

A Study on the Concentration Characteristics of Atmospheric Polycyclic Aromatic Hydrocarbons (PAHs) in Chongju

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Abstract

Ambient polycyclic aromatic hydrocarbons (PAHs) were measured during the winter and summer of 2002 in Chongju. A filter pack and polyurethane foam (PUF) system was employed to collect simultaneously the particulate and gas phase PAHs. The samples were then analyzed using a gas chromatograph equipped with mass spectrometer detectors (GC/MSD).

A total of 29 samples were collected and 11 PAH species were identified. The lower molecular weight PAH compounds (3~4 rings) dominated the total PAH mass. The higher molecular weight PAH compounds (5~6 rings) were less abundant. The PAHs were showed to exhibit seasonal variations. The concentrations of all compounds were significantly higher in winter than summer. The lower molecular weight PAHs were mostly found in the gas phase whereas the heavier ones were mainly associated with particulate phase. Vehicle emissions are likely to be the primary contributor of PAHs in Chongju. This study also demonstrated that it is necessary to perform simultaneously particulate and gas phase measurements to determine the accurate concentrations of ambient PAHs.

Key words : Particulate and gas phase PAHs, PM_{2.5}, Seasonal variation, Phase distribution, Chongju

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous constituents of urban airborne particulates and are of major health concern mainly due to their well-known carcinogenic and mutagenic properties (Kado *et al.*, 1996; Nielsen *et al.*, 1996; Kao, 1994; Manzie *et al.*, 1994). The PAHs are formed by incomplete com-

bustion or pyrolysis of organic material. Combustion-generated PAHs are initially generated at source in the gaseous state, a proportion of which then adsorb onto existing particles upon cooling of the emissions (Smith and Harrison, 1996). Forest fires and volcanoes contribute to the PAH burden, but by far, anthropogenic sources are responsible for the majority of the PAH input to the atmosphere, which in turn contributes to depositional loadings to aquatic and terrestrial systems. The largest anthropogenic sources of PAHs are vehicular emissions from both gasoline and diesel powered

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vehicles, coal and oil combustion, petroleum refining, natural gas consumption, and municipal and industrial incinerators. Motor vehicles are a significant source of PAHs in the atmosphere, accounting for about one-third of total PAH emissions in the United States (Khalili *et al.*, 1995).

The heavier PAHs (5~7 rings) covering most of the carcinogens are associated with particles in the atmosphere. Part of three- and four-rings PAHs is also bound to particles (Lyll *et al.*, 1988; Cautreels and Van Cawenberghe, 1978). The PAHs are normally associated with small particles which generally have long residence times in the atmosphere and therefore have the potential to be transported to quite long distance. Atmospheric PAH concentrations are strongly dependent upon the size of airborne particulate matter, with the highest concentration being in the respirable size range: about 95% of total PAH is associated with a size class less than 3 μm in diameter (Kaupp and McLachlan, 1999; Venkataraman and Friedlander 1994; Back *et al.*, 1991; Pistikopoulos *et al.*, 1990).

It is a well-known fact that atmospheric PAHs are partitioned between the particulate matter and gas phase. The lighter PAHs (2~3 rings), which are generally not carcinogenic, are mostly found in the gas phase while the heavier ones are mainly associated with airborne particles (Nicolaou *et al.*, 1984; Vaeck and Cawenberghe, 1978). Accurate measurements of gas and particulate phase distributions of semi-volatile organic compounds such as PAHs are needed in order to assess their health effects since lung deposition patterns differ between the gas and particulate phases. Similarly, environmental fates of semi-volatile organic species are phase dependent because atmospheric reactions and transport and deposition processes differ for gaseous and particulate semi-volatile species (Bidleman, 1988).

Despite of the importance in understanding the fate and behavior of PAHs in the atmosphere, there have only been undertaken few field studies to investigate composition characteristics of PAHs in $\text{PM}_{2.5}$ in Korea (Baek and Choi, 1996).

The aim of this study was to simultaneously determine particulate and gas phase concentrations of PAHs for two seasons (winter and summer) using a filter pack and polyurethane foam (PUF) system in Chongju and to investigate the phase distribution of PAHs.

2. METHODS

2.1 PAH sampling

The samples were taken on the roof of a two-story building (6 m above ground level), which is located in a residential area and the middle part of Chongju city. There are no large buildings around the sampling site to disrupt wind flow patterns. Atmospheric samples were taken in the winter (January and February) and summer (July and August) of 2002. The data set was collected on 29 different days with a sampling period of 24 h (from 9:00 a.m. to 9:00 a.m. the next day).

As shown in Fig. 1, ambient samples were collected using the filter pack and polyurethane foam (PUF) (diameter 22 mm, length 3.8 cm) trap (URG) consisting of a teflon-coated aluminum cyclone to remove coarse particles ($> 2.5 \mu\text{m}$ at a flow rate of 10 L/min) and one annular denuder to maintain laminar flow.

The particulate phase PAHs were collected on a 47 mm pre-cleaned quartz fiber filter (URG) that has been thermally treated to remove any organic contaminants. The pre-cleaned quartz filter was sealed and stored at -18°C until sampling. The gas phase PAHs were collected in a trap containing PUF. This trap collects the PAHs in the gas phase as well as the PAHs which evaporate from the filter preceding the PUF trap. The PUF was pre-cleaned with dichloromethane (DCM) for 24 h using a Soxhlet extraction apparatus and dried in a vacuum oven at 70°C and then stored in sealed glass-ware at -18°C until sampling. Pre-fired quartz fiber filter and cleaned PUF were transported to the field in their containers without allowing exposure to ambient air. After sampling, filter and PUF were placed into their containers and stored in a refrigerator at -18°C until they were analyzed.

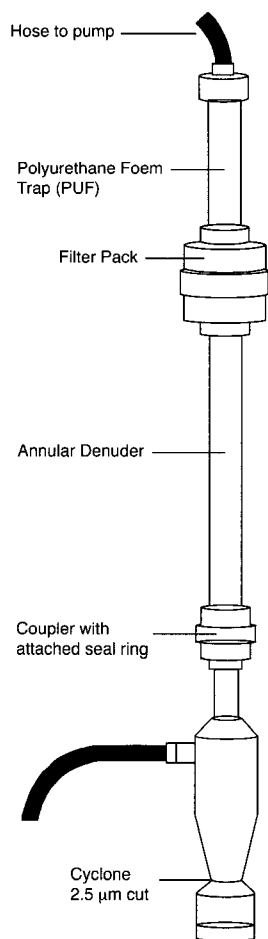


Fig. 1. Schematic view of sampling system.

2. 2 Analysis of PAH samples

The analytical method was used on the basis of the U.S. EPA method TO-13 (US EPA, 1988). The sampled filters were ultrasonically extracted with DCM. The PUF samples were also extracted with DCM for 24 h using a Soxhlet extractor. All extracts were concentrated to approximately 2 mL using 200 mL of DCM with a rotary evaporator (BUCHI, R-144) at 40°C. A silica column packed with 3 g activated silica and sodium sulfate (Na₂SO₄) was used to remove any polar organics that might interfere during analysis. Due to higher boiling point of toluene (111°C) than DCM (b.p. 40°C), the samples could be finally concentrated to 100

Table 1. GC/MSD operating parameters for PAH analysis.

Detector	MSD
Column	HP-5MS capillary column (5% diphenyl & 95% dimethylpolysiloxane) length 30 m × 0.25 mm, film thickness 0.25 μm
Inlet	split
Auxiliary temperature	300°C
Injection volume	1 μl
Temperature program	60°C for 1 min 20°C/min to 130°C 4°C/min to 300°C
Carrier gas	He (99.9999%)
Total run time	62 min

μl using 100 μl of toluene and 100 μl of 4, 4-dibromooctafluorobiphenyl under a gentle stream of purified nitrogen concentrator.

Samples were analyzed using a gas chromatograph (HP 6890) connected to mass spectrometer detectors (HP5973) (GC/MSD). The auto injector was programmed to make 1 μl split injections of all samples. The mass spectrophotometer was programmed in a selective ion monitoring (SIM) mode. The PAHs were identified by comparing retention times and ion ratios to those of a reference standard. The GC/MSD operating parameters are shown in Table 1.

2. 3 Quality control/Quality assurance (QA/QC)

The QA/QC of the data was determined using limit of detection (LOD), recovery, and linearity. The detection limit was obtained by spiking blank sample with the calibration standard. The LOD is defined as three times of the average mass plus 3 standard deviations (3σ). The detected PAHs were Acenaphthylene (ACNPL), Phenanthrene (PHEN), Fluoranthene (FLRTH), Pyrene (PYR), Benz (a)anthracene (BaA), Chrysene (CHRY), Benzo (b) fluoranthene (BbF), Benzo (a)pyrene (BaP), Dibenz(a, h)anthracene (DahA), Benzo (g, h, i) perylene (BghiP), and Indeno (1, 2, 3, cd) pyrene (I123P). The quantification of the PAHs in the samples

Table 2. Limit of detection (LOD) for PAHs.

PAHs	Abbreviation	Number of rings	LOD (ng/m ³) ^a
Acenaphthylene	ACNPL	3	0.0030
Phenanthrene	PHEN	3	0.0060
Fluoranthene	FLRTH	4	0.0062
Pyrene*	PYR	4	0.0070
Benz (a) anthracene	BaA	4	0.0006
Chrysene*	CHRY	4	0.0060
Benzo (b) fluoranthene*	BbF	5	0.0200
Benzo (a) pyrene*	BaP	5	0.0011
Dibenz (a, h) anthracene*	DahA	5	0.0010
Benzo (g, h, i) perylene	BghiP	6	0.0040
Indeno (1, 2, 3, cd) pyrene*	I123P	6	0.0040

^aValue calculated by sampling volume of 14.4 m³

*Suspected carcinogen

was achieved by comparing the measured peak heights with those of the standards of known concentration. All samples were spiked with known amounts of PAH standards prior to extraction in order to determine analytical recovery efficiencies. The recovery efficiencies were relatively reasonable, ranged from 68 to 95%. The linearities of calibration standards were calculated by regression analysis and the values were 0.99~1.00 (R²) for all PAHs. Table 2 shows the LOD for the 11 PAH compounds. For the low concentrations below the detection limit, they were excluded in final data. However, it was not nearly found for the entire sampling period except for DahA.

3. RESULTS AND DISCUSSION

The data set was collected on 15 days in winter (9 January to 17 February 2002) and on 14 days in summer (31 July to 25 August 2002). Distributions of PAH concentrations between the particulate and gas phases are summarized in Table 3. They include a group of suspected carcinogens such as PYR, CHRY, BbF, BaP, DahA, and I123P. The mean concentrations were ranged from 0.056 ng/m³ for DahA to 3.666 ng/m³ for ACNPL in winter and from 0.010 ng/m³ for DahA to 1.999 ng/m³ for PHEN in summer. The PAH concentrations were higher during the winter months and

lower during the summer months. Average temperature and relative humidity measured from the National Observatory of Chongju were also included in Table 3.

3.1 Ambient PAH Concentrations

The PAH levels in Chongju were much lower than other urban sites (Liu *et al.*, 2001; Simcik *et al.*, 1999; Odabasi *et al.*, 1999; Coleman *et al.*, 1997; Harrison *et al.*, 1996; Broman *et al.*, 1991). Based on annual mean concentration (sum of winter and summer data), PHEN (2.667 ng/m³) and ACNPL (1.854 ng/m³) were much higher than those of other PAHs (Table 3). The main source of these compounds is known from mobiles. From the previous studies (Kulkarni and Venkataraman, 2000; Harrison *et al.*, 1996; Khalili *et al.*, 1995), the following PAHs have been identified as markers for various sources in urban atmosphere: ACNPL and PYR from gasoline powered vehicles; ACNPL and PHEN from diesel powered vehicles; and FLRTH, PYR, and CHRY from industrial-oil burning. The annual mean concentrations of FLRTH (1.068 ng/m³) and PYR (0.955 ng/m³) were relatively high. It indicates that vehicular emissions are likely to be the primary contributor to PAH concentrations in Chongju with an additional contributor such as industrial-oil burning. This is consistent with that the sampling site is located in a residential area near a major roadway and a industrial complex (approximately 1.5 km from the sampling site in northwestern direction).

The ratios of specific particulate phase PAHs are also characteristics of different sources (Simcik *et al.*, 1999). The common ratios used include benzo (a) pyrene to benzo (ghi) perylene (BaP/BghiP) and benz (a)anthracene to chrysene (BaA/CHRY). The mean values of these ratios in Chongju are presented along with the values for several other sources in Table 4. The BaP/BghiP ratio from Chongju lies in the range of the values for the vehicles and diesel. The BaA/CHRY ratio is similar to the vehicles and gasoline. Based on these ratios, the major source of PAHs in Chongju is also consistent with the signals provided by vehicle emissions.

Table 3. Summary of mean PAH concentrations measured in Chongju (ng/m³).

Compound		Winter (n = 15)			Summer (n = 14)			Average ^a
		Particle	Gas	Total ^b	Particle	Gas	Total	Total
Acenaphthylene (ACNPL)	avg	0.017	3.649	3.666	ND ^c	0.174	0.174	1.854
	range	0.004~0.046	0.566~11.397			0.0059~0.352		0.006~11.406
	SD ^d	0.012	3.330			0.090		2.886
Phenanthrene (PHEN)	avg	0.477	3.016	3.493	0.022	1.977	1.999	2.667
	range	0.113~1.078	1.011~8.557		0.008~1.078	1.093~3.252		0.312~8.933
	SD	0.247	2.167		0.290	0.633		1.817
Fluoranthene (FLRTH)	avg	1.345	0.397	1.742	0.019	0.054	0.073	1.068
	range	0.367~3.008	0.072~1.476		0.008~0.055	0.146~0.937		0.155~4.220
	SD	0.785	0.372		0.015	0.209		1.015
Pyrene (PYR)	avg	1.272	0.245	1.517	0.023	0.347	0.370	0.955
	range	0.111~2.927	0.041~0.940		0.011~0.068	0.118~0.813		0.136~3.746
	SD	0.895	0.249		0.016	0.197		0.946
Benz(a)anthracene (BaA)	avg	0.632	ND	0.632	0.025	ND	0.025	0.350
	range	0.110~1.552			0.005~0.071			0.005~1.552
	SD	0.490			0.021			0.469
Chrysene (CHRY)	avg	0.980	ND	0.980	0.051	ND	0.051	0.531
	range	0.166~2.448			0.011~0.148			0.011~2.448
	SD	0.713			0.042			0.691
Benzo(b)fluoranthene (BbF)	avg	0.987	ND	0.987	0.063	ND	0.063	0.541
	range	0.258~2.158			0.031~0.184			0.031~2.158
	SD	0.620			0.040			0.643
Benzo(a)pyrene (BaP)	avg	0.565	ND	0.565	0.022	ND	0.022	0.313
	range	0.104~1.451			0.001~0.075			0.001~1.451
	SD	0.481			0.023			0.443
Dibenz(ah)anthracene (DahA)	avg	0.056	ND	0.056	0.010	ND	0.010	0.049
	range	0.014~0.155			0.003~0.023			0.003~0.155
	SD	0.043			0.012			0.043
Benzo(ghi)perylene (BghiP)	avg	0.995	ND	0.995	0.037	ND	0.037	0.636
	range	0.178~3.058			0.007~0.100			0.007~3.058
	SD	0.850			0.030			0.815
Indeno(1, 2, 3, cd)pyrene (I123P)	avg	0.326	ND	0.326	0.013	ND	0.013	0.175
	range	0.072~0.912			0.006~0.044			0.006~0.912
	SD	0.255			0.010			0.240
Temperature (°C)		0.9			25.1			
Relative humidity (%)		62.9			75.6			

^aSum of winter and summer data

^bSum of particle and gas

^cNot detected

^dStandard deviation

Table 4. Diagnostic PAH ratios in atmospheric particles (Simcik *et al.*, 1999).

Sources	BaP/BghiP	BaA/CHRY
Vehicles	0.3~0.78	0.63
Gasoline	0.3~0.4	0.28~1.2
Diesel	0.46~0.81	0.17~0.36
Coal	0.9~6.6	1.0~1.2
This study	0.56	0.68

The higher molecular weight PAH compounds, such as BaA, CHRY, BbF, BaP, DahA, BghiP, and I123P, were less abundant in Chongju (Table 3). These results are similar to those measured in urban sites (Lee *et al.*, 2001; Smith and Harrison, 1996). The BaP has been the most extensively measured PAH in urban areas around the world due to its high carcinogenic property

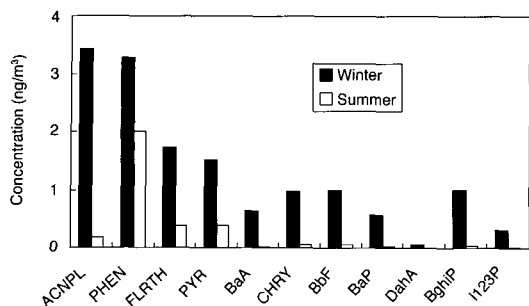


Fig. 2. Seasonal distribution patterns of the 11 PAHs in Chongju.

and was used as an indicator for the total PAH concentrations (Lee *et al.*, 2001). The total mean concentrations of BaP were 0.565 and 0.022 ng/m³ in winter and summer, respectively. The BaP levels in Chongju were much less than in Taegu commercial area (1.2 and 0.4 ng/m³ in the winter and summer, respectively) (Baek and Choi, 1996). Very high BaP levels were observed in Jakarta (4.37 ng/m³), Seoul (1.17 ng/m³), and Bangkok (0.98 ng/m³) (Lee *et al.*, 2001).

3.2 Seasonal comparison of PAH concentration

From this study, it was demonstrated that the PAH concentrations in summer were significantly lower than those found in winter (Table 3 and Fig. 2). The relatively high PAH concentrations in winter may be due to greater PAH emissions related to the use of fossil fuels during the cold season and meteorological conditions which are less favourable for dispersion in winter. It can be also explained by the heavy and frequent precipitation in summer. The previous work has shown that the particulate PAH concentrations can be diminished by 60% due to precipitation (Kaupp and McLachlan, 1999; Masclet *et al.*, 1988).

The mean ratio of winter to summer for the total PAHs was 5.3. This is in accordance with the results reported in other studies: the winter levels, as stated in several investigations carried out in Europe and USA, were generally higher by a factor of 1.5~5.5 than in

summer (Gigliotti *et al.*, 2000; Caricchia *et al.*, 1999; Harrison *et al.*, 1996).

For particulate phase in winter, FLRTH (1.345 ng/m³) and PYR (1.272 ng/m³) have shown higher concentration than other species, and in summer, the concentrations of BbF (0.063 ng/m³) and CHRHY (0.051 ng/m³) were relatively higher. For gas phase in winter, ACNPL (3.649 ng/m³) and PHEN (3.016 ng/m³) have shown higher concentration than other compounds, and in summer, the concentrations of PHEN (1.977 ng/m³) and PYR (0.347 ng/m³) were higher.

For the PAHs with 4~6 rings which are primarily bound to particles throughout the whole year (winter and summer), the concentrations in the winter samples were about 21 times higher than in summer. The carcinogen BaP shows marked seasonal fluctuation, with the winter concentration 25-fold higher than that of summer. The BaP level (0.565 ng/m³) in winter exceeded the U.K. air quality standard value (0.25 ng/m³) (Dimashki *et al.*, 2001).

3.3 Phase distribution

An accurate phase distribution of PAHs must be known in order to assess their health effects since the lung deposition patterns depend on the distribution of PAH between the gas and particulate phases. In the atmosphere, the PAHs depend on the physical characteristics of the compounds themselves and the environmental situation such as temperature and humidity. The lower molecular weight PAH species are present in air mainly as vapors, on the other hand higher molecular weight species being physically adsorbed on particulate surfaces (Pupp *et al.*, 1976).

The particulate/gas phase distribution varied widely as shown in Fig. 3. The gas phase percentage generally decreased with increasing molecular weight. The gas phase percentage of ACNPL and PHEN were 99.5 and 86.3% in winter and 100 and 98.9% in summer, respectively. FLRTH and PYR were mainly found in the gas phase (74 and 93.8%, respectively) in summer but in the particulate phase (77.2 and 83.8%, respectively) in winter as shown in Table 3 and Fig. 3. Mean tempera-

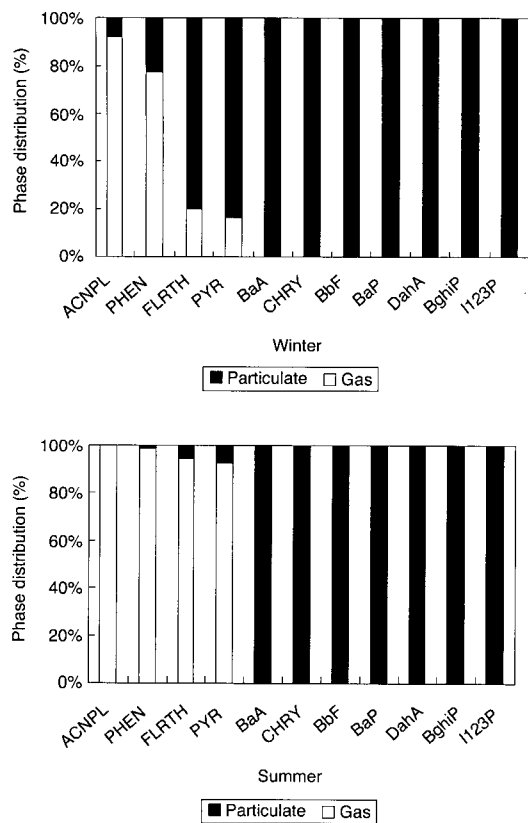


Fig. 3. The phase distribution of PAHs in winter and summer.

tures in winter and summer were 0.9 and 25.1°C, respectively. It was found that temperature was a factor altering the gas/particle distribution of lower molecular weight species. At higher temperature, during the summer months, The lower molecular weight PAHs are associated with the gaseous phase due to the increased volatilization (Panther *et al.*, 1999). The loss of more volatile compounds in summer, such as ACNPL and PHEN, can cause an underestimation of particle concentration.

The heavier PAHs, such as BaA, CHRY, BbF, BaP, DahA, BghiP, and I123P, were mostly found in the particulate phase during the winter and summer sampling studies (Table 3 and Fig. 3). These results were generally consistent with the particle/gas phase distrib-

utions reported in the previous studies (Liu *et al.*, 2001; Odabasi *et al.*, 1999; Baek and Choi, 1998; Harrison *et al.*, 1996; Smith and Harrison, 1996; Broman *et al.*, 1991).

4. SUMMARY

To evaluate the concentration characteristics and phase distribution of PAHs in Chongju, a study of PAH concentrations was conducted during 2002. The PAH levels in Chongju were much lower than other urban sites. The PHEN and ACNPL concentrations were much higher than those of other PAHs. The lower molecular weight PAH compounds, such as PHEN, ACNPL, FLRTH, and PYR, dominate the total PAH mass. The heavier PAHs, such as BaA, CHRY, BbF, BaP, DahA, BghiP, and I123P, were less abundant in Chongju. The BaP (known as one of the most carcinogen of the PAHs) levels in Chongju was less than those in Taegu commercial area. Vehicular emissions may be the primary contributor to PAH concentrations in Chongju with an additional contributor such as industrial-oil burning. All PAHs have higher concentrations in winter than summer. It may be due to greater PAH emissions from anthropogenic combustion sources and less dispersion in winter. There were the marked variations of gas-particulate distributions for each PAH, reflecting the effect of ambient temperature. ACNPL and PHEN exist exclusively in the gas phase regardless of season. FLRTH and PYR were mainly found in the gas phase in summer but in the particulate phase in winter. The heavier molecular weight PAHs, such as BaA, CHRY, BbF, BaP, DahA, BghiP, and I123P, were absolutely found in the particulate phase during the whole sampling period.

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