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Seasonal Change in the Atmospheric Concentration of Particulate Polycyclic Aromatic Hydrocarbons in Ho Chi Minh City, Vietnam

Masao Kishida · Chihiro Mio · Keiichi Fujimori · Kiyoshi Imamura · Norimichi Takenaka · Yasuaki Maeda · Tran Thi Ngoc Lan · Yasuhiko Shibutani · Hiroshi Bandow

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Abstract We analyzed atmospheric particulate polycyclic aromatic hydrocarbons (PAHs) in Ho Chi Minh City, Vietnam, for 19 months. The average concentrations of total PAHs at dry and rainy seasons were 4.28 ± 2.83 and 15.71 ± 8.21 ng m^{-3}, respectively. The use of motorcycles without catalytic converters, estimated to be main emission sources of PAHs, would be higher during the dry season. PAH concentrations show a negative correlation with sunshine duration ($r = -0.51$). Furthermore, the ratio of average PAH concentration in the dry season to that in the rainy season shows a positive correlation with photolytic half-life ($r = 0.94$). Thus, seasonal changes in PAH concentrations are attributable to their photolytic degradation.

M. Kishida · K. Imamura
Research Institute for Environment, Agriculture, and Fisheries, Osaka Prefectural Government, 1-3-62 Nakamichi, Higashinari-ku, Osaka 537-0025, Japan

C. Mio · K. Fujimori · Y. Shibutani
College of Engineering, Osaka Institute of Technology, 5-16-1 Ohmiya, Asahi-ku, Osaka 535-8585, Japan

N. Takenaka · Y. Maeda · H. Bandow
Graduate School of Engineering, Osaka Prefecture University, 1-1 Gakuen-cho, Naka-ku, Sakai, Osaka 599-8531, Japan

T. T. N. Lan
College of Natural Science, Vietnam National University of Ho Chi Minh City, 227 Nguyen Van Cu Str., Dist. 5, Ho Chi Minh City, Vietnam

M. Kishida (✉)
Environmental Management Division, Department of Environment, Agriculture, and Fisheries, Osaka Prefectural Government, 2-1-2 Otemae, Chuo-ku, Osaka 540-0008, Japan
e-mail: kishida82477@iris.eonet.ne.jp

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Polycyclic aromatic hydrocarbons (PAHs) in the atmosphere are important because of their carcinogenicity and/or mutagenicity for human health (Fang et al. 2006). PAHs result from the incomplete combustion of organic matter, and are emitted into the environment via human activities such as cooking, heating, and the combustion of fossil fuels in motor vehicles. With the aim of understanding the distribution and toxicity of atmospheric particulate PAHs, many surveys have been undertaken in developed countries since the 1970s, as high-molecular-weight PAH compounds such as benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and benzo[*ghi*]perylene, which mainly exist in particulate matter (PM) in the atmosphere due to their low vapor pressure (Lao et al. 1973), are considered to be carcinogenic and/or mutagenic. Detailed surveys of PAHs have also recently been undertaken in Asian countries such as Taiwan (Fang et al. 2006), China (Oda et al. 2003), and Nepal (Kishida et al. 2009).

Ho Chi Minh City (HCMC), the largest city in Vietnam, is located in Southeast Asia, with a subtropical climate and high atmospheric temperatures throughout the year. The dry season extends from November to April, and the rainy season from May to October. In recent years, HCMC has become increasingly industrialized and urbanized, with a marked increase in the number of motorcycles, especially in urban areas. Consequently, severe air pollution has resulted from the exhaust gas of motorcycles (Lan et al. 2004). To observe air quality in HCMC, a monitoring program has been established to track concentrations of PM, CO, SO_2, ozone, and NO_2 in the atmosphere. Recent

studies have focused on carcinogenic and mutagenic PAHs in HCMC (Hien et al. 2007a, b) and Hanoi (Kishida et al. 2008). In particular, Hien et al. (2007b) reported that the concentrations of 11 PAHs in PM increased during the rainy season and decreased during the dry season; however, there exist insufficient data on particulate PAHs in HCMC to understand their behavior. Therefore, in the present study we monitored total suspended particulates (TSPs) and 45 particulate PAH compounds in HCMC for 19 months with the aim of understanding seasonal changes in PAH concentrations.

Materials and Methods

This study considered PAH compounds with molecular weights of ≥178. These compounds were classified into five categories depending on the number of aromatic rings in the molecule: (1) three rings: 9,10-dihydrophenanthrene (9,10-DiHyPh), 9,10-dihydroanthracene (9,10-DiHyAnt), dibenzothiophene (DiBTh), fluorenol, phenanthrene (Ph), anthracene (Ant), 1-, 2-phenylnaphthalene (1-, 2-PN), *o*-, *m*-, *p*-terphenyl, 1-, 2-, 3-, 4-, 9-methylphenanthrene (1-, 2-, 3-, 4-, 9-MePh), 2-, 9-methylanthracene (2-, 9-MeAnt), 3,6-dimethylphenanthrene (3,6-DiMePh), and 9,10-dimethylanthracene (9,10-DiMeAnt); (2) four rings: 1,2,3,4-tetrahydrofluoranthene (TeHyFlu), 4H-cyclopenta[*def*]phenanthrene (4H-CdefP), pyrene (Py), fluoranthene (Flu), benzo[*b*]fluorene, 1,1-binaphthyl, 9-phenylanthracene (9-PAnt), benz[*a*]anthracene (BaA), triphenylene (Tri), chrysene (Chr), and 7-methylbenz[*a*]anthracene (7-MeBaA); (3) five rings: benzo[*b*]fluoranthene (BbF), benzo[*j*]fluoranthene (BjF), benzo[*k*]fluoranthene (BkF), benzo[*e*]pyrene (BeP), benzo[*a*]pyrene (BaP), perylene, 3-methylcholanthrene (3-MeCh), 7-methylbenzo[*a*]pyrene (7-MeBaP), 9,10-diphenylanthracene (9,10-DiPAnt), and dibenz[*a,h*]anthracene (DiBahA); (4) six rings: indeno[1,2,3-*cd*]pyrene (INcdP), benzo[*ghi*]perylene (BghiP), and anthanthrene; and (5) seven rings: coronene (Cor).

The air samples were collected between one and three times per month from January 2003 to July 2004 at Vietnam National University, HCMC (10°45′46″N, 106°40′58″E), located in a residential area of HCMC. Eleven air samples were collected during the dry season (January to April 2003, and December 2003 to April 2004), and 16 during the rainy season (May to October 2003, and May to July 2004).

Total suspended particulates was collected on quartz-fiber filter (QFF, 203 × 254 mm; Advantec, Tokyo, Japan) at a flow rate of 1.0 m^3 min^{-1} for 24 h using a high-volume (HV) air sampler. Circular filters ($\phi = 60$ mm) were subjected to extraction with dichloromethane (DCM) using Soxhlet apparatus for 24 h. The extract was then concentrated and dissolved in hexane, and purified with 5 g of silica (deactivated by 5% distilled water) by gel column chromatography. The first fraction was eluted with 14 ml of hexane, and the second with 80 mL of 1% acetone/hexane. The latter fraction was concentrated to 1 mL under a gentle stream of pure nitrogen gas after the addition of 50 ng of a deuterated internal standard, Flu-d_{12}. One microliter of each concentrate was analyzed using a high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) (HP5890; Agilent, DE, USA/JMS700D; JEOL, Tokyo, Japan) equipped with an HP-1MS capillary column (15 m × 0.25 mm i.d. × 0.25 μm film thickness; J&W, DE, USA). The HRGC/HRMS analysis was conducted under low-resolution mode ($R \geq 1{,}000$). The methods used to extract, clean up, and analyze the collected air samples were described previously (Kishida et al. 2009). Meteorological data used in the present study were provided by the General Statistics Office, Government of Vietnam.

Results and Discussion

Table 1 lists the average concentrations of PAHs and TSP in HCMC during the dry and rainy seasons. The total concentration of 45 PAH compounds (∑45PAHs) was 1.44–9.21 ng m^{-3} (average: 4.28 ± 2.83 ng m^{-3}) during the dry season, and 5.52–31.51 ng m^{-3} (average: 15.71 ± 8.21 ng m^{-3}) during the rainy season. These values are comparable with those reported for Shizuoka, Japan (Ohura et al. 2004), but lower than those for Beijing, China (Oda et al. 2003), Taichung, Taiwan (Fang et al. 2006), and Kathmandu, Nepal (Kishida et al. 2009). TSP concentrations were 5.7–16.2 μg m^{-3} (average: 11.1 ± 2.8 μg m^{-3}) during the dry season, and 4.2–12.3 μg m^{-3} (average: 8.4 ± 2.2 μg m^{-3}) during the rainy season. We observed no correlation between ∑45PAHs in PM and TSP ($r = -0.06$).

Among the 4–7-ring PAHs, the concentrations of 3-ring PAHs were lowest during both seasons (Table 1). Most of the PAHs would have passed through the filter or evaporated from the surface of TSPs collected on the filter due to their high vapor pressure (Yamasaki et al. 1982; Park et al. 2001). The PAH compounds collected on the filters were mainly 4–7-ring PAHs. The dominant PAH compounds collected during the dry season were 5–7-ring PAHs, including Cor, BghiP, INcdP, BeP, BbF + BjF, and BkF; their average ratios to ∑45PAHs were 25.6% ± 4.3%, 24.9% ± 2.5%, 15.2% ± 1.2%, 9.2% ± 1.6%, 7.5% ± 1.9%, and 3.5% ± 0.9%, respectively. The dominant PAHs during the rainy season were also 5–7-ring PAHs, including BghiP (29.7% ± 3.9%), Cor (21.5% ± 3.4%), INcdP (16.7% ± 1.7%), BeP (10.4% ± 1.9%),

Table 1 Average concentrations of TSP and PAH compounds during the 2003–2004 dry and rainy seasons in Ho Chi Minh City, Vietnam

	Dry season	Rainy season
TSP (μg m^{-3})	11.1 ± 2.8	8.4 ± 2.2
PAH concentrations (*ng* m^{-3})		
Σ3-ring PAHs[a]	0.16 ± 0.10	0.25 ± 0.10
Flu	0.07 ± 0.05	0.09 ± 0.04
Py	0.13 ± 0.11	0.20 ± 0.10
benzo[b]fluorene	0.02 ± 0.01	0.05 ± 0.03
BaA	0.03 ± 0.03	0.14 ± 0.10
Tri + Chr	0.04 ± 0.02	0.09 ± 0.04
Σ4-ring PAHs[a]	0.30 ± 0.21	0.59 ± 0.28
BbF + BjF	0.33 ± 0.27	1.08 ± 0.52
BkF	0.15 ± 0.14	0.63 ± 0.38
BeP	0.40 ± 0.29	1.58 ± 0.85
BaP	0.08 ± 0.06	0.41 ± 0.30
Perylene	0.01 ± 0.01	0.06 ± 0.06
7-MeBaP	0.01 ± 0.01	0.04 ± 0.04
DiBahA	0.04 ± 0.03	0.14 ± 0.07
Σ5-ring PAHs[a]	1.02 ± 0.79	3.96 ± 2.11
INcdP	0.67 ± 0.48	2.64 ± 1.39
BghiP	1.10 ± 0.78	4.86 ± 2.91
DiBahA	0.04 ± 0.03	0.14 ± 0.07
Σ6-ring PAHs[a]	1.77 ± 1.25	7.56 ± 4.33
Cor	1.04 ± 0.62	3.36 ± 1.82
Σ7-ring PAH[a]	1.04 ± 0.62	3.36 ± 1.82
Σ45PAHs[b]	4.28 ± 2.83	15.71 ± 8.21

[a] ΣN-ring PAHs: Total N-ring PAH compounds, $N = 3–7$

[b] Total for the 45 PAH compounds described in the text

BbF + BjF (7.4% ± 2.0%), and BkF (4.1% ± 1.1%). Thus, the individual PAHs show similar patterns to each other during both seasons; however, lower concentrations were recorded during the dry season than during the rainy season. This observation suggests that the main emission sources of PAHs are similar during both seasons, and that seasonal differences in PAH concentrations are attributable to factors other than the appearance of additional emission sources of PAHs during the rainy season.

Many surveys have analyzed the diagnostic molecule ratios of PAHs to estimate the emission sources of PAHs (Park et al. 2001; Ohura et al. 2004; Zielinska et al. 2004; Fang et al. 2006; Hien et al. 2007a; Kishida et al. 2008, 2009). Hien et al. (2007a) reported INcdP/BghiP values of 0.4 for gasoline engines and 1.0 for diesel engines, while BghiP/BeP values are 2.9 ± 0.9 for gasoline engines and 1.3 ± 0.8 for diesel engines. In the present study, the average value of INcdP/BghiP was 0.61 ± 0.06 during the dry season and 0.58 ± 0.07 during the rainy season; average values of BghiP/BeP were 2.78 ± 0.57 during the dry season and 3.10 ± 1.46 during the rainy season. These values for HCMC are largely constant throughout the year, consistent with emission sources from gasoline engines. Asian Development Bank (2004) reported that the number of motorcycles in Vietnam exceeded 11 million in 2003, with 122,000 passenger vehicles. Therefore, the main emission source of particulate PAHs in both seasons is considered to be motorcycles.

Kishida et al. (2008) reported that Cor is one of the most abundant PAHs emitted from non-catalyzed engines, and that the Cor/(Cor + BghiP) value for PM emitted from non-catalytic engines is 0.42; the equivalent value for engines with catalytic converters is 0.19. The average Cor/(Cor + BghiP) values for PM in HCMC are 0.50 ± 0.05 during the dry season and 0.42 ± 0.05 during the rainy season, indicating that many motorcycles in the city are not fitted with catalytic converters. In addition, Cor/(Cor + BghiP) values during the dry season were slightly higher than those during the rainy season. This finding suggests that the number of motorcycles in HCMC without catalytic converters is greater during the dry season than during the rainy season.

Polycyclic aromatic hydrocarbon concentrations were higher during the rainy season than during the dry season; however, PAH discharge appears to be highest during the dry season. Hien et al. (2007a) reported that traffic jams are commonly caused by the greater use of passenger cars on rainy days, thereby explaining the increase in PAH concentrations recorded during the rainy season; however, this interpretation is not supported by the fact that PAH discharge in HCMC is dominated by motorcycles rather than passenger cars. Our results suggest that seasonal differences in PAH concentrations are strongly influenced by factors such as meteorological conditions rather than traffic conditions.

Table 2 summarizes the meteorological conditions in HCMC during the sampling period, revealing the typical features of a subtropical climate and pronounced differences between the dry and rainy seasons. In particular, the average monthly rainfall was much higher during the rainy season (281.9 mm) than during the dry season (2.6 mm). During the rainy season, sunshine duration is shorter, atmospheric temperature is slightly lower, and relative humidity is higher compared with the dry season.

Table 2 Average monthly values of meteorological factors during the 2003–2004 dry and rainy seasons in Ho Chi Minh City, Vietnam

	Dry season	Rainy season
Temperature (°C)	28.8 ± 1.5	28.2 ± 0.7
Relative humidity (%)	69 ± 2	79 ± 2
Rainfall (mm)	2.6 ± 4.4	281.9 ± 55.8
Sunshine duration (h)	209.8 ± 5.7	163.9 ± 24.6

In general, PM in the air is thought to be scavenged due to the washout process on rainy days, when PAHs associated with PM would decrease in accordance with the scavenged PM. Indeed, we observed a negative correlation between average monthly TSP concentrations and monthly rainfall ($r = -0.35$); in contrast, average monthly $\sum$45PAH concentrations show a positive correlation with monthly rainfall ($r = 0.57$). Greenfield (1957) noted that washout is mainly caused by Brownian expansion and inertial impaction, with the former being effective for very small particles (<0.01 μm in diameter), and the latter being effective for large particles (>1.0 μm in diameter). PM with a diameter of 0.01–1.0 μm falls in the "scavenging gap" (Andronache 2003). Many previous studies have shown that particulate PAHs mainly occur in fine particles, whereas most PM consists of coarse particles (Zielinska et al. 2004; Hien et al. 2007a). In particular, Hien et al. (2007a) reported that approximately 80% of particulate PAHs in HCMC occur in fine particles of <1.1 μm in diameter, whereas approximately 80% of PM consists of PM of >1.1 μm in diameter. Thus, washout would be effective in reducing PM concentrations, but not PAH concentrations.

Valerio and Lazzarotto (1985) reported reduced PAH concentrations due to photolytic degradation under intense radiation. In the present study, the average monthly $\sum$45PAH concentrations showed a negative correlation with monthly sunshine duration ($r = -0.51$). The lowest $\sum$45PAH concentration (1.44 ng m^{-3}) was observed in March 2003, which recorded the highest monthly sunshine duration (254.9 h) during the sampling period. This observation suggests that the decrease in PAH concentrations during the dry season could be attributable to photolytic degradation due to longer sunshine duration. A negative correlation was observed between average monthly $\sum$45PAH concentrations and average monthly temperature ($r = -0.31$). It is therefore possible that the slightly higher temperatures during the dry season accelerate the photolytic degradation of PAHs (Hien et al. 2007a) and/or evaporation of PAHs from PM (Yamasaki et al. 1982).

To examine the above proposal, in Fig. 1 we plot the ratios of average PAH concentrations in the dry season ($A\text{-}_{dry}$) to those in the rainy season ($A\text{-}_{rainy}$) vs. the photolytic half-lives of ten PAH compounds (Ph, Ant, Flu, BaA, BkF, BaP, INcdP, DiBahA, BghiP, and perylene) whose half-lives are well understood (Lu et al. 2007), revealing a positive correlation ($r = 0.94$; Student's t-test, $p < 0.01$). In particular, PAH compounds with short photolytic half-lives of 0.36–0.97 hrs (e.g., BaA, BkF, BaP, perylene, INcdP, DiBahA, and BghiP) show low $A\text{-}_{dry}/A\text{-}_{rainy}$ values of <0.30. In contrast, PAH compounds with longer photolysis half-lives of 2.94–4.51 (e.g., Ph, Flu, and Py) show high $A\text{-}_{dry}/A\text{-}_{rainy}$ values of 0.62–0.85. These data

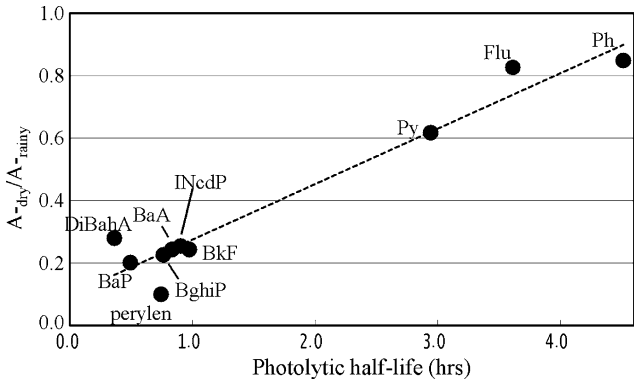

Fig. 1 Relationship between $A\text{-}_{dry}/A\text{-}_{rainy}$ values and photolysis half-lives of 10 PAH compounds. $A\text{-}_{dry}$ average PAH concentration in the dry season, $A\text{-}_{rainy}$ average PAH concentration in the rainy season. *Ph* phenanthrene, *Ant* anthracene, *Flu* fluoranthene, *BaA* benz[*a*]anthracene, *BkF* benzo[*k*]fluoranthene, *BaP* benzo[*a*]pyrene, *INcdP* indeno [1,2,3-*cd*]pyrene, *DiBahA* dibenz[*a,h*]anthracene, *BghiP* benzo[*ghi*] perylene

strongly support the proposal that PAH concentrations in HCMC decreased during the dry season due to photolytic degradation.

In the present study, we monitored 45 PAH compounds associated with PM for 19 months in HCMC, Vietnam. The average $\sum$45PAH concentration was higher during the rainy season than during the dry season, and the average TSP concentration was higher during the dry season than during the rainy season. In both seasons, motorcycles without catalytic converters are suspected to be the main emission source of PAHs. We observed a negative correlation between TSP concentration and rainfall, and a positive correlation between PAH concentration and monthly rainfall. Because PAH compounds associated with PM mainly occurred in the size range of the "scavenging gap," washout would have been ineffective in reducing PAH concentrations. In contrast, PAH concentrations show a negative correlation with sunshine duration. We also observed a positive correlation between $A\text{-}_{dry}/A\text{-}_{rainy}$ values and the photolytic half-lives of ten PAH compounds. These findings strongly support the proposal that the photolytic degradations of PAHs is accelerated by the longer sunshine duration of the dry season.

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