

Air-Soil Partitioning of PCBs in Rural Area

Hyun-Gu Yeo*, Minkyu Choi, Man-Young Chun¹⁾, and Young Sunwoo

Department of Environmental Engineering, Konkuk University

¹⁾*Department of Environmental Engineering, National Hankyong University*

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Abstract

The soil concentrations of polychlorinated biphenyls (PCBs) were measured at 12 sites in Ansung, Kyonggi province, Korea. Correlation coefficient (r) between total PCBs and organic matter content (OM) was significant ($r = 0.562$, $p < 0.05$). It suggests that organic matter may be a key factor of soil absorption of PCBs. The PCB concentrations of low chlorinated congeners with high vapor pressure were relatively abundant in air but high chlorinated congeners with low vapor pressure were mainly dominated by soil. The results indicated the influence of physicochemical properties of PCBs such as vapor pressure, octanol-air partition coefficient (K_{OA}). The calculated soil/air fugacity quotients suggested that the soil may be a source of heavier molecular PCBs (> penta-CBs) to the atmosphere, where lighter molecular PCBs appear to be affected by a movement from air to soil, especially tetra-CBs. Therefore, PCB homologs with low vapor pressure might have been influenced by re-volatilization from soil.

Key words : PCBs, Air, Soil, Fugacity quotients, Source

1. INTRODUCTION

Soil is the primary environmental reservoir for semi-volatile organic compounds (SOCs) in the terrestrial environment. It has been estimated that the soil contains the vast majority (>90%) of the total UK environment burden of PCBs (Harrad *et al.*, 1995), PAHs (Wild and Jones, 1995) and PCDD/F (Duarte-Davidson *et al.*, 1997). Once deposited to the soil, many persistent and strongly sorbed SOC may residue for many years, but the soil may not a permanent sink for these compounds. Leaching to groundwater and biodegradation can be important soil removal processes for some

compounds, but for non-ionic persistent SOC losses by volatilization are likely to dominate. Soils can therefore become a source of some SOC (e.g. PCBs) to the atmosphere (Jones, 1994). The exchange of SOC between the soil and air is important because although soil is the major sink of SOC in the terrestrial environment, air is the primary vector for human exposure (Hippelein and McLachlan, 1998). Two important pathways influencing human exposure to SOC are air-plant-cow-human (direct adsorption by plants of SOC from the air) and soil-air-plant-cow-human (adsorption of SOC by plants that have volatilized from the soil) (Fries, 1995). Therefore, air-soil exchange is critical in controlling the risk posed to humans by SOC in the environment. Furthermore, air-soil exchange will play an important role in controlling the

* Corresponding author.

Tel : 016-755-5174, E-mail : hgyeo@alumni.konkuk.ac.kr

regional and global transport and redistribution of SOCs and thus amounts entering Arctic food chains (Wania and Mackay, 1993). The main processes controlling to air-soil exchange of SOCs are dry deposition (both by vapor adsorption and with particles), wet deposition (by rain and snow scavenging of vapor- and particles-sorbed chemicals) and volatilization from the soil. Gaseous deposition (vapor adsorption) to the soil and re-emission of vapors from the soil are essentially the same process and are key to controlling the exchange of SOCs. Re-entrainment of SOCs sorbed to soil particles into the atmosphere is a possible additional transport route for SOCs from soil to the atmosphere, but is relatively insignificant compared to gaseous emissions, even for the least volatile compounds (PCDD/F) (Duarte-Davidson *et al.*, 1997). Atmospheric deposition of SOCs occurs directly to the soil surface or to the surfaces of vegetation growing in the soil (Duarte-Davidson *et al.*, 1997). Chemicals scavenging by vegetation may enter the soil when the vegetation falls to the soil surface and decays or may volatile from the vegetation and re-enter the atmosphere.

The purpose of this study intends to estimate the potential source of PCBs in the atmosphere with identifying soil concentration in the vicinities of air sampling site.

2. EXPERIMENT

2.1 Sample extraction and analysis

The soil samples were taken in vicinity of Ansong, Hankyong University (Fig. 1). The samples were collected using a stainless steel hand-held corer that was cleaned before and after each sample using hexane. The first two cores were always discarded. The sampling depth was 0~5 cm after removal of the litter layer. Three cores, taken over an area of several square meter, were bulked together to form one sample. The samples were stored in glass vial pre-cleaned hexane and dichloromethane in 3 times to minimize the possibility for

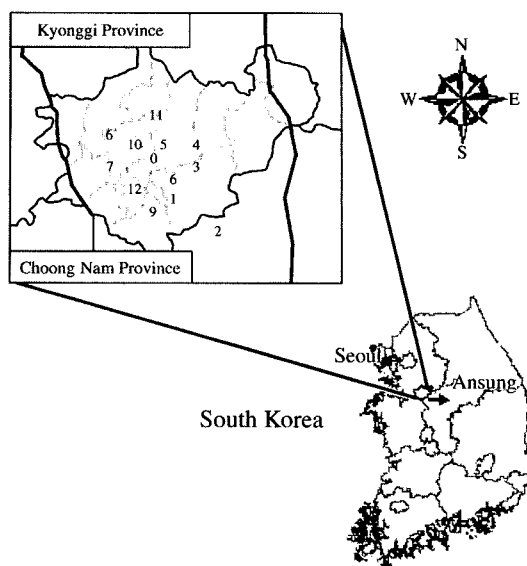


Fig. 1. Locations of soil sampling site in the vicinity of the air sampling site (O).

contamination. The samples were immediately transferred to a freezer (-26°C) where they were until analysis. The three cores mixed together after which a subsample of about 15 g wet weight of soil (corresponding to between 10 and 14 g dry weight (DW)) was weighed out and mixed with Na_2SO_4 to remove water. The sample was then transferred to a pre-extracted thimble in a Soxhlet apparatus and spiked with a surrogate standards ($^{13}\text{C}_{12}$ PCB isotopes: PCB 28, PCB 52, PCB 101, PCB 138, PCB 180, and PCB 209). The samples were extracted for 36 h using dichloromethane.

2.2 Quality control and quality assurance (QA/QC)

Recoveries were calculated using $^{13}\text{C}_{12}$ PCB isotopes added to prior to extraction. Average recoveries ($n = 18$) were $89 \pm 7\%$ for PCB 28, $71 \pm 9\%$ for PCB 52, 86 ± 5 for PCB 101, 74 ± 18 for PCB 153, 100 ± 13 for PCB 138, 101 ± 11 for PCB 180, and $82 \pm 7\%$ for PCB 209. The duplicates were collected as bulk samples (sample-1, sample-2). Mean concentrations of sample -1 and sample -2 were 6.72, 7.25 (pg/g dry weight

[DW]), respectively. The regression analysis between duplicate samples showed quite good correlation ($p < 0.001$): [Sample-2] = 0.95 [Sample-1] + 0.89 ($R^2 = 0.83$, $p < 0.001$). Significant correlation between duplicate samples implies that there are no any other problems for quantity PCBs on soils except for PCB congeners measured in high concentration (Fig. 2).

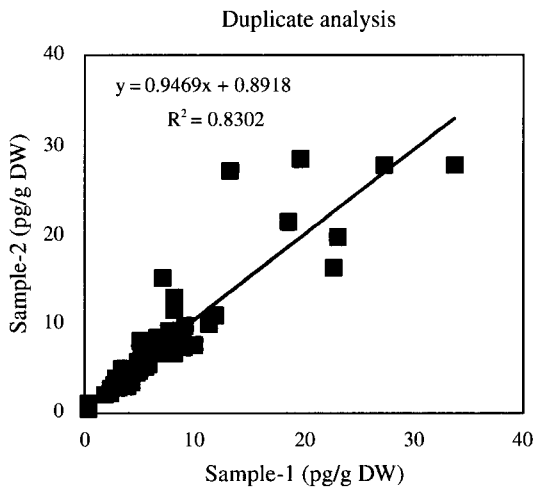


Fig. 2. Duplicate analysis of PCB congeners detected in soils.

3. RESULTS AND DISCUSSION

3.1 Characteristics of PCBs in soil

3.1.1 Physicochemical properties

Soil properties affecting soil-air partitioning include water content, organic matter content, texture, structure and porosity. Hippelein (1997) has measured soil-air partitioning coefficients (K_{SA}) of SOCs at the wide range of soil moisture content. They found that K_{SA} was greatly affected at low soil moisture content (0.3 ~ 0.8%), but that soil moisture did not affect K_{SA} at soil moisture content from 1.9 to 12%. Although soil mixture (above a certain level) does not have a significant effect on sorption of non-ionic SOCs to the surface by convective transport in soil water (Hippelein and McLachlan, 1998). The fraction of soil organic matter is the most important soil factor for sorption of weakly polar or non-ionic organic compounds. Soil-air partitioning coefficients (K_{SA}) will be proportional to the soil organic matter content.

There was very wide in soil properties, notably in organic matter (OM) content, water content, in sample set. OM varied between 0.5% and 5.8% for all soils,

Table 1. Sample information and soil concentration of PCBs (pg/g dry weight).

Site	Soil type	Soil properties		Soil concentration of PCB homologs(pg/g dry weight(DW))						
		OM (%)	WC (%)	Tetra	Penta	Hexa	Hepta	Octa	Deca	Total
1	V.F ¹⁾	2.94	19.13	38.71	57.66	60.58	17.54	4.00	1.63	176.42
2	B.G ²⁾	1.32	13.08	39.39	67.69	53.82	15.80	3.12	1.63	177.72
3	V.F	1.67	10.33	49.97	75.13	62.95	17.71	4.58	1.95	206.94
4	V.F	3.96	14.74	50.59	75.77	61.76	16.68	3.64	2.29	206.99
5	P.G ³⁾	0.54	4.55	59.07	68.93	62.13	20.70	4.13	2.95	210.77
6	R.F ⁴⁾	4.64	24.64	48.97	77.02	86.97	27.53	4.39	3.70	239.31
7	R.F	4.01	22.87	55.62	75.02	86.17	32.83	5.20	2.92	250.39
8	R.F	5.07	27.78	65.52	82.77	93.93	33.25	5.54	3.69	273.07
9	R.F	3.74	21.39	62.00	113.99	105.63	38.86	5.09	3.79	318.91
10	V.F	2.16	4.06	69.91	108.81	109.86	41.91	5.56	24.94	348.07
11	R.F	3.98	25.08	69.61	127.71	130.63	38.82	7.12	3.50	365.19
12	R.F	5.82	28.86	59.41	285.56	382.97	165.44	19.86	13.27	861.78
	Mean	3.32	18.04	55.73	101.27	108.12	38.92	6.07	5.52	302.92
	S.D ⁵⁾	1.61	8.60	10.51	61.64	89.80	40.99	4.48	6.86	187.05
	Min	0.54	4.06	38.71	57.64	53.82	15.80	3.12	1.63	176.42
	Max	5.82	28.86	69.91	285.36	382.97	165.44	19.86	24.93	861.78

¹⁾Vegetation field, ²⁾Background area, ³⁾Playground, ⁴⁾Rice field, ⁵⁾Standard deviation

Table 2. Correlation coefficients of PCB congener concentrations in soil between sites.

	Site 1	Site 2	Site 3	Site 4	Site 5	Site 6	Site 7	Site 8	Site 9	Site 10	Site 11	Site 12
Site 1	1.00											
Site 2	0.94**	1.00										
Site 3	0.72**	0.72**	1.00									
Site 4	0.76**	0.80**	0.98**	1.00								
Site 5	0.62**	0.58**	0.94**	0.82**	1.00							
Site 6	0.90**	0.74**	0.80**	0.68**	0.81**	1.00						
Site 7	0.90**	0.75**	0.70**	0.59**	0.75**	0.97**	1.00					
Site 8	0.86**	0.69**	0.68**	0.61**	0.73**	0.96**	0.96**	1.00				
Site 9	0.73**	0.60**	0.70**	0.55**	0.72**	0.86**	0.85**	0.88**	1.00			
Site 10	0.65**	0.52**	0.44*	0.39*	0.55**	0.76**	0.77**	0.79**	0.80**	1.00		
Site 11	0.75**	0.66**	0.79**	0.65**	0.79**	0.88**	0.87**	0.65**	0.95**	0.74**	1.00	
Site 12	0.15	0.10	0.27	0.05	0.43*	0.55**	0.61**	0.62**	0.60**	0.59**	0.53**	1.00

** : $p < 0.01$, * : $p < 0.05$

also water content varied between 4% and 29%. Correlation coefficients (r) between total PCBs and OM, water content were 0.562 ($p < 0.05$), 0.472 ($p = 0.16$), respectively. In other words, there was no relationship between total PCBs and water content, but significant correlation coefficient was found in total PCBs and organic content matter. Therefore, organic matter content may be a key factor of absorption on soil for PCBs. Ian *et al.* (1997) reported that the relationship between the soil organic matter content verse total PCBs and total PAHs content of soil and therefore, linear regressions were carried out between these two variables. Total PCBs and total PAHs content tend to correlate well with the soil organic matter content at all four sites, with correlation coefficients (r) varying between 0.77 and 0.97 (Ian *et al.*, 1997). Examination of the probability values indicates that all of the correlations are statistically significant ($p < 0.01$), with many highly statistically significant ($p < 0.001$). No statistically significant correlations could be found between other soil properties (e.g. soil water content, soil porosity, and soil mineral matter content) (Ian *et al.*, 1997). Table 1 summarizes sample information for all sites, including location, soil properties, and homolog concentrations.

3. 1. 2 General comments on soil concentrations

PCBs congeners were detected 27 kinds (i.e. tetra-CBs (47, 52, 60), penta-CBs (87, 101, 105, 110, 114, 118, 126,)), hexa-CBs (138, 141, 149, 151, 153, 155,

156, 157, 167, 170), hepta-CBs (180, 183, 185, 187, 189), octa-CBs (194), and deca-CBs (209)). Results for individual PCB congeners are available in supporting information. In general, differences in concentration between sites were 1 order (except tetra-CBs) of magnitude for PCB homologs. Generally, the highest total PCB concentrations was found in site 12. The lowest total PCB concentrations was also shown in site 1.

3. 1. 3 Comparison of PCB congener concentrations in soil between sites

Table 2 was correlation coefficients of PCB congener concentrations in soil between sites. PCB congener concentrations amongst the sites are statistically very significant ($0.59 < R^2 < 0.97$, $p < 0.01$ except site 12) which imply that spatial distributions of PCB congeners in soil were mostly affected by similar sources. That is, within soil samples a relative homogeneity of composition was observed.

3. 2 Relationship between air and soil

3. 2. 1 PCB homologs profiles

The concentrations of PCB homologs in air and soil were shown in Figure 3. Mean concentrations of PCB homologs in air were calculated from June, 2000 to July, 2002 in Hankyong University, Ansong. Mean concentration of PCB homologs in air showed $57.46 \pm$

48.55 $\mu\text{g}/\text{m}^3$ of tri-CBs, 34.34 \pm 38.70 $\mu\text{g}/\text{m}^3$ of tetra-CBs, 11.42 \pm 7.45 $\mu\text{g}/\text{m}^3$ of penta-CBs, 5.71 \pm 3.59 $\mu\text{g}/\text{m}^3$ of hexa-CBs, 3.23 \pm 2.70 $\mu\text{g}/\text{m}^3$ of hepta-CBs, 0.32 \pm 0.17 $\mu\text{g}/\text{m}^3$ of octa-CBs, and 0.18 \pm 0.11 $\mu\text{g}/\text{m}^3$ of deca-CBs, respectively. The concentration pattern of PCB homologs in air increased with decreasing degree of chlorine (Cl). Also, mean concentrations of PCB homologs in soil were calculated using measurements of 12 sampling sites in the vicinity of air sampling site. Mean concentration of PCB homologs in soil showed 55.73 \pm 10.51 $\mu\text{g}/\text{g}$ DW of tetra-CBs, 101.27 \pm 61.64 $\mu\text{g}/\text{g}$ DW of penta-CBs, 108.12 \pm 89.80 $\mu\text{g}/\text{g}$ DW of hexa-CBs, 38.92 \pm 40.99 $\mu\text{g}/\text{g}$ DW of hepta-CBs, 6.07 \pm 4.48 $\mu\text{g}/\text{g}$ DW of octa-CBs, and 5.52 \pm 6.86 $\mu\text{g}/\text{g}$ DW of deca-CBs, respectively. In general, the concentration pattern of PCB homologs in

soil increased with increasing degree of chlorine (except octa- and deca-CBs). As to above result, the concentration pattern of PCB homologs in air was quite different compared to that of soil. In particular, tri-CBs that appeared the largest fraction (%) of total PCBs in air showed below limit of detection values in soil. That is, tri-CBs concentrations measured in air have been a little influenced by re-volatilization of PCBs in soil compared to other homologs. Soil in rural areas may be more rapidly depleted in the lighter PCBs (e.g. tri-CBs) due to their higher volatility, whereas heavier, less volatile PCBs (e.g. penta- through hepta-CBs) are more likely to have maintained a high soil concentration (Harner *et al.*, 1995; Alock *et al.*, 1993).

3.2.2 PCB congeners profiles

The concentrations of PCB congeners in air and soil were shown in Figure 4. Number of detected PCB congeners in air was 38 compounds: tri-CBs (PCB 18, 27, 28, 30, 33, and 38), tetra-CBs (PCB 44, 47, 49, 52, 60, 61, 66, and 74), penta-CBs (PCB 87, 101, 105, 110, 114, 118, and 126), hexa-CBs (PCB 138, 141, 149, 151, 153, 155, 156, 157, 169, and 170), hepta-CBs (PCB 180, 183, 185, 187, and 189), octa-CBs (PCB 194), and deca-CBs (PCB 209). Whereas, number of detected PCB congeners in soil appeared 27 compounds: tri-CBs (< limit of detections), tetra-CBs (PCB 47, 52, and 60), penta-CBs (PCB 87, 101, 105,

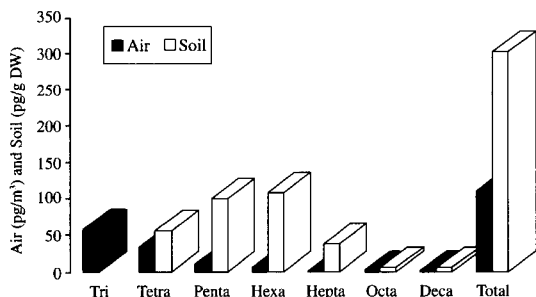


Fig. 3. Concentrations of PCB homologs in air and soil.

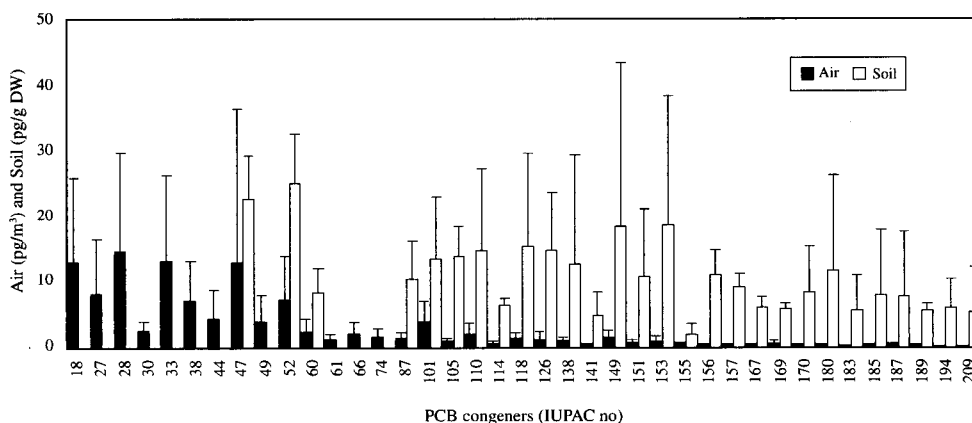


Fig. 4. Concentrations of PCB congeners in air and soil.

110, 114, 118, and 126), hexa-CBs (PCB 138, 141, 149, 151, 153, 155, 156, 157, 169, and 170), hepta-CBs (PCB 180, 183, 185, 187, and 189), octa-CBs (PCB 194), and deca-CBs (PCB 209). Most of congeners detected both air and soil were very similar except tri- and tetra-CBs. The low molecular congeners with having a high vapor pressure in air tends to have a high concentration compared to heavier ones. Especially, contribution (%) of tri-CBs (sum of PCB 18, 27, 28, 30, 33, 38) and tetra-CBs (sum of PCB 44, 47, 49, 52, 60, 61, 66, 74) in air, with having a high vapor pressure, approximately 52.4% and 31.3%, respectively. Whereas, contributions (%) of tri-(not detected) and tetra-CBs (sum of PCB 47, 52, and 60) in soil was about 18.4%. Contributions (%) of penta-, hexa-, hepta-, octa-, and deca-CBs in air were approximately 9.6, 5.2, 1.2, 0.1, and 0.2%, respectively. However, contributions (%) of penta-, hexa-, hepta-, octa-, and deca-CBs in soil were approximately 29.2, 35.7, 12.8, 2.0, and 1.8%, respectively. As a result of PCB congener contributions, low chlorinated congeners were relatively abundant in air but high chlorinated congeners were dominant in soil. These results was associated with K_{OA} , vapor pressure etc.

3. 2. 3 Soil-air partitioning

A less well-established partition coefficient is the soil-air partition coefficient (K_{SA}). In the absence of measured values, the current practice is to calculate

(K_{SA}) from the equilibrium soil-water partition coefficient (K_d) and the Henry's law constant (H) (Mackay *et al.*, 1992a):

$$K_{SA} = K_d/H' \quad (1)$$

$$K_{SA} = 0.411 f_{oc} \frac{1}{\rho} K_d/H' \quad (2)$$

Table 3 shows the scavenging coefficients of soil, together with K_{OA} and K_{SA} at the same time detected congeners in both air and soil. K_{SA} , in this study, was calculated using equation (2) and K_{OA} used a literal review (Lancaster university, 1999). Several workers have proposed the use of octanol-air partition coefficient (K_{OA}) and soil-air partition coefficient (K_{SA}) in helping to model air-soil transfer of the gas phase SOCs (Spencer and Cliath, 1973; Mackay *et al.*, 1992a).

Figure 5 presents log scavenging coefficients (m^3 air g^{-1} soil dry weight (DW)) against log octanol-air partition coefficients ($\log K_{OA}$) and log soil-air partition coefficient ($\log K_{SA}$). The regression analyses between $\log Cs/Ca$ and $\log K_{OA}$ and $\log K_{SA}$ provide very significant values ($p < 0.01$) which imply that equilibrium has been attained between the gaseous concentration between air and soil. However, the slope of plot between $\log Cs/Ca$ and $\log K_{OA}$ was lower than that of plot between $\log Cs/Ca$ and $\log K_{SA}$, which would imply that octanol was not an ideal surrogate for soil-air partitioning. The calculated K_{SA} values in using

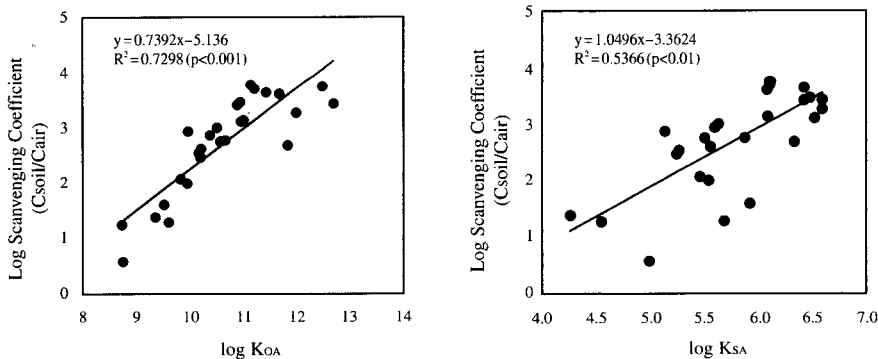


Fig. 5. Log scavenging coefficients against log octanol-air partition coefficients ($\log K_{OA}$) and log soil-air partition coefficient ($\log K_{SA}$).

Table 3. The scavenging coefficients of soil, together with K_{OA} and K_{SA} at the same time detected congeners in both air and soil.

Congener	$\log C_s/C_a^{1)}$ [m ³ air/g soil DW]	$\log K_{OA}$ [liter review]	$\log K_{SA}$ [calculated]
47	0.56	8.75	4.99
52	1.25	8.73	4.54
60	1.37	9.35	4.26
87	2.07	9.82	5.47
101	1.29	9.60	5.69
105	2.86	10.37	5.14
110	2.00	9.94	5.55
114	2.54	10.17	5.28
118	2.47	10.19	5.26
138	2.77	10.66	5.88
141	2.76	10.57	5.51
149	2.61	10.22	5.57
151	2.94	9.97	5.60
153	3.00	10.51	5.64
155	1.60	9.52	5.92
156	3.78	11.14	6.12
157	3.71	11.20	6.11
167	3.15	11.01	6.09
169	2.69	11.83	6.33
170	3.62	11.68	6.08
180	3.65	11.43	6.43
183	3.43	10.88	6.43
185	3.47	10.93	6.49
187	3.12	10.95	6.53
189	3.28	12.00	6.60
194	3.76	12.48	6.10
209	3.44	12.69	6.66

¹⁾C_s: soil concentration (pg/g dry weight), C_a: air concentration (pg/m³)

physicochemical properties (e.g. f_{oc} (organic matter content in soil), K_d (soil-air partition coefficient), ρ_s (density in soil)) may be well account for soil-air partitioning.

3. 2. 4 Assessing the equilibrium condition using fugacity quotients

One approach to determine whether or not compartments are in equilibrium with each other is to look at a snap-shot by comparing the fugacities of the compounds in the compartments. Fugacities can be calculated from the concentrations and fugacity capacities of the compounds in the compartment. If the fugacities are more or less equal then the compartments are close to equilibrium, but if there is a large disparity then there will be a tendency for a chemical to move from

on compartment to the other in order to establish equilibrium conditions. By dividing the fugacity of one compartment to another a fugacity quotient is calculated. Fugacity quotients are a convenient way of expressing the relative fugacities of two environmental compartments and have been used previously in the literature (Duarte-Davidson *et al.*, 1997; McLachlan, 1996).

Soil and air concentrations of PCBs measured in the sampling site used to calculate fugacities of the PCBs in the air and soil compartments. All the necessary physicochemical data were taken from Mackay *et al.* (1992a). The fugacity of PCBs in a gaseous phase can be calculated from the concentration, C (g m⁻³), using:

$$f = \frac{C}{zM} \quad (3)$$

where M is the molecular mass (g mol⁻¹) and z is the fugacity capacity of the phase for the PCBs (mol m⁻³ Pa⁻¹). The fuacity capacity for air is defined by:

$$z_A = \frac{1}{RT} \quad (4)$$

where R is the gas constant (8.314 J mol⁻¹ K⁻¹) and T is the absolute temperature. The fugacity capacity of soils can be estimated using:

$$z_s = f_{oc} K_{oc} z_w \frac{1}{\rho_s} \quad (5)$$

where f_{oc} is the fraction of organic content (3.2% for soils in this study), ρ_s is the density (assumed to be 1.5 g cm⁻³ for all calculations) and K_{oc} is the soil organic carbon/water partition coefficient, defined according to Karickhoff (1981) using:

$$K_{oc} = 0.41 K_{ow} \quad (6)$$

z_w , the fugacity capacity of water, is calculated from:

$$z_w = \frac{1}{H} \quad (7)$$

where H is the Henrys Law constant (Pa m³ mol⁻¹).

Figure 6 was calculated soil/air fugacity quotients for PCB congeners measured in air and soil. Recent

studies (Lead *et al.*, 1996; Alcock *et al.*, 1994) have shown that exposure of samples to air can cause either contamination or loss of PCBs, depending on the equilibrium position of the soil. Therefore, it follows that in the environment, depending on the equilibrium position between the air and the soil. The soil can be either a sink for SOCs deposited there or a secondary source of SOCs is from the soil to the air. Then, it suggests that primary sources of PCBs to the atmosphere have been substantially reduced and air concentrations are now controlled by secondary sources (or, that there are natural sources of the compounds from the soil).

Soil/air fugacity quotients were calculated by dividing the calculated soil fugacities by calculated air fugacities. Fugacity quotient values near to one show equilibrium between air and soil, since when two phases are in equilibrium their fugacities are equal. A fugacity quotient greater than one indicate a tendency for a compound to volatilize from soil, whereas a value of less than one imply a tendency to remain in the soil and a capacity for the air to supply the soil with more PCBs (Cousins and Jones, 1998).

Cousins and Jones (1998) examined that for the spiked soils with SOCs (i.e. PCBs, PAHs, and PCDD/Fs) the fugacity quotients are far larger than one at the start and end of the experiment for all compounds. They reported that this is consistent with observed losses during the field experiment. For unspiked soil with SOCs, however, most of the fugacity quotients are much nearer unity. All of the PCBs appear to be close to equilibrium between soil and air, although the fugacity quotients are slightly less than one.

Wendy *et al.* (1998) reported that soil/air fugacity quotients for PCB 52, 101, 118, and 153 were predicted to be from soil to air. These data can be interpreted as showing that soil have a tendency to outgas the lighter molecular PCBs. In contrast, for PCB 138 and 180 movement is predicted to be from air to soil.

In this study, soil/air fugacity quotients were ranged from 0.06 (PCB 52) to 56.49 (PCB 209). Soil/air fugacity quotients of PCB 47, 52 and 60, with having a high vapor pressures, were very lower values than those of

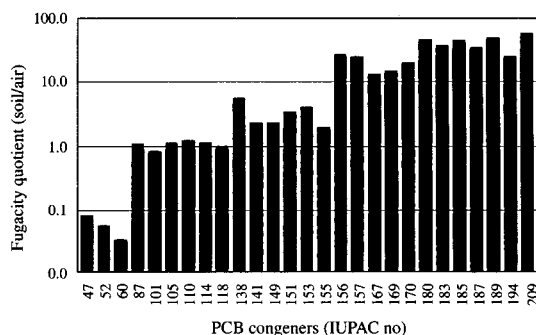


Fig. 6. Calculated soil/air fugacity quotients for PCB congeners measured in soil and air.

heavier molecular PCBs (hepta-CB through deca-CBs). And, soil/air fugacity quotient values of penta-CBs (i.e. PCB 101, 105, 110, 114, and 118), with having a middle range of vapor pressure, were approximately one which might be shown in equilibrium between air and soil. Whereas, soil/air fugacity values of hexa- through deca-CBs, with having a lower vapor pressure, marked from 2.29 to 56.49 which indicates a tendency for PCBs to volatilize from soil to air. MaLachlan (1996) hypothesized that diffusive transfer to PCBs from the earths surface to the atmosphere determined levels of PCBs in the atmosphere. This hypothesis was originally developed to explain the observed seasonal cycling of gaseous PCB concentrations in air characterized by higher levels in summer and lower levels in winter. Referring to above our results, soil/air fugacity quotient values of tetra-CBs did not approach equilibrium between soil and air which indicate a tendency for PCBs to transport from another site. However, for the hexa- through deca-CBs, these homologs measured in soil might have influenced the PCB concentration in air. The heavier molecular PCBs with low vapor pressure tend to easier exist in soil than lighter ones because of difference in K_{OA} value.

4. CONCLUSIONS

This study might be intended to identify the PCBs

sources in air using soils in the vicinity of air sampling site, and to investigate factors of air-soil partitioning.

The significant correlation coefficient was found in total PCBs and organic content matter which may be a key factor of absorption on soil for PCBs. PCB congener concentration amongst the sites are statistically very significant values ($0.59 < R^2 < 0.97$; $p < 0.01$) which imply that spatial distributions of PCB congeners in soil were mostly affected by similar sources.

The soil/air fugacity quotient values of tetra-CBs did not approach equilibrium between soil and air which indicate a tendency for PCBs to transport from another site. However, heavier molecular PCBs, such as hexa- through deca-CBs, measured in air might have been more affected by re-volatilization from soil than lighter ones.

REFERENCES

- Alcork, R.E., A.E. Johnston, S.P. McGarth, M.L. Berrow and K.C. Jones (1993) Long-Term Changes in the Polychlorinated Biphenyl Content of United Kingdom Soils, *Environ. Sci. Technol.*, 27, 1918-1923.
- Alcock, R.E., C.J. Halsall, and C.A. Harris (1994) Contamination of Environmental Samples Prepared for PCB Analysis, *Environ. Sci. Technol.*, 28, 1838-1842.
- Cousins, I.T. and K.C. Jones (1998) Air-Soil Exchange of Semi-volatile Organic Compounds (SOCs) in the UK, *Environmental Pollution*, 102, 105-118.
- Duarte-Davidson, R., A. Sewart, R.E. Alcock, I.T. Cousins, and K.C. Jones (1997) Exploring the Balance between Sources, Deposition, and the Environmental Burden of PCDD/Fs in the U.K., *Terrestrial Environment: an Aid to Identifying Uncertainties and Research Needs*, *Environ. Sci. Technol.*, 31, 1-11.
- Fries, G.F. (1995) A Review of the Significance of Animal Food-Products as Potential Pathways of Human Exposure to Dioxin, *Journal of Animal Science*, 73, 1639-1650.
- Harner, T., D. Mackay, and K.C. Jones (1995) Model of the Logn-term Exchnge of PCB between Soil and the Atmosphere in the Southern UK, *Environ. Sci. Technol.*, 29, 1200-1209.
- Harrad, S.J., A. Sewart, and R.E. Alcock (1995) Polychlorinated Biphenyls in the British Environment Sinks, Sources, and temporal trends, *Environmental Pollution*, 88, 91-108.
- Hippelein, M. (1997) PhD Thesis.
- Hippelein M. and M.S. McLachlan (1998) Soil/Air Partitioning of Semivolatile Organic Compounds, Part 2: Method Development and Influence of Physical-Chemical Properties, *Environ. Sci. Technol.*, 32, 310-316.
- Ian, T.C., G. Bondi, and C.J. Kevin (1997) Measuring and Modelling the Vertical Distribution of Semi-Volatile Organic Compounds in Soils. 2: PCB and PAH Soil Core Data, *Chemosphere*, 39, 2507-2518.
- Jones, K.C. (1994) Observations on Long-term Air-Soil exchange of Organic Contaminants, *Environmental Science Pollution Researches Institute*, 1, 171-177.
- Spencer, W.F. and M.M. Cliath (1973) Pesticide Volatilisation as Related to Water Loss from Soil, *J. Environ. Qual.*, 2, 284-289.
- Karickhoff, S.W. (1981) Semi-empirical Estimation of Sorption of Hydrophobic Pollutants on Natural Sediments and Soils, *Chemosphere*, 10, 833-849.
- Lancaster University (1999) PCB QA/QC Protocol-Second Draft.
- Lead, W.A., E. Steinnes, and K.C. Jones (1996) Atmospheric Deposition of PCBs to Moss (*Hylocomium splendens*) in Norway between 1977 and 1990, *Environ. Sci. Technol.*, 30, 524-530.
- Mackay, D., W.Y. Shiu, and K.C. Ma (1992a) Illustrated Handbook of Physical-chemical Properties and Environmental Fate for Organic Chemicals, Vol. 1, Monoaromatic Hydrocarbons, Chlorobenzenes and PCBs, Chesea/MI: Lewis Publishers.
- McLachlan, M.S. (1996) Bioaccumulation of Hydrophobic Chemicals in Agricultural Food Chain, *Environ. Sci. Technol.*, 30, 252-259.
- Wania, F. and D. Mackay (1993) Global Fractionation and Cold Condensation of Low Volatility Organochlorine Compounds in Polar Regions, *Ambio*, 10-18.
- Wendy, A., A.J. Ockenden, H.F. Sweetman, E.S. Prest, and C.J. Kevin (1998) Toward an Understanding of the Global Atmospheric Distribution of Persistent Organic Pollutants: The Use of Semipermeable Membrane Devices as Time-Integrated Passive Sampler, *Environ. Sci. Technol.*, 32, 2795-2803.
- Wild, S.R. and K.C. Jones (1995) Polynuclear Aromatic Hydrocarbons in the United Kingdom environment: a Preliminary Source Inventory and Budget, *Environmental Pollution*, 88, 91-108.