

Effects of Particle Size and Characteristics on the Gas-particle Partitioning of PAHs in the Air

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대기중 입자의 크기와 특성이 다환방향족탄화수소류 화합물의 증기상-입자상간의 분배에 미치는 영향

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요 약

다환방향족탄화수소류 화합물의 증기상-입자상간의 분배평형의 설명을 위해 흔히 입자상의 흡착지점이 균질하고 총흡착면적은 TSP에 비례한다는 가정을 사용하는데 본 연구의 목적은 이러한 가정의 타당성을 평가하는 것이다.

본 연구를 위해 도심에서 6단의 다단계 대기중입자채집기를 사용하여 대기 중의 입자를 포집하였으며 이들 입자에 흡착된 phenanthrene, anthracene, fluoranthene, pyrene을 분석하여 입경별 분포를 측정하였다. 특히 연구기간 중에 황사현상이 일어나 입경분포나 입자의 기원이 매우 다른 경우에 대한 연구가 가능하였다.

주요연구결과로서 우선 야마사키가 제안한 분배평형의 온도의존식은 제한된 범위에서 사용되어야 한다는 것이 관측되었다. 즉, 황사현상이 일어나는 경우와 같이 입자의 흡착특성과 입경분포가 보통때와 다른 경우에는 $\log K_p$ 와 $1/T$ 의 관계에서 선형성이 상당히 저하되었다. 또한 특히 낮은 온도에서는 입자의 입경분포가 달라지면 전체적인 분배평형이 달라지게 되는 것으로 평가되었으며 입자의 흡착특성도 분배평형의 온도의존성에 결정적인 영향을 줄 수 있는 것으로 나타났다. 따라서 입자의 기원이 다양하거나 입경분포가 달라지면 흡착평형이 바뀌기 때문에 흡착특성의 균질성과 단순한 TSP를 전제로 하는 분배평형의 평가나 예측은 실제 대기조건에서는 정확하지 않을 수가 있으며 제한적인 조건에서 사용되어야 할 것이다.

Key words : Gas-particle partitioning, Particle size distribution, PAHs

INTRODUCTION

Partitioning of PAHs between gas and particle pha-

ses in air has been the subject of numerous studies (Finizio *et al.*, 1997; Gustafson and Dickhut, 1997; Baek and Choi, 1998; Harner and Bidleman, 1998). Yamasaki *et al.* (1982) first suggested a temperature dependence of the partition equilibrium coefficient ($\log K_p = \log (C_g/(C_p/TSP)) = m/T + b$, K_p : gas-par-

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ticle partition equilibrium coefficient, C_g : gas phase concentration, C_p : particle phase concentration, TSP: total suspended particulates, m and b : slope and intercept). Two assumptions were critical to the derivation of the temperature dependence: 1) the adsorption sites are homogeneous regardless of the particle size and origin and 2) the total sorption area available is proportional to TSP. Extensive review and theoretical attachment have followed (Dickhut, 1997; Baek and Choi, 1998; Gustafson and Harner and Bidleman, 1998; Simcik *et al.*, 1998) for the suggested temperature dependence. Little attention, however, has been paid to the potential limitation that the two assumptions might have for adequate description of the field observation of the temperature dependence. It has been reported that the sorption characteristics of PAHs vary with particle size and characteristics because of their different physicochemical properties, thereby changing the partitioning of PAHs between gas and particle (Allen *et al.*, 1996; Mader *et al.*, 1997) Particularly, concentrations of PAHs in the particulate phase varied with the particle size (Schnell *et al.*, 1995; Allen *et al.*, 1996; Baek and Perry, 1996; Kaupp and McLachlan, 1999). However, quantitative analysis of the particle size effects has remained limited.

The principal objective of the present study was therefore to evaluate the two assumptions by taking advantage of the YS phenomenon. YS refers to the occasions where TSP drastically increases typically in April and May by the particles long-range transported from China. The phenomenon can also significantly alter the size distribution of particulates in air for a few days (Chung and Park, 1998), thereby, providing a unique opportunity to investigate the effects of significant variation in both the size distribution and sorption characteristics of the particulates on the partitioning equilibrium in the field conditions

EXPERIMENTAL METHODS

Sample collection

All the samples were collected on the roof of the

engineering building at Ewha Women's University, Seoul, Korea, in the year 2000. The sampling site represents a typical metropolitan area with numerous anthropogenic PAH sources such as automobiles and commercial and residential buildings. Sampling was conducted by using a high volume sampler equipped with a six-stage cascade impactor (SA236, Andersen, USA, cut-off diameters of 0.41 μm , 0.73 μm , 1.4 μm , 2.1 μm , 4.2 μm , and 10.2 μm) and a cartridge serially loaded with two pieces of polyurethane foam (PUF) (9 cm \times 5 cm, Kimoto, Japan). The air was taken at a constant rate of 560 L/min during the sampling. To minimize the temperature variation, the sampling was maintained during either daytime or night. The temperature and relative humidity were measured every three minutes and logged into a data logger (HOBO-RH, StowAway XTI, USA).

Materials and analysis

The solvents used were pesticide grade dichloro-

Table 1. Target and qualifier ions in the GC/MSD analysis

Abbreviation	PAHs	Target ion	Qualifier ion
	Naphthalene-d8	136.0	
NAP	Naphthalene	128.1	127.2
ACE	Acenaphthylene	152.2	151.1
	Acenaphthene-d10	164.2	
ACT	Acenaphthene	153.2	154.2
FLN	Fluorene	166.2	165.2
	Phenanthrene-d10	188.2	
PHE	Phenanthrene	178.2	176.1
ANT	Anthracene	178.2	176.1
FL	Fluoranthene	202.2	200.2
	Pyrene-d10 (GC-Internal)	212.2	
PY	Pyrene	202.2	200.2
BaA	Benzo (a)anthracene	228.2	226.2
	Chrysene-d12	240.3	
CHR	Chrysene	228.2	226.2
BbF	Benzo (b)fluoranthene	252.2	253.2
BkF	Benzo (k)fluoranthene	252.2	253.2
BaP	Benzo (a)pyrene	252.2	253.2
	Perylene-d12	264.3	
IcdP	Indeno (1, 2, 3-cd)pyrene	276.2	138.2
DahA	Dibenzo (a, h)anthracene	278.3	279.3
BghiP	Benzo (g, h, i)perylene	276.2	127.2

methane (DCM) (Caledon, Canada) and pentane of 99.99% purity (Caledon, Canada). All glass wares were cleaned with detergent, sonicated, and rinsed three times with deionized water, and finally heated at 450°C over 4 hours.

The samples were preserved at -25°C till the analysis. The National Oceanic and Atmospheric Administration (NOAA) method (NOAA, 1993) was used for the PAH analysis. Briefly, each sample was Soxhlet-extracted with DCM (200 mL for particle samples and 600 mL for each PUF) for 24 hours. The extract was concentrated to 1 mL and cleaned up with a silica-alumina column using a DCM and pentane mixture (1 : 1 v/v). The extract after clean-up was concentrated to 0.5 mL for the instrumental analysis with gas chromatograph/mass selective detector (GC/MSD) (Hewlette Packard 6890/5973, USA). In Table 1 listed the 16 PAHs and deuterated surrogate and internal standards. Recoveries of all the samples analyzed were in the ranges of 40% to 120%, which met the quality control (QC) limits.

RESULTS AND DISCUSSION

The weather conditions and the particle size distribution are summarized in Table 2. The cutoff diame-

ter is defined as the value at 50% collection efficiency for spherical particles with unity mass density at 25°C and 760 mmHg at an air flow rate of 560 L/min.

The particle size distribution was typical of bimodal distribution for both daytime and night as shown in Fig. 1. At night, the relative humidity was higher but temperature was lower than at daytime. Partitioning to particles of hydrophobic substances decreases with relative humidity and temperature (Pankow *et al.*, 1993; Sanusi *et al.*, 1999). The conditions at night of higher relative humidity and lower temperature could act in opposite directions for the partition equilibrium. In the present study, however, the relative hum-

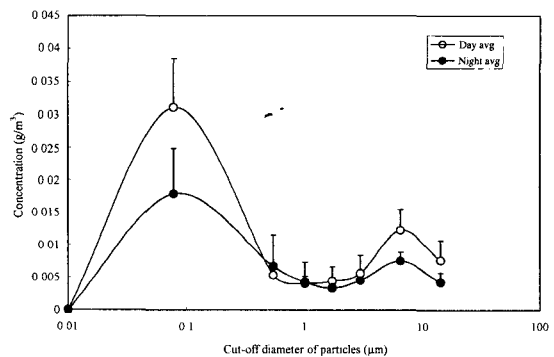


Fig. 1. Particle size distribution (Error bar denotes a standard deviation).

Table 2. Weather conditions and particle mass concentrations as captured by impactor stages (unit: $\mu\text{g}/\text{m}^3$ for TSP and St_i)

Sample ID ^a	Temp (°C)	Relative humidity (%)	TSP	St7 ^b	St6	St5	St4	St3	St2	St1
0406D ^c	11.1	28.0	278	58.9	41.1	24.2	33.9	36.1	45.0	39.2
0406N ^c	6.3	39.2	30	45.0	41.0	41.3	45.0	39.8	48.9	43.2
0423D	13.5	36.2	20	100.7	10.0	6.5	13.8	21.5	40.5	15.9
0423N	9.1	64.2	99	39.9	4.4	1.3	7.7	12.4	24.3	9.3
0508D	20.8	35.2	195	81.5	15.4	12.7	16.2	17.4	21.8	30.2
0508N	17.1	60.2	94	23.7	12.5	9.0	8.6	10.9	19.2	10.2
0601D	23.8	53.8	136	60.0	14.1	11.4	3.9	6.8	27.7	12.1
0601N	20.4	65.7	163	68.0	20.9	15.2	9.0	13.2	18.9	17.9
0621D	29.7	48.6	159	66.9	13.7	10.2	10.2	9.9	31.0	17.2
0621N	25.8	52.7	170	62.7	35.7	21.3	11.4	12.2	18.2	8.5

^a D and N denote daytime and night, respectively.

^b St_i denotes the mass concentration of the particles collected in the cascade impactor stage *i* with the following cut-off diameter:

St1: $D_p > 10.2 \mu\text{m}$, St2: $4.2 \mu\text{m} < D_p < 10.2 \mu\text{m}$, St3: $2.1 \mu\text{m} < D_p < 4.2 \mu\text{m}$, St4: $1.4 \mu\text{m} < D_p < 2.1 \mu\text{m}$, St5: $0.73 \mu\text{m} < D_p < 1.4 \mu\text{m}$, St6: $0.41 \mu\text{m} < D_p < 0.73 \mu\text{m}$, St7: $D_p < 0.41 \mu\text{m}$

^c Samples taken when "Yellow Sand" occurred.

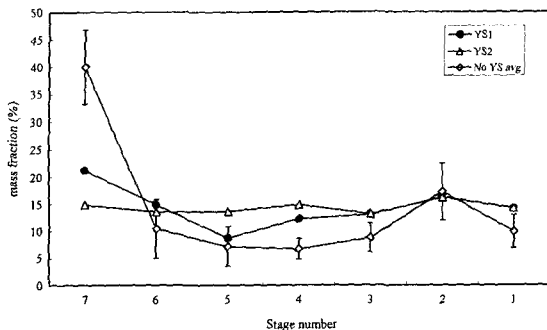


Fig. 2. Mass fraction of particulates in each stage (YSi: yellow sand, No YS avg: average of observations with no yellow sand, error bars denote one standard deviation).

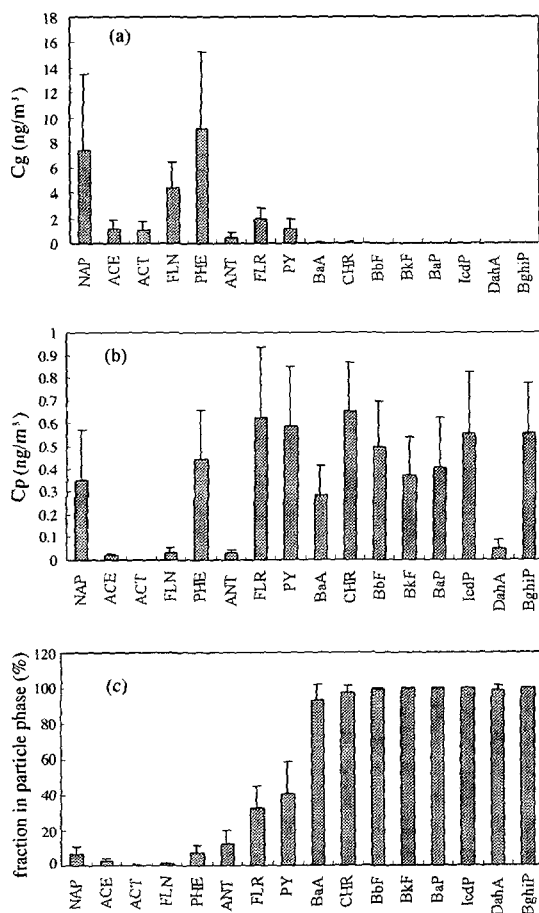


Fig. 3. The PAH levels in (a) gas phase, (b) particulate phase, and (c) their fractions in the particulate phase in air.

idity (28~66%) was in such a range that might not significantly influence the partition equilibrium (Mader and Pankow, 2000). Therefore, the temperature effect is deemed more pronounced at night.

TSP was significantly higher ($278 \mu\text{g}/\text{m}^3$ and $304 \mu\text{g}/\text{m}^3$) during the YS period as compared to an average of $153 \mu\text{g}/\text{m}^3$ for the YS absent periods. As shown in Fig. 2, the fractions of mass concentration of each stage significantly varied when YS occurred. The fraction of the finest particulates, St7, was notably lower while those of St3, St4, and St5 appeared higher.

The PAH levels in the gas and the particle phases are shown in Fig. 3a and 3b, respectively. The levels were typical of urban areas (Cotham and Bidleman, 1995; Smith and Harrison, 1996; Colman *et al.*, 1997; Caricchia *et al.*, 1999). As shown in Fig. 3c, only a few PAHs partitioned between the two phases to the extents suitable for the partitioning study. Furthermore, the PAHs lighter than phenanthrene were excluded in the present study as the analysis of the back-up PUF indicated occurrences of break-through. Consequently, only four PAHs including phenanthrene, anthracene, fluoranthene, and pyrene were selected for the equilibrium study.

In Fig. 4, the impact of YS is shown on the relationship between the log K_p and $1/T$. Linear relationships appeared to hold for all the four PAHs only if the YS cases are excluded. The YS data significantly deviate from the linear fits, indicating that the partitioning to the particulates did not vary in proportion to the variations in TSP ($\mu\text{g}/\text{m}^3$) introduced by YS. Obviously, such observations strongly suggest that the Yamasaki approach simply on the basis of TSP has limited applicability. The values of K_p would be greater at the same temperature for YS than those estimated in the absence of YS. The greater K_p values are a result of less sorption capacity of the particulates and are consistent with the fact that particulates of YS is of primarily natural origin. Two questions were raised from the observation. First, the sorption characteristics of the active sites might vary significantly depending upon the particle size and/or origin. Seco-

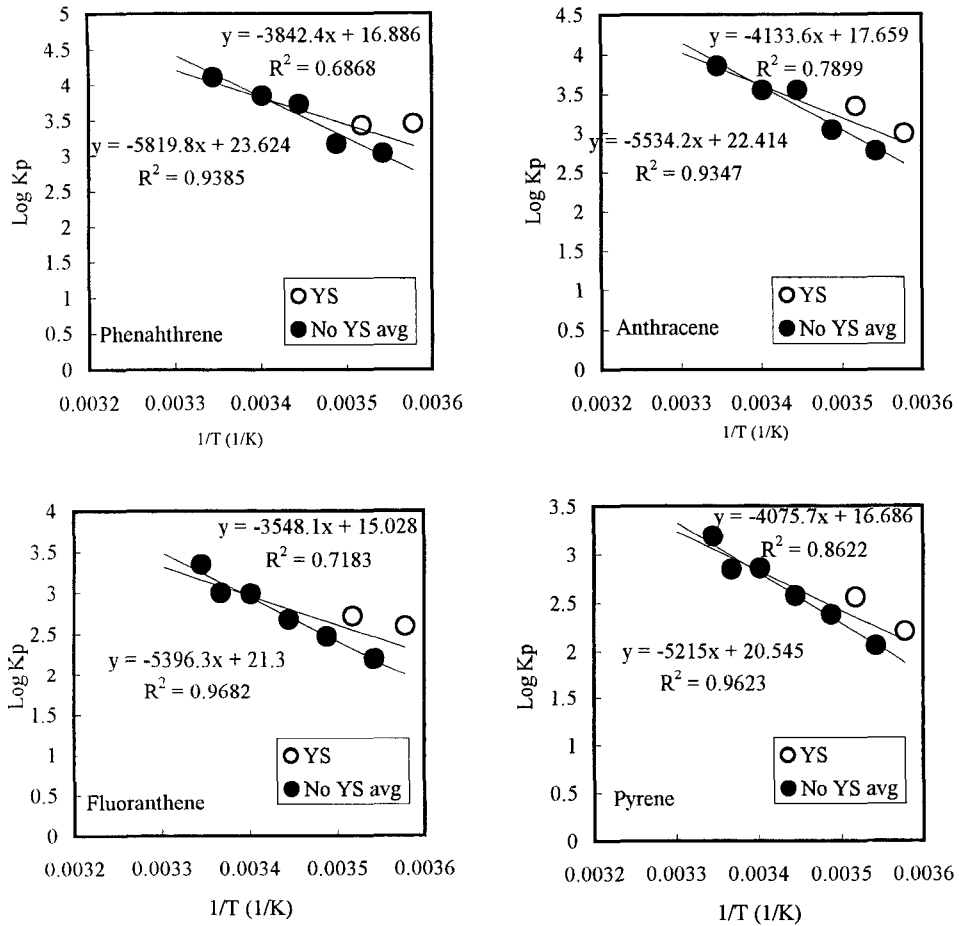


Fig. 4. Comparison of the PAH partition equilibria for the cases in the absence of (No YS avg).

Table 3. Linear regression parameters for the temperature dependence of PAHs

	Stage	7	6	5	4	3	2	1
PHE	slope	4226	9887	10852	6660	6682	341	4400
	y-intercept	18.2	37.1	40.2	26.2	26.6	15.6	19.1
	r ²	0.91	0.95	0.999	0.95	0.96	0.9	0.98
ANT	slope	4007	9857	9625	7100	5234	*	*
	y-intercept	17.1	36.8	36	27.5	21.3	*	*
	r ²	0.9	0.97	0.917	0.92	1 ^a	*	*
FLR ^a	slope	4825	9443	10441	4033	2843	3131	3875
	y-intercept	19.3	34.7	38	16.4	12.7	14	16.6
	r ²	0.97	0.95	0.981	0.6	0.55	0.92	0.57
PY	slope	4795	9040	10109	3849	2686	3224	4273
	y-intercept	19.1	33.2	36.8	15.6	12.1	14.2	17.5
	r ²	0.98	0.95	0.975	0.57	0.51	0.76	1 ^a

The numbers of data used for the regression for PHE, ANT, FLR, and PY were 4, 4, 5, and 5, respectively.

^aOnly two data points were used.

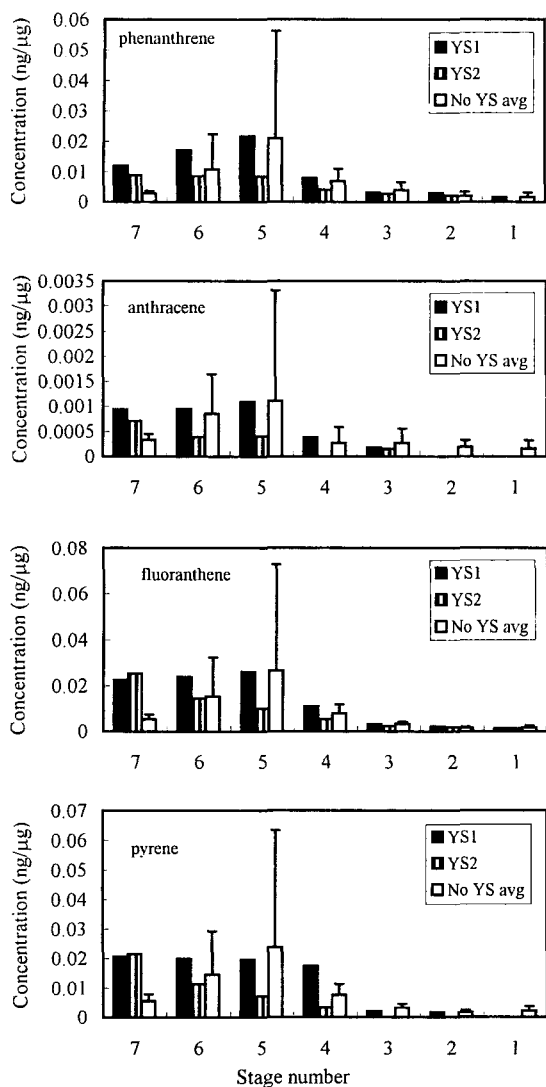


Fig. 5. PAH concentrations in the particulate phase (YS1: yellow sand, No YS avg: average of observations with no YS).

nd, even under the assumption that the sorption characteristics are uniform in all active sites, the value of K_p could still depend on the particle size distribution because the number of active sites per unit mass of the particles would be a function of the particle size. Therefore, the K_p derived from the basis of TSP values might lead to oversimplification.

As shown in Fig. 5, the average PAH mass concen-

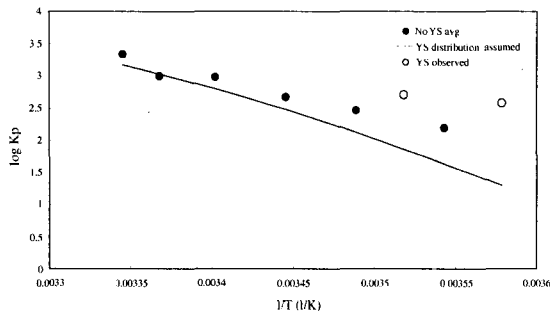
tration per unit mass of the particulate, i.e., sorbing capability, significantly varied with the particle size even in the absence of YS. If the sorption characteristics were homogeneous at all sizes, the higher sorbing capability would be associated with the finer particulates. The sorbing capabilities, however, were not a monotonous function of the particle size as their magnitudes were consistently in the order of $St_5 > St_6 > St_4 > St_7 > St_3 > St_2 \sim St_1$ for the four PAHs. Furthermore, the St_7 particulates were consistently higher for YS than for the no YS cases in their sorbing capability for all the four PAHs. Although greater concentrations were generally observed in the finer particulates, no consistent pattern across the PAHs was observed with the particle size of YS. Therefore, it was likely that not only the size but the sorbing natures of the particulates affect the partitioning. The non-uniform partition equilibrium was further shown by its temperature dependence that varied with the particle size. As summarized in Table 3, the slope and the y-intercept of the plot of $\log K_p$ vs. $1/T$ changed with the particle size. Therefore, it is clear that the altered partition equilibrium for YS resulted from the combined effects of particulates with different sorbing natures and the particle size distribution.

The significance of the particle size distribution could be examined by introducing hypothetical variations in the size distribution pattern and the resulting change in the slope of the plot of $\log K_p$ with $1/T$. To estimate the change in the K_p value, following relationships were used.

- (1) $\sum CP_i = C_p$ (ng/m^3) where C_{p_i} is the concentration of PAH in the particle phase of stage i .
- (2) $\sum TSP_i = TSP$ ($\mu g/m^3$) where TSP_i is the concentration of suspended particles of stage i .
- (3) $C_g + C_p = C_T$ where C_g is the gas-phase PAH concentration and C_T is the total PAH concentration in the air.
- (4) $K_{p_i} = C_g / (C_{p_i} / St_i)$ where K_{p_i} and St_i are the equilibrium partition coefficient and mass concentration of the particles, respectively, of stage i .
- (5) $\log K_{p_i} = -m_i / T + b_i$ where m_i is the slope and b_i is y intercept of stage i , respectively, and T is

Table 4. Change in overall Kp value at different temperatures with a change of \pm one standard deviation in particulate mass at each stage (unit: %)

	St7	St6	St5	St4	St3	St2	St1
282.1 K	± 2.2	± 16.0	± 22.5	± 0.9	± 2.2	± 5.2	± 3.0
286.5 K	± 1.4	± 13.0	± 15.7	± 0.3	± 1.8	± 4.8	± 2.8
290.1 K	± 0.7	± 9.5	± 10.4	± 0.3	± 1.4	± 4.4	± 2.6
293.8 K	± 0.2	± 5.7	± 5.8	± 0.9	± 0.8	± 3.8	± 2.4
296.8 K	± 0.2	± 3.0	± 2.9	± 1.4	± 0.3	± 3.3	± 2.2
298.8 K	± 0.3	± 1.4	± 1.4	± 1.7	± 0.0	± 3.0	± 2.1

**Fig. 6.** Effects of particle size distribution and sorbing nature (No YS avg: average of the observations with no yellow sand; YS: yellow sand cases).

temperature in K.

From (4) and (5),

$$(6) C_{p_i} = 10^{(m_i/T - b_i)} \times St_i \times C_g = A_i \times C_g \text{ where } A_i \text{ denotes } 10^{(m_i/T - b_i)} \times St_i.$$

Plugging (6) into (1),

$$(7) \sum A_i \times C_g = C_T$$

Plugging (7) into (3) gives,

$$(8) (\sum A_i + 1) \times C_g = C_T$$

A_i and C_T are determined from field measurements, all other parameter values can be estimated.

Fluoranthene was chosen for the analysis as its Kp showed the most consistent temperature dependence and existing data were available to be compared with Baek and Choi (1998), Yamasaki *et al.* (1982), and Keller and Bidleman (1984). The value of A_i was calculated by using the fluoranthene concentration in each stage and temperature data. C_T was assumed to be 3 ng/m^3 , which approximated the average of the observed ones.

Under the assumption that the partition characteristics of the particles remain unchanged, the sensitivity of Kp to the change in the particle size distribution was estimated by varying the mass fraction of particles of each stage by \pm one standard deviation from its average value. As shown in Table 4, the change in overall Kp value was more sensitive to the change in the mass fraction of stages 5 and 6 at the lower temperatures. This is anticipated because the sorbing capacity was greater with the particulates of stages 5 and 6 as previously shown in Fig. 5. As temperature rises, the temperature effect becomes less sensitive probably because adsorption decreases with temperature. During the YS period, variations greater than one standard deviation were observed at stages 3, 4, 5 and 7 (Fig. 5) and the ambient temperature ($5 \sim 15^\circ\text{C}$) was in the range that can influence the Kp value. Therefore, the changes in the particle size distribution of YS would affect the temperature dependence of Kp for YS. The line in Fig. 6 illustrates a hypothetical impact to the temperature dependence by the observed variation in particle size distribution of YS if YS is assumed to have the same sorbing nature as that of no YS cases. As the line indicates, the Kp value should have decreased by more than a factor of few as compared to that of no YS cases if only the particle size distribution had impacted. This indicates that the particle size distribution could have significant influence on the partition equilibrium. Contrarily, however, the observed Kp increased for YS, suggesting that the size distribution alone could not account for the observed temperature dependence and influence of different sorbing nature should also exist for

YS. In the present study, the impact of sorbing nature exceeded the effect of the particle size distribution to result in significant increases in K_p values.

CONCLUSION

It was observed in the field conditions that the Yamasaki-type temperature dependence of partition equilibrium was of limited applicability. In the presence of the YS particulates of different sorbing nature and of different size distribution, linear relationships between $\log K_p$ and $1/T$, which have frequently reported in other studies, do not hold for all the four PAHs investigated in this study. Given certain partition characteristics of particulates, it was shown that a change in the particle size distribution could have a significant effect on the overall partition equilibrium particularly at low temperature. Also, the sorbing characteristics of the particulates appear to impact significantly the temperature dependence of the partition equilibrium. Therefore, the two assumptions, 1) adsorption sites are homogeneous regardless of the particle size and origin and 2) the total available sorbent area is proportional to TSP, now widely accepted in describing the temperature dependence of K_p , could be inadequate for predicting the partitioning in widely varying environmental conditions of particle size and characteristics such as the YS cases and should carefully be applied within limited field conditions.

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