

## Gas/Particle Level and Dry Deposition Flux of Atmospheric PCBs

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**Abstract :** Atmospheric samples were conducted from September 2001 to July 2002 with GPS-1 PUF sampler in rural site to concentration distributions of gas/particle PCBs and to calculate dry deposition flux of PCBs. ΣPCBs concentrations of gas/particle PCBs were  $59.29 \pm 48.83$ ,  $6.56 \pm 6.59$   $\mu\text{g}/\text{m}^3$ , respectively. Gas contribution (%) of total PCBs (gas + particle) was 90% which existed gas phase in the atmosphere. The particle contribution (%) of PCB congeners increased relatively more of the less volatile congeners with the highest chlorine number. The correlation coefficients (r) between total PCBs and temperature ( $^{\circ}\text{C}$ ) showed negative correlation in - 0.62 ( $p < 0.01$ ) for particle phase, positive correlation in 0.63 ( $p < 0.01$ ) for gas phase. In other word, particle phase PCBs is enriched in colder weather which could be due to greater in corporation of condensed gas phase at low temperature. The calculated dry deposition of total PCBs (gas + particle) was  $0.008 \pm 0.008$   $\mu\text{g m}^{-2} \text{day}^{-1}$  which showed maximum dry deposition flux in December, minimum data in July Bs in the atmosphere. The calculated dry deposition fluxes of total PCBs were influenced by particle phase PCBs even though PCBs in the atmosphere were present primarily in the gas phase.

**Keywords :** PCBs, gas phase, deposition velocity, contribution, dry deposition flux

### Introduction

The atmosphere is an important pathway for the transport and the global distribution of many substances of toxicological relevance. Organic compounds partition between atmospheric particles and the corresponding gas phase according to ambient temperature, compound properties and the particle surface available for sorption<sup>1,2</sup>. This process has a decisive influence on transport, deposition, and degradation processes and, hence, on the atmospheric residence times of the compounds<sup>3,4</sup>.

Atmospheric Dry deposition is the transfer of atmospheric gases and particles to the earth's surface, including soil, water, and vegetation where they are removed<sup>5</sup>. The removal rate by dry deposition is a function of the physical and chemical properties of the pollutant, meteorological conditions (temperature, wind speed, atmospheric stability), and surface characteristics. Since the mid-1980s it

has been commonly accepted that the impact of atmospheric deposition of semi-volatile organic compounds (SOCs) to the Great Lakes is large<sup>6</sup>. Historically, most efforts to determine the loadings of SOC<sub>s</sub> have monitored the concentrations of these compounds in precipitation<sup>7</sup>. Current dry deposition estimation methods often use measured air concentrations and modeled dry deposition velocities. These models assume the dry deposition flux of particles ( $F_p$ ) can be estimated by using an overall particle dry deposition velocity ( $V_p$ ) and particle phase air concentration ( $C_p$ ):

$$F_p = V_p \cdot C_p \quad (1)$$

To date there has been no consensus on the appropriate dry deposition velocity to use in these types of models. Estimated<sup>6</sup> and experimental<sup>8</sup> dry deposition velocities of SOC<sub>s</sub> range over an order of magnitude. One of the reasons for this discrepancy is that deposition velocity is a function of particle size. Gravitational settling has a significant effect on the deposition of coarse particle, while Brownian motion dominates the deposition of very fine particles ( $< 0.1 \mu\text{m}$ )<sup>5</sup>. As the particle diameter

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increases the magnitude of flux increases due to higher deposition velocities. A recent study has shown that a multi-step modeling approach, which divides the fine and coarse particle distributions into a number of intervals and assigns an appropriate deposition velocity to each interval, gives a better estimate of dry deposition than the approach shown in equation (1)<sup>8)</sup>.

The objectives of this study were to measure the gas/particle concentration and to estimate dry deposition flux of polychlorinated biphenyls(PCBs) in a rural atmosphere.

## Materials and Method

### Sampling programs

Atmospheric samples were taken from September 2001 to July 2002 with GPS-1 PUF sampler (General Metal Workes Inc., Ohio). The sampling program was conducted in a rural area of Kyonggi-do in South Korea. The sampling site is located at the National Hankyong University campus, which is about 1 km northeast of downtown Ansong, and 10 km west of Kyongbu express-way. The population of Ansong is about 130,000, population density is 882 (people/km<sup>2</sup>), and agricultural fields make up 33% of the total area in the sampling site. There is a complex mix between agricultural and industrial regions on the south side of Seoul (Fig. 1). Samples (N=28) were collected using a high-

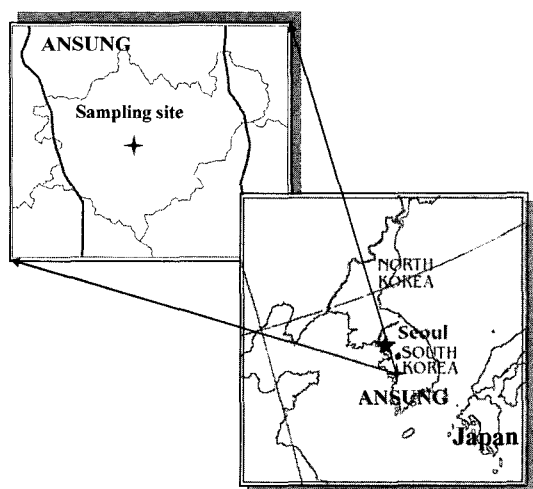


Fig. 1. Map of the sampling site.

volume air sampler. The air was drawn through a glass fiber filter (GFF, length 10 cm) to collect particles and then through a polyurethane form (PUF) plug (length 8.0 cm, diameter 7.5 cm) to collect compounds present in the gas phase. The duration of sampling was 48 hr, average flowrate was 0.23 m<sup>3</sup>/min and the sample volume was approximately 600-800 m<sup>3</sup>. Meteorological data such as temperature, wind speed, wind direction and relative humidity were obtained from a meteorological tower (AWS) located on top of a building located in the Hankyong University campus.

### Chemical analysis

The GFFs were precombusted at 450°C for 24 hours in loosely wrapped aluminum foil envelopes then they were sealed and stored at 4°C until sampling. The PUFs were precleaned by Soxhlet with hexane: dichloromethane (9:1 v/v) and then placed in a vacuum dry oven to dry and stored in sealed glassware at 4°C until sampling. After sampling, the GFFs and PUFs were all wrapped in glassware and stored at -26°C until analysis. PUFs were spiked with PCBs surrogate standards prior to sampling to determine analytical recovery. The surrogate standards were made up of PCB 28, 52, 101, 138, 153, 180, 209, and 4,4'-dibromooctafluorobiphenyls (Supelco, USA) were used as the internal standard. The GFF and PUF samples were extracted and analyzed separately in order to study the gas/particle distribution of PCBs. The samples were extracted in Soxhlet apparatus with hexane: dichloromethane (9:1 v/v) for 48 hours. Extracts were concentrated to approximately 2 ml in a rotary evaporator (BUCHI, R-124). The first clean-up of PCBs was eluted with hexane 15 ml, hexane: dichloromethane (1:1 v/v) 12 ml and dichloromethane 25 ml on a silica column (activated silica 3 g at 350°C overnight, sodium sulfate at 450°C overnight) to remove any polar organics that might interfere with the analysis. The second clean-up and fractionation were eluted with hexane: dichloromethane (1:1 v/v) on a GPC (gel permeation chromatography, BUCHI, B-688) column. The samples were then concentrated under a gentle stream of purified nitrogen concentrator to 50 µl by using dodecane (SIGMA, D-4259) which was added to the extracts to prevent losses during the volume reduction to 50 µl using N<sub>2</sub>

concentrator and analyzed on a Hewlett Packard 6890 equipped with MS detector (HP 5973). PCBs were analyzed by GC/MS with HP-5MS (5% diphenyl & 95% dimethylpolysiloxane) column [30 m (length), 0.25 mm (i.d), 0.25  $\mu\text{m}$  (film thickness)]. The temperature program was as follows: 150°C for 2 minutes, 30°C/min to 170°C, 4°C/min to 200°C for 1.3 minutes, 4°C/min to 268°C, 70°C/min to 320°C for 4.1 minutes.

#### Quality assurance/Quality control (QA/QC)

The analytical quality of the data was determined using limit of detection (LOD), recovery, reproducibility, linearity and by checking sampling artifacts. The detection limit was obtained by spiking a blank sample with the calibration standard at a concentration of 1-5 times the expected LOD. LOD is defined as three times the average mass plus 3 standard deviations ( $3\sigma$ ). The detection limit of the PCB standards ranged from 0.005 (PCB 18) to 0.04  $\text{pg}/\text{m}^3$  (PCB 52) with normalizing average sampling volume (800  $\text{m}^3$ ), respectively. The materials used in the present study were assessed for possible analytical contaminants. The purpose of blank test is to check whether and how much contamination of PCBs can be found in experiment materials. Due to the many experimental materials used in this study, it was necessary to check the background concentration level of PCBs in a considerable number of individual matrices. The solvent-cleaned and pre-combusted sampling materials, glass fiber filters, filters for PUF samplers, and PUF plug, were Soxhlet extracted by hexane: dichloromethane (9:1 v/v). The results of the blank tests did not show any interference peaks that would inhibit quantifying the PCB congeners in PUFs and GFFs. The recoveries ( $n=5$ ) of PCBs surrogate standards were  $77.2 \pm 5.5\%$  for PCB 28,  $62.9 \pm 7.3\%$  for PCB 52,  $78.0 \pm 9.2\%$  for PCB 101,  $100.8 \pm 9.7\%$  for PCB 153,  $106.1 \pm 8.2\%$  for PCB 138,  $116.6 \pm 10.2\%$ , and  $116.0 \pm 13.4\%$  for PCB 209. The linearity of calibration standards (Ultra Scientific Inc.) were calculated by regression analysis with values ranging from 0.9917-1.0000 ( $R^2$ ) for PCBs. Sampling artifacts associated with the GFF and PUF can affect the apparent gas-particle distributions of PCBs. The GFF may exhibit two such artifacts with counteracting effects on

the distribution. First, gas phase PCBs may adsorb to the filter surface and particles collected on the filter<sup>9-11</sup>. Second, the more volatile compounds may be stripped from the filter by continuing gas flow if the gas phase concentration decreases, the temperature increases during the sampling periods, or due to gas phase reactions on the filter<sup>12,13</sup>. The extent of gas adsorption is often estimated using a second filter. A backup filter was used on 5 samples collected in Ansong city. The percent mass on the second filter for individual PCBs was either below 5% or not detected at all. Therefore, the mass from the backup filter was neither subtracted from the particle phase concentration nor added to the gas phase concentration. Also, volatilization from the filter has the opposite affect. This was not determined, but Zhang and McMurry<sup>13</sup> have suggested that this impact is usually less than or equal to 10%.

Spilt PUFs were collected to assess gas phase breakthrough. The bottom half of the spilt PUFs contained an average of 12% ( $N=3$ ) of the total mass, indicating minimal breakthrough. Therefore, the gas phase concentrations in this study were adjusted accordingly. Simcik *et al.*<sup>14</sup> examined gas phase breakthrough in the same manner with this study in Chicago and over Lake Michigan during July when the ambient temperature was highest. Individual PCBs on the bottom half of the spilt PUFs were all less than 15% of their total PUF masses. We concluded that the GFF- and PUF-associated concentrations of PCBs are a good estimate of the true gas and particle concentrations in this study.

## Results and Discussion

### Gas/particle distribution of PCBs

PCBs are SOCs which are found in the atmosphere as gases and are associated with suspended particle matter. The gas to particle ratio is important in the atmospheric removal by rain and dry deposition<sup>15</sup>. The distributions of PCBs between gas and particle phases were dependent on temperature, humidity, adsorption surface available, molecular weight and vapor pressure of compounds.

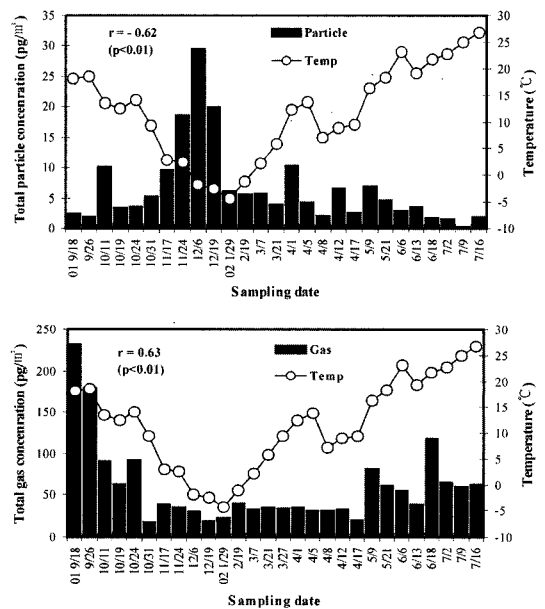
The mean concentrations of gas and particle PCBs in this study were  $59.29 \pm 48.83$ ,  $6.56 \pm 6.59$

**Table 1.** Concentration of PCB homologs in the gas and particle phase

Homologs	Particle phase (pg/m <sup>3</sup> )			Gas phase (pg/m <sup>3</sup> )		
	Mean	Min	Max	Mean	Min	Max
Tri-CBs	0.65	0.15	2.36	24.15	5.73	70.46
Tetra-CBs	1.38	0.12	5.12	25.39	5.48	144.7
Penta-CBs	1.86	0.08	9.72	8.59	2.92	22.88
Hexa-CBs	1.46	0.07	7.40	3.59	0.96	9.84
Hepta-CBs	0.70	0.08	3.02	0.89	0.35	2.93
Octa-CBs	0.20	0.02	0.75	0.14	0.06	0.21
Deca-CBs	0.67	0.04	3.16	0.16	0.06	0.34
Total PCBs <sup>1)</sup>	6.56	0.41	29.52	62.59	17.30	232.8

<sup>1)</sup>Total PCBs : summation of homologs.

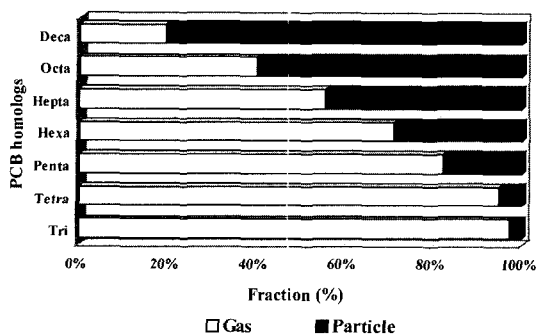
pg/m<sup>3</sup>, respectively. About 90% of total PCBs (gas + particle) showed the gas phase, which was agreement with previous studies<sup>3,16</sup>. Also, gas phase contributions (%) of total PCBs were investigated in Tawain of urban and rural areas which consist of 63% for urban and 59% for rural area<sup>17</sup>. Higher concentration in the particle phase showed from congeners with six and ten chlorine atoms (Table 1). Lower concentration of lighter molecular congeners with three and five chlorine atoms was appeared than in the corresponding gas phase. The maximum  $\Sigma$ PCBs (24 congeners) values in the gas phase (232.8 pg/m<sup>3</sup>) were obtained in September, and the minimum value (17.30 pg/m<sup>3</sup>) was obtained in October. The maximum  $\Sigma$ PCBs concentration in particle (29.52 pg/m<sup>3</sup>) measured in December, and the minimum  $\Sigma$ PCB concentration (0.41 pg/m<sup>3</sup>) was measured in July. Mandalakis *et al.*<sup>18</sup> reported  $\Sigma$ PCBs (38 congeners) mean concentrations in the urban center of Athens which were 344.9 pg/m<sup>3</sup> for gas phase and 3.7 pg/m<sup>3</sup> for particle phase. And, the  $\Sigma$ PCBs concentration at Penteli, considered as a background station for the Athens area, was 80.5 pg/m<sup>3</sup> and the corresponding concentration of the coastal area (Saronikos gulf) was 184.4 pg/m<sup>3</sup>.<sup>18</sup> The mean  $\Sigma$ PCBs concentration (92 pg/m<sup>3</sup>) measured in a rural site of Greece approached that of Penteli<sup>18</sup>. The mean concentration of  $\Sigma$ PCBs measured in this study were considerably lower than that of urban area in Athens, were similar to background area in Athens. Fig. 2 shows that correlation coefficients of total PCBs between gas, particle phase and temperature during the sampling



**Fig. 2.** Concentrations of  $\Sigma$ PCBs in gas and particle phase during the sampling periods.

periods. The correlation coefficients ( $r$ ) between total PCBs and temperature ( $^{\circ}\text{C}$ ) showed negative correlation in  $-0.62$  ( $p < 0.01$ ) for particle phase, positive correlation in  $0.63$  ( $p < 0.01$ ) for gas phase. In other word, particle phase PCBs is enriched in colder weather which could be due to greater in corporation of condensed gas phase at low temperature.

Fig. 3 shows the contribution of each PCB congener to total PCBs in gas and particle phase.



**Fig. 3.** The average homolog fractions (%) of total PCBs during the sampling periods.

The general pattern of contribution (%) for PCB congeners to total PCBs in gas phase increased with decreasing chlorine substitution. The particle contribution (%) of PCB congeners increased relatively more of the less volatile congeners, i.e., with the highest chlorine number. Similar observations on the increasing importance of the particle phase PCBs with increasing molecular weight have also been made for SOCs<sup>19</sup>. Lee *et al.*<sup>17</sup> also reported that higher chlorinated PCB homologs were found to have significant fraction in particle phase due to their molecular weight and low vapor pressure.

### Dry deposition of total PCBs (gas + particle) and homologs

Like other semivolatile organic compounds, PCBs dry deposition flux was a combination of both the gas and the particle phase flux, and is given by

$$F_T = F_G + F_P = C_G \cdot V_{D,G} + C_P \cdot V_{D,P} \quad (2)$$

Where  $F_T$  is the PCB dry deposition flux ( $\mu\text{g m}^{-2} \text{day}^{-1}$ ) contributed by the summation of both the gas phase and the particle phase,  $F_G$  is the PCB dry deposition flux ( $\mu\text{g m}^{-2} \text{day}^{-1}$ ) contributed by the gas phase,  $F_P$  is the dry deposition flux ( $\mu\text{g m}^{-2} \text{day}^{-1}$ ) contributed by the particle phase,  $C_G$  is the PCB concentration in the gas phase ( $\text{ng m}^{-3}$ ) which is measured by the PUFs of the high volume PUF sampler,  $V_{D,G}$  is the dry deposition velocity ( $\text{cm s}^{-1}$ ) of the gas phase PCBs,  $C_P$  is the PCB concentration in the particle phase ( $\text{ng m}^{-3}$ ) which is measured by the glass fiber filter of the high volume PUF sampler, and  $V_{D,P}$  is the dry deposition velocity ( $\text{cm s}^{-1}$ ) of the particle phase PCBs.

In the atmosphere, naphthalene (Nap) is one of SOCs. It is primarily in the gas phase. Less than 2.1% of Nap mass was found in the particle phase. The mean dry deposition velocity of Nap ( $0.011 \text{ cm s}^{-1}$ ) was estimated by Sheu *et al.*<sup>20</sup> Dry deposition of gas phase SOCs is mainly by diffusion, while the particle phase SOCs is mainly by the gravitational settling. Compared with particle phase, the dry deposition velocity of gas phase SOCs is fairly constant. A selected value ( $0.010 \text{ cm s}^{-1}$ ) of gas phase PAH dry deposition velocity ( $V_{D,G}$ ) was proposed by sheu *et al.*<sup>20</sup> Here, this value ( $0.010 \text{ cm s}^{-1}$ ) can also be used for the approximate calculation of PCB dry deposition flux contributed by its gas phase. Also, the dry deposition velocity of particle phase PCBs used the value of measuring in the rural atmosphere of Taiwan by Lee *et al.* (1996). By using equation (2), the calculated dry

**Table 2.** Particle Dry deposition fluxes of  $\Sigma$ PCBs measured at various locations

Flux ( $\mu\text{g}/\text{m}^2 \cdot \text{day}$ )	Year	location	Sampling method	Reference
< 0.05-0.15	73-74	Southern California	glass plate, mineral oil	Young <i>et al.</i> , 1975
0.05-1.7	74	Los Angeles basin	glass plate, mineral oil	Young <i>et al.</i> , 1975
< 0.3-0.42	73-75	Kingston, RI	aluminum pans, glycerin-water	Bidleman <i>et al.</i> , 1981
< 0.035-0.45	76-79	Columbia, SC	aluminum pans, glycerin-water	Bidleman <i>et al.</i> , 1981
4.5	89-90	Chicago, IL	filter, wetted with propane	Holsen <i>et al.</i> , 1991
0.87	92	Door Co., WI	Mylar strips, Apezion L grease	Thomas <i>et al.</i> , 1998
0.08	93-95	Offshore Chicago, IL	Mylar strips, Apezion L grease	Thomas <i>et al.</i> , 1998
0.06	93-95	Sleeping Bear Dunes, MI	Mylar strips, Apezion L grease	Thomas <i>et al.</i> , 1998
0.008	01-02	Ansung city	glass filter filter	This study

deposition of total PCBs (gas + particle) showed approximately  $0.008 \pm 0.008 \mu\text{g m}^{-2} \text{day}^{-1}$  which was down to 1-2 orders of magnitude lower than those measured at Lake Michigan (South Haven MI, and Steeping Bear Dunes, MI), Chicago IL, and Greck Lake (Table 2)<sup>8,17,21</sup>.

The mean particle dry deposition fluxes of PCB homologs in this study were calculated by the values of Lee *et al.*<sup>17</sup> These velocities of particle dry deposition proposed by Lee *et al.*<sup>17</sup> were 0.32, 0.47, 0.57, 0.70, 0.94, 0.84, and 0.98  $\text{cm s}^{-1}$  for the tri-, tetra-, penta-, hexa-, hepta-, octa-, and deca-CBs, respectively. This is due to the fact that the low molecular PCB homologs are enriched in the gas phase. Also, deposition velocity of gas phase used in value ( $0.010 \text{ cm s}^{-1}$ ) proposed by sheu *et al.*<sup>20</sup>.

The monthly calculated dry deposition of PCB homologs shown in Fig. 4. The calculated dry deposition flux of total PCBs (gas + particle) were varied between 0.0013 in the July and 0.0143  $\mu\text{g m}^{-2} \text{day}^{-1}$  in the December. The mean dry deposition flux of PCB homologs were 0.00032  $\mu\text{g m}^{-2} \text{day}^{-1}$  for tri-CBs, 0.00068  $\mu\text{g m}^{-2} \text{day}^{-1}$  for tetra-CBs, 0.00103  $\mu\text{g m}^{-2} \text{day}^{-1}$  for penta-CBs, 0.00094  $\mu\text{g m}^{-2} \text{day}^{-1}$  for hexa-CBs, 0.0058  $\mu\text{g m}^{-2} \text{day}^{-1}$  for hepta-CBs, 0.00014  $\mu\text{g m}^{-2} \text{day}^{-1}$  for octa-CBs, and 0.00058  $\mu\text{g m}^{-2} \text{day}^{-1}$  for deca-CBs, respectively. All PCB homologs showed maximum dry deposition flux in December, minimum value in July which was highly associated with the particle phase

variation of PCBs in the atmosphere ( $r=0.99$ ,  $p<0.001$ ). That is, since particle have much higher deposition velocities than gases they may be important in understanding the dry deposition of PCBs.

## Summary

This study was conducted to investigate the gas/particle distributions and dry deposition flux of PCBs in rural atmosphere from September 2001 to July 2002.

Mean  $\Sigma$ PCBs concentrations of gas and particle phase were  $59.29 \pm 48.83$ ,  $6.56 \pm 6.59 \text{ pg/m}^3$ , respectively. About 90% of total PCBs existed gas phase in the atmosphere of Korea. The particle contribution (%) of PCB congeners increased relatively more of the less volatile congeners with the highest chlorine number.

The calculated dry deposition of total PCBs (gas + particle) was  $0.008 \pm 0.008 \mu\text{g m}^{-2} \text{day}^{-1}$  which was down to 1-2 orders of magnitude lower than other studies. All PCB homologs showed maximum dry deposition flux in December, minimum value in July which was highly associated with the particle phase variation of PCBs in the atmosphere ( $r=0.99$ ,  $p<0.001$ ). The calculated dry deposition fluxes of total PCBs were influenced by particle phase PCBs even though PCBs in the atmosphere were present primarily in the gas phase.

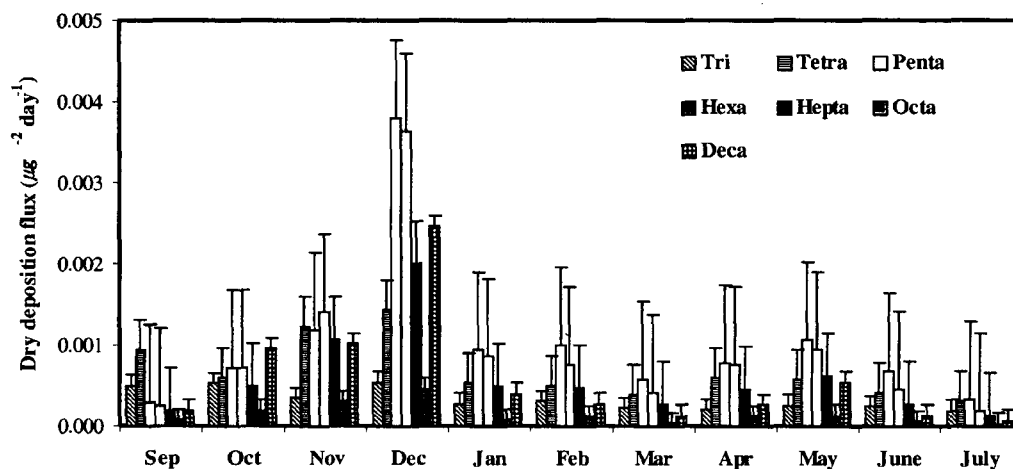


Fig. 4. Monthly calculated dry deposition flux (gas + particle) of PCB homologs.

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