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Concentration and Seasonal Variation of Particle PCBs in Air

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Abstract

Atmospheric concentrations of PCBs were monitored in Ansung-city, Kyonggi province during the 2001/2002 to characterize the concentration distribution and seasonal variation of particle polychlorinated biphenyls (PCBs). Average concentration of particle bound PCB showed maximum value for penta-CBs and minimum value for octa-CBs. Seasonal contributions (%) of total particle PCBs showed the highest value in winter months and lowest value in summer month. This result indicated that concentration of total particle PCBs increased with decreasing temperature in the atmosphere. Therefore, particle PCBs were easily formed by the condensation of gas phase PCBs in winter months.

The total particle PCBs exhibited an inverse correlation with temperature ($p < 0.01$) which suggested that particle PCBs were easily formed by condensation of gaseous PCBs in winter months.

Key words : PCBs homologs, Seasonal contribution, Particle bound, Temperature

1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are considered as significant environmental contaminants due to their ecotoxicological and human health implications. PCBs are included in the broad category of persistent organic pollutants (POPs) that is a group of compounds with specific properties: persistency, high bioaccumulative coefficient, ability for transmission to large distances (Harrad *et al.*, 1994). As our understanding of semi-volatile organic compounds (SOCs) in the environment has improved it has become increasingly important not only to measure the atmospheric concentrations of these compounds but also to gain an understanding of the role of the atmosphere in the deposition, degradation, transportation (Cotham and Bidleman, 1995). Polychlorinated biphenyls (PCBs) production and use were phased out and banned by the mid-1970s in most countries. PCBs became ubiquitous pollutants in all environmental compartments, including seawater, air, vegetation, soil and animals, all over the world. The major source of PCBs to the atmosphere is volatilization from sites where they have been disposed or stored and incineration of PCB-containing materials (Simcik *et al.*, 1998). The partitioning of SOCs to atmospheric aerosols is an important factor in their fate and transport in urban and rural areas, and globally. PCBs in the air exist mainly in the gas phase and there is a wide range of reported values on the gas and particle distribution percentages. For example, a study in Bloomington, IN showed that the gas phase contained 99% and

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90% of the total PCBs in the summer and winter periods, respectively (Hermanson and Hites, 1989). Also, the contribution of gas and particle phase in Toronto, Ontario showed 57~86%, 14~43%, respectively (Robert *et al.*, 2000). Chen *et al.* (1996) reported that contribution of gas and particle phase in Southern Taiwan showed 60.9% and 39.1%, respectively. Also, concentration distribution for particle and gas phase PCBs was investigated in many foreign countries. Kouimtzis *et al.* (2002) studied that particle-bound PCBs was monitored at two sites in northern Greece to discuss the relationship with possible source influence and prevailing meteorology and calculate the dry deposition of particle bound PCBs. As to the above researchers, the contributions of particle phase have a function of ambient temperature, source characteristics, and meteorological conditions. Kaupp and McLachlan (1999) studied to establish the baseline PCBs contamination with gas and particle phase of the environment prior to the start-up of a new municipal incinerator, air samples were collected continuously over a period of one year. Lohmann *et al.* (2000) compared the partitioning of PCDD/Fs, PAHs, and PCBs at different locations in Northern England to better understand the importance of source types, the proximity to sources, and influence of meteorology on the gas-particle distributions of these important compound classes. Also, different types of atmospheric particles will likely exhibit different properties for sorption of SOCs from the gas phase (Venkataraman and Friedlander, 1994; Pankow and Bidleman, 1992). However, the researches of SOCs in the environmental matrixes are still at an early stage in Korea. A number studies have been conducted for PCDD/Fs, PAHs (Park and Kim, 2002; Park *et al.*, 2002; Oh *et al.*, 2002) and PCBs (Yeo *et al.*, 2004; 2003a,b, Ikonomou *et al.*, 2002; Shin *et al.*, 2002; Yeo *et al.*, 2002a,b,c; Kong *et al.*, 1999) in the atmosphere of Korea. For instance, Yeo *et al.* (2002a) was investigated to study concentration distribution of PCBs and to identify source characteristics of each PCB homolog in rural area. Yeo *et al.* (2002b) was studied to investigate relationship between the atmospheric concentration of PCBs and temperature were measured at urban site. Shin *et al.* (2002) studied the size distribution of particle PCBs to investigate spatial distribution of PCBs at 6 sites. Ikonomou *et al.* (2002) investigated PCB levels from Korean municipal waste incinerator to determine possible groupings of similar emissions and which congeners are significant in defining such group. Kong *et al.* (1999) investigated PCBs sources, levels and distributions to the environment such as stack gas, fly ash, soil, and human milk to study toxic evaluation for PCBs.

Concentration distribution and seasonal variation of particle PCBs in this study were investigated in order to better understand temperature dependence of particle PCBs in the atmosphere. And similar grouping of PCB homologs to identify source characteristics during the sampling periods was examined in using correlation analysis and principal component analysis (PCA). Furthermore, concentration and seasonal variation of particle PCBs measured in this study offer the useful information to obtain particle PCBs source profiles for a number of possible local sources such as motor vehicles, domestic heating facilities, open burning of refuse, open burning of biomass and fly ash for another in future researches. Therefore, This work presented novel approaches to understand atmospheric behavior of particle PCBs depending on temperature variation regardless of gaseous PCBs concentration.

2. EXPERIMENT

Sampling was performed in rural atmosphere of Kyonggi-do in Korea. The sampling site is located at the Hankyong National University campus, which is about 1 km northeast of downtown Ansung, and 10 km west of Kyongbu-express way. Air Samples were taken between 2001 and 2002 on a meteorological tower at a height of 10 m from the ground. The population is about 130,000 and population density (population/km^2) is 882 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. Air

samples (N = 27) were collected using high-volume sampler (GPS-1, General Metal Workes Inc., Ohio). The air was drawn through a glass fiber filter (GFF) to collect particles for 48 hr and the sample volume was approximately 600~800 m^3. Meteorological Data such as temperature, wind speed, wind direction and relative humidity were obtained from meteorological tower (AWS) located on top of a building located in the Hankyong National University campus.

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. After sampling, the GFFs were all wrapped in glassware and stored at −26°C until analysis. GFFs were spiked with PCBs surrogate standards (^{13}C) prior to extraction to determine analytical recovery. The surrogate standards were composed of PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, PCB 180, PCB 209. The GFFs were extracted in sonicator apparatus with hexane: dichloromethane (9 : 1 v/v) for 3 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 3g 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_2 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% dimethypolysiloxane) column [30 m (length), 0.25 mm (i.d), 0.25 μm (film thickness)]. The temperature program of PCBs was as follows: 150°C for 2 minutes, 30°C/min to 170°C, 4°C/min to 200°C for 13 minutes, 4°C/min to 268°C, 70°C/min to 320°C for 4.1 minutes.

The analytical quality of the data was determined using LOD (limit of detection), recovery, reproducibility and linearity. LOD of filter 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 (mean±3σ). The detection limit of the PCB standards ranged from 0.011 (PCB 18)-0.124 pg/m^3 (PCB 52). The recoveries (n = 5) of PCB surrogate standards were 77.2±5.5% for PCB 28, 62.9±7.3% for PCB 52, 78.0±9.2% for PCB 101, 100.8±9.7% for PCB 153, 106.1±8.2% for PCB 138, 116.6±10.2% for PCB 180, and 116.0±13.4% for PCB 209. The linearity of calibration standards were calculated by regression analysis with values ranging from 0.9917~1.0000 (R^2) for PCBs. Full detailed of QA/QC were detailed in Yeo *et al.* (2003a,b).

3. RESULTS AND DISCUSSION

3. 1 Concentration distribution

Table 1 shows the collection data and concentration of total particle PCBs (sum of 25 congeners: PCB 18, 28, 47, 52, 60, 77, 87, 101, 105, 110, 123, 126, 138, 141, 149, 153, 155, 170, 180, 183, 189, 194, and 209) in rural site during the sampling periods. This study was taken about 48 hour sampling time to determine particle PCB concentrations. A separate sampling train consisting of a single glass fiber filter was used for total suspended particle determination (TSP in μg/m^3). The filter pre-weighted after equilibrium in a constant humidity chamber for 24 hour. Average temperatures in this study were apparent seasonal variation: Monthly average temperature was 12.7±6.9°C for fall (from September to November), −2.5±1.4°C for winter (from December to February), 10.4±4.9°C for spring (from March to May), and 23.1±2.6°C for summer (from June to July). Monthly average TSP concentrations were 69.0±23.2 μg/m^3 for fall, 86.6±7.8 μg/m^3 for winter, 287.5±358.6 μg/m^3 for spring, and 60.8±34.1 μg/m^3 for summer.

Concentration distributions of PCB homologs in

Table 1. Concentration of total particle PCBs, TSP and ambient temperature.

Sampling date	Temp (°C)	TSP (μg/m^3)	Total particle PCBs (pg/m^3)
9/18~9/20, 01	18.7	79.8	2.45
9/26~9/28, 01	18.5	67.2	1.92
10/11~10/13, 01	13.4	45.6	10.2
10/19~10/21, 01	12.5	67.1	3.60
10/24~10/26, 01	14.0	114.8	3.73
10/31~11/2, 01	9.4	50.1	5.28
11/17~11/19, 01	2.9	-	9.77
11/24~11/26, 01	2.5	58.4	18.56
12/6~12/8, 01	−1.8	80.0	29.52
12/9~12/11, 01	−2.5	-	20.08
1/29~1/31, 02	−4.4	84.4	6.19
2/19~2/21, 02	−1.2	95.2	5.63
3/7~3/9, 02	2.2	85.4	5.83
3/21~2/23, 02	5.8	1135.4	4.11
4/1~4/3, 02	12.4	141.8	10.46
4/5~4/7, 02	13.8	-	4.32
4/8~4/10, 02	7.2	616.1	2.15
4/12~4/14, 02	8.9	106.7	6.73
4/17~4/19, 02	9.5	168.2	2.71
5/9~5/11, 02	16.3	106.6	7.04
5/21~5/23, 02	18.3	112.5	4.76
6/6~6/8, 02	23.1	107.0	3.06
6/13~6/15, 02	19.2	60.2	3.75
6/18~6/20, 02	21.7	96.9	1.77
7/2~7/4, 02	22.8	33.4	1.61
7/9~7/11, 02	24.9	24.1	0.45
7/16~7/18, 02	26.9	43.5	1.85
Mean	11.6	149.2	6.58
S.D	9.02	238.9	6.58
Min	−4.4	24.1	0.45
Max	26.9	1135.5	29.52

− : not measured

particle phase were shown in Fig. 1. Due to various factors during meteorological conditions and atmospheric transport process, the range of particle PCBs concentration was rather scattered being 0.46±0.60 pg/m^3 for tri-CBs, 1.38±1.32 pg/m^3 for tetra-CBs, 1.86±1.94 pg/m^3 for penta-CBs, 1.46±1.58 pg/m^3 for hexa-CBs, 0.70±0.67 pg/m^3 for hepta-CBs, 0.20±0.19 pg/m^3 for octa-CBs, and 0.67±0.81 pg/m^3 for deca-CBs, respectively. Mean concentration of particle PCB homologs showed penta-CB > hexa-CBs > tetra-CBs > hepta-CBs > deca-CBs > tri-CBs > octa-CBs. Kouimtzis *et al.* (2002) studied PCBs (sum of six congener: PCB28, 52,

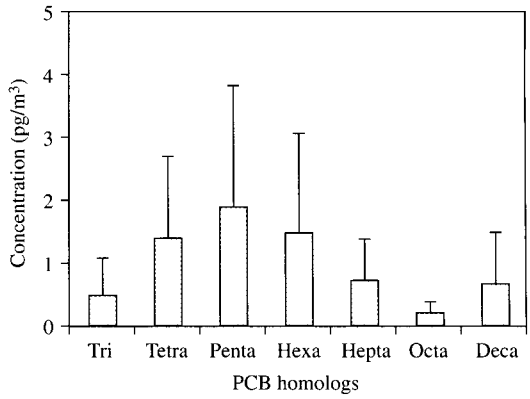

Fig. 1. Concentration distribution of PCB homologs in the particle phase (error bar: standard deviation).

101, 138, 153, 180) in airborne particle matter of the greater Thessaloniki area, N. Greece. There is a distinct different in PCB concentrations between urban (mean: 9.8 pg/m^3) and rural site (mean: 3.0 pg/m^3). Generally, urban areas are more polluted with PCBs in comparison to rural areas.

The PCB mass adsorbed on the atmospheric particle and normalized by the particle mass was called particle bound PCB composition. The compositions of total particle bound PCBs was 0.08± 0.09 μg/g. Lee *et al.* (1996) reported the total particle bound PCBs in Taiwan which 11.3±31.0 μg/g for petroleum refinery known by potential PCBs sources, 11.1±70.9 μg/g for urban site, and 9.87± 32.1 μg/g for rural site. Also, total particle bound PCBs compositions was 0.1~9.6 μg/g in Bloomington, IL (Hermanson and Hites, 1989), 30~50 μg/g in Chicago, IL (Holsen *et al.*, 1991). This value in this study was 2~3 orders of magnitude lower than those measured at urban and rural areas by previous studies. This result was rather affected by Yellow Sand Phenomenon with the TSP in 1135 μg/m^3 (March, 2002) and in 616 μg/m^3 (April, 2002). The relationship between the particle bound PCB composition and measured TSP concentration varies rather significantly ($p < 0.05$) because the partitioning will differ extensively depending on site, meteorological conditions and particle characteristics (Falconer and Bidleman, 1994).

3. 2 Seasonality

Seasonal variations of particle PCBs were noted during the study. The seasonal contributions (%) of PCB homologs during the sampling periods were shown in Table 2. The seasonal mean contribution of PCB homologs for total particle PCBs was calculated in order to make a direct comparison of PCB homologs distribution between seasons (Fig. 2). Seasonal mean contribution was divided based on similar monthly temperature: winter includes December-February, spring include March-May, summer includes June-July, and fall includes September-November. Maximum contribution of PCB homologs appeared 45.2% for tri-CBs, 43.3% for penta-CBs, 46.3% for hexa-CBs, in winter month except tetra-CBs and octa-CBs. However, minimum contribution of PCB homologs showed in summer month: 6.8% for tri-CBs, 8.4% for tetra-CBs, 10.4% for penta-CBs, 7.7% for hexa-CBs, 9.2% for hepta-CBs, 6.7% of octa-CBs, 4.3% for decta-CBs. Most of PCB homologs showed maximum contribution in winter, minimum in sum-mer. Grenier and Chevreuil (1997) mentioned a clear augmentation of particle PCBs in winter, that can be linked both to a drop of temperature and an increase in suspended particles. In other words, particle matter is enriched in PCBs in colder weather. This could be due to greater incorporation of condensed gas phase material at low temperature. Also, Table 3 showed the concentration of ICES (International Council for the Explanation of the Seas) congeners in particle phase compared to other researchers.

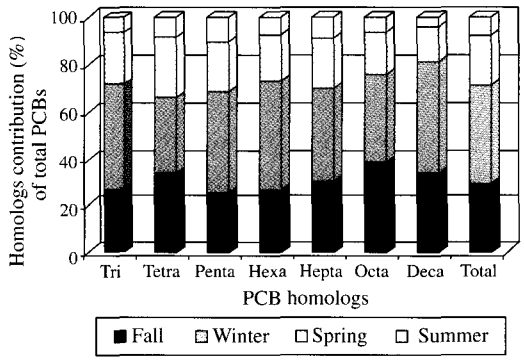

Fig. 2. Seasonal contribution of PCB homologs in the particle phase.

Table 2. Seasonal contributions of PCB homologs in the particle phase.

Contribution (%)	Tri	Tetra	Penta	Hexa	Hepta	Octa	Deca	Total
Fall	26.5	33.4	24.8	26.4	30.0	38.5	33.7	28.8
Winter	45.2	32.2	43.3	46.3	39.8	37.0	47.1	41.7
Spring	21.4	26.0	21.4	19.7	21.0	17.8	14.9	21.2
Summer	6.8	8.4	10.4	7.7	9.2	6.7	4.3	8.3

The ΣICES concentrations were 1.96 pg/m^3 for rural site in this study, 9.80 pg/m^3 for urban site (Tessaloniki, Greece) surrounded by several residential communities, while an extended industrial area such as oil refining, petrochemical, fertilizer production is located northwesterly (Kouimtzis *et al.*, 2002), 3.00 pg/m^3 for semi-rural site was located at the outskirts of the residential community Halastra, about 20 km west of Thessaloniki, Greece (Kouimtzis *et al.*, 2002), and 3.15 pg/m^3 for rural site where impacted by anthropogenic inputs of SOCs from the adjoining metropolitan area and, in the case of PCBs, from the Upper Hudson River (Totten *et al.*, 2001). The correlation coefficients of ICES concentrations between this study and other researchers (Kouimtzis *et al.*, 2002; Totten *et al.*, 2001) were very significant ($0.73 < r < 0.92$, $p < 0.01$), which suggested that concentration patterns of ICES congeners in the atmosphere were similar because of their physicochemical properties such as

Table 3. Concentrations of ICES congeners in the particle phase (pg/m^3)

ICES congeners	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	Total ICES
This study [Rural site]	0.41	0.14	0.43	0.39	0.34	0.25	1.96
Kouimtzis *et al.* (2002) [Urban site]	1.07	2.06	2.10	1.66	1.91	1.58	9.80
Kouimtzis *et al.* (2002) [Semi-rural site]	0.87	1.41	1.20	0.73	0.73	0.53	3.00
Totten *et al.* (2001) [Rural site]	0.23	0.95	0.59	0.65	0.46	0.27	3.15

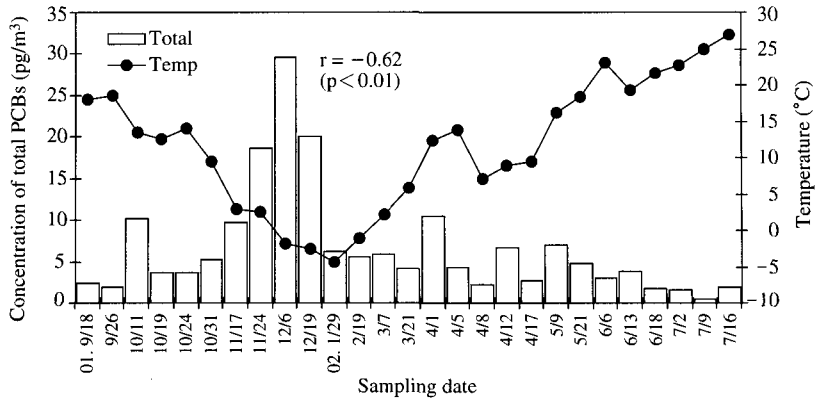

Fig. 3. Daily variation of total particle PCBs.

Table 4. Correlation coefficients between PCB homologs during the sampling periods.

	Tri-CBs	Tetra-CBs	Penta-CBs	Hexa-CBs	Hepta-CBs	Octa-CBs	Deca-CBs
Tri-CBs	1.00						
Tetra-CBs	0.75 ($p<0.01$)	1.00					
Penta-CBs	0.79 ($p<0.01$)	0.74 ($p<0.01$)	1.00				
Hexa-CBs	0.79 ($p<0.01$)	0.81 ($p<0.01$)	0.97 ($p<0.01$)	1.00			
Hepta-CBs	0.82 ($p<0.01$)	0.85 ($p<0.01$)	0.94 ($p<0.01$)	0.98 ($p<0.01$)	1.00		
Octa-CBs	0.67 ($p<0.01$)	0.84 ($p<0.01$)	0.83 ($p<0.01$)	0.90 ($p<0.01$)	0.94 ($p<0.01$)	1.00	
Deca-CBs	0.64 ($p<0.01$)	0.64 ($p<0.01$)	0.81 ($p<0.01$)	0.84 ($p<0.01$)	0.84 ($p<0.01$)	0.86 ($p<0.01$)	1.00

K_{OA}(octanol-air partition coefficient), vapor pressure(Yeo *et al.*, 2003b)

Figure 3 showed the daily variations of PCBs homologs in particle phase. Total particle PCBs concentration varied between 0.45 and 29.52 pg/m^3 (mean: 6.58±6.58 pg/m^3). In Athens, mean total particle PCBs (38 congeners) concentrations were 3.7 pg/m^3 for urban area, 3.3 pg/m^3 for coastal area, and 2.0 pg/m^3 for background area (Mandalakis *et al.*, 2002). Total particle PCBs in this study was higher than those of Athens. Also, the highest and lowest concentration of total PCBs showed December and July, respectively. The reasons why winter months marked higher concentration of total particle PCBs than summer months indicated that first, any other sources of gas phase PCBs were decreasing with freezing or covering the snow on the ground and second, gas/particle partitioning progresses to the particle phase with decreasing temperature ($r = -0.62$, $p<0.01$). This tendency easily found in middle number of chloride for PCBs such as penta-CBs through hepta-CBs having a similar fraction between gas and particle phase

The correlation coefficients between PCB homologs were shown in Table 4. In this study, a strong correlations ($r>0.9$) were observed: first, between penta-CBs and other homologs (such as hexa-CBs, hepta-CBs) and second, between hexa-CBs and hepta-CBs, octa-CBs, and third, between hepta-CBs and octa-CBs. A weaker correlation was only found between tri-CBs and octa-CBs, deca-CBs.

Principle components analysis (PCA) was performed using SPSS 10 statistical package. The scope of the PCA analysis is to group the compounds according to the similarities of variations in the PCB homologs concentration during the sampling periods. Twenty seven samples containing seven variables were subjected to factor analysis

with the varimax rotation. Only those factors corresponding to correlation matrix eigenvalues of >1 were retained as a factor. From the PCA analysis for PCB homologs, it was shown in one group to account for 85% of total sample variances. Overall correlation coefficients and PCA analysis between PCB homologs were very significant ($p<0.01$) which suggests that the source characteristics remained relatively constant over the sampling periods. Therefore, PCB homologs measured in this study were affected by similar sources during the sampling periods.

3. 3 Temperature dependence

The correlation coefficients between concentrations of PCB homologs and inverse absolute temperature (1000/T) were shown in Fig. 4.

Correlation coefficients (r) between PCB concentration in particle phase and inverse temperature showed positive significant correlations ($p<0.05$) for all PCB homologs: $r=0.64$ for tri-CBs, $r=0.65$ for tetra-CBs, $r=0.68$ for penta-CBs, $r=0.70$ for hexa-CBs, $r=0.69$ for hepta-CBs, $r=0.54$ for octa-CBs, and $r=0.54$ for deca-CBs ($r=0.72$ for total PCBs). These results suggested that PCBs concentrations in particle phase increased with decreasing ambient temperature especially in penta-CBs through hepta-CBs. Generally, relationship between particle phase PCBs and temperature showed negative correlation coefficients compared to those

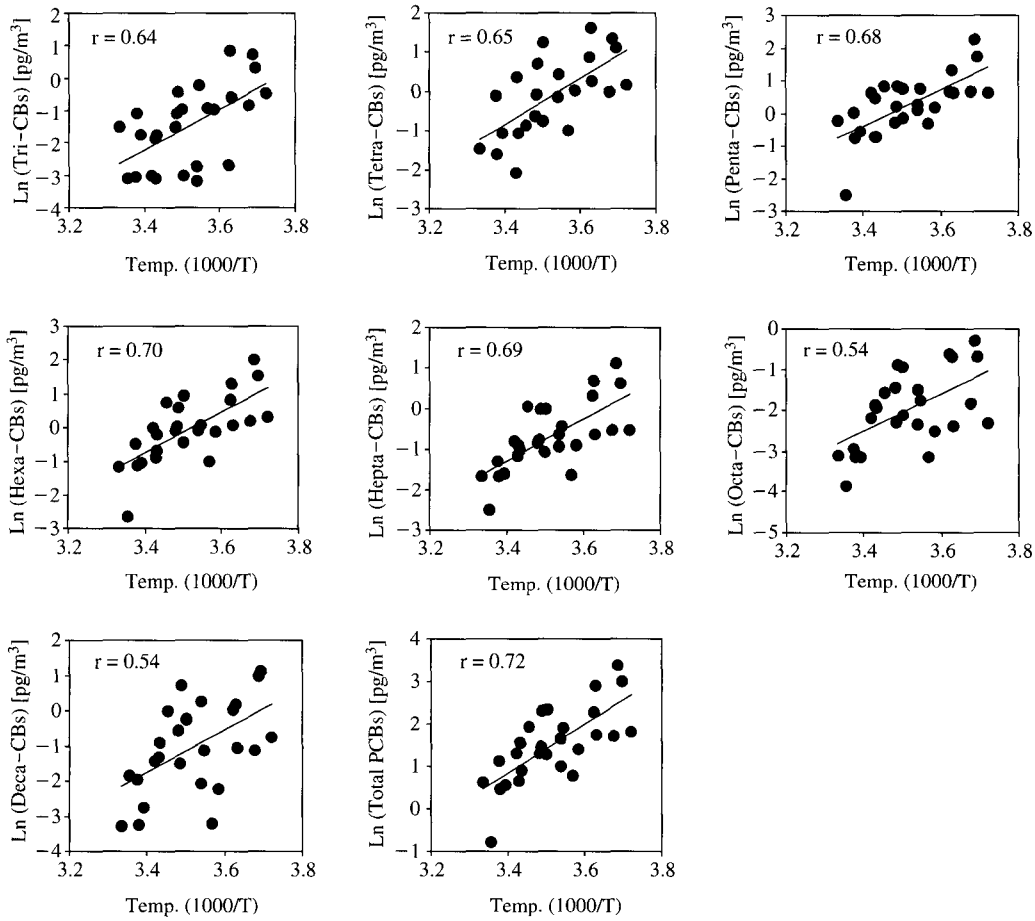

Fig. 4. Correlation coefficients between concentrations of PCB homologs and inverse absolute temperature.

measured previous studies of gas phase PCBs which have a positive correlation with ambient temperature.

4. CONCLUSION

This study was conducted to investigate seasonal variation and temperature factoring of particle PCBs in the atmosphere. Mean concentration of particle PCB homologs showed maximum in penta-CBs and miminum in octa-CBs. Most of PCB homologs showed maximum contribution in winter, minimum in summer. This could be due to greater incorporation of condensed gas phase material at low temperature. Correlation coefficients (r) between PCB concentration in particle phase and inverse temperature showed positive significant correlations ($p < 0.05$) for all PCB homologs which suggested that PCBs concentrations in particle phase increased with decreasing ambient temperature. As a result of this study, PCBs measured in this study were affected by similar sources during the sampling periods.

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