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Short communication

# Polycyclic aromatic hydrocarbons in the airborne particulate matter at a location 40 km north of Bangkok, Thailand

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## Abstract

Total suspended particulate matter in ambient air was sampled by high-volume samplers at four sites at the Asian Institute of Technology campus, west of the Phahonyothin Road, Phatumthani Province, 40 km North of Bangkok, Thailand. The concentrations of 18 polycyclic aromatic hydrocarbons (PAH), were measured by gas chromatography with flame ionisation and/or liquid chromatography with fluorescence detection. The PAH profile with relatively high concentrations of benzo(ghi)perylene and coronene, decreasing with the distance from the road, suggested a substantial contribution from the traffic. The concentrations in the core of the campus were in the same range as those reported for residential areas in the Bangkok Metropolitan. © 2000 Elsevier Science Ltd. All rights reserved.

*Keywords:* PAH; Particulate matter; Bangkok

## 1. Introduction

Polycyclic aromatic hydrocarbons (PAH) constitute a major group of carcinogens and mutagens in the environment. PAH are products of incomplete combustion and released from various sources, both natural and anthropogenic. In the latter category burning of fossil fuels, refuse and agro-residues, were estimated to contribute more than 90% of the PAH to the environment (Hutzinger and Reischl, 1990).

PAH are initially emitted as gases during combustion processes. Heavier PAH (with more than three rings) are rapidly attached to existing particles, usually soot particles, by adsorption or condensation upon cooling of fuel gas (Kamens et al., 1995). A considerable fraction of

the more volatile PAH will escape particulate incorporation and exist in the gas phase. Upon long-term ageing the aerosols grow in size hence particle size distribution of PAH undergoes a slight shift toward larger particles (Vaeck and Cauwenberghe, 1985).

The concentration of PAH in the atmosphere is in the order of  $\text{ng m}^{-3}$ . The majority of PAH (70–90%) are sorbed on suspended particles at ambient temperatures. The lighter PAH (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. Moreover, PAH are mostly sorbed on small inhalable particles with a high concentration on airborne particles of sub-micron diameters (Vaeck and Cauwenberghe, 1978; Nicolaou et al., 1984), which can be deposited in the respiratory tract, hence increasing the potential health effects. During their atmospheric residence time (a few days to weeks) the fine PAH-carrying particles may be transported over large distances. Thus PAH become widespread in the

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environment (Masclat et al., 1988). PAH are planar and relatively inert, but in the giant reactor of the atmosphere they participate in various chemical reactions and decompose. Most PAH are readily photo-oxidised when exposed to UV light of 300–420 nm. They undergo thermal decomposition and react with a number of atmospheric chemicals producing derivatives, which can be more toxic than the original compounds (Nicolaou et al., 1984).

Due to the toxic and carcinogenic effects, as well as the ubiquity of PAH in the environment, the compounds have received a considerable research interest. With the increase in fossil fuel combustion, resulting from the industrial expansion, traffic and population growth, over the last few decades, the atmospheric concentrations of PAH in Asian countries are expected to be high. Presently, air-quality-monitoring activities have just started in the Asian developing countries and are mainly restricted to common parameters such as suspended particulate matter, carbon-, sulphur- and/or nitrogen-oxides. Much less attention is given to monitoring for PAH, which requires more elaborate analytical protocols. As a result, environmental data on these toxic compounds in the region are still scarce.

This study was a part of the efforts to fill up the gap in the air-quality data. It was designed to initiate the monitoring activities of PAH in the region. The obtained database will be useful for the establishment of efficient air-quality management programs to reduce health risks resulted from the pollutants.

## 2. Materials and methods

### 2.1. Study area

The Asian Institute of Technology (AIT) is located on a 160-ha area, west of the Phahonyothin Road, the main traffic artery to and from the north and northeast of Thailand to Bangkok. The road traffic was composed of both gasoline and diesel vehicles. The average traffic density during rush hours was 6600 vehicles  $\text{h}^{-1}$  at an average speed of 38  $\text{km h}^{-1}$  (Boontharawara et al., 1994) with a daily average of 60,000 vehicles (Anonymous, 1994a). The high traffic density at rush hours and regular U-turns at street level during the monitoring periods partly explain the low average speed, and increased vehicle emission. On campus sources include cooking facilities, vehicles on campus roads, diesel-fuelled back-up energy supply, a petrol station, and laboratory exhaust. Some off-campus adjacent activities may also contribute to the contaminants in the AIT air. These would include the dense traffic in the Bangkok city centre (40 km south), the transport at Don Muang airport (17 km south) and a small industrial estate (6 km north).

### 2.2. Sampling

High-volume samplers, HVC-1000, SIBATA, Japan, and Precision Scientific Co., USA, were used to collect total suspended particulate matter at four sites. Site 1 was located at the academic building area while site 2 was located between the student cafeteria and a gasoline station around 400 m from site 1. Site 3, a roadside, was at the AIT gate, next to the Phahonyothin Road in the southeast corner of the campus and around 800 m from site 1. Site 4 was in the green field, near the meteorological station at the northwest of the campus area and around 2000 m from site 1. Sampling was done in June 1996, October–November 1996 and April 1997 during the days when there was no rain. The average daily temperature was 27.5–29°C with a diurnal temperature fluctuation of 6.0–8.5°C. Each sample was collected for a period of around 24 h with an average sampled air volume of 2200  $\text{m}^3$ . Extraction of the PAH from the total suspended particles (TSP) retained on the glass microfibre filters (Whatman, 934-AH<sup>TM</sup>, cat. No 1827 110) was proceeded immediately after the sample collection.

### 2.3. Reagents

All reagents were tested in procedural blanks, and of chromatographic-grade quality, from the J.T. Baker Company, except for the anhydrous sodium sulphate (p.a.), and silica gel (chromatographic grade), 60–230 mesh, which were purchased from the Merck Company.

### 2.4. Analytical equipment and procedure

The analytical method was developed on the basis of the US EPA method TO-13 (US EPA, 1988) schematically presented in Fig. 1. The details on sample extraction and clean-up procedures are given in Kim Oanh et al. (1999).

The samples collected at sites 1 and 2 were analysed by gas chromatography with flame ionisation detection (GC-FID), Shimadzu GC14B. Due to the limited availability of PAH standards at the time only three compounds were analysed: benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP) and indeno(1,2,3-cd)pyrene (IP). A glass column of 1.7 m length and 2.6 mm ID was packed with Chromosorb W (AW-DMCS, 100/120 mesh) coated with 3% OV-17. The oven temperature program was as follows: 100°C (4 min) and increased at 8°C  $\text{min}^{-1}$  to 280°C (15 min). The  $\text{N}_2$  carrier gas flow rate was of 50  $\text{ml min}^{-1}$ .

The samples from sites 3 and 4 were analysed for 18 PAH including 16 US EPA priority PAH plus coronene (COR) and benzo(e)pyrene (BeP) by a Hewlett-Packard high-performance liquid chromatography, HP 1050, using the same analytical method and equipment as presented in Kim Oanh et al. (1999).

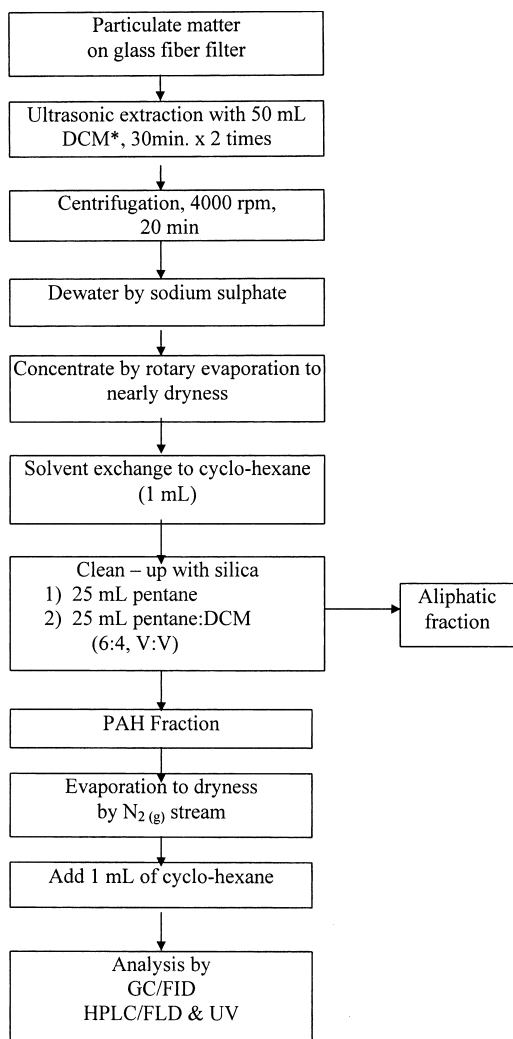


Fig. 1. Analytical procedures. \*DCM: dichloromethane.

External PAH standards (18 PAH in a mixture) were used to quantify the individual analytes, from both the GC-FID and HPLC-FLD analyses. The linear correlation between the compound concentrations, and both peak height and peak areas had  $R^2$  values in the range of 0.98–0.99 for all PAH analysed.

The recovery test was done by spiking known amounts of PAH onto pre-extracted TSP retained on glass fibre filters. The spiked sample was then sealed in a Petri dish and was analysed 24 h afterward using the method given in Fig. 1. Four tests were conducted for each analytical procedure. The average recovery of the GC-FID method was 99, 91 and 92% for BkF, BaP and IP, respectively. The obtained average recovery of HPLC was as follows:

fluorene (FLU), 32%; phenanthrene (PHE), 101%; anthracene (ANT), 38%; fluoranthene (FA), 84%; pyrene (PY), 103%; benzo(a)anthracene (BaA), 84%; chrysene (CHR), 98%; BeP, 96%; benzo(b)fluoranthene (BbF), 98%; BkF, 63%; BaP, 81%; dibenzo(a,h)anthracene (DBaA), 90%; benzo(g,h,i)perylene (BghiP), 102%; IP, 95% and COR, 108%. The recovery was low for the more volatile compounds including the first three compounds (naphthalene, acenaphthylene and acenaphthene) which, as seen later, were not detected in the analysed samples plus FLU and ANT. This may also be due to the gas-filter paper partitioning of the compounds rather than the analytical procedure alone. The recovery was, therefore, not included in the results reported in this study.

The analytical protocol used in both the HPLC-FLD and GC-FID applications had, however, previously proved satisfactory in an inter-calibration exercise of the partly certified urban dust Standard Reference Materials, SRM 1649 (Kim Oanh, 1999).

### 3. Results and discussions

The average concentrations of the selected PAH and TSP are presented in Fig. 2. Site 1 located in the academic building area, distant from principal combustion sources was found with the noticeable lower levels of the BaP and IP as compared to site 3. The particulate matter concentration at this site was the lowest ( $56 \mu\text{g m}^{-3}$ ). The PAH levels at site 2 were slightly higher than those at site 1 though they still can be considered to be in the same range. The vicinity of site 2 to the gasoline station and the traffic within the campus, as well as cooking activities in the cafeteria may be the main reasons for the higher PAH levels. It is noted that the GC columns are generally not able to separate BkF, BbF and benzo(j)fluoranthene (BjF). Hence the results reported for BkF at sites 1 and 2, for which the GC was used, may be the sum of these three compounds and the actual BkF levels may be lower than the reported levels.

Among the 18 PAH analysed (Fig. 2) at sites 3 and 4, the more volatile compounds, naphthalene (NAPH), acenaphthylene (ACY) and acenaphthene (ACE) were not detected on the TSP at the former site plus FLU at the latter. However, these PAH of lower ring number may be present in the gas phase, especially at the high ambient temperatures during the sampling period (27–29.5°C shadow values). As a matter of fact, for the same range of ambient temperatures, at a residential sampling site in Bangkok city Hatairatana (1999) found PY, which is heavier than the four above-listed PAH, almost entirely in gaseous phase (91%) while heavier compounds were mostly sorbed on particulate matter: 77% for BaA and 79% for BeP and 100% for BaP, DBaA and BghiP.

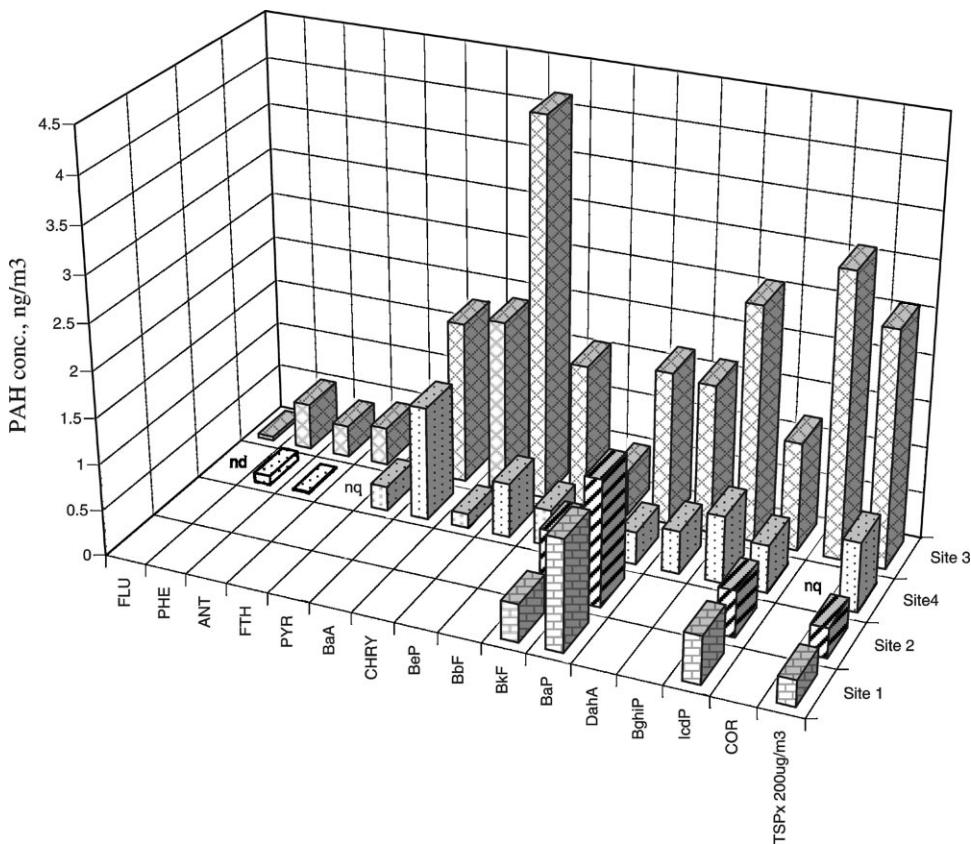


Fig. 2. Concentration of PAH on airborne particulate matter ( $\text{ng m}^{-3}$ ) and TSP (in  $200 \mu\text{g m}^{-3}$ ) in ambient air samples; blank cells: not analysed; nd: not detected; nq: not quantified.

Thus, at the high temperature at the monitoring locations the fractions of low-ring PAH present in the gas phase are possibly substantial. This suggested that the information on gas-phase PAH is important. The possibility of more heavy PAH present in gas phase at high temperatures and the longer atmospheric residence time of gaseous PAH emphasise the strong need to monitor the gaseous phase of the compounds.

The concentrations of individual PAH, in fractions of BaP (Table 1) show high ratios for BeP, COR, BghiP at site 3. At site 4 high ratios for BaA, BghiP, BeP and IP were obtained, however, FA and COR were identified but not quantified, due to the generally low PAH concentrations.

Relatively high concentrations of BghiP and COR on particulate matters are reported to be markers of vehicular emissions (Gordon and Bryan, 1973; Greenberg et al., 1981; Kiss et al., 1996). Harrison et al. (1996) noted that BghiP and COR are indicative for gasoline while PHE and benzo(b)naphtho(2,1-d)thiophene (BNT) for diesel vehicles. In addition, Duval and Friedlander (1981), as cited by Harrison et al. (1996), noted that BbF, in addi-

tion to BghiP, COR, FA and PY, are indicators of diesel-powered vehicles.

The PAH-profile obtained in this study and the increase in PAH and TSP concentrations from field site (4) to roadside (3) indicate that the road traffic is the main contributor of the pollutants at the sites.

Details on the highway traffic composition, i.e. fraction of gasoline and diesel vehicles, were not available. The low ratio of PHE to BaP associated with the particulate matter phase ( $\sim 0.3$  at both sites 3 and 4) is not, however, indicative of a low contribution from diesel vehicles. The fact that PHE at tropical temperatures mostly remains in the gas phase as discussed above suggests that the gas-phase data are required for a more complete interpretation.

Emission from domestic cooking using oil, gas, wood, etc. is reported to have a relatively high fraction of lower molecular weight PAH, including ANT, PHE, FA and PY (Harrison et al., 1996; Kiss et al., 1996). Kim Oanh et al. (1999), who studied PAH emission from charcoal and wood also found a high fraction of low-ring PAH (from NAPH to PY). Bottled gas is the only principal fuel used

Table 1  
Average individual PAH to BaP ratios

Sites (no. of samples)	PAH compounds														
	FLU	PHE	ANT	FA	PY	BaA	CHR	BeP	BbF	BkF	BaP	DBahA	BghiP	IP	COR
Site 1 (5) Academic building										0.34 <sup>a</sup>	1 [1.2]			0.43	
Site 2 (5) Cafeteria & gas station										0.51 <sup>a</sup>	1 [1.4]			0.36	
Site 3 (3) AIT gate, roadside	0.02	0.3	0.21	0.25	0.13	1.06	1.12	2.47	0.94	0.29	1 [1.7]	0.94	1.53	0.71	1.82
Site 4 (3) Meteorological station	nd	0.29	0.03	nq	0.77	3.43	0.46	1.67	1.11	0.46	1 [0.35]	1.31	2.08	1.46	nq

<sup>a</sup> BkF values may include BbF and BfF.

nd: not detected; nq: not quantified; blank cell: not analysed. Concentration of BaP in ng m<sup>-3</sup> are given in parentheses [..].

Analysed compounds and the minimum detectable quantity of the method (HPLC-FLD/UV) are given as follows:

- NAPH: naphthalene (UV-0.41 ng)
- FLU: fluorene (FLD-0.005 ng)
- FA: fluoranthene (FLD-0.015 ng)
- CHR: chrysene (FLD-0.019 ng)
- BkF: benzo(k)fluoranthene (FLD-0.012 ng)
- BghiP: benzo(g,h,i)perylene (FLD-0.062 ng)
- ACY: acenaphthylene (UV-0.35 ng)
- PHE: phenanthrene (FLD-0.013 ng)
- PY: pyrene (FLD-0.032 ng)
- BeP: benzo(e)pyrene (UV-0.066 ng)
- BaP: benzo(a)pyrene (FLD-0.02 ng)
- IP: indeno(1,2,3-c,d)pyrene (UV-0.15 ng)
- ACE: acenaphthene (UV-0.22 ng)
- ANT: anthracene (FLD-0.008 ng)
- BaA: benzo(a)anthracene (FLD-0.014 ng)
- BbF: benzo(b)fluoranthene (UV-0.038 ng)
- DBahA: dibenzo(a,h)anthracene (FLD-0.02 ng)
- COR: coronene (FLD-0.72 ng)

Table 2  
Comparison between selected data of the present study and others

Sampling site	Concentration, ng m <sup>-3</sup>		
	BkF	BaP	IP
EE Building (site 1)	0.4 <sup>a</sup>	1.2	0.5
Cafeteria (site 2)	0.7 <sup>a</sup>	1.4	0.5
AIT Gate (site 3)	0.5	1.7	1.2
Meteorological station (site 4)	0.2	0.4	0.5
<i>Data for comparison</i>			
Beijing, Residential <sup>b</sup>	na	29.3	na
Beijing, Clean area <sup>b</sup>	na	4.9	na
Bangkok, Roadside <sup>c</sup>	1.1	2.5	na
Bangkok, Residential <sup>c</sup>	0.5	0.7	na
Stockholm centre <sup>d</sup> , winter	0.5	0.2	0.4
New Jersey, summer <sup>e</sup>	0.03–0.2	0.06–0.23	0.09–0.46
New Jersey, winter <sup>e</sup>	0.28–0.97	0.32–1.63	0.79–2.9

<sup>a</sup>May include BbF and BjF; na: not available.

<sup>b</sup>Zhao and Sun (1986).

<sup>c</sup>Hathairatana (1999).

<sup>d</sup>Broman et al. (1991).

<sup>e</sup>Greenberg et al. (1985).

for cooking on campus. Some diesel oil, around 15,000 l yr<sup>-1</sup>, is also used for the power backup system. These on-campus stationary sources thus could principally contribute to the fraction of lower molecular weight PAH.

The green AIT campus, as well as the surrounding green belt of trees and bushes, and the golf course, serve as an effective barrier to prevent pollutants, especially those associated with particulate matter, from reaching deep inside the campus. This may be the main reason for the low levels of PAH and TSP found inside the campus area.

As compared to a residential site in Bangkok the PAH levels inside the AIT campus (sites 1–2, Table 2) are higher for BaP but in the same range for BkF (taking into account the fact that BkF for these sites may include BbF and BjF as well). Site 3 at AIT gate, which is a roadside, has lower PAH levels than the roadside inside the Bangkok City. In the Stockholm centre the BkF and IP concentrations were in the same range as those of sites 1 and 2, but with lower BaP level. The PAH levels found at AIT are higher than those reported for New Jersey during summer but of the same magnitude as the winter levels.

There are not yet many air-quality standards established for PAH in the world. In Italy, for example, BaP must not exceed 2.5 ng m<sup>-3</sup>, which would be decreased to 1.0 ng m<sup>-3</sup> from 1 January 1999 (Anonymous, 1994b).

For the carcinogenic compounds the general consensus is that there is no threshold level for induction. This means that any non-zero exposure is an added risk. There are also mounting evidences that skin absorption may be an overlooked and underestimated way of exposure, which may be of particular importance in tropical countries (Quinlan et al., 1995).

#### 4. Conclusions

PAH and TSP concentrations were found to decrease from the Pahonyothin Road in the east to the campus field in the west. A typical traffic pattern of PAH suggested a substantial contribution from vehicles. The green belt surrounding the campus may to some extent protect the campus area from intrusion of particulate pollutants. The levels of PAH found in the campus core were compatible with the levels in residential areas in Bangkok City though BaP at AIT was somewhat higher.

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