Measurement of Atmospheric Dry Deposition and Size Distribution of Particulate PCBs in 1999 at Seoul

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Abstract

Ambient particle size distributions of PCBs and their dry deposition fluxes were measured at a site in Seoul to quantify dry deposition fluxes of PCBs and size characteristics of PCBs in the air, and to estimate ambient concentrations of gaseous PCBs and dry deposition fluxes. The dry deposition plate was used to measure dry deposition fluxes of particulate mass and PCBs and a cascade impactor and rotary impactor were used to measure ambient particle size distributions for small $(D_p < 9 \, \mu m)$ and large $(D_p > 9 \, \mu m)$ particles, respectively. Six sample sets were collected from April to July 1999. The fluxes of particulate total PCBs (the sum of 43 congeners) ranged from 160 to 607 ng m^{-2} day^{-1}. The size distribution of total PCBs was bimodal with two peaks in small particle size $(D_p \sim 0.6$ and 6 μm, respectively) and, thus, mass concentration being dominant in small particles. The mean particulate PCBs concentration was 6.9 μg PCBs/g. The concentrations of PCB homologues in the gas phase were estimated based on the particle/gas partition coefficient (K_p) with the measured values of particulate PCBs in this study and they were comparable to those observed in other previous studies. Dry deposition fluxes were estimated by calculating dry deposition velocities.

Key words : Dry deposition flux, PCBs, Size distribution, Gaseous PCBs concentration, Dry deposition model

1. INTRODUCTION

PCBs (PolyChlorinated Biphenyls) are a class of persistent organic pollutants (POPs) that degrade slowly and tend to bio-accumulate. In Korea, there are no PCBs point source emissions from industries into the atmosphere since PCBs was banned to produce in 1998. However, the levels of PCBs in the air, water, and soil are not negligible. Atmospheric transport is an important way for the transfer of PCBs from land to natural waters (Strachan *et al.*, 1988). Previous reports estimated that major fraction of the PCBs entering water bodies originated in the atmosphere and that the atmosphere may be the main source of PCBs in remote ocean regions (Atlas *et al.*, 1986). Atmospheric deposition is an impor-

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tant mechanism controlling the fate of airborn materials and their transfer from the atmosphere to the natural surfaces. Atmospheric deposition of particles to ecosystem takes place via both wet and dry processes. Dry deposition occurs by several processes, such as Brownian motion of particles, sedimentation, and impaction.

However, few measurement data of dry particle deposition or size distributions of PCBs are available, especially, in Korea primarily because of the difficulty in making these measurements for PCBs with low atmospheric concentrations. Yeo et al. (2002a, b) measured both particulate and gaseous PCBs at urban and rural area by using PUF samplers between July 1999 and January 2000. They applied the Clausius-Clapeyron equation to the data to quantify the temperature dependence of gas/particle partitioning of PCBs. Shin et al. (2002) measured ambient size distributions and dry deposition fluxes of PCBs at seven sites in Korea in summer 1999. They estimated dry deposition fluxes from the ambient size distributions and meteorological conditions and compared them with the measured fluxes.

The objectives of this study are (1) to quantify the dry deposition fluxes of PCBs into land surface, (2) to characterize the size distribution of particulate total PCBs and homologues, (3) to estimate the concentration of gaseous PCBs in Seoul, the capital city of Korea with about ten million inhabitants, and (4) to estimate dry deposition fluxes and compare them with the measured fluxes.

2. EXPERIMENTAL METHODS

2.1 Sampling

Atmospheric samples were taken from April to July, 1999 in Seoul. The sampling site is on the roof of a five-story building ($\sim$15 m height) in Seodaemungu. It is within a mixed institutional, residential, and commercial area in the west region of Seoul. Buildings around the sampling site are mainly low-rise. The sampling equipments and meteorological tower on the roof were outside the roof wake boundary. Meteorological parameters such as tempera-

Table 1. Summary of sampling information.

Sample No.	Sampling date (1999)	Sampling time (min)	Avg. wind speed (m s^{-1})	Avg. temp. (°C)
1	4/18~4/20	2029	2.40	22.18
2	4/21~4/25	2194	2.40	22.09
3	4/29~5/1	1953	2.24	19.05
4	5/4~5/6	2202	2.77	19.95
5	5/10~5/12	2073	2.67	20.88
6	6/24~7/4	3447	1.87	28.73

ture, wind speed, wind direction, and relative humidity were obtained from a meteorological tower next to the samplers (Table 1).

The dry deposition fluxes were measured using a smooth greased plate. It was made of poly-vinyl chloride (PVC) and was 21.5 cm long, 7.6 cm wide, and 0.65 cm thick with a sharp leading ($<10°$) that was pointed into the wind direction by a wind vane. Each of plates was covered on top with two Mylar strips (7.6 × 5 cm) coated with Apezion L grease (thickness $\sim$5 μm) to collect impacted particles. The strips were weighed before and after exposure to determine the total mass of particles collected. Then, the strips were extracted and analyzed. Details on the plate sampler are given in Holsen et al. (1991). The dry deposition sampling duration was determined based on the likely detectable mass from the samplers. Daily samples were usually taken from 9:00a.m. to 8:00p.m. (day time only). When there was no rain, the sampling period was about five consecutive days.

Ambient particle size distribution was measured with a cascade impactor and a coarse particle rotary impactor (CPRI). Cascade impactor provides information on the small particles ($D_p \leq 9$ μm) and CPRI provide information on the large particles ($D_p > 9$ μm). The cut-size diameter of cascade impactor are; 9-5.8, 5.8-3.3, 3.3-2.1, 2.1-1.1, 1.1-0.65, 0.65-0.43, and <0.43 μm. The CPRI was operated at 320 rpm, which produces theoretical aerodynamic (particle density of 1 g cm^{-3}) cut-size diameter of 6.5, 11.5, 24.7, and 36.5 μm for stages A, B, C, and D, respectively. The overall average sampling volumes were about 71.6 m^3 for the cascade impactor and were 46.7, 120.7, 401.1, 401.1 m^3 for stages A-D of CPRI,

respectively. These samples were obtained concurrently at the site where the deposition samples were being taken. Atmospheric size distributions were measured continuously. Details on the CPRI are given in Noll and Fang (1986) and Noll et al. (1990).

2.2 Analysis

The analytical procedure used in this study was jointly developed by the Gray Freshwater Biological Institute and in the department of Civil and Mineral Engineering, University of Minnesota, Navarre, and Geochemical and Environmental Research Group (GERG) at Texas A&M University in USA and at the Atmospheric Environment Service in Canada (NOAA, 1993).

The analysis of PCBs consisted of three steps: (1) extraction of the target compounds from the sample matrix, (2) clean-up of the extract, i.e. isolation of the target analytes from interfering compounds present in the extract, and (3) qualitative and quantitative instrumental analysis with a gas chromatography (GC). Quality assurance and quality control (QA/QC) procedure of the data was performed for the chemical and instrumental analysis of the samples.

Glasswares were washed several times in hot water with Alconox, rinsed a number of times with tap water, and then sonicated in hot tap water with Alconox for 30 minutes. Then, they were rinsed a couple of times with tap water and with distilled water. They were wrapped with aluminum foil and placed in an oven at ~450°C for at least 4 hours to remove any residual organic compound. Then they were removed from the oven and kept as wrapped with aluminum foil. Inside of the glassware was rinsed twice with dichloromethane (DCM) right before usage. Exposure to the laboratory air was kept to a minimum at all times.

After final weighing of the samples, they were placed into the bottle containing 150 mL of extraction solvent, which is a mixture of petroleum ether (PE) and DCM in 4 : 1 volume ratio. The bottle was sonicated for 15 minutes to separate particles from the strips after surrogate standards were added to the sample. After the sonication, PCBs in the parti-

cles were extracted by the standard Soxhlet method. Extraction continued for 24 hours and each cycle time was about 45 to 60 minutes. Then, the Kuderna -Danish (K-D) evaporator purged with nitrogen gas was used to reduce the amount of solvent to 5 mL. Then, 15 mL of hexane was added and it was again evaporated until the final volume was down to 2 mL.

The clean-up column, 1.5 cm i.d. × ~30 cm length with a Teflon stopcock, was rinsed twice with DCM before use. Clean glass wool was plugged to the bottom of the column using a glass rod. Then, the column was packed by adding 6 g of deactivated silica, 4 g of alumina, and 2 cm of Na_2SO_4 in DCM-slurry in the given order. The packed column was then pre-washed with 20 mL of DCM and then with 20 mL of PE. Before the top of the column became dry, the sample was added. In order to collect the fraction containing the PCBs, 25 mL of PE was then added and collected in a flask. Finally, solvent was exchanged to hexane and the GC internal standard, Tetrachloro-m-xylene was spiked. A Hewlett Packard (HP) 6890 GC with electron capture detector (ECD) and HP 7673 auto sampler were used.

Individual PCBs were quantified by using the internal standard method. Response factors relative to the internal standards were updated daily using a calibration standard. Aroclor mixture standard was used as the calibration standard (Aroclor 1016 : 1242 : 1254 : 1260 = 1 : 1 : 1 : 1). Five-point calibration was routinely performed for these compounds. This solution contains 43-targeted PCB congeners at known concentration levels which were majority of the PCBs found in the ambient air as shown in

Table 2. PCB congeners analyzed in this study (IUPAC Numbers Grouped by Homologues).

Group	Congeners
Tri	17+15 (17), 16+32 (16), 25, 31+28, 33
Tetra	45, 52, 49, 41+64 (41), 74, 70, 60+56 (56)
Penta	66+95 (95), 90+101 (101), 99, 87+115 (87), 77+110 (110), 118
Hexa	151, 123+149 (149), 132+153+105 (153), 160+138 (138)
Hepta	129+126+178 (178), 202+171+156 (171), 180

Table 2.

Surrogate standards were used to monitor the analytical recoveries of the PCB congeners. The surrogate standards were 4, 4'-Dibromooctafluorobiphenyl (DBOFB), PCB 103, and PCB 198. The standard was added to each sample and blank prior to extraction. It is important to note that both surrogate and internal standards are similar to the PCB congeners but they are not present in the environment. In all cases the surrogate recovery efficiencies were in the acceptable range (between 50 and 120%).

Accuracy is used to determine if a measured or computed value represents the true value of the analyte. In this study, accuracy was assessed by evaluation of recoveries using surrogate spikes in each sample and matrix and analysis of Standard Reference Material (SRM). The SRM used in this study was 1649a Urban Dust from National Institute of Standards and Technology, USA for an atmospheric particulate material collected in an urban area. Analytical values of SRM must fall within ±35% of reference values. Analytical mean values of SRM were fallen within the acceptable range with relative standard error (RDS) of 0.004~0.324. Method Detection Limit (MDL) tests were performed by analyzing 6 of spiked blank samples. Each MDL values were created by standard deviation of each congener multiplied by 3. The range of the MDLs was between 0.561 for PCB 87/15 and 11.84 for PCB 60/56.

Field blanks were taken to determine the background contamination which might be caused from the methods used. All blank materials were cleaned and prepared with the same procedures as the real samples. Field blanks were transported to the field in order to expose them to the same environment as the actual samples were exposed to before and after the sampling. After these blank samples were brought to the laboratory, they were spiked with the surrogate standards and the other necessary steps for PCB analysis were performed. These results were used to calculate either concentrations or fluxes by considering air volumes or area and sampling times.

Table 3. Comparison of particulate dry deposition fluxes of PCBs with other study results.

Sampling site	Year	Fluxes (ng m^{-2} day^{-1})	Reference
Seoul, Korea (n=6)	April~July, 1999	160~607 (360)	This study
Seoul, Korea	August, 1999	131	Shin et al. (2002)
Chicago, US	1989~1990	4500	Holsen et al. (1991)
Chicago, US	1993~1995	105~390 (240)	Yucel (1997)
South Heaven, US	1993~1995	140	Yucel (1997)
Taiwan	1994	3500~6800	Chen et al. (1996)

3. RESULTS AND DISCUSSION

3.1 The dry deposition fluxes

The particulate dry deposition fluxes of total PCBs ranged from 160 to 607 ng m^{-2} day^{-1} with the geometric mean value of 360 ng m^{-2} day^{-1}. Also, the particulate mass fluxes were from 0.087 to 0.53 μg m^{-2} day^{-1}. Table 3 shows comparison of this result with other PCBs dry deposition flux measurement results. Though it is difficult to directly compare these results due to different sampling and calculation techniques, the flux values in this study are comparable to those reported for the same site but different period (Shin et al., 2002) or Chicago in USA but higher than a suburban area in USA (South Heaven) and lower than in Taiwan.

Figure 1 shows the relationship between the particulate homologue PCBs flux (the sum of the 43 PCB congener fluxes) and atmospheric conditions: temperature and wind speed. The particulate total PCBs flux was not correlated well with the average ambient air temperatures or wind speeds. This reason for this lack of correlation is not clear but probably due to long sampling time.

Among congeners, the flux of PCB 180 (2, 2', 3, 4, 4', 5, 5'-heptachlorobiphenyl) was the highest, accounting for 27.8% of total PCB flux. Note that in Chicago, the flux of PCB 101 (2, 2', 4, 5, 5'-pentachlorobiphenyl) was the highest (Yucel, 1997). Among the 43 congeners, 10 congeners were under
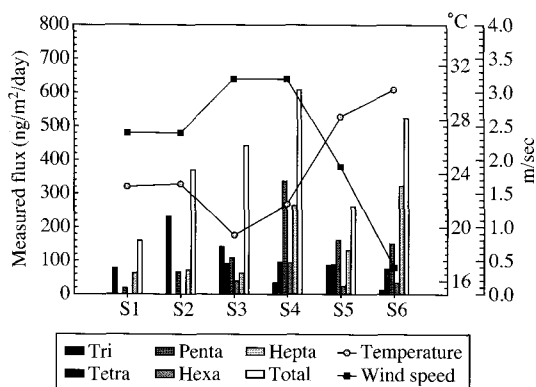
Fig. 1. Histogram of the PCB fluxes of congeners grouped by homologues for each samples (six samples) and temperature and wind speed.

Fig. 2. A typical size distribution of total particulate PCBs.

the detection limit. Generally, the fluxes were high for medium molecular weight (MW) and high MW PCB congeners. Low MW PCBs were detected in all flux measurements. Due to their high vapor pressure, in theory the low MW PCBs cannot be on the particulate phase but in the gas phase at the ambient conditions. Possible explanations for this result could be (1) capture of some gaseous PCBs by grease applied to the strips and/or (2) some gaseous PCBs might have been associated with particles and then deposited on the plates (so-called positive artifacts among semi-volatile species).

3.2 Size characteristics

In total, six size distributions were measured in this study. In Figure 2, a typical size distribution of total PCBs between 0.1 and 100 µm in diameter is shown. The distribution was bimodal with two peaks in the small particle range ($D_p \sim 0.6$ and 6 µm, respectively). This result was comparable to the previous study by Chen et al. (1996). They observed that the particle size distribution of total PCBs was bimodal in small particle size range. In particular, they reported that the industrial particles (i.e., particles collected near to the source) were dominant in the fine particle mode ($D_p < 2.5$ µm), while particles in the urban site were dominant in the coarse particle mode ($2.5 < D_p < 10$ µm). The ambient concentrations of particulate PCBs were in the range of
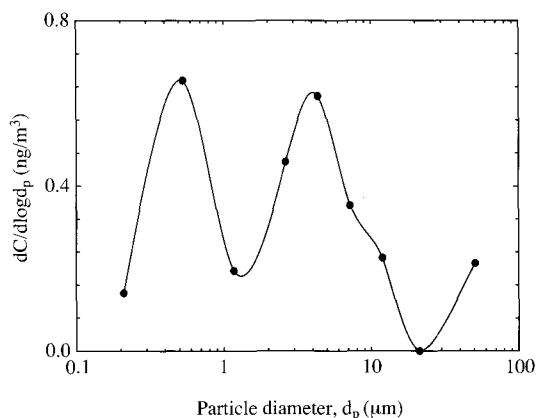
0.05 to 0.74 ng m^{-3}. The fraction of PCBs in the coarse particles was less than 3% of the total PCBs concentrations due probably to the preferential absorption of PCBs in organic-rich fine fraction of particles (Pankow et al., 1997).

Particle-bound PCBs concentration (mass PCBs per mass particles) was calculated with the measured TSP and PCBs concentrations and compared with other measurements in Table 4 along with the particulate total PCBs concentrations. The relationship between the particle-bound PCBs concentration and the measured particulate PCBs concentration varies significantly based on the sampling location because the partitioning would differ extensively depending on the sites, meteorological conditions, and particle characteristics. However, the particle-bound PCBs concentration and particulate PCBs concentration in this study were similar to those of rural areas in US (Holsen et al., 1991; Hermanson, 1989) and southern Taiwan (Chen et al., 1996).

The ratios of total PCBs and homologues to small ($D_p \leq 9$ µm) and large ($D_p > 9$ µm) particles are shown in Table 5. As a whole, most PCB homologues were dominant on the small particle with 72% of total particulate PCB concentration. But, high MW PCBs such as penta, hexa, and heptachloro biphenyls existed exclusively in the fine particles (not shown in Table 4).

Table 4. Comparison of the particle-bound PCBs concentrations with other study results.

Site	PCBs mass per particle mass (μg PCBs/g TSP)	Ambient PCBs concentration (ng PCBs m^{-3} air)	Reference
Seoul, Korea	6.9	1.10	This study, 1999
Bloomington, US	0.1~9.6	0.65~2.53	Hermanson, 1989
Chicago, US	30~50	7.55~20.26	Holsen et al., 1991
Southern Taiwan	–	–	Chen et al., 1996
-Rural	10.3	2.50	
-Urban	13.9	4.51	
-Industrial	9.3	5.91	

Table 5. Comparison of particle-bound PCB homologue concentrations with particle size.

	Homologue	Small particle (μg PCBs/g particle)	Large particle (μg PCBs/g particle)
TSP (μg m^{-3})		118.6	38.3
Low molecular PCBs	Tri-CBs	2.5 (76%)	0.8 (24%)
	Tetra-CBs	1.9 (84%)	0.4 (16%)
High molecular PCBs	Penta-CBs	0.8 (73%)	0.3 (27%)
	Hexa-CBs	0.2 (83%)	0.04 (17%)
	Hepta-CBs	2.7 (65%)	1.5 (35%)
	Total PCBs	8.2 (72%)	3.1 (28%)

3.3 Estimation of gaseous PCBs

PCBs in the air exist mainly in the gas phase, but the concentrations of gaseous PCBs were not measured in this study. Therefore, the concentration of gaseous PCBs (C_g) was estimated with the measured average concentration of particulate PCBs (C_p) and the reported particle/gas partition coefficient (K_p) of each homologue based on the following simple equation (Pankow et al., 1997).

$$\log K_p = \log[(C_p/TSP)/C_g] \tag{1}$$

Where, C_p, C_g are in ng m^{-3}, respectively and TSP in μg m^{-3}. The result is shown in Table 6. The K_p value used in the calculation were from Falconer et al. (1995). Along with their measurement results of the gaseous PCBs concentrations for an urban

Table 6. Estimation of gaseous PCBs concentrations.

Congener	Homologue	log K_p at 25°C	Measured particulate PCBs (ng/m^{-3})	Estimated gaseous PCBs (ng m^{-3})	Gaseous PCBs levels measured by Falconer et al. (1995) (ng m^{-3})
37	Tri-CBs	−5.03	0.33	226	183~1100
49	Tetra-CBs	−5.33	0.25	335	19~382
101	Penta-CBs	−4.70	0.11	36	39~1890
138	Hexa-CBs	−3.94	0.03	2	15~18
171	Hepta-CBs	−3.64	0.37	10	71~95

area. With the increase of chlorine homologues, the particle/gas partition coefficient values increase. Thus, the higher MW PCBs are dominant in the particle phase. The estimated gaseous PCBs concentrations are in the range of the measured ranges.

3.4 Estimation of dry deposition fluxes of particulate PCBs

The predicted fluxes were calculated with the ten-step model in conjunction with the measured size distributions of total PCBs concentrations and the deposition velocities obtained from Sehmel-Hodgson model (Sehmel, 1973). This model combines the effects of eddy diffusion and particle inertia on particle motion by an "effective" eddy diffusion coefficient directed toward the surface. Input parameters for the Sehmel-Hodgson model are wind speed, ambient temperature, particle density ($\rho = 1$ g cm^{-3}), and particle size distribution. The dry deposition velocity depends strongly on particle size. The dry deposition velocities of small particles were below 0.2 cm sec^{-1} while those of large particles ranged from 1 to 10 cm sec^{-1} as shown in Figure 3. Thus, this model predicts that for particles larger than approximately 10 μm in diameter, their deposition velocity equals to the settling velocity (Sehmel, 1973).

The calculated flux of total particulate PCBs can be obtained by using the following equation from the multi-step model.

$$\text{Total particulate PCB flux in particle phase} = \sum_{i=1}^{10} V_{d,i} C_i \tag{2}$$
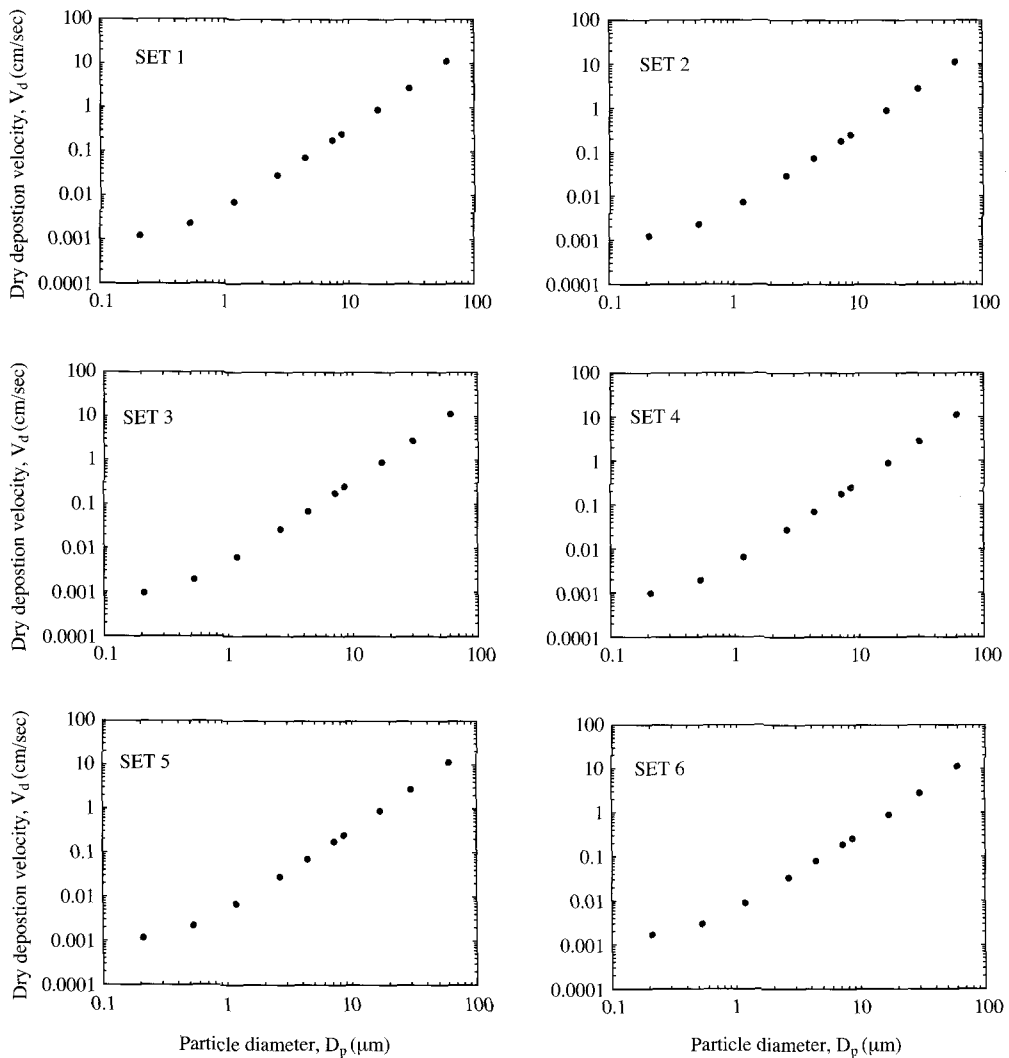
Fig. 3. The dry deposition velocity calculated from the Sehmel-Hodgson model for the six size distribution measurement results.

Where $V_{d,i}$ is the dry deposition velocity of the i-th size range and C_i is the ambient particulate PCBs concentration in the i-th size range. First, the entire particle size range was divided into ten intervals as in the measurement. Then, the appropriate deposition velocities for each interval size were calculated by the Sehmel-Hodgson model. Finally, the calculated fluxes for each interval were summed to calculate the total flux.

Based on the model calculation it was found that

PCBs associated with fine and coarse particles contribute from 2.5 to 42% and from 58 to 97.5% to the total PCB dry deposition flux. This can be explained by the higher dry deposition velocity for the coarse particles (10~100 times) than the one for the fine particles.

The measured total PCBs fluxes were compared to those calculated from the multi-step model (Eq. (2)) in Figure 4. The calculated fluxes agree well with the measured ones though the calculated fluxes
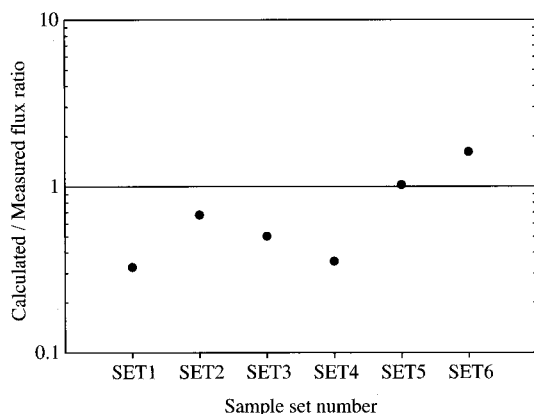
Fig. 4. The ratio of the calculated and measured total particulate PCBs fluxes.

were generally smaller than the measured ones. This was probably due to the underestimation of dry deposition velocity calculated from the Sehmel-Hodgson model.

4. SUMMARY

PCBs are one class of the persistent organic pollutants (POPs). Though PCBs are harmful to human, its behavior in the air, especially in Korea, is not well understood. In particular its dry deposition characteristics, an important mechanism of transferring PCBs from the atmosphere to water body or earth system, are poorly understood. A very important parameter that determines dry deposition fluxes is its particle size distribution. Ambient particle size distributions of PCBs and their dry deposition fluxes were measured at a site in Seoul.

The dry deposition plate was used to measure dry deposition fluxes of particulate mass and PCBs, a cascade impactor and rotary impactor were used to measure ambient particle size distributions for small $(D_p \leq 9\ \mu m)$ and large $(D_p > 9\ \mu m)$ particles, respectively. Six samples were collected from April 1999 to July 1999. Special emphasis was put on the sampling and analysis procedures adopted in this study.

The fluxes of particulate total PCBs (the sum of 43 congeners analyzed in this study) ranged from

$160 \sim 607$ ng m^{-2} day^{-1} and those of total particle mass from 0.087 to 0.53 μg m^{-2} day^{-1}. This result is comparable to those in Chicago for the PCBs. The size distribution of total PCBs was bimodal with the mass concentration being dominant in small particles. The mean particulate PCBs concentration was 6.9 μg PCBs/g TSP which was similar to the values measured at Bloomington, US.

The concentrations of PCB homologues in the gas phase were estimated based on the particle/gas partition coefficient (K_p) with the measured values of particulate PCBs in this study and they were comparable to those observed in other previous studies. The dry deposition fluxes were calculated by using the Sehmel-Hodgson model and the calculated fluxes were comparable to the measured ones.

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