

# Polychlorinated biphenyl congeners in soils and lichens from King George Island, South Shetland Islands, Antarctica

HYUN PARK<sup>1</sup>, SANG-HWAN LEE<sup>2</sup>, MINKYUN KIM<sup>3</sup>, JEONG-HOON KIM<sup>1</sup> and HYOUN SOO LIM<sup>1\*</sup>

<sup>1</sup>Korea Polar Research Institute, Korea Ocean Research and Development Institute, Songdo-dong 7-50, Yeosu-gu, Incheon 406-840, Korea

<sup>2</sup>Technology Research Center, Mine Reclamation Corporation, Seoul 110-727, Korea

<sup>3</sup>Research Institute of Industrial Science & Technology, Pohang 790-600, Korea

\*tracker@kopri.re.kr

**Abstract:** The levels and distribution of polychlorinated biphenyl (PCB) congeners were analysed in fourteen soil and eight lichen (*Usnea aurantiaco-atra*) samples from King George Island, West Antarctica. A total of 32 PCB congeners were found in five soil samples collected in 2006, and the mean concentration of total PCBs was 20.4 pg g<sup>-1</sup> dry weight (range, 8.0–33.8 pg g<sup>-1</sup> dry weight). The most abundant PCB isomers in soil samples were di-, tri-, and penta-CBs, which accounted for more than 75% of the total residues. Twelve dioxin-like PCBs were also detected in nine soil and eight lichen samples, and the levels of dioxin-like PCBs were 5-fold higher in lichens than in soil. PCBs were detected at very low levels in most soil and lichen samples. The highest congener concentrations were found for PCB 118 (6.63 and 21.93 pg g<sup>-1</sup> in soil and lichen, respectively) among dioxin-like PCBs. PCB levels in air samples were highly correlated with those in soil and lichen samples, as were PCB levels in soil and lichen samples collected at the same site. Long-range atmospheric transport is thought to be the main source of PCBs on King George Island. However, PCB levels in soil and lichen samples were also apparently influenced by local sources of PCBs.

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**Key words:** environmental contamination, persistent organic pollutants, *Usnea aurantiaco-atra*

## Introduction

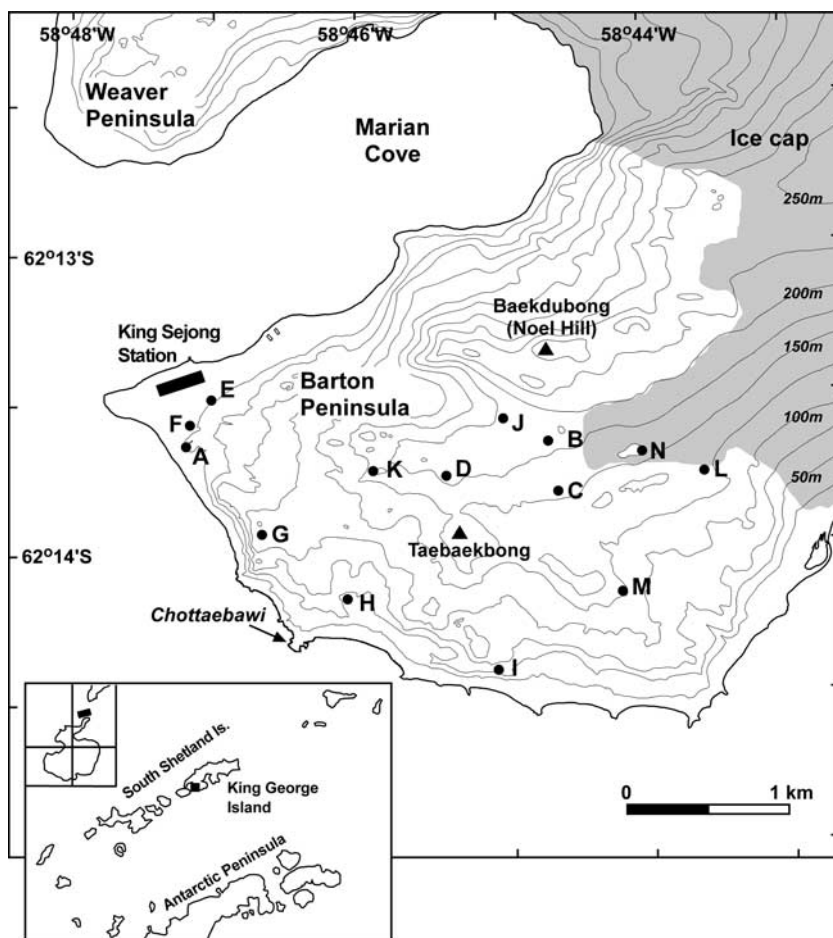
Polychlorinated biphenyls (PCBs) are among the major persistent organic pollutants (POPs). In the past, commercial mixtures of PCBs were widely used in numerous industrial applications, including the production of dielectric fluid for electrical transformers, flame retardants, plasticizers, resins, waxes, paints, copy paper, and sealants for wood and cement surfaces (FAO/UNEP 1992). PCB congeners are stable, persistent, highly lipophilic, and show particle reactivity and low vapour pressure. As a result, these toxic compounds are ubiquitously distributed throughout both aquatic and terrestrial environments and tend to bioaccumulate through the food chain, thus threatening ecosystems and human health.

PCBs were first reported in Antarctica in the 1960s and 1970s (Risebrough *et al.* 1968, 1976). Their presence in this relatively isolated region is thought to be the result of long-range transport in the atmosphere, precipitation, and cold condensation (Wania & Mackay 1993, Loganathan & Kannan 1994). On the other hand, several studies pointed out the pollution from local sources of POPs around research stations (Risebrough *et al.* 1990, Larsson *et al.* 1992, Borghini *et al.* 2005, Klanova *et al.* 2008). Therefore, increasing PCB levels in various environmental matrices in Antarctica are a matter of growing concern. Several reports have documented PCB levels in Antarctic soil, seawater,

and sediment (Fuoco *et al.* 1996, Montone *et al.* 2001b, Negoita *et al.* 2003), as well as in various organisms from various trophic levels (Reinhardt & Van Vleet 1986, Focardi *et al.* 1995, Montone *et al.* 2001a, Weber & Goerke 2003).

The accumulation of PCBs occurs mainly in soil, which plays an important role in their global fate, and this process occurs via mass flow in the atmosphere. In addition, vegetation is the primary site of bioaccumulation via the uptake of lipophilic organic compounds from the air into plant tissues. In Antarctica, the distribution of terrestrial vegetation is limited to ice free areas, mainly coastal regions, and biodiversity is restricted to mostly lichens and mosses. Lichens are slow-growing organisms with large surface areas that facilitate the direct absorption of pollutants from the atmosphere. Therefore, lichens have been widely used for biomonitoring air pollution. However, there have been few studies on PCBs in Antarctic lichens (Focardi *et al.* 1991, Negoita *et al.* 2003).

This study was performed to determine the distribution and concentration profiles of PCBs in the air, soil, and lichen collected in the maritime Antarctic. To evaluate the potential risk to the Antarctic ecosystem, the concentrations of 12 dioxin-like PCB congeners listed by the World Health Organization (WHO) were measured in soil and in the lichen *Usnea aurantiaco-atra* (Jacquinot) Bory, which has



**Fig. 1.** Locations of the study area and sampling sites. For details refer to Table I.

**Table I.** Description of samples and sampling locations.

Site	Sampling location (GPS)	Material <sup>a</sup>	Description
A	62°13'37.7"S; 58°47'15.6"W	soil	380 m from the station. Brown soil. Fruticose lichens are dominant.
B	62°13'38.2"S; 58°47'37.7"W	soil	Dark brown soil. No vegetation. Collected from the centre of sorted circles.
C	62°13'48.3"S; 58°47'39.5"W	soil	Dark brown soil. Collected from centre of sorted circles. Lichen and moss are abundant.
D	62°13'43.4"S; 58°47'25.8"W	soil	Dark brown soil. Covered by volcanic breccias. Fruticose lichens are common.
E	62°13'30.7"S; 58°47'6.8"W	soil	240 m from the station. Light brown soils in slope. Fruticose lichens dominant.
F	62°13'33.6"S; 58°47'16.9"W	soil	290 m from the station. Olive gray soil. Collected from the centre of sorted circles.
G	62°13'55.6"S; 58°47'45.1"W	lichen	Moss and fruticose lichen are abundant.
		soil	Light brown soil. Collected from the center of sorted circles. Fruticose lichens are dominant.
H	62°13' 9.8"S; 58°47'8.5"W	soil	Light reddish brown soil in slope. Soil sample was collected from the newly exposed surface.
		lichen	Sea-birds feathers and droppings are common (near the penguin rookery)s.
I	62°13'23.6"S; 58°47'3.7"W	soil	Dark brown soil. Moss is dominant (wet condition). Frequent by seabirds and penguind.
		lichen	
J	62°13'34.6"S; 58°47'0.9"W	soil	Dark brown soil. Covered by fruticose lichen. Flat area between Seorakbong and Sabong.
		lichen	
K	62°13'43.2"S; 58°47'55.4"W	soil	Dark brown soil. Collected from centre of sorted circles. Fruticose lichens are dominant.
		lichen	
L	62°13'44.8"S; 58°47'35.2"W	soil	Brown soil. Covered by volcanic breccias. Fruticose and crustose lichens are common.
		lichen	Near the ice cap (1 km from the Potter Cove).
M	62°13'8.0"S; 58°47'11.7"W	soil	Dark brown soil. Fruticose lichens are dominant.
		lichen	
N	62°13'40.9"S; 58°47'2.5"W	soil	Near the ice cap. No vegetation. Covered by volcanic rocks. Not frequented by birds.

<sup>a</sup>lichen: *Usnea aurantiaco-atra*.

a circumpolar distribution with the greatest abundance along the Antarctic Peninsula.

## Materials and methods

### Sample collection

King George Island, the largest island in the South Shetland Islands, is located *c.* 120 km north of the Antarctic Peninsula (Fig. 1). Most of the island is covered with glaciers, and outcrops are exposed only along the shorelines in restricted areas. The island has a cold oceanic climate and with an average annual temperature of  $-1.8^{\circ}\text{C}$  to  $-1.6^{\circ}\text{C}$ , relative humidity of 89%, precipitation of 437.6 mm, and a mean wind velocity of  $7.9\text{ m s}^{-1}$ , predominantly from the north-west and south-west, as recorded at the Korean Antarctic Research Station (King Sejong Station,  $62^{\circ}13'\text{S}$ ,  $58^{\circ}47'\text{W}$ ) on Barton Peninsula (Lee *et al.* 2007). Most of the ice free area is covered by relatively abundant vegetation, dominated by cryptogamic species. The flora includes two flowering plants, 33 bryophytes, and 62 lichens (Lee 1992, Kim *et al.* 2006). The Chottaebawi area on the southern coast of Barton Peninsula is one of the most important penguin colonies in the maritime Antarctic region (Fig. 1). In addition, the cape area near the King Sejong Station is populated by seabirds, mainly south polar skua (*Catharacta maccormicki* Saunders) and penguins (*Pygoscelis papua* Forster and *Pygoscelis antarctica* Forster *etc.*). In total, 14 soil samples were collected near the King Sejong Station. Five soil samples for the analysis of all PCB congeners were collected in January 2006, and nine additional soil samples and eight lichen samples were collected for the analysis of 12 dioxin-like PCB congeners in January 2008 in the ice free area (Fig. 1). After collection, all samples were stored at  $-20^{\circ}\text{C}$  until extraction. Locations and detailed descriptions of sampling sites are shown in Table I and Fig. 1.

### Analytical procedure

Samples were treated, extracted, and analysed according to the US Environmental Protection Agency method 1668A (US EPA 1999). Samples of approximately 50 g of soil were spiked with  $^{13}\text{C}$ -labelled PCB internal standards (1, 3, 4, 15, 19, 37, 54, 77, 81, 104, 105, 114, 118, 123, 126, 155, 156, 157, 167, 169, 188, 189, 202, 205, 206, 207, and 209), and extracted for 16 h using a hot Soxhlet manifold with 300 ml of toluene. The extract was collected and concentrated to *c.* 2 ml using a rotary evaporator, and further purified using a column filled with silica gel. The column consisted of (from bottom to top) quartz glass wool, 0.9 g of silica gel, 3 g of 2% KOH silica gel, 0.9 g of silica gel, 4.5 g of 44%  $\text{H}_2\text{SO}_4$  silica gel, 6 g of 22%  $\text{H}_2\text{SO}_4$  silica gel, 0.9 g of silica gel, 3 g of 10%  $\text{AgNO}_3$  silica gel, and 6 g of sodium sulphate. The column was washed with 50 ml of hexane immediately prior to use. The extract was passed through the column and eluted with 120 ml of hexane.

**Table II.** Concentrations of PCB congeners (pg/g dry weight) in five soil samples from Barton Peninsula<sup>a</sup>.

Congener	Site				
	A	B	C	D	E
MonoCB1	0.470	0.674	0.449	0.329	0.539
MonoCB2	0.194	0.268	0.200	0.120	0.233
MonoCB3	0.755	1.141	0.895	0.553	1.012
DiCB4	1.324	2.255	0.782	0.671	1.538
DiCB15	2.311	3.345	2.014	1.149	3.359
TriCB18	3.700	6.517	2.893	1.305	5.122
TriCB19	0.411	0.842	0.367	0.238	0.487
TriCB28	4.679	10.755	4.392	1.464	8.873
TriCB37	2.008	2.092	0.839	0.312	2.333
TetraCB52	2.109	2.563	1.053	0.393	1.303
<b>TetraCB77</b> <sup>b</sup>	0.303	0.207	0.063	0.082	ND
<b>TetraCB81</b>	0.020	ND	ND	ND	0.058
PentaCB101	1.104	0.817	0.414	0.196	ND
<b>PentaCB105</b>	0.414	0.381	0.107	0.069	0.450
PentaCB111	1.004	ND	0.598	0.329	0.000
<b>PentaCB114</b>	0.600	0.055	0.205	0.199	0.118
<b>PentaCB118</b>	0.753	0.765	0.244	0.156	1.440
<b>PentaCB126</b>	ND	0.063	ND	ND	ND
HexaCB128	0.121	0.064	0.032	0.050	0.306
HexaCB153	0.512	0.337	0.158	0.117	0.327
HexaCB155	0.035	0.053	0.026	0.014	ND
<b>HexaCB156</b>	0.071	0.195	0.021	0.021	0.036
<b>HexaCB157</b>	0.027	0.037	0.000	0.011	0.260
<b>HexaCB167</b>	0.064	0.087	0.015	0.016	0.052
<b>HexaCB169</b>	0.030	0.121	0.047	0.043	0.044
HeptaCB178	0.018	ND	ND	ND	ND
HeptaCB180	0.300	0.030	0.076	0.098	0.040
<b>HeptaCB189</b>	0.015	ND	ND	ND	ND
OctaCB202	0.015	0.016	0.011	0.006	0.017
NonaCB206	0.018	0.051	ND	ND	0.123
DecaCB209	0.092	0.121	0.223	0.079	0.000
$\Sigma\text{PCB (pg g}^{-1}\text{)}$	23.476	33.853	16.123	8.019	28.420
$\Sigma\text{dioxin-like PCB}$	2.298	1.912	0.701	0.598	0.893

<sup>a</sup>ND = Not detected. <sup>b</sup>Coplanar (dioxin-like) congeners are shown in bold.

The eluate was concentrated to 5 ml using a rotary evaporator and then reduced to 0.5 ml under a gentle stream of nitrogen gas. Finally, 1.2 ng of  $^{13}\text{C}_{12}$ -labelled injection standards (91, 521, 1011, 1381 and 1941) were added as internal standards prior to analysis. Dioxin-like PCB congeners were identified and quantified by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS) on a DFS mass spectrometer (Thermo Fisher Scientific, Bremen, Germany) using EPA method 1668A (US EPA 1999) for PCB congeners. The mass spectrometer was operated in electron impact mode (36 eV) with helium gas as a carrier. The GC column was a DB-5MS fused silica column (60 m  $\times$  0.32 mm i.d., 0.20  $\mu\text{m}$  film thickness). The column oven temperature was programmed to increase at a rate of  $20^{\circ}\text{C min}^{-1}$  from an initial temperature of  $120^{\circ}\text{C}$  (3 min hold) to a temperature of  $220^{\circ}\text{C}$  (5 min hold), then at  $4^{\circ}\text{C min}^{-1}$  to  $260^{\circ}\text{C}$  (17 min hold). The injector, transfer line, and ion source were all at a temperature of  $260^{\circ}\text{C}$ . The identification of dioxin-like PCBs was performed based on the retention times of the  $^{13}\text{C}$ -labelled

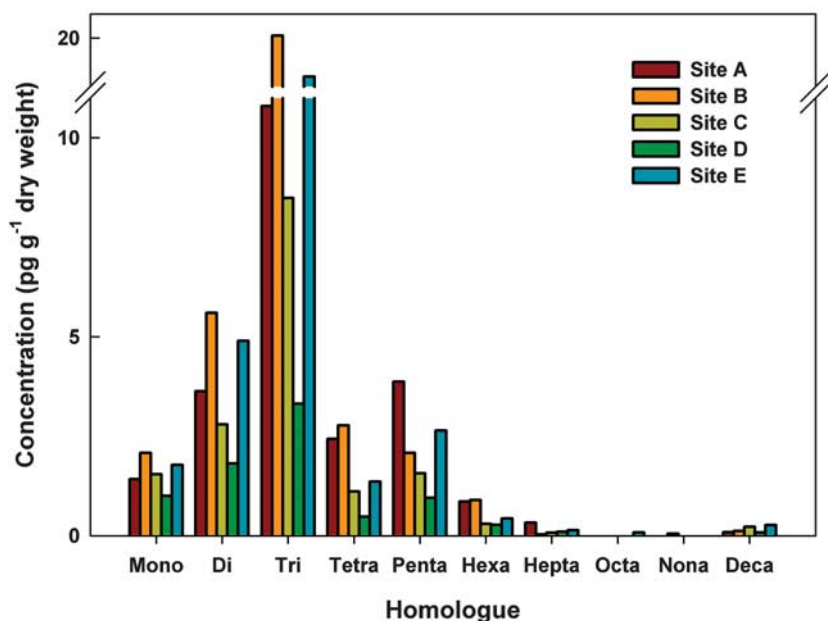


Fig. 2. PCB homologue patterns in Antarctic soil.

standard and isotope ratios  $M/(M+2)$  or  $(M+2)/(M+4)$ . The concentrations of native analytes with corresponding  $^{13}\text{C}$ -labelled surrogate standards were calculated using the isotope dilution method of quantification based on EPA method 1668A (US EPA 1999). The average recovery of internal standards was  $89 \pm 27\%$ . The limits of detection for selected major PCBs found in samples were  $0.001\text{--}0.041 \text{ pg g}^{-1}$  dry weight.

The total carbon (TC) content of the nine soil samples collected in 2008 were analysed using an element analyser (FlashEA 1112; Thermo Fisher Scientific) by measuring  $\text{CO}_2$  generated by combustion at  $950^\circ\text{C}$ . The total inorganic carbon (TIC) content was analysed using a  $\text{CO}_2$  coulometer (UIC Inc, Joliet, IL, USA) by measuring the  $\text{CO}_2$  gas generated by reaction of *c.* 50 mg of powdered bulk samples with 42.5% phosphoric acid at  $80^\circ\text{C}$  for 10 min. The total organic carbon (TOC) content was determined from the difference between the TC and TIC content.

## Results and discussion

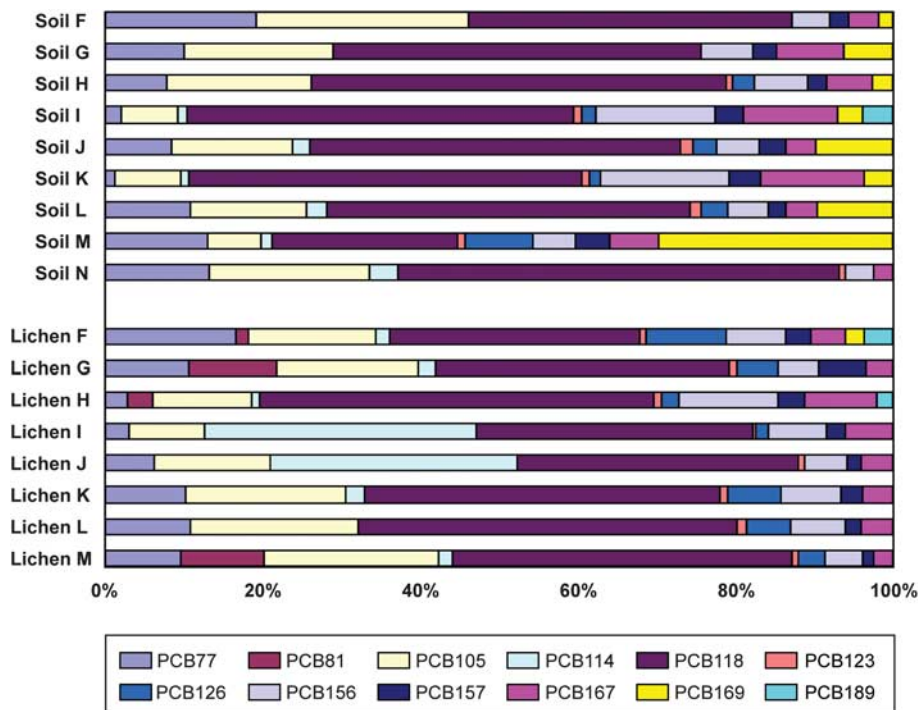
### PCB congener distribution and concentrations in soil

At five sampling location (Fig. 1, Sites A–E), the distribution and the concentration of each PCB congener in soil were analysed. Results of detected PCBs ( $\Sigma\text{PCB}$ ) in five soil samples collected in 2006 are shown in Table II. Thirty-two PCB congeners (IUPAC numbers 1, 2, 3, 4, 15, 18, 19, 28, 37, 52, 77, 81, 101, 105, 111, 114, 118, 126, 128, 153, 155, 156, 157, 167, 169, 178, 180, 189, 202, 205, 206, and 209) were identified in soil samples taken from Sites A–E. The mean concentration of  $\Sigma\text{PCB}$  in these five samples was  $22.0 \text{ pg g}^{-1}$  dry weight (range,  $8.0\text{--}33.8 \text{ pg g}^{-1}$  dry weight; Table II). The soil samples taken from Sites B

and D showed the highest and lowest  $\Sigma\text{PCB}$  concentrations, respectively. The PCB levels of surface soils from Barton Peninsula were comparable to those in soils from other areas in Antarctica, but much lower than those in soils from James Ross Island, West Antarctica (Klanova *et al.* 2008), and slightly lower than those in soils from East Antarctic coastal regions (Negoita *et al.* 2003). The presence of PCBs in Antarctica is thought to be the result of global contamination through long-range transport, as well as local contamination by human activity at the research stations. Recently, Choi *et al.* (2008) reported that the atmospheric levels of 31 PCB congeners detected in this area using passive air samplers were  $2.1\text{--}8.07 \text{ pg m}^{-3}$ , and they observed that PCB levels in atmospheric samples decreased with increasing distance from the main building of King Sejong Station. The study area is located downwind from not only ten other research stations operating on King George Island, but also from a Chilean air force base, thus, it is probable that atmospheric PCB contamination also occurred through local emissions. But PCB concentration in soil samples of this study did not show any significant correlation with distance from the research station. The most abundant classes of PCB isomers were of low molecular weight. All soil samples (Sites A–E) had similar homologue patterns, with high fractions of di-, tri-, and penta-CBs accounting for more than 75% of the total homologue concentrations (Fig. 2). This PCB profile might be suggestive of long range transport from distant sources to the polar region. In contrast, two soil samples collected near the research station (Sites A and E) showed elevated highly chlorinated homologues (penta-CBs and higher) among five samples, implying the potential influence of local pollution. In addition, the significant levels of highly persistent congeners, such as PCBs 118, 153, and 180, may







**Fig. 4.** PCB homologue compositions (%) in soil and lichen samples from Barton Peninsula.

and N show high  $\Sigma_{12}\text{PCB}/\text{TOC}$  values, although their  $\Sigma_{12}\text{PCB}$  concentrations were lower than mean  $\Sigma_{12}\text{PCB}$  concentration. The soil sample from Site F (near the station) shows the lowest TOC content (0.15 wt.%) but highest  $\Sigma_{12}\text{PCB}/\text{TOC}$  ratio, suggestive of local contamination caused by human activities near the station. The soil sample collected from site N also shows relatively high  $\Sigma_{12}\text{PCB}/\text{TOC}$  value compared with its low TOC content (0.26 wt.%). It can be explained by the fact that no vegetation was present at site N because it is newly exposed due to recent glacial retreat.

#### *Dioxin-like PCBs in lichen*

Lichen is the dominant type of terrestrial vegetation in Antarctica, and *U. aurantiaco-atra* is commonly found not only in and around the coastal region of the ice free area, but also in adjacent island areas. The concentrations of pollutants measured in lichens reflect absorption from the atmosphere, which is integrated over a long period and should be correlated with the mean atmospheric concentration of pollutants because of the lack of root-like structures in lichen (Villeneuve *et al.* 1988). However, there have been only a few reports regarding the presence of POPs in lichens (Villeneuve *et al.* 1988, Negoita *et al.* 2003, Yogui & Sericano 2008). In this respect, the concentration of dioxin-like PCB congeners in *U. aurantiaco-atra* collected on Barton Peninsula were measured (Table III). The mean concentration of  $\Sigma_{12}\text{PCB}$  was  $17.133 \text{ pg g}^{-1}$  dry weight (range  $5.49\text{--}43.765 \text{ pg g}^{-1}$  dry weight). In general, the PCB concentrations in lichen samples decreased with increasing distance from the research station, except at Sites H and I (Table III). These

observations were in good agreement with a previous atmospheric analysis from the King Sejong Station (Choi *et al.* 2008). However, the lichen samples from Sites H and I showed very high levels of PCBs ( $43.8 \text{ pg g}^{-1}$  and  $34.4 \text{ pg g}^{-1}$ , respectively), suggesting the influence of local pollution. As these two sites are located near a penguin rookery and skua nests, respectively, the high PCB levels in these samples were thought to be influenced by wildlife, such as seabird droppings and carcasses. The dominant PCB congeners in the lichens were PCB 118 and PCB 105. Interestingly, the PCB 114 congener was detected in over 30% of lichen samples from Sites I and J, but was detected at markedly lower levels in soil samples from the same sites. PCB 118 was also detected at higher levels than other congeners in soil and atmospheric samples (Choi *et al.* 2008). Focardi *et al.* (1995) reported that the most abundant congeners were penta-CB, hexa-CB, and hepta-CB in Antarctic fish, skua, penguins, and Weddell seals, because lower chlorinated biphenyl homologues are the most biodegradable PCB congeners. Comparison of the compositions of PCB congeners in soil and lichen samples showed some important differences (Fig. 4). The soil samples showed markedly higher concentrations of hexa-CBs, but the lichen samples contained large amounts of penta-CBs because of the lack of PCB 169.

#### *Comparison of dioxin-like PCBs in soil, lichen, and air*

The greater proportion of PCB compounds found in Antarctica were probably transported through the atmosphere and deposited in the soil and lichen. Kallenborn *et al.* (1998)

**Table IV.** Correlations between PCB congener concentrations in air and soil samples, air and lichen samples, and soil and lichen samples collected at the same site.

	site	correlation coefficient
Soil-lichen	F	0.949**
	G	0.924**
	H	0.974**
	I	0.628*
	J	0.693*
	K	0.892**
	L	0.965**
M	0.409	
Air-soil	F	0.879**
Air-lichen	F	0.859**

\*\* $P < 0.01$ , \* $P < 0.05$ .

reported the atmospheric long-range transport of POPs from South America to Signy Island in the South Orkney Islands. Furthermore, Montone *et al.* (2003) reported a similar trend for PCBs on King George Island and Choi *et al.* (2008) recently discussed the potential relationship between atmospheric PCB levels and long-range transport in the same region examined here. The sampling site F in this study is the same site that Choi *et al.* (2008) collected air samples with passive air samplers. The relationship between the atmospheric and soil/lichen PCB profiles (Site F) was investigated by calculating Spearman's correlation coefficients (Table IV). The PCB congener concentrations showed a strong correlation between air and soil ( $r = 0.879$ ,  $P < 0.01$ ), suggesting that these congeners accumulated in soil by partitioning from the atmosphere. A high correlation coefficient was also observed between air and lichen concentrations ( $r = 0.859$ ,  $P < 0.01$ ), which can be explained by the direct absorption of PCBs from the atmosphere due to the absence of root-like structures in lichen. The  $\Sigma_{12}$ PCB level was about 5-fold higher in lichen than in soil. Soil and lichen samples collected from the same sites also showed significant correlations in PCB congener concentrations, except at Site M. Despite the limited number of sites these correlations suggest that atmospheric transport may be the main source of PCBs in the study area.

In conclusion, the surface soil and lichen samples collected on King George Island showed very low levels of PCBs. Several pathways for PCB contamination in soil and lichen samples were suggested. Long-range atmospheric transport is thought to be the main source of PCBs detected on King George Island. However, PCB levels in soil and lichen samples also appeared to be influenced by local sources, such as the presence of wildlife. Our results suggested that, on a local scale, bioaccumulation and biologically mediated transport of PCBs could cause greater contamination than human impact from the research station. Therefore, further comprehensive studies on bioaccumulation are needed to clarify the pathways of PCB contamination in this area.

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