximately 100–200 km north of the present position of the white spruce treeline, in the tundra zone, where vegetation is characterized by dwarf shrubs, herbs, mosses, and lichens. Permafrost is extensive but discontinuous. Archean granitic rocks characterize the geology of the region, and acidic rock outcrops and cryosols are typical of the surface material (Environment Canada, 2000). Evidence of glaciation in the form of cirques, deep U-shaped valleys, fjords, and bouldery morainal deposits is seen throughout the region (Solomon, 1999).

Local weather patterns influence the degree and direction of contaminant dispersal from a local source (Lee and Jones, 1999). Prevailing wind direction and the direction of winds during major storm events influence the pattern of vapor-phase and particulate-bound contaminant dispersal (Lee and Jones, 1999; Reid et al., 2000). Wind data collected from a meteorological station at the Saglek site summit (~762 m a.s.l.) from 1989 to 1998 indicated that during the summer months, the winds blow moderately (20 km h<sup>-1</sup>) from the north and south, 20% and 15% of the time, respectively (Knowlton, 2000). In winter and spring, winds are primarily out of the west. High winds typical of storm events are out of the northeast and southeast, but these occur infrequently.

### SAMPLE COLLECTION

Soil and plant samples were collected in the months of July and August, 1997 to 1999. Lakes were cored in 1997 and 1998. All samples were collected at increasing distances from the three areas contaminated with PCBs at concentrations exceeding 50 µg g<sup>-1</sup> (site summit, Antenna Hill, beach) and to a maximum of 27 km (Figs. 3 and 4). During the 3-yr sampling period, extensive site remediation involving soil excavation and stockpiling took place.

At the site, plant/soil pairs were collected along transects up to 1.6 km long that extended out from each of the three contaminated areas (Fig. 3). At the beach, east and west transects were sampled at 0, 0.1, 0.2, 0.4, 0.6, 0.8, and 1.0 km. At the site summit, samples were collected at 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 km along one transect that extended southeast of the contaminated area. At Antenna Hill, samples were collected at 0.001, 0.003, 0.005, 0.01, 0.5, 0.1, 0.2, 0.4, 0.6, 0.8, and 1.0 km on two transects bearing south and west from the contaminated area. On the west Antenna Hill transect, samples were also collected at 1.2, 1.4, and 1.6 km. Eleven additional soil/plant pairs were collected beside 11 of the 16 lakes from which sediment cores were collected, and 3 soil/plant pairs were collected from the islands north of the station in Saglek Bay (Big Island, BI [n = 2] and Shuldham Island, SI [n = 1], Fig. 4).

Soil samples (n = 58) were obtained from the top 10 cm of substrate using a stainless-steel scoop (prewashed with hexane and dichloromethane, baked at 300–400°C, and stored in baked aluminum foil) and placed in a 125-mL amber jar (Proclean<sup>TM</sup>) fitted with a Teflon-lined lid.

Approximately 20 g of plant tissue were collected within 1 m<sup>2</sup> of the corresponding soil sample and consisted of roots, shoots, and leaves from a single species. Plants were washed thoroughly with locally available water (sampled and analyzed separately) to remove all visible soil and then laid out on and patted dry with Kim<sup>TM</sup> towels. All plant samples were wrapped in baked aluminium foil and placed in clean, labeled Ziploc<sup>TM</sup> bags.

Plant samples (n = 62) consisted primarily of willows (*Salix* spp., n = 24) and dwarf birch (*Betula glandulosa*, n = 18), which are common components of the Northern Labrador plant community. Vegetation sampled when willows and birch were unavailable included sedges (*Carex bigelowii*, n = 6), bluegrass (*Poa* spp., n = 5), wood rushes (*Luzula confusa*, n = 3), bilberry (*Vaccinium uliginosum*, n = 3), alpine bearberry (*Arctostaphylos alpina*, n = 2) and cottongrass (*Eriophorum callitrix*, n = 1). Plant identifications were made using Porsild and Cody (1980) and Scoggan (1979); nomenclature follows the standard of Scoggan (1979).

All sediment cores were retrieved from near the center of each lake using a 7.6-cm-diameter Glew (1991) gravity corer. The large internal diameter of this corer provided a sufficient volume of sediment to perform analyses of Pb-210 dates, PCBs, and siliceous microfossils on the same core (results of paleolimnological analyses are presented in Paterson et al., 2003). Standard physical and environmental characteristics of each lake were recorded at the time of sampling and are presented in Table 2.

Cores were sectioned as soon as possible after collection (typically at lakeside) using a vertical extruding device (Glew, 1988). The top 10 cm of each core was sectioned at 0.5-cm intervals, and the remainder of each core was sectioned at 1-cm intervals. Each slice was carefully scraped into a labeled Whirlpak<sup>®</sup> bag using a clean plastic spatula. The spatula was cleaned with distilled water and laboratory-grade Kim<sup>TM</sup> towels between samples to prevent cross-contamination between samples. Corresponding soil and plant samples were collected as close to the lakes as possible (within 50 m of the shore) and outside any obvious drainage channels. Soil, plant, and sediment samples were stored in coolers

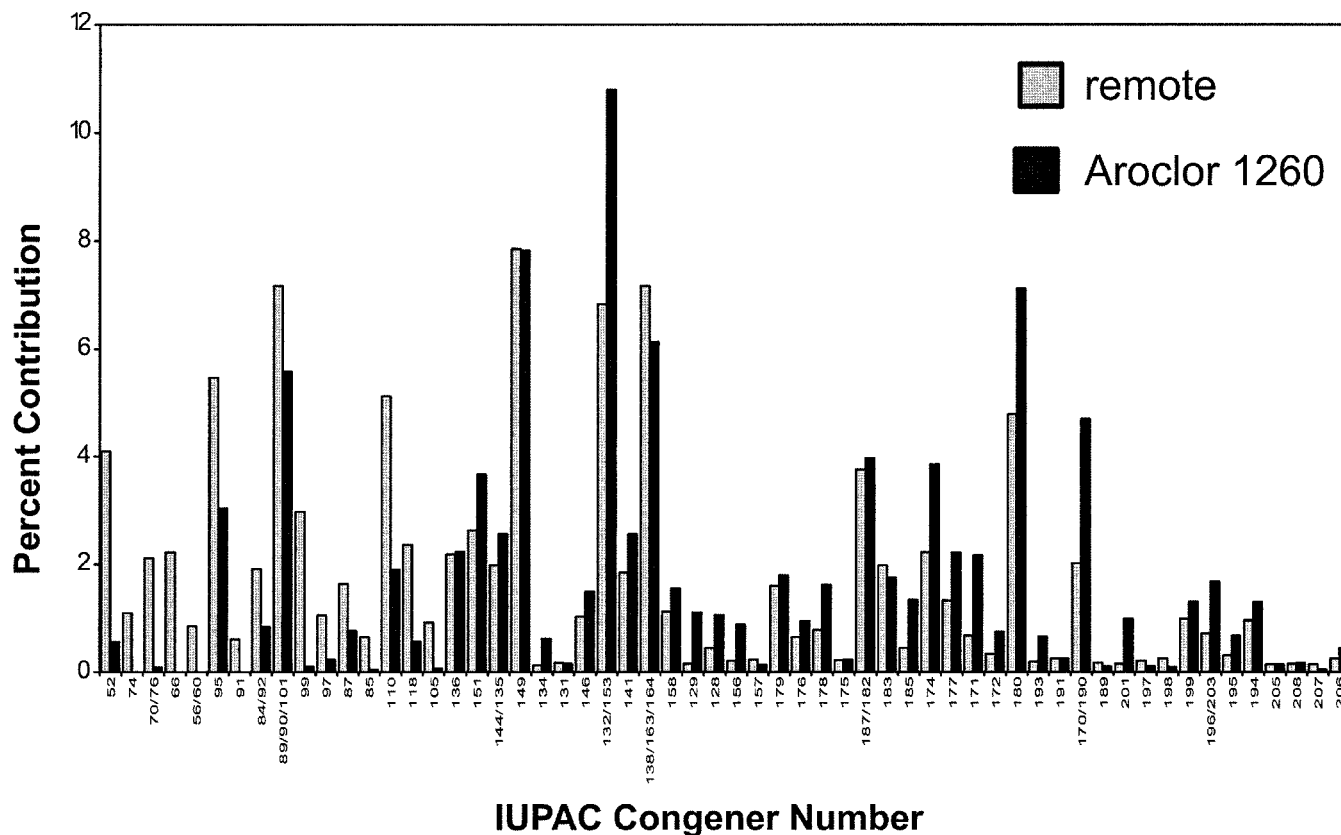


FIGURE 2. Percent congener compositions of Aroclor 1260 industrial mixture and that of a representative remote background sample (plant from Hot Weather Creek, Ellesmere Island, Nunavut).

and shipped to Axys Analytical Services Ltd., where they were stored frozen until analyzed.

Sample locations were recorded with a Global Positioning System (GPS) (Ashtech SCA/12 using the "Precision" option) to an accuracy of within 1.0 m. GPS data were postprocessed using software by Reliance™.

#### SAMPLE HANDLING AND ANALYTICAL PROCEDURES

Sample handling and analytical methods used in the determination of congener-specific PCB concentrations are described in detail elsewhere (Pier et al., 2002). Briefly, Axys Analytical Services Ltd. in Sidney, British Columbia, Canada, conducted the analyses. Percent lipid determination was performed by standard gravimetric analysis in duplicate on plant tissue extracts, prior to any chromatographic cleanup procedures. Samples were analyzed by gas chromatography/mass spectrometry using a modified method of the U.S. Environmental Protection Agency (EPA 680/8270). Detection limits for individual congeners ranged from 0.001 to 5.3 ng g<sup>-1</sup> (dependent on total PCB concentration in sample). Quality control included the monitoring of accuracy by including a certified reference (NRC standard) or spiked blank and a procedural blank in each batch of 8 samples. Average recoveries of spiked matrices were very good (85–109% for sediment and 85–115% for tissues, respectively). PCBs were undetected in most blanks, and when present (25% of blanks for both sediment and tissue) were at concentrations just above detection; thus, no corrections were made. Precision was assessed using 58 tissue and 19 soil/sediment analytical duplicates. Results for duplicates were consistently within the acceptable range ( $\pm 30\%$  precision).

#### DATING OF SEDIMENT CORES

A single sediment core (Monument Lake [ML]) was selected for detailed chronological analysis of PCBs (by Axys Analytical Services Ltd.) and for Pb-210 alpha spectrometry analysis (by Flett Research Ltd., Winnipeg, Manitoba) following the methods of Eakins and Morrison (1978).

#### COMPUTATIONAL METHODS AND STATISTICAL ANALYSES

Statistical analyses employed the Systat Statistical Software Package (Version 8.0, SPSS Inc., 1998). Reported statistics are geometric means and 95% confidence intervals. The null hypothesis was rejected if  $p \leq 0.05$ . The Student's t-Test was employed to assess whether differences between soil and plant samples from the same locations were significant. Analysis of variance (ANOVA) determined differences in the sum of the PCB congeners (PCB) among samples from different locations. Analysis of covariance (ANCOVA) tested interactions among genera and distance from a contaminated area for PCB in plants. The genus *Eriophorum*, represented by only 1 sample, was excluded from this statistical test.

Of the up to 91 congener peaks reported by Axys Analytical Services Ltd., 69 congeners or groups of congeners (58 peaks) were detected in at least 50% of samples. As a result, total PCB concentrations (PCB) were calculated based on the sum of these congeners. Congener concentrations less than the detection limit were replaced with a random number between the detection limit and one-half the detection limit (Helsel, 1990; Travis and Land, 1990). Soil and sediment data are expressed on a dry-weight basis (ng g<sup>-1</sup> dw), while PCB concentrations in plants are expressed in nanograms per gram lipid (ng g<sup>-1</sup> lipid).

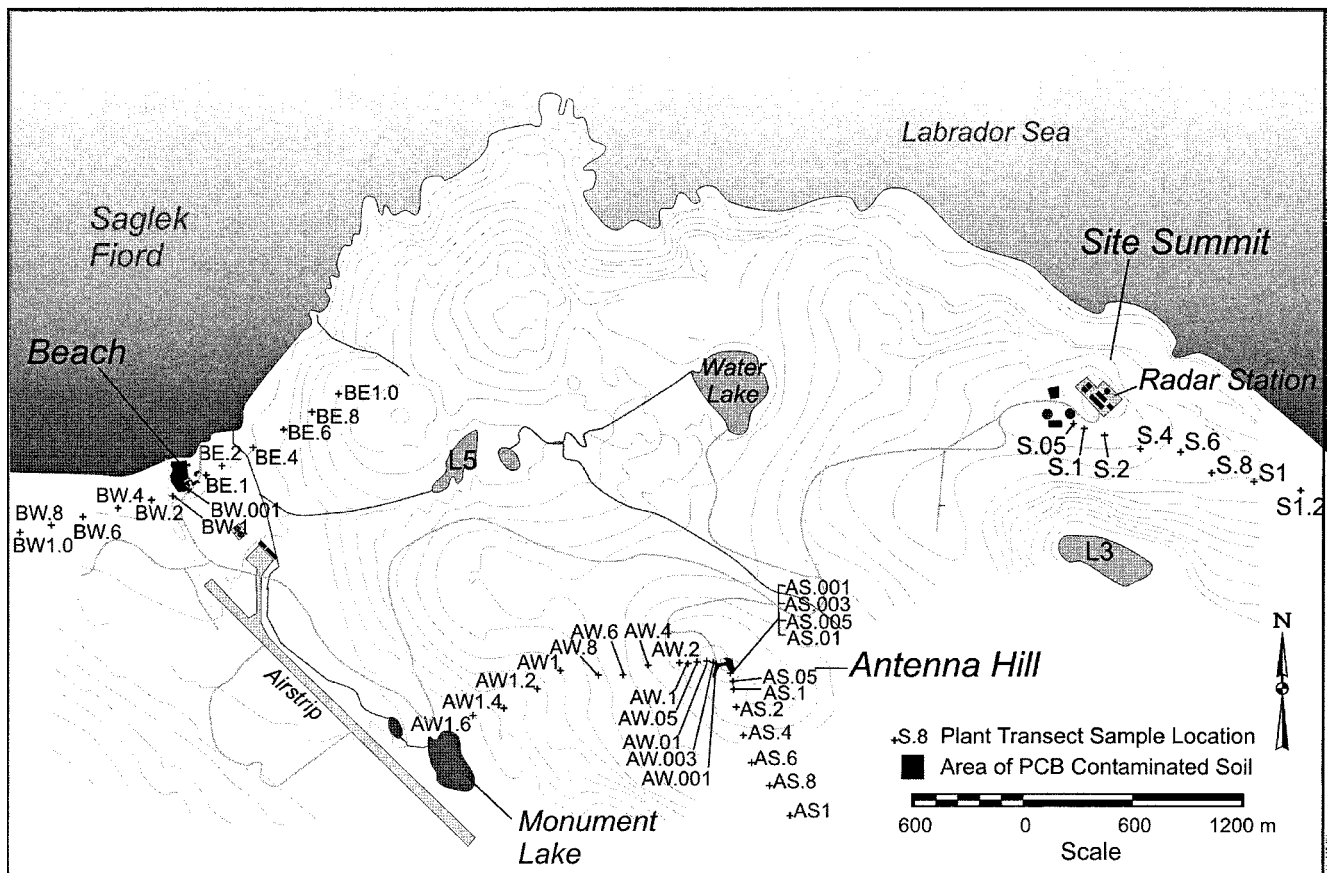


FIGURE 3. Locations where samples under the direct influence of contaminated areas at Saglek were collected. All samples were located within 3 km of one of three contaminated areas: Beach, Antenna Hill, Site Summit. BW = Beach West; BE = Beach East; S = Site Summit; AS = Antenna Hill South; AW = Antenna Hill West transect samples; L = Lake samples.

Distances from a source used in regression analyses were to the closest of three contaminated areas as determined on georeferenced maps created in Autocad Map. Data were  $\log_{10}$  transformed prior to conducting parametric statistics. Regression analyses required that distance measures be  $\log_{10}(x + 1)$  transformed.

For comparison purposes, samples were divided into those within the immediate influence of the local source of PCB contamination at Saglek (<3 km from a contaminated area) and those more distant from the site (>3 km) and therefore capable of receiving inputs of PCBs from Saglek only by aerial transport processes.

## Results

### DOWNCORE ANALYSIS OF PCBs

The Pb-210 profile indicated that the sediment core had not experienced any marked bioturbation or other mixing in the recent sediments and therefore was a standard decay profile. An iterative best-fit model predicted sediment accumulation rates of  $0.0252 \text{ g cm}^{-2} \text{ yr}^{-1}$  ( $r^2 = 1.00$ ). The core covers a period of approximately 200 yr.

PCBs were detected in every sample analyzed from the Monument Lake core. A subsurface peak of  $550 \text{ ng g}^{-1} \text{ dw}$  PCBs was detected 1–1.5 cm down the core, corresponding to around the mid-1970s (Fig. 5). PCB concentrations then declined steadily below 3–3.5 cm (ca. 1945–1950), after which concentrations ( $5.5\text{--}13.3 \text{ ng g}^{-1} \text{ dw}$ ) were comparable to reported background values (Jensen et al., 1997) reported for lake sediments elsewhere in the Canadian Arctic

(Muir et al., 1995, 1996). Congener signatures of all slices were dominated by congener 180 and, in general, resembled the Aroclor 1260 signature, but slices from >5-cm depth exhibited some enrichment with lower-chlorinated congeners.

### OVERVIEW OF TOTAL PCBs IN SURFACE SEDIMENT, SOIL AND PLANTS

Total PCB concentrations in each medium were not significantly influenced by year of collection (ANOVA,  $p > 0.05$ ). Neither soil nor plant sample PCB concentrations were significantly different among the three contaminated areas at Saglek (ANOVA,  $df = 3$ , plants  $p = 0.208$ , soil  $p = 0.358$ ). Differences in PCB among plants of different genera were significant (ANOVA,  $df = 7$ ,  $p < 0.0005$ ), but these differences were accounted for when “distance from a source” was considered as a covariate (ANCOVA,  $df = 6$ , genera  $p = 0.689$ ,  $\log_{10} \text{ distance } p = 0.005$ ).

As expected, PCB concentrations in soil, plant, and sediment samples collected within 3 km of contaminated areas at Saglek were significantly greater than those detected in samples farther removed from the site (Table 3). PCB concentrations in soils collected <3 km from the site (range =  $0.69\text{--}7842 \text{ ng g}^{-1} \text{ dw}$ ) were significantly elevated, exceeding concentrations in soils collected >3 km from the site (range =  $0.146\text{--}79 \text{ ng g}^{-1} \text{ dw}$ ) by a factor of 236 ( $t = -6.158$ ,  $df = 52$ ,  $p < 0.0005$ ; Table 3).

PCB concentrations in plants collected at the site were 242 times the average concentration in plants removed from the direct influence

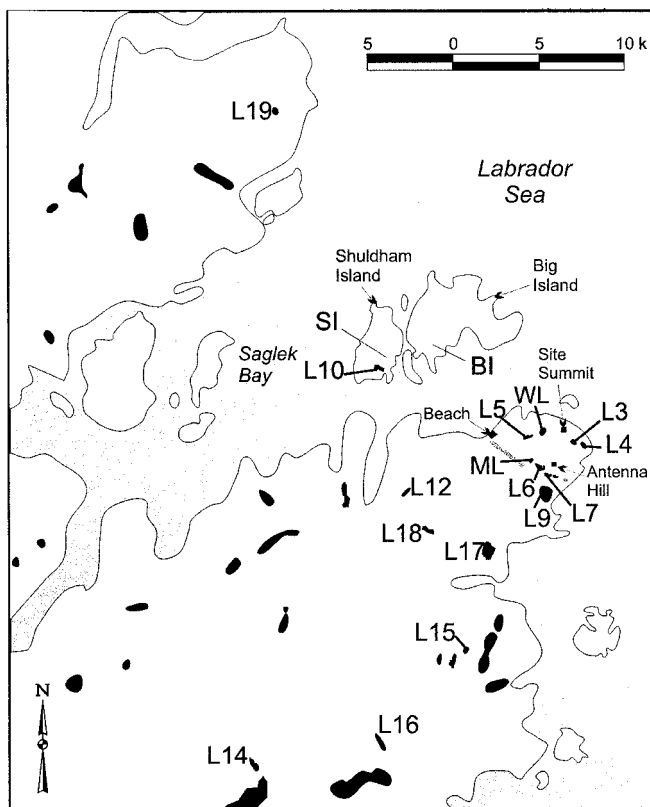


FIGURE 4. Sample locations for lakes (sediments  $n = 16$ ; soils and plants  $n = 11$ ), Shuldham Island ( $n = 1$  for soils and plants) and Big Island ( $n = 2$  soils and plants).

of the site ( $t = -5.616$ ,  $df = 60$ ,  $p < 0.0005$ ). A sample of *Betula glandulosa* collected beside L15 contained the lowest concentration of PCBs overall ( $0.11 \text{ ng g}^{-1}$  lipid). The highest plant PCB concentration ( $12,380 \text{ ng g}^{-1}$  lipid) was detected in a willow (*Salix arctica*) growing within the contaminated area at the beach.

PCB concentrations in surface sediments from lakes at the site (range =  $12\text{--}720 \text{ ng g}^{-1}$  dw) were 48 times mean concentrations detected in sediment from distant lakes (range =  $0.68\text{--}23 \text{ ng g}^{-1}$  dw) ( $t = -6.642$ ,  $df = 14$ ,  $p < 0.0005$ ).

#### COMPARISON OF THE SPATIAL DISTRIBUTION OF PCBs IN SOILS, PLANTS AND SEDIMENTS

##### Total PCBs

Total PCB concentrations in soils, plants, and sediments exhibited similar log-linear relationships to distance from a source (Fig. 6). The slopes of the relationship of PCB to distance were not significantly different among the 3 media (GLM,  $p = 0.607$  for "matrix  $\times$   $\log_{10}$ —distance" interaction), but if distance at which samples were collected was included in the analysis as a covariate, sediments were significantly more contaminated than corresponding soils and plants (ANCOVA,  $df = 2$ ,  $p < 0.0005$ ).

Based on upper 95% confidence limits determined for the regression relationships, several soil and plant samples from the south Antenna Hill and site summit transects had PCB concentrations that were greater than expected based on their distance from a source (Fig. 6a and b). Plants and soils from the south Antenna Hill transect had concentrations of PCBs that were significantly greater than those detected in samples from the west beach and west Antenna Hill transects (ANOVA,  $df = 4$ , plants  $p = 0.023$ , soil  $p = 0.003$ ).

##### Congener Signatures

Variations in the congener signatures of soils, plants, and sediments were examined to provide additional information about the pattern of redistribution of PCBs from Saglek to the surrounding environment. Congener signatures of representative samples are presented in Figure 7. Overall, soil/plant pairs had similar congener profiles, but the signatures of the soil samples tended to be more variable.

PCB signatures of plants growing within 3 km of the site (e.g., Monument Lake in Fig. 7a) included a high proportion of congeners dominant in Aroclor 1260 commercial mixtures, the source of PCBs at Saglek (e.g., 149, 153, 138/163/164, 187/182, 180, and 170/190; Schulz et al., 1989). Plant samples collected at lakes removed from the direct influence of the site (L10, L14, and L19) had signatures that reflected a greater contribution of lower-chlorinated congeners consistent with inputs from long-range atmospheric transport (Fig. 8b, c, and d).

In general, as noted earlier for plants, lake sediment congener signatures exhibited increasing proportions of lower-chlorinated congeners with increasing distance from the site. Sediment from lakes directly connected via drainage pathways to contaminated areas (e.g., ML) had PCB loads clearly influenced by the Aroclor 1260 source but exhibited some enrichment with higher-chlorinated congeners relative to the corresponding plant samples (Fig. 7a).

Sediment congener signatures from L14, L19, and, in particular, L10 departed from the pattern seen in plants (Fig. 7b, c, and d). L10 and L14 sediments contained congeners dominated by lower-chlorinated forms. The signature of the sediment from L10, located just 7.4 km northwest of the contaminated beach, contained a high proportion of lower-chlorinated congeners. The congener signature of the L19 sediment contained several congeners (180, 170/190, 196/203) at concentrations suggestive of the Aroclor 1260 influence. The plant from L19, unlike the co-occurring sediment, contained relatively more lower-chlorinated congeners.

Congener profiles of the samples were further compared using Principal Components Analysis (PCA). PCA generated three principal components that explained 91% of total variance in the data (68%, 17%, and 6%, respectively). Plots of sample scores on PC(1) and PC(2) produced two groupings in which sediments were separated from soil and plant samples.

Despite the observed separation of sediment from plants and soils by PC(1), samples were distributed more evenly when plotted based on their PC(2) and PC(3) scores (Fig. 8). In this manner, soils separated into two loose groupings (Fig. 8a). Most of the soils from the beach and south Antenna Hill transects scored high on PC(2), whereas soils from the west Antenna Hill transect, the lakes, and Big Island scored low on this component.

Plant samples separated into four fairly distinct groups (categorized in Fig. 8b according to the location from which most samples originated). South Antenna Hill transect samples plotted beside the Aroclor 1260 congener signature, reflecting a tighter relationship between the congener signatures of plants from this transect and the source of contamination than was exhibited by plants from any other transect. West Antenna Hill transect samples and L4 and L6 plants plotted together and Monument Lake, Big Island, and the beach transect samples formed a third group. Finally, plant samples collected at the greatest distances from the site (5.6–27 km) scored lower in both components.

Sediment samples, like plants, separated into four distinct groups based on their PC(2) and PC(3) scores (Fig. 8c). The sediments from lakes receiving drainage directly from contaminated areas (ML, WL, L3, and L5) scored highest in PC(2) and closest to the Aroclor 1260 signature. Sediments from all other lakes scored low in PC(2) but fell into three distinct groups based on their PC(3) scores. Lakes at the site,

TABLE 2  
Physical and environmental characteristics of the 16 study lakes

Lake Name	Location	Distance from Source (km)	Sediment PCB (ng g <sup>-1</sup> )	Altitude (m)	Max. Depth (m)	Secchi Depth	Surface Water Temp. (°C)	pH*	Sp. Cond. (µS)	% Veg. Cover
Water	N58-29.30'									
Lake Monument	W62-37.15'	1.2	20	229	7.5	7.5	13	6.8	67.40	75
L3	N58-28.12'									
L3	W62-39.20'	1.4	385	76	4.0	4	11	6.5	86.00	65
L3	N58-29.50'									
L3	W62-35.10'	0.72	720	152	25	>15	15	6.9	66.50	75
L4	N58-28.30'									
L4	W62-34.30'	1.3	140	168	22	11	14	6.8	31.50	70
L5	N58-29.10'									
L5	W62-38.30'	1.4	180	137	6.5	6.5	15	7.0	98.00	85
L6	N58-28.00'									
L6	W62-37.45'	1.4	53	76	6.0	6.0	8	7.1	48.30	75
L7	N58-27.45'									
L7	W62-37.00'	1.4	12	76	8.5	6.3	10	6.5	31.60	90
L9	N58-27.15'									
L9	W62-36.10'	2.2	23	46	5.0	5.0	11	6.5	61.70	95
L10	N58-31.10'									
L10	W62-46.45'	7.4	5.1	15	2.5	2.5	13	7.0	252.70	100
L12	N58-27.374'									
L12	W62-44.888'	5.6	0.87	274	11	10	11	6.2	53.00	15
L14	N58-18.525'									
L14	W62-54.907'	25	4.2	305	11	7.0	13	6.6	17.10	80
L15	N58-21.737'									
L15	W62-42.360'	15	2.1	259	4.0	4.0	14	6.1	—	55
L16	N58-19.135'									
L16	W62-46.622'	20	0.87	259	12	11.2	12	6.4	—	65
L17	N58-25.451'									
L17	W62-40.641'	6.7	0.68	107	7.8	7.8	12	5.9	—	80
L18	N58-25.428'									
L18	W62-44.470'	8.0	1.6	305	16	12	11	5.8	—	10
L19	N58-40.810'									
L19	W62-53.778'	27	1.7	396	30.0	16	12	5.9	—	5

\* pH = average of three measurements.

but lacking a direct connection via surface drainage paths to one of the contaminated areas (L4, L6, L7, and L9), scored highest in PC(3). Six of the sediment samples from lakes located between 5.6 and 27 km from the site had intermediate scores on PC(3), but sediments from L10 and L14, which are located 7.4 and 25 km, respectively, from the site, had much lower scores in PC(3) and were thereby grouped separately from the other lake sediments.

Congener loadings scores on PC(2) and PC(3) indicate that samples scoring higher in PC(2) have signatures in which congeners making the greatest contribution to the Aroclor 1260 signature predominate (138/163/164, 153, 158, 180, 170/190) (Fig. 8d) and confirms that sediments from the connected lakes (ML, WL, L3, and L5) were enriched with these congeners. Conversely, samples scoring low on PC(2) contain a higher proportion of lower-chlorinated congeners.

## Discussion

### SEDIMENT CORE PCB PROFILE

Sediment cores are often used for long-term monitoring programs because they provide an invaluable historical record of contaminant inputs to the local environment. In the current investigation, we used the sediment core from Monument Lake to examine the pattern of PCB

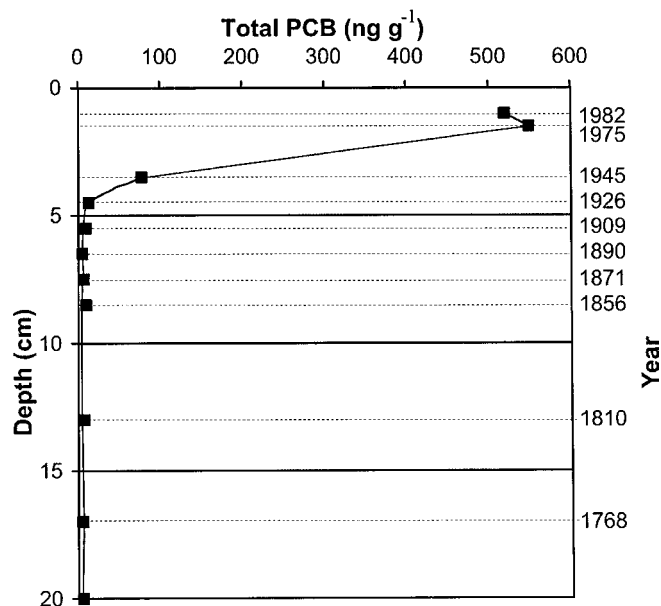


FIGURE 5. Monument Lake core PCBs, associated depths, and Pb-210 dates.



TABLE 3

Total PCB concentrations (sum of 69 congeners) in freshwater lake sediment, terrestrial soil, and plants within 3 km of Saglek, Labrador, and in locations up to 27 km away. Values are geometric means and 95% confidence intervals (in parentheses). Statistical comparisons were by ANOVA within each matrix

Location	Soil			Plants			Sediment <sup>a</sup>		
	N	PCB (ng g <sup>-1</sup> dw)	P < 0.05	N	PCB (ng g <sup>-1</sup> lipid)	P < 0.05	N	PCB (ng g <sup>-1</sup> dw)	P < 0.05
Antenna Hill	23	340 (140–810)	a	24	130 (51–340)	a	—	—	—
Site Summit	8	104 (24–451)	a	8	48 (9–250)	a	—	—	—
Beach	12	44 (13–146)	a	16	45 (14–140)	a	—	—	—
Lake Samples <sup>b</sup>	3	36 (3–393)	ab	4	4.8 (0.5–48)	ab	8	82 (40–170)	—
<3 km from Saglek	46	100 (59–185)	a	52	63 (33–120)	a	8	82 (40–170)	a
>3 km Distant	8	0.44 (0.11–1.7)	b	10	0.26 (0.06–1.14)	b	8	1.7 (0.8–3.5)	b

<sup>a</sup> Data obtained from top section of each core.

<sup>b</sup> Soils and plants were collected at Monument Lake (ML), L4, L5, L6, L10, L12, L14, L15, L16, L17, L19; 3 additional plant were collected on Shuldham Island (n = 1) and Big Island (n = 2).

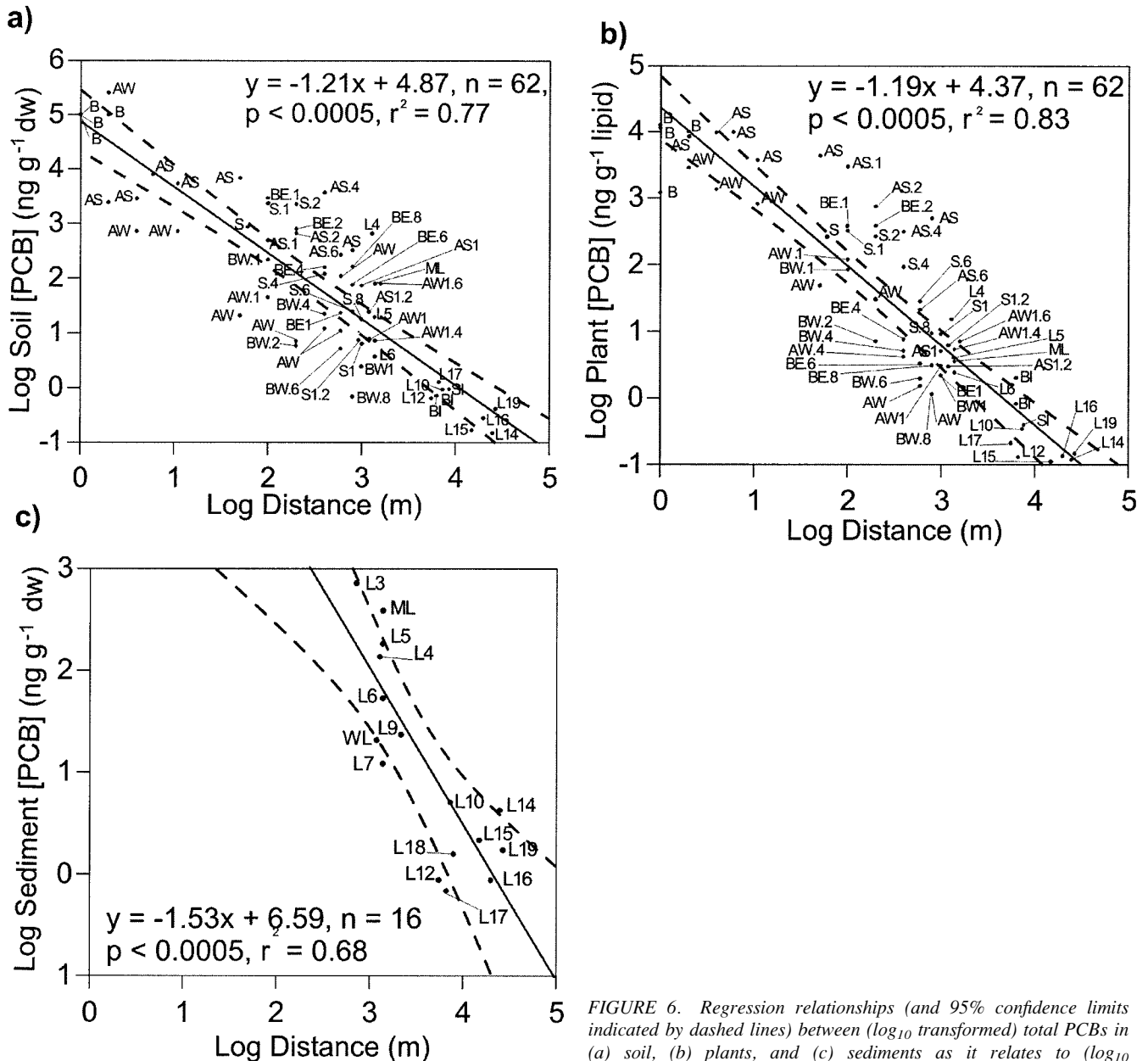


FIGURE 6. Regression relationships (and 95% confidence limits indicated by dashed lines) between ( $\log_{10}$  transformed) total PCBs in (a) soil, (b) plants, and (c) sediments as it relates to ( $\log_{10}$  transformed) distance from a source.

inputs to the environment surrounding Saglek over the estimated 200-yr period that the core encompassed. PCBs were detected in all of the core samples analyzed, including those corresponding to years predating the manufacture of PCBs. Lead-210 dating of the core indicated that it had not experienced any marked bioturbation or other mixing in the recent sediments. Very high concentrations of PCBs in the uppermost sediment layers appear to have diffused downward into the earlier-deposited sediments (Gunkel and Nolte, 1994; Schindler et al., 1995; Muir et al., 1996). Enrichment of layers deeper in the core with less hydrophilic, lower-chlorinated congeners supports this theory.

PCBs were introduced in large quantities to the North with the construction of the DEW Line and associated facilities such as the Polevault Line. This introduction corresponds with a global increase in PCB manufacture and usage. As a result, it is not possible to differentiate between the relative influence of short- versus long-range transport of PCBs on local contaminant loads from gross measurements of total PCBs alone. Examination of congener profiles, as demonstrated in the current investigation, is necessary to determine the dominant pathway of contaminant input.

#### SOILS, PLANTS AND SEDIMENTS AS INDICATORS OF ENVIRONMENTAL CONTAMINATION

Evidence of the Aroclor 1260 commercial PCB mixture that contaminated soil at Saglek, Labrador, was present in soils, plants, and surficial freshwater lake sediments up to 27 km away from the site. Concentrations of congeners reflecting the Aroclor 1260 signature (149, 132/153 138/163/164, and 180) exceeded those typically seen (DIAND, 1997) in samples receiving inputs only from long-range transport, even in the samples collected 25 and 27 km from the site (L14 and L19, respectively). Therefore, the halo of PCB contamination around Saglek may extend up to 27 km from the site (i.e., a halo with diameter >50 km) and is larger than has been demonstrated previously for contaminated sites in the Arctic (Bright et al., 1995; Dushenko et al., 1996).

Variations in congener signatures among the three sample media revealed differences in their ability to reflect ambient contaminant concentrations. In general, soils and plant congener signatures were dominated by the Aroclor 1260 congeners in samples collected near the site and exhibited progressively higher proportions of lower chlorinated congeners with increasing distance from the source. Overall, however, soil congener signatures were more variable than those of plants.

Higher concentrations of PCBs detected in sediments as compared to the corresponding soils or plants reflect the tendency of lake sediments to receive relatively greater amounts of PCBs during snowmelt and rain events that transport particulate-bound PCBs from the area constituting a lake's entire watershed (Schindler et al., 1995). Not surprisingly, sediments from lakes directly receiving drainage from the contaminated areas (ML, WL, L3, and L5) had the highest PCB concentrations and congener profiles consistent with Aroclor 1260 (Figs. 6c and 8c). Other site lakes not directly connected to a contaminated area (L4, L6, L7, and L9) generally had lower PCB concentrations and were enriched in higher-chlorinated congeners (Fig. 8c). These hydrophobic, highly chlorinated congeners have a greater tendency to be bound to sediments relative to hydrophilic, lower-chlorinated congeners that more easily partition into the water column (Karickhoff et al., 1979; Burgess et al., 1996). Lakes located more than 3 km from Saglek (with the exception of L10 and L14) have very similar congener compositions indicative of similar PCB inputs.

Sediments from L14 and L19 had congener distributions that were dissimilar despite their collection nearly equal distances southwest and northwest, respectively, from the site. L14 sediments contained

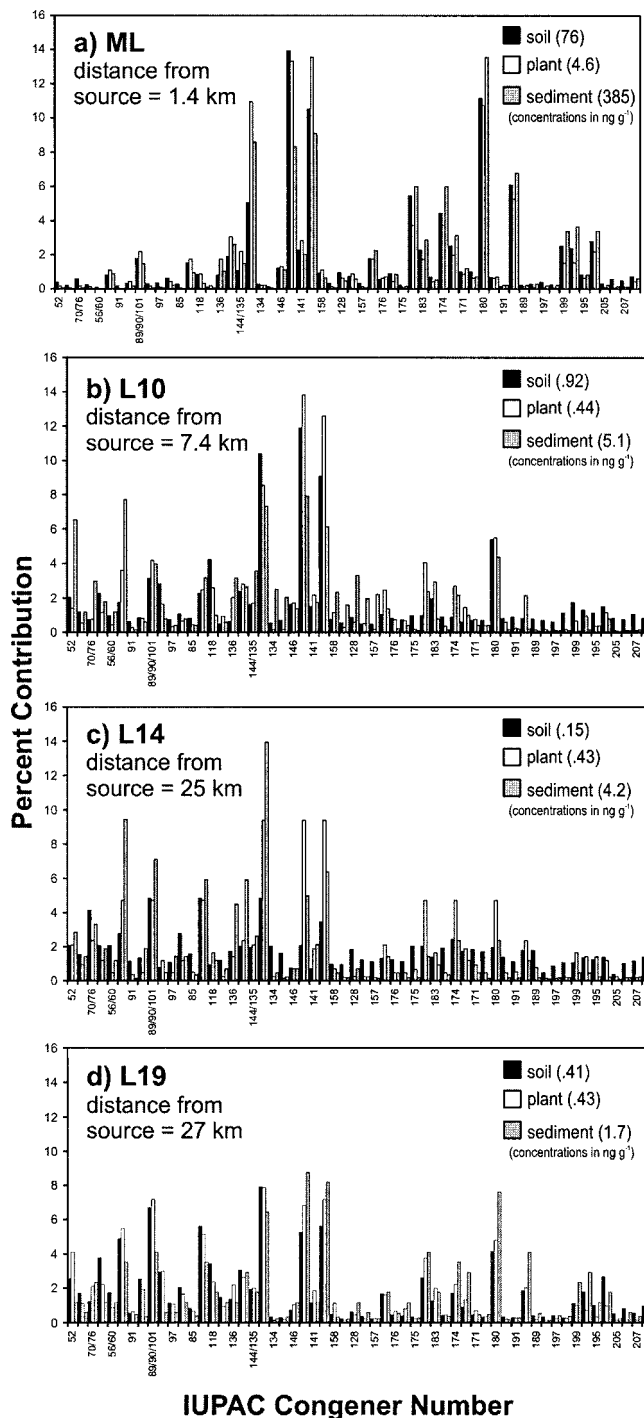


FIGURE 7. PCB congener compositions (percent contribution to totals) of representative plant and sediment samples from Saglek, Labrador, and environs. Total PCB concentrations for each matrix are given in parentheses.

congeners dominated by lower-chlorinated forms (Fig. 8c), similar to signatures typical of lakes receiving PCB inputs primarily by long-range transport. In contrast, the L19 sediment contained several congeners (132/153, 138/163/164, 180, and 170/190) at relatively higher proportions suggestive of the Aroclor 1260 influence. The differences in the sediment congener profiles reflect a greater influence of the local source at Saglek on L19, potentially due to large differences in the topography surrounding both lakes. The cirque in which L19 is located may funnel wind currents from the south into the lake,

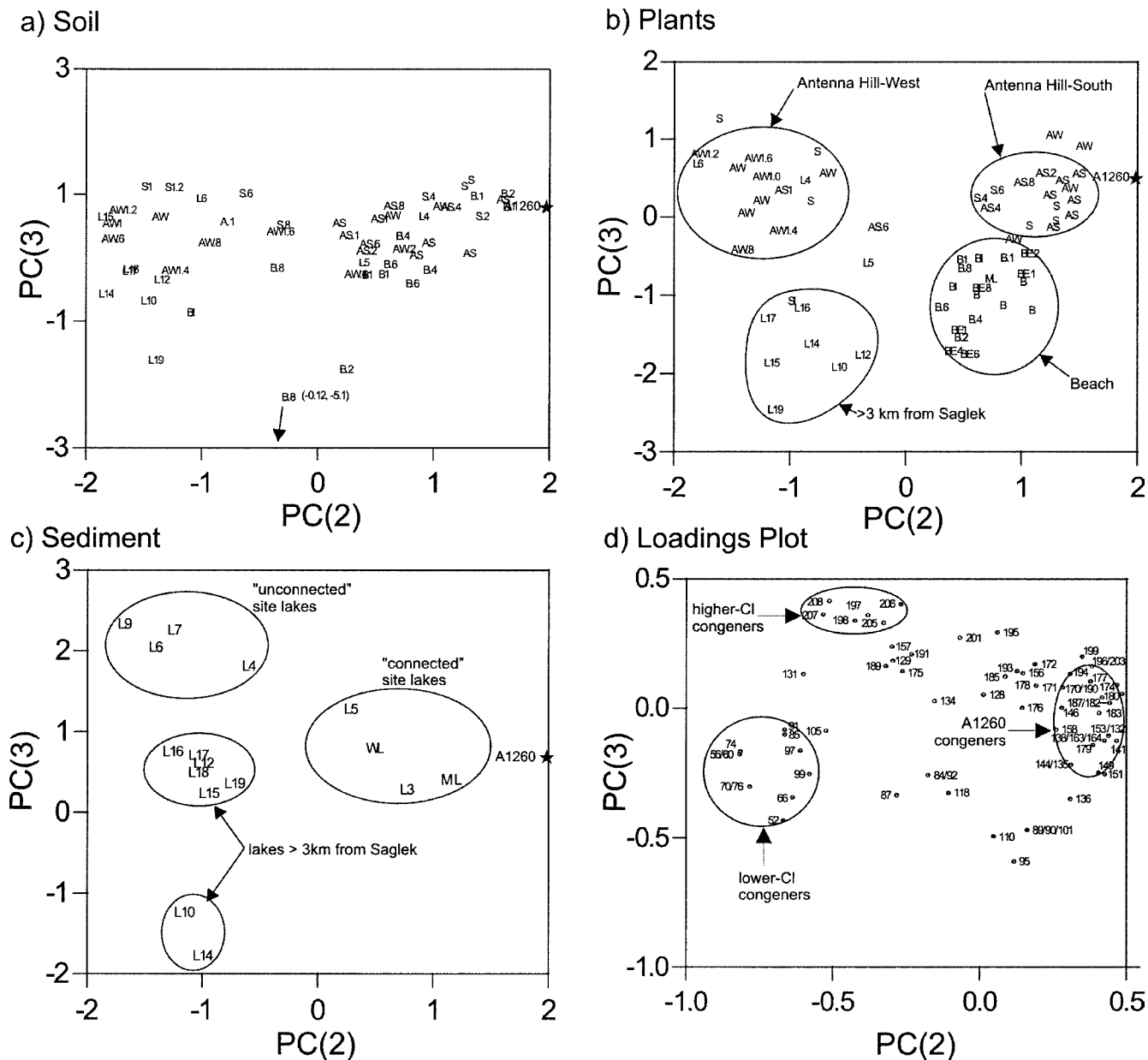


FIGURE 8. Principal components scores and loadings plot (d) of the congener composition of (a) soil, (b) plants, and (c) freshwater lake sediments near Saglek. Samples are coded according to lake name/number or location where samples were obtained (AW = Antenna Hill west, AS = Antenna Hill south, B = beach, BI = Big Island, ML = Monument Lake, S = Site Summit, SI = Shuldhham Island, WL = Water Lake). Numbers following B, AW, AS, and S sample designations represent distance in kilometers from the nearest source.

whereas land rising to heights greater than 600 m between Saglek and L14 has likely reduced the influence of the site on this lake over the long term.

L10 sediments contained a much higher proportion of lower-chlorinated congeners than expected from their distance from the source alone (Fig. 8c). This discrepancy is not easily attributed to a single phenomenon. This sample likely included sediment deposited over double the time period of sediments from the other lakes (it was from the top 2 cm of the core, whereas all other samples were from the top 1 cm of deposited lake sediments). The water in L10 had a higher specific conductance and salinity than any of the other lakes sampled (Table 2); qualitative assessment suggested that the sediments contained a higher proportion of organic carbon; and the water in L10 likely had higher concentrations of dissolved organic carbon (DOC), as suggested by its brown color. It is known that patterns of surface runoff from the

surrounding watershed, lake DOC content (MacDonald and Metcalfe, 1989), and differences in watershed character (geology, surface area, aspect, percent vegetative cover) (Schindler et al., 1995; Paterson et al., 1998) all influence the sequestering of organic compounds.

PCB concentrations in plants decreased with distance from contaminant source, both along transects within the site and on a more regional scale (Fig. 6b); indeed, plants showed a stronger relationship than either soils or sediments (Figs. 6a and 6b). PCBs were generally found at higher concentrations (Fig. 6b), and with a congener profile similar to Aroclor 1260 (Fig. 8b), in plants obtained from those ends of the transects in closest proximity to the source. The congener signature of many of the plant samples from Antenna Hill west were enriched with more highly chlorinated congeners (Fig. 8b); this higher level may reflect a combination of drainage and wind patterns. All of the plants collected adjacent to lakes located more than 3 km from the site

(L10, L12, L14, L15, L16, L17, and L19) formed a distinct cluster and are relatively enriched with lower-chlorinated congeners. The plants, unlike the sediments, appear to be similarly influenced by a combination of long- and short-range transport and therefore appear to be better indicators of ambient PCB concentrations.

## Conclusions

The current study of PCB concentrations in terrestrial plants, soils, and freshwater lake sediments collected near Saglek, Labrador, suggests that the environment within 27 km of the site has been influenced by the presence of this local source of PCB contamination. Soil and sediment samples exhibited contaminant loads that reflected differences in exposure such as the duration over which material was deposited, location relative to drainage pathways connected to contaminated areas, and, in the case of sediments, the size and character of the lake's watershed.

Sediments are often used in environmental studies because they are invaluable for assessing changes in the patterns of historical inputs. However, they may not provide the resolution necessary to assess recent or more short-term environmental contamination. Plants, on the other hand, consistently reflect ambient PCB exposures. Annual leaf-drop effectively wipes the slate clean each year, thereby eliminating interseason variability.

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