

Concentrations of Atmospheric Polycyclic Aromatic Hydrocarbons in Particulate Matter and the Gaseous Phase at Roadside Sites in Hanoi, Vietnam

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Abstract We analyzed the concentrations of polycyclic aromatic hydrocarbons (PAHs) in both particulate matter (PM) and the gaseous phase at 10 roadside sites in Hanoi, Vietnam. The average concentrations of 47 PAHs ($\sum 47\text{PAHs}$) were $63 \pm 82 \text{ ng m}^{-3}$ in PM and $480 \pm 300 \text{ ng m}^{-3}$ in the gaseous phase. The PAHs mainly originated from motorcycles without catalytic converters. The highest concentrations of $\sum 47\text{PAHs}$ in both PM and the gaseous phase were observed at a terminal for buses and trucks. The operation of large commercial vehicles led to increased PAH pollution at the terminal site.

Keywords Gaseous phase · Hanoi · Particulate matter · Polycyclic aromatic hydrocarbons (PAHs)

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The occurrence of polycyclic aromatic hydrocarbons (PAHs) as pollution in the atmosphere is of great concern in terms of human health (Waller 1952; Commins 1962; Lao et al. 1973). Most of the high-molecular-weight PAHs, including benz[*a*]anthracene, chrysene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, and benzo[*ghi*]perylene, are carcinogenic and/or mutagenic (Commins 1962; Lao et al. 1973), and most occur as particulate matter (PM) in the atmosphere because of their low vapor pressure (Yamasaki et al. 1982). Many previous studies have analyzed particulate PAHs collected using filters with the aim of understanding their distribution and toxicity. In contrast, the low-molecular-weight PAHs such as phenanthrene and pyrene occur mainly within the gaseous phase because of their high vapor pressure (Yamasaki et al. 1982). These PAHs are considered to be less harmful to human health than high-molecular-weight PAHs; however, they are able to react with other pollutants such as ozone and NO_x to form highly toxic compounds (Park et al. 2001). Investigations of particulate and gaseous PAHs in the atmosphere have been performed in developed countries such as Japan (Yamasaki et al. 1982), the USA (Park et al. 2001), and Greece (Mandalakis et al. 2002); however, there are few studies of gaseous PAHs compared with the number of studies that have analyzed particulate compounds.

Hanoi, the capital and second-largest city in Vietnam, is locating in the north of the country, with a population of 3 million. Since 1986, the economy of Vietnam has shown rapid growth due to the Doi Moi reforms that introduced free market economics to the previously socialistic country. The resulting industrialization has been accompanied by a dramatic increase in the number of motorcycles in urban areas, leading to concerns regarding air pollution in the form of PM, nitrogen oxide (NO_x), sulfur oxide (SO_x), and volatile organic compounds (VOCs) (Lan et al. 2004).

Hien et al. (2007a, b) studied particulate PAHs in an urban area in Vietnam; however, gaseous-phase PAHs have yet to be investigated in this country. In the present study, we investigated the distributions of atmospheric PAHs in both PM and the gaseous phase at 10 roadside sites in Hanoi. We also determined the sources of atmospheric PAHs based on diagnostic ratios of PAH compounds.

Materials and Methods

We analyzed PAH compounds with molecular weights of ≥ 178 . These compounds are classified into the following five categories depending on the number of aromatic rings in the molecule: 3-ring: 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; 9,10-dimethylanthracene; 9,10-dihydroanthracene (9,10-DiHyAnt); 9,10-dihydrophenanthrene (9,10-DiHyPh); and dibenzothiophene (DiBT); 4-ring: 1,2,3,4-tetrahydrofluoranthene; 4H-cyclopenta[def]phenanthrene (4H-CdefP); pyrene (Py); fluoranthene (Flu); benzo[*b*]fluorene; 1,1-binaphthyl; 9-phenylanthracene; benz[*a*]anthracene (BaA); triphenylene (Tri); chrysene (Chr); naphthacene; and 7-methylbenz[*a*]anthracene (7-MeBaA); 5-ring: benzo[*b*]fluoranthene (BbF); benzo[*j*]fluoranthene (BjF); benzo[*k*]fluoranthene (BkF); benzo[*e*]pyrene (BeP); benzo[*a*]pyrene (BaP); perylene; 3-methylcholanthrene; 7-methylbenzo[*a*]pyrene (7-MeBaP); 9,10-diphenylanthracene; and dibenz[*a,h*]anthracene; 6-ring: indeno[1,2,3-*cd*]pyrene (INcdP); benzo[*ghi*]perylene (BghiP); anthanthrene; naphtho[2,3-*a*]pyrene; and 3,4,8,9-dibenzopyrene; and 7-ring: coronene (Cor).

Air samples were collected at 10 roadside sites in Hanoi (Table 1) using mini-pumps (MP- $\Sigma 500$; Shibata, Japan) at a flow rate of 5 L min^{-1} over a period of 24 h. The samples were first passed through quartz fiber filters (QFFs) (Supelco, USA; $\phi = 32 \text{ mm}$) to collect particulate PAHs and then through two layers of polyurethane foam (PUF) plugs (Supelco; $\phi = 22 \text{ mm}$, height = 76 mm) to obtain gaseous PAHs. Prior to sampling, the QFFs were combusted at 600°C for 4 h, and the PUF plugs were washed with warm water, rinsed with acetone, and Soxhlet extracted with acetone for 8 h and then with dichloromethane (DCM) for 16 h. After sampling, the QFFs and PUF plugs were sealed in plastic cases and a glass container, respectively, and stored at 4°C . In this study, particulate PAHs are defined as those collected on the QFFs, whereas gaseous PAHs are defined as those collected on the two layers of PUF plugs.

Prior to extraction, 10 ng of each of five deuterated surrogate standards (Ph- d_{10} , Chr- d_{10} , BaP- d_{12} , BghiP- d_{12} , and Cor- d_{12}) were spiked in both the QFF and PUF plugs to enable corrections related to the recovery of PAH compounds throughout the extraction and cleanup procedures. The QFF and PUF plugs were separately extracted with DCM using Soxhlet apparatus for 24 h and ultrasonic extraction for 10 min (repeated 3 times), respectively. The extract was concentrated and then dissolved into hexane. The concentrate was purified with 5 g of 5% hydrous silica gel column chromatograph. The first fraction was eluted with 10 mL of hexane, and the second with 80 mL of 1% acetone/hexane. The second fraction was concentrated to 0.2 mL under a gentle stream of pure nitrogen gas after the addition of 10 ng of each of two deuterated internal standards (Flu- d_{12} and perylene- d_{12}). PAH compounds were

Table 1 Descriptions of the 10 sampling sites in Hanoi, Vietnam

Site no.	Site description		Geographic coordination		Sampling dates
			North	East	
1	Cau Moi Bridge	Intersection	21°00.085'	105°49.091'	5–6/Aug/2005
2	De La Thanh and Nguyen Chi Thanh	Intersection	21°01.506'	105°48.686'	9–10/Aug/2005
3	Lieu Gai Street	Roadside	21°02.444'	105°49.950'	10–11/Aug/2005
4	Ton Duc Thang and Kham Thien	Intersection	21°01.144'	105°49.819'	11/Aug/2005
5	Dai Co Viet and Giai Phong	Intersection	21°00.433'	105°50.496'	15–16/Aug/2005
6	Tran Nhan Tong and Ba Trieu	Intersection	21°01.038'	105°50.959'	16/Aug/2005
7	Tran Hung Dao and Le Duan	Intersection	21°01.438'	105°50.497'	27–28/Aug/2005
8	Le Hong Phong and Dien Bien Phu	Intersection	21°02.006'	105°50.298'	28/Aug/2005
9	Hanoi Opera House	Roadside	21°01.460'	105°51.453'	30–31/Aug/2005
10	Long Bien intersection	Terminal	21°02.465'	105°50.999'	29–30/Aug/2005

analyzed using a high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) (HP5890; Agilent, USA; JMS700D; JEOL, Japan) equipped with an HP-1MS capillary column (15×0.25 mm i.d. \times 0.25 μ m film thickness). The injection port was kept at 300°C, and 1 μ L of each concentrate was injected in splitless mode followed by a 90 s purge. The column temperature was held at 70°C for 1 min, then programmed to increase at 15°C/min to 130°C and then at 6°C/min to 300°C before being held for 2 min. HRGC/HRMS analyses were conducted under high-resolution mode ($R \geq 10000$).

Results and Discussion

Figure 1 shows the total concentrations of 47 PAHs compounds ($\Sigma 47\text{PAHs}$) in PM and gaseous phases at the 10 sampling sites. The average $\Sigma 47\text{PAHs}$ were 63 ± 82 ng m⁻³ in PM and 480 ± 300 ng m⁻³ in the gaseous phase. The obtained values of particulate $\Sigma 47\text{PAHs}$ are higher than those reported previously for residential areas in Ho Chi Minh City (HCMC), Vietnam (Hien et al. 2007a), an industrial area in Texas, USA (Park et al. 2001), and urban areas in Athens, Greece (Mandalakis et al. 2002), and are similar to those reported for a roadside site in HCMC, Vietnam (Hien et al. 2007b). The obtained values of gaseous $\Sigma 47\text{PAHs}$ are also higher than those reported for the industrial area in Texas, USA (Park et al. 2001) and the urban areas in Athens, Greece (Mandalakis et al. 2002). Thus, the values of $\Sigma 47\text{PAHs}$ in PM and the gaseous phase measured at roadside sites in Hanoi are relatively high, and particulate $\Sigma 47\text{PAHs}$ make up approximately $10 \pm 3.5\%$ of the combined particulate and gaseous $\Sigma 47\text{PAHs}$.

The highest concentrations of particulate and gaseous $\Sigma 47\text{PAHs}$ were recorded at Site 10 (290 and 1300 ng m⁻³, respectively), located near a terminal for buses and trucks. At this site, particulate $\Sigma 47\text{PAHs}$ made up approximately 19% of the total $\Sigma 47\text{PAHs}$, being the highest contribution among the 10 sites. These observations indicate that the frequent use of large commercial vehicles in the area of the terminal has a strong influence on PAH pollution.

Table 2 shows the average concentrations of PAH compounds with 3–7 aromatic rings in PM and gaseous phases. The dominant PAH compounds in PM at Sites 1–9 were 5–7-ring PAHs, including BghiP (8.3 ± 4.1 ng m⁻³), Cor (7.7 ± 4.5 ng m⁻³), and INcdP (7.5 ± 3.4 ng m⁻³). In contrast, the dominant PAH compounds in the gaseous phase at the nine sites were 3–4-ring PAHs, including Ph (150 ± 54 ng m⁻³), Py (65 ± 30 ng m⁻³), Flu (36 ± 14 ng m⁻³), and MePh isomers (42 ± 13 ng m⁻³). These findings are consistent with those of previous studies (Yamasaki et al. 1982; Park et al. 2001; Mandalakis et al. 2002).

At Site 10, the predominant PAH compounds in PM were 4–6-ring PAHs, including BaP (52 ng m⁻³), INcdP (37 ng m⁻³), BkF (33 ng m⁻³), BbF + BjF (30 ng m⁻³), BghiP (29 ng m⁻³), Tri + Chr (27 ng m⁻³), BeP (22 ng m⁻³), and Cor (17 ng m⁻³). For gaseous PAHs, the predominant compounds at Site 10 were 3–4-ring PAHs, including Ph (300 ng m⁻³), Py (270 ng m⁻³), Flu (180 ng m⁻³), MePh isomers (110 ng m⁻³), and Ant (96 ng m⁻³). Thus, the patterns of particulate and gaseous PAHs observed at Site 10 are different from those at the other sites, with higher concentrations of 4-ring PAHs in PM and the gaseous phase than at other sites.

In estimating the emission sources of PAHs, many surveys have employed diagnostic molecule ratios of PAHs (Rogge et al. 1993; Park et al. 2001; Yunker et al. 2002; Zakaria et al. 2002). The values of five diagnostic ratios of PAHs, as determined in the present study, are listed in Table 3.

Zakaria et al. (2002) reported that the value of $\Sigma \text{MePh}/\text{Ph}$ for petrol (petrogenic origin) is greater than 2.0, whereas that for combustion exhaust (pyrogenic origin) is <1.0 . The average $\Sigma \text{MePh}/\text{Ph}$ values obtained for the 10 roadside sites analyzed in the present study were 0.75 ± 0.20 for PM and 0.29 ± 0.04 for the gaseous phase, indicating dominantly pyrogenic sources.

Yunker et al. (2002) estimated the emission sources of PAHs based on the value of Flu/(Flu + Py). The authors proposed that values of <0.2 , 0.2 – 0.5 , and >0.5 correspond to petrogenic origins, exhaust gases from gasoline and

Fig. 1 Concentrations of particulate and gaseous $\Sigma 47\text{PAHs}$ measured at 10 roadside sites in Hanoi, Vietnam

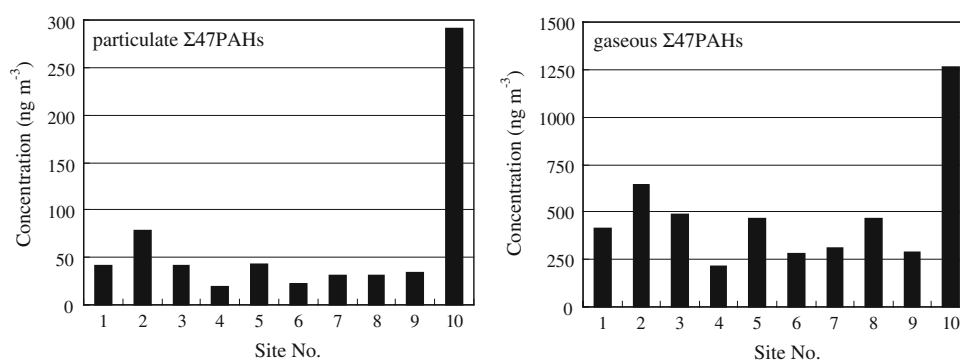


Table 2 Individual PAH concentrations (ng m^{-3}) included in PM and the gaseous phase

	Average concentrations at sites 1–9		Concentrations at site 10	
	Particulate PAHs	Gaseous PAHs	Particulate PAHs	Gaseous PAHs
DiBT	0.03 ± 0.01	17 ± 5.1	0.05	28
Fluorenel	N.D. ^a	8.0 ± 3.0	N.D. ^a	51
Ph	0.23 ± 0.11	150 ± 54	0.40	300
1-MePh	0.05 ± 0.02	11 ± 3.7	0.08	36
2-MePh	0.06 ± 0.03	17 ± 5.7	0.10	37
3-MePh	0.04 ± 0.01	10 ± 2.9	0.06	26
4- + 9-MePh	0.01 ± 0.01	4.4 ± 1.4	0.03	11
9,10-DiHyPh	0.02 ± 0.05	11 ± 8.6	N.D. ^a	13
Ant	N.D. ^a	15 ± 6.1	N.D. ^a	96
2-MeAnt	0.02 ± 0.02	3.7 ± 1.7	0.06	13
9-MeAnt	0.30 ± 0.11	2.8 ± 1.5	0.01	13
9,10-DiHyAnt	0.04 ± 0.10	6.4 ± 5.5	N.D. ^a	11
1-PN	N.D. ^a	5.8 ± 1.5	N.D. ^a	24
2-PN	0.02 ± 0.02	4.7 ± 1.2	0.06	24
Σ 3-ring PAHs ^b	0.85 ± 0.34	270 ± 78	0.90	690
Flu	0.66 ± 0.24	36 ± 14	1.2	180
Py	1.2 ± 0.41	65 ± 30	2.2	270
BaA	1.0 ± 0.50	1.8 ± 0.78	13	13
Tri + Chr	0.53 ± 0.21	1.2 ± 0.40	27	3.6
7-MeBaA	0.78 ± 0.40	1.0 ± 0.44	7.3	1.4
Σ 4-ring PAHs ^c	4.3 ± 1.7	130 ± 56	51	570
BbF + BjF	2.3 ± 1.0	0.12 ± 0.04	30	0.28
BkF	1.0 ± 0.49	0.11 ± 0.04	33	0.24
BeP	1.8 ± 0.86	0.09 ± 0.05	22	0.19
BaP	1.6 ± 1.0	0.13 ± 0.19	52	0.93
Σ 5-ring PAHs ^d	7.2 ± 3.6	0.49 ± 0.31	150	2.0
IncdP	7.5 ± 3.4	0.12 ± 0.18	37	0.62
BghiP	8.3 ± 4.0	0.40 ± 0.60	29	1.0
Σ 6-ring PAHs ^e	16 ± 7.7	0.54 ± 0.83	76	2.0
Cor	7.7 ± 4.5	0.34 ± 0.52	17	0.53
Σ 7-ring PAHs ^f	7.7 ± 4.5	0.34 ± 0.52	17	0.53
Σ 47PAHs	38 ± 17	400 ± 130	290	1300

^a Not detected^b Total 3-ring PAH compounds^c Total 4-ring PAH compounds^d Total 5-ring PAH compounds^e Total 6-ring PAH compounds^f Total 7-ring PAH compound

diesel engines, and the combustion of coal, grass, and wood, respectively. The average Flu/(Flu + Py) values obtained for the 10 roadside sites analyzed in the present study were 0.35 ± 0.03 for PM and 0.37 ± 0.02 for the gaseous phase, indicating a source comprising exhaust gases from gasoline and diesel engines.

In a study of particulate PAH compounds emitted from diesel engines, Rogge et al. (1993) reported that the concentrations of PAHs with 4 aromatic rings (e.g., Py) were higher than those with 5–7 aromatic rings. The Py/BaP ratio was close to 13 for compounds emitted from diesel engines, with the value for gasoline engines being approximately 1.3. The average Py/BaP value for PM at Sites 1–9 in the present study was 0.92 ± 0.39 , similar to that for PAHs from gasoline engines. Given that the Asian

Development Bank (2004) reported 11 million motorcycles in Vietnam compared with just approximately 122,000 passenger cars, the above findings indicate that the atmospheric PAHs recorded in the present study mainly originated from motorcycles. The Py/BaP value for Site 10 was 0.04 because of high BaP concentrations in PM (52 ng m^{-3}), being more than 30 times higher than the average concentration at the other sites ($1.6 \pm 1.0 \text{ ng m}^{-3}$).

The BaP/BeP value in PM has been used to estimate the degree of stability of PAHs in the environment (Cotham and Bidleman 1995; Hien et al. 2007a). BaP is a highly reactive compound because of its short photochemical half-life, whereas the long half-life of BeP makes it relatively stable (Hien et al. 2007a; Lu et al. 2007). The average BaP/BeP value for Sites 1–9 in the present study was 0.85 ± 0.20 ,

Table 3 Diagnostic ratios of PAH compounds at roadside sites in Hanoi, Vietnam

	Average ratios for sites 1–9		Ratios for site 10	
	Particulate PAHs	Gaseous PAHs	Particulate PAHs	Gaseous PAHs
$\sum \text{MePh/Ph}$	0.76 ± 0.21	0.29 ± 0.04	0.70	0.36
$\text{Flu}/(\text{Flu} + \text{Py})$	0.35 ± 0.04	0.37 ± 0.02	0.35	0.40
Py/BaP	0.92 ± 0.39	–	0.04	–
BaP/BeP	0.85 ± 0.20	–	2.31	–
$\text{Cor}/(\text{Cor} + \text{BgHiP})$	0.48 ± 0.02	–	0.37	–

higher than values reported previously for residential areas in HCMC (Hien et al. 2007b). The high values at Sites 1–9 reflect the fact that the main emission sources of atmospheric PAHs were situated close to the sampling sites (Cotham and Bidleman 1995). At Site 10, the value of BaP/BeP was 2.31, more than twice as high as the values measured at other sites. In general, BaP/BeP values are higher at nighttime than at daytime (Hien et al. 2007a). In Hanoi, large commercial vehicles are prohibited from entering the central city; most buses leave from or arrive at the bus and truck terminal (Site 10). Furthermore, many trucks arrive at the terminal from the suburbs to supply goods to central Hanoi during the nighttime. These observations suggest that trucks make a significant contribution to the high BaP/BeP value recorded at Site 10. A positive correlation observed between BaP/BeP values and particulate $\sum 47\text{PAHs}$ ($r = 0.9245$, Student's t -test, $p < 0.01$) indicates that the extremely high concentrations of $\sum 47\text{PAHs}$ might be related to nighttime traffic.

Finally, $\text{Cor}/(\text{Cor} + \text{BgHiP})$ values were used to estimate the emission sources of the measured PAHs. Rogge et al. (1993) reported that BgHiP and coronene are the most abundant PAHs emitted from non-catalyzed engines. The authors calculated that the $\text{Cor}/(\text{Cor} + \text{BgHiP})$ values for PM emitted from non-catalytic engines are 0.42, whereas those for engines with a catalytic converter are 0.19. The average $\text{Cor}/(\text{Cor} + \text{BgHiP})$ value for PM at the 10 roadside sites analyzed in the present study was 0.46 ± 0.04 , suggesting that many motorcycles in Hanoi are not equipped with catalytic converters.

In the present study, atmospheric PAHs in both PM and the gaseous phase were investigated at 10 roadside sites in Hanoi. The obtained concentrations are higher than those reported previously from other countries. Approximately 90% of atmospheric PAHs occurred in the gaseous phase. The measured atmospheric PAHs mainly originated from motorcycles without catalytic converters, as indicated by the high concentrations of particulate PAHs with 6–7 aromatic rings (BgHiP, Cor, and INcdP) and gaseous PAHs with 3 aromatic rings (Ph). The highest levels of $\sum 47\text{PAHs}$ in both PM and the gaseous phase were recorded at a bus and truck terminal. The concentrations of PAHs with 4 aromatic rings (in both PM and the gaseous

phase) were highest at this site because of the operation of large commercial vehicles in the area of the terminal. In particular, nighttime traffic around the terminal site makes a significant contribution to PAH pollution.

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