# Characterization and Carcinogenic Risk Assessment of Polycyclic Aromatic Hydrocarbons in the Respirable Fraction of Airborne Particles in the Bangkok Metropolitan Area

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In this study, the characteristics of polycyclic aromatic hydrocarbons (PAHs) in airborne particles equal to or less than  $10 \, \mu m$  (PM10), collected from the Bangkok urban air, were investigated. Sixteen PAHs content in PM10 were simultaneously measured. High molecular weight PAHs (four-, five-, and six-ring) were more abundant in airborne particles (91.7% of total PAHs) than those of low molecular weight PAHs (two-, and three-ring). Further, 71.4% of the total PAHs found in the study sites are potentially carcinogenic PAHs. Benzo[a]pyrene (BaP) was a prominent carcinogenic compound for PAH mixtures found in the area. Correlation analysis revealed that there is a close correlation between the concentration of carcinogenic PAHs and amount of PM10. This is due to their absorptivity property on the surface of the particles. In this study the lifetime lung cancer risk was estimated from the seven carcinogenic PAHs using the toxicity equivalent factor (TEF). The probable number of lung cancer cases in Bangkok Metropolitan was estimated at 27 cases/year. The concentration of carcinogenic PAHs found in Bangkok city in 2002/2003 is comparable to values in many other cities.

Key words — polycyclic aromatic hydrocarbon, risk assessment, airborne particle

#### INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are the principal pollutants from incomplete combustion, and are of special interest due to their toxicity, carcinogenicity, and ubiquitous presence in the environment. Major PAH sources to the atmosphere include motor vehicles, home heating, fossil fuel combustion in energy and industrial processes, biomass burning, and municipal and medical incinerators. Ambient atmospheric PAH concentrations vary greatly with high concentrations most often found in urban areas and low concentrations in rural and remote regions. The association of PAH with the respirable fraction of airborne particle matter is of particular importance in terms of human health ef-

fects. Furthermore, ambient urban air levels of PAH are of concern because they are produced from the combustion of fossil fuels. In this study, an investigation on airborne particle PAH was conducted in metropolitan Bangkok.

Metropolitan Bangkok is the largest city in Thailand with a population of about 10 million. As in other large cities, Bangkok is faced with serious air quality problems. Particles matter equal to or less than 10  $\mu$ m in diameter (PM10) often exceeds the Thai ambient air quality standard of 120  $\mu$ g/m³ over a 24 hr period.<sup>4</sup>) Major sources of particulate matter have been attributed to construction activities and motor vehicle traffic. This severe situation suggests air pollution control technologies currently in use may be ineffective for inhaled particles. Various issues have been investigated recently to mitigate air quality concerns in Bangkok.<sup>4</sup>)

Studies that focus on carcinogenic substances such as PAHs in inhaled particles are still scare in Thailand and other Asean countries, even through

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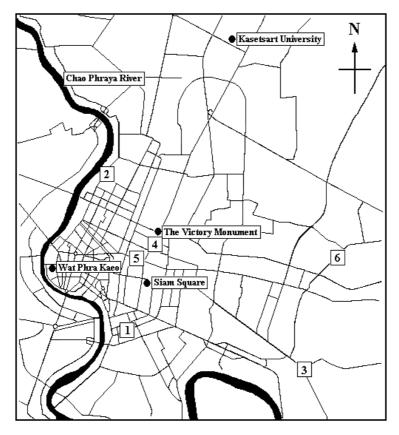


Fig. 1. Bangkok Sampling Sites

statistics on cancer in Thailand have shown that lung cancer is the most common form of cancer in areas where the air pollution levels are considered dangerous.<sup>5</sup> The concentrations and distribution of airborne PAH in a tropical environment are expected to be different from those in a temperate one where a number of studies have been undertaken<sup>6–8)</sup> due to variations in type and scale of emission sources as well as air temperatures. The objective of this study was to investigate the amount and characteristic of PAHs in inhaled dust and its carcinogenic potential.

#### **MATERIALS AND METHODS**

Sample Collection — Ninety six samples were collected from six sampling stations in the Bangkok metropolitan by high volume sampler. To avoid wet removal of airborne PAH, the study was done during the dry season (Nov. 2002 to April 2003) when suspended particle matter pollution in Bangkok is the most serious. Sampling stations were located along the roadsides throughout most of the inner part of the Bangkok metropolitan area as shown in Fig. 1.

Station 1 & 2 and 3 & 4 were selected as representative of low density traffic sites (< 30000 cars/day) and dense traffic sites (30000–90000 cars/day), respectively. While station 5 & 6 were the highest density traffic sites with the traffic volume greater than 90000 cars/day.<sup>9)</sup> The high volume sampler used in this study was equipped with a quartz filter ( $18 \times 23$  cm<sup>2</sup> and pore size 0.3  $\mu$ m) and PM10 inlet. After sampling, exposed filters were folded in half and wrapped in aluminum foil and kept together in a capped plastic bag. Filter samples were dried in a dark desiccator for 24 hr and weighed. Samples were stored at 4°C in the dark until extraction and analysis for PAHs.

Chemical Analysis — Analysis of PAH content of each filter paper was begun by cutting it into small pieces and placing them in a screw cap centrifuge tube (20 ml) with a Teflon liner. Dichloromethane (20 ml) was added to the tube which was then sealed and placed in an ultrasonic bath for 30 min at  $10^{\circ}$ C. The tube was then centrifuged at 3200 rpm for 5 min and 15 ml of the supernatant transferred to a small test tube to which  $50 \mu l$  of dimethylsulfoxide (DMSO) was added to retain PAH. The test tube was

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Response compound	Abbreviation	Ex	Em	MDL
		[nm]	[nm]	$(ng/m^3)$
Naphthalene	Naph	220	330	0.002
Acenaphthene	Ace	220	315	0.001
Fluorene	Flu	249	350	0.002
Phenanthrene	Phen	250	390	0.002
Anthracene	Ant	286	463	0.002
Fluoranthene	Flt	237	386	0.002
Pyrene	Pyr	277	370	0.002
Benz[a]anthracene	BaA	265	399	0.002
Chrysene	Crys	265	399	0.002
Benzo[ $e$ ]pyrene	BeP	235	435	0.001
Benzo[b]fluoranthene	BbF	255	405	0.001
Benzo[k]fluoranthene	BkF	270	399	0.002
Benzo[a]pyrene	BaP	270	399	0.002
Dibenz(a,h)anthacene	DahA	295	415	0.002
Benzo[ghi]perylene	BghiP	295	402	0.002
Indeno[1,2,3-cd]pyrene	Ind	250	500	0.003

**Table 1.** Fluorescence Excitation (Ex) and Emission (Em) Wavelengths and Minimum Detection Limits (MDL) of PAH

then set in a dry thermostat unit at about  $30^{\circ}$ C and the dichloromethane evaporated under a gentle nitrogen stream. The residue in the test tube was dissolved in 1 ml of acetonitrile. Finally, the extracted samples were filtered prior to separation analysis with a  $0.2 \ \mu m$  pore Nylon filter.

The analysis for sixteen PAHs was accomplished by HPLC (Hewlett Packard, 1100 series, CA, U.S.A.) which consisted of a main column (Vydac, C18,  $\phi$  4.6 × 250 mm, CA, U.S.A., 28°C) and fluorescence detector (Hewlett Packard, 1046A) for PAH separation. Excitation, emission wavelength and minimum detection limit of individual PAHs are shown in Table 1.

Ultra pure grade (99.999%) nitrogen gases used in this study were purchased from Thai Industrial Gas Co. Ltd., Bangkok, Thailand. Acetonitrile, dichloromethane, dimethylsulfoxide (Merck, chromatographic grade) were used in analysis. Standard Reference Material (Urban Dust, SRM 1649a) obtained from the National Institute of Standards and Technology (NIST), CA, U.S.A. was run to check the reliability of the analysis. The resulting values were in good agreement with certified values given for this material (see Table 2).

#### **RESULTS AND DISCUSSION**

# Characterization of Polycyclic Aromatic Hydrocarbons in PM10

Field sampling was conducted in the Bangkok metropolitan area during the dry season. Ambient temperatures ranged from 17.5 to 36.6°C and relative humidity ranged from 73 to 85%. During the sampling period, the prevailing wind direction was N/NE (November–December) and S/SW (January–April) with wind speeds ranging from 1.7 to 2.3 m/sec.

The analyses of PAH was performed on samples of PM10 which is recognized to be a more appropriate indicator of adverse health effects than total suspended particles. The gas phase was not investigated because higher-molecular PAHs, like target PAHs concerned in this study, are present in air almost exclusively as particle-bound compounds. <sup>10)</sup>

Among the sixteen PAH analysed as shown in Fig. 2, the lighter PAHs (2–3 rings) that are generally not carcinogenic, such as Naph, Ace, Flu, Phen and Ant were detected at the low concentration in the study sites. This is due to its high volatility especially at the high ambient temperatures during the sampling period (17.5–36.6°C). Therefore, most of these compounds are commonly found in the gas phase. Heavier PAHs such as Benzo[ghi]perylene (BghiP), Benzo[b]fluoranthene (BbF), Benzo[e]pyrene (BeP), Indeno[1,2,3-cd]pyrene (Ind), Benzo-

Table 2	2. Mean C	Concentrations	of PAH	Analyzed in	Reference	Urban Dust	(SRM 1649a)	. Average in 10 Runs
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PAH	mg/kg $\pm$ (S.D.)	Certified Concentration (mg/kg)	% Recovery
Phenanthrene	$3.72 \pm 0.27$	$4.14 \pm 0.37$	89.9
Anthracene	$0.38 \pm 0.09$	$0.43 \pm 0.08$	88.4
Fluoranthene	$5.61\pm0.11$	$6.45\pm0.18$	87.0
Pyrene	$5.04 \pm 0.15$	$5.25\pm0.25$	96.0
Benz[a]anthracene	$1.89 \pm 0.10$	$2.21\pm0.07$	85.5
Chrysene	$2.53\pm0.07$	$3.05\pm0.06$	83.0
Benzo[e]pyrene	$2.86 \pm 0.13$	$3.09 \pm 0.19$	92.6
Benzo[b]fluoranthene	$1.25\pm0.07$	$1.36\pm0.05$	91.9
Benzo[k]fluoranthene	$1.57\pm0.08$	$1.91\pm0.03$	82.2
Benzo[a]pyrene	$2.38 \pm 0.12$	$2.51 \pm 0.09$	90.8
Dibenz $(a,h)$ anthacene	$0.21 \pm 0.08$	$0.29 \pm 0.02$	72.4
Benzo[ghi]perylene	$3.55\pm0.73$	$4.01 \pm 0.91$	88.5
Indeno[1,2,3-cd]pyrene	$2.87 \pm 0.53$	$3.18\pm0.72$	90.3

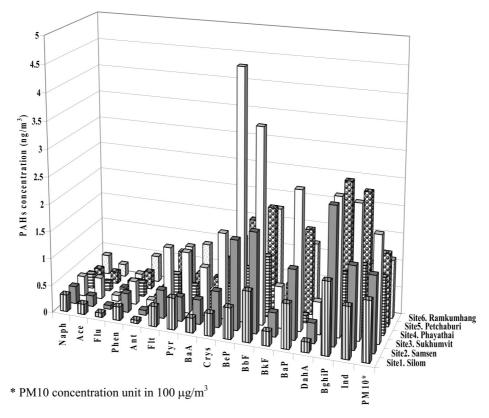


Fig. 2. Concentration of PAH on PM10 in Ambient Air Samples of the Bangkok Metropolitan Area

[a]pyrene (BaP), Chrysene (Crys), Fluoranthene (Flt), Pyrene (Pyr), Benz[a]anthracene (BaA), Benzo[k]fluoranthene (BkF), and Dibenz(a,h)-anthacene (DahA) that mostly sorb on small inhalable size particles were found in higher concentration on airborne particles than lighter PAHs. This is in accordance with previous studies of Kim et al., 11) also in the Bangkok but in different region

than the present study. These sorbed compounds are deposited in the human respiratory tract, increasing potential health effects.

All of the PAHs tended to be more concentrated at site 3 (Sukhumvit Road) than other sites consistent with the dense traffic conditions. Road traffic was included both gasoline and diesel fueled vehicles from a local bus terminal. Moreover, canopy over

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Table 3. Mean (± S.D.) PAH Concentrations and Their Contribution to the Total PAH in PM10 from Bangkok Metropolitan Area

PAHs Compound	Relative Molecular	Rings	Carcinogenic	Mean $\pm$ S.D.	% of Total PAH
	Weight		$potency^{a)}$	$(ng/m^3)$	
Low molecular weight:					
Naphthalene	128.18	2	D	$0.30 \pm 0.11$	2.4
Acenaphthene	154.20	3	_	$0.22 \pm 0.10$	1.8
Fluorene	166.23	3	D	$0.09\pm0.03$	0.7
Phenanthrene	178.24	3	D	$0.35 \pm 0.11$	2.8
Anthracene	178.24	3	D	$0.08 \pm 0.03$	0.7
			Sum	$1.05\pm0.07$	8.3
High molecular weight:					
Fluoranthene	202.26	4	D	$0.68 \pm 0.34$	5.4
Pyrene	202.26	4	D	$0.58 \pm 0.33$	4.6
Benz[a]anthracene	228.3	4	B2	$0.47\pm0.25$	3.7
Chrysene	228.3	4	B2	$0.77\pm0.43$	6.1
Benzo[e]pyrene	252.32	5	_	$1.75\pm1.37$	13.9
Benzo[b]fluoranthene	252.32	5	B2	$1.76 \pm 0.92$	14.0
Benzo[k]fluoranthene	252.32	5	B2	$0.44 \pm 0.19$	3.5
Benzo[a]pyrene	252.32	5	B2	$1.30 \pm 0.72$	10.3
Dibenz(a,h)anthacene	278.35	5	B2	$0.33 \pm 0.18$	2.6
Benzo[ghi]perylene	276.34	6	D	$1.92 \pm 0.90$	15.3
Indeno[1,2,3-cd]pyrene	276.34	6	B2	$1.54 \pm 0.86$	12.3
			Sum	$11.55\pm0.59$	91.7
Total				$12.59 \pm 0.94$	

a) US EPA carcinogenic classification.<sup>16</sup> B2: Probable human carcinogen: D: Not classifiable as to human carcinogenicity.

Sukhumvit road constructed to accommodate Bangkok's Mass Transit System, restricts air ventilation and PAH distribution from vehicle emissions. We also detected high concentrations of BeP, Pyr and Crys that are molecular markers for diesel vehicle emission at this study sites. Ind and BghiP, both considered as good markers for gasoline vehicle emission<sup>12)</sup> were found in abundance at most study sites. This suggests that the PAH products found along Sukhumvit road were a result of the direct emission of fuel combustion from gasoline and diesel powered vehicles.

To assess PAH homolog distribution for each collected sample, we further classified sixteen PAHs into two categories: low molecular weight (LM-PAHs, containing two- to three-ringed PAHs), and high molecular weight (HM-PAHs, containing four-to six-ringed PAHs) (Table 3). This study revealed that low molecular weight PAHs represented only a small portion (8.3%) of the total in airborne particles with most PAHs being of high molecular weight (91.7%).

Furthermore, information from this study indi-

cated that five ringed PAHs were the predominant homolog followed by six, four, three and two ringed forms, respectively (Fig. 3). Unfortunately, the predominant five ringed PAHs are regarded as having the highest carcinogenic potency. Similar patterns of distribution as shown in Fig. 3 were found at every study site in the Bangkok metropolitan area. This suggests the PAH residues found at the study sites may have all originated from the combustion of gasoline and diesel fuels.

Relatively high concentrations of BeP and BghiP on airborne particles collected from the outer and inner part of Bangkok metropolitan are reported by many investigators. <sup>11,13,14</sup> The PAH profiles obtained from those investigators are in accord with the present study. The results of these studies also indicated that BbF, in addition to BghiP, BkF, Flt and Pyr are mostly contributed from diesel powered vehicles.

Table 4 shows that total and carcinogenic PAHs are strongly correlated with the mass of PM10 but weakly correlated with area traffic volume. This is probably due to the sorptive property of the airborne

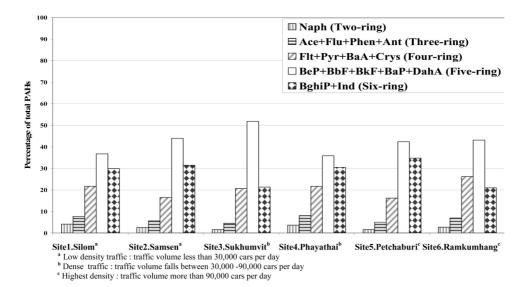


Fig. 3. Distribution of PAHs at Each Sampling Site, as a Percentage of the Total, in the Bangkok Metropolitan Area on the Basis of the Number of Aromatic Rings

**Table 4.** Multiple Correlation between Mass of PM10, Carcinogenic PAHs (cPAHs), Total PAHs (ΣPAH), and Traffic Volume in the Bangkok Metropolitan Area

	cPAHs	SPAH	Traffic volume
PM10	0.78	0.79	0.48
Traffic volume	0.41	0.74	
ΣΡΑΗ	0.98		

Significant level of correlation was p < 0.01, n = 96.

particles that increases directly with airborne particle concentration.

Moreover, considering that several PAH compounds are known as human carcinogens, we also classified them into two categories depending on their carcinogenicity, such as carcinogenic (BaA, Crys, BbF, BkF, BaP, DahA and Ind) and non-carcinogenic PAHs (Naph, Ace, Flu, Phen, Ant, Flt, Pyr, BeP, and BghiP). This indicated that 71.4% of the total PAHs found in the study sites are carcinogenic PAHs whereas 28.6% are non-carcinogenic PAHs (Fig. 4.).

### **Risk Assessment of Carcinogenic PAH**

Several PAH compounds are known as human carcinogens such as BaA, Crys, BbF, BkF, BaP, DahA and Ind. The information obtained from the present study indicated that carcinogens like BbF (0.92–3.56 ng/m³), BaP (0.73–2.54 ng/m³), Ind (0.93–2.44 ng/m³), and Crys (0.41–1.58 ng/m³) are the most predominant compounds in the Bangkok metropolitan area as shown in Figs. 5 and 6.

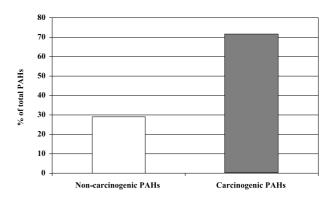


Fig. 4. Non-Carcinogenic and Carcinogenic PAHs as a Percentage of the Total at the Study Sites of the Bangkok Metropolitan Area

However, health risk assessment of carcinogenic PAH can not be related only to overall concentration. Rather, each PAH has a different carcinogenic potential. Health risk assessment associated with inhalatory PAH uptake is often estimated on the basis of the BaP concentrations in air. The development and the establishment of a toxicity equivalency factor (TEF) is used in the assessment of mixtures containing PAHs. Therefore, the carcinogenic potencies of individual carcinogenic PAHs have to be considered by multiplying their concentration with the appropriate TEF. Concentrations of individual PAHs were calculated in terms of benzo[a]pyrene equivalents (BaP<sub>eq</sub>) and are presented in Table 5.

Based on the calculated BaP<sub>eq</sub> values for individual carcinogenic PAHs, we can also calculate

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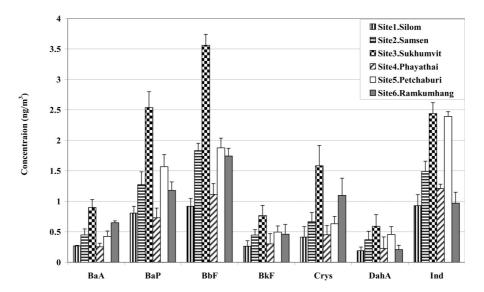


Fig. 5. Carcinogenic PAH Concentrations at the Sampling Sites in the Bangkok Metropolitan Area

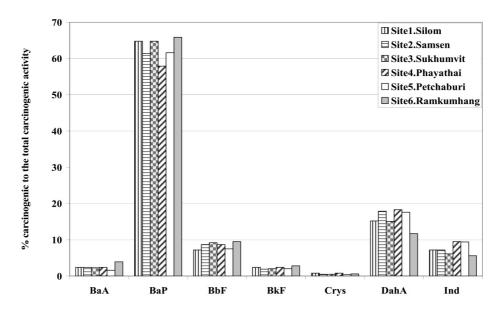


Fig. 6. Relative Contribution in Carcinogenic Activity of Individual PAHs at Sampling Sites, Expressed as a Percentage of Total Carcinogenic PAH

percent contribution in carcinogenic activity of the individual PAHs for each sampling sites as shown in Fig. 6. The contribution of the carcinogenic potency of BaP alone is in the range of 57.9–65.9% of the total carcinogenic activity. BaP is the highest carcinogenic contributor in every study site follow by DahA, BbF, and Ind respectively. This result underlines and confirms the importance of BaP as a prominent carcinogenic compound of PAH mixtures in air.

Regarding the lung cancer risk via the inhala-

tion route, WHO  $(1987)^{15}$  suggested the unit risk of  $8.7 \times 10^{-5}$  (ng/m³)<sup>-1</sup> for the lifetime 70 years PAHs exposure, assuming one exposed to the averaged level of one unit BaP concentration  $(1 \text{ ng/m}^3)$ . The above unit risk was proposed to estimate the lung cancer risk caused by a lifetime exposure. Therefore, it has been adopted by a recent study for assessing lung cancer risks by adults exposed to ambient atmospheric PAHs. In this study, the sum of BaP<sub>eq</sub> levels at each study site was used to estimate the corresponding lifetime lung cancer risks for

PAHs	TEFs <sup>16)</sup>	Site 1. Silom	Site 2. Samsen	Site 3. Sukhumvit	Site 4. Phayathai	Site 5. Petchaburi	Site 6. Ramkumhang	Average
		$BaP_{eq}$	$BaP_{eq}$	$BaP_{eq}$	$BaP_{eq}$	$BaP_{eq}$	$BaP_{eq}$	$BaP_{eq}$
		$(ng/m^3)$	$(ng/m^3)$	$(ng/m^3)$	$(ng/m^3)$	$(ng/m^3)$	$(ng/m^3)$	$(ng/m^3)$
BaA	0.10	0.03	0.05	0.09	0.03	0.04	0.07	0.05
Crys	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01
BbF	0.10	0.09	0.18	0.36	0.11	0.19	0.17	0.18
BkF	0.10	0.03	0.04	0.08	0.03	0.05	0.05	0.05
BaP	1.00	0.81	1.27	2.54	0.73	1.57	1.18	1.35
DahA	1.00	0.19	0.37	0.59	0.23	0.45	0.21	0.34
Ind	0.10	0.09	0.15	0.24	0.12	0.24	0.10	0.16
Sum		1.25	2.07	3.92	1.26	2.55	1.79	2.14
Annual number of cancer case (persons per 10 <sup>6</sup> )		16	26	49	16	32	23	27

Table 5. Airborne PAH Concentrations (in BaPeq) at Sampling Sites in the Bangkok Metropolitan Area

**Table 6.** Airborne Carcinogenic PAH Concentrations (in BaP<sub>eq</sub>, ng/m<sup>3</sup>) in Bangkok City During 2002/2003 (present study) Compared with Other Cities and a Previous Study in 1996/1997

PAHs	Nagasaki <sup>8)</sup>	Copenhagen <sup>6)</sup>	Rome <sup>7)</sup>	Bangkok <sup>13)</sup>	Bangkok <sup>a)</sup>
	1997/1998	1992	1996/1997	1996/1997	2002/2003
BaA	0.05	0.41	0.71	0.36	0.05
Chrys	0.02	0.08	_	0.07	0.01
BbF	0.13	_	_	0.85	0.18
BkF	0.06	0.93	0.26	0.39	0.05
BaP	0.71	4.4	1.22	4.11	1.35
DahA	0.25	_	0.16	_	0.34
Ind	0.07	0.45	0.13	1.05	0.16
Total	1.29	6.27	2.48	6.83	2.13

a) Present study

Bangkok residents. The annual number of lung cancer case (persons per million) in the Bangkok metropolitan area (population = 10 millions) can be estimated by the following equation.<sup>16)</sup>

Annual number of lung cancer case (persons per million)

$$= \frac{\text{Unit Risk} \times \text{Sum BaP}_{\text{eq}} \times \text{Population (million of residents)}}{\text{Life expectancy}}$$

The findings of this study indicate that the annual number of lung cancer cases among Bangkok residents attributable to these carcinogenic PAH compounds in 2002/2003 was estimated at 27 persons per million (Table 5). This is about three times lower than the previous estimate for Bangkok residents of 78 persons/million made in 1996/1997 by Ruchirawat *et al.*,<sup>13)</sup> and may indicate a significant

improvement of air quality in the metropolitan area. In 1996/1997 Bangkok had a severe problem with the diesel fuel used by motor vehicles. Recent adoption of the national industrial standard for the diesel engine has resulted in improved efficiency of combustion motors and may have caused a decline in PAH emissions. However, with an increase in the number of vehicles in the Bangkok metropolitan area, PAH abatement measures are still needed.

Table 6 compares airborne carcinogenic PAH concentrations (in BaP<sub>eq</sub>) between Bangkok city and other cities. The concentration of carcinogenic PAHs found in Bangkok city in 2002/2003 is comparable to concentrations in Nagasaki and Rome and is considerably lower than those reported in Copenhagen in 1992. This likely reflects, at least in part, the influence of temperature on volatilization of PAHs. The temperature during the sampling period in

Copenhagen was only 2.7°C while that in Bangkok was about 27°C. Overall, the aromatic PAHs in tropical countries can be expected to be volatilized faster than those in temperate environments.

In conclusion, this study focused on the characterization and carcinogenic risk assessment posed by PAH exposure via inhalation of the respirable fraction of airborne particles in the Bangkok metropolitan area. BghiP, BbF, BeP, Ind, and BaP were the predominant PAHs in this area followed by Crys, Flt, Pyr, BaA, Phen, BkF, DahA, Naph, Ace, Flu and Ant, respectively. The results of the study revealed that low molecular weight PAHs represented only a small portion of the total with most being of high molecular weight. Our results indicated that 71.4% of the total PAHs found in the study sites have carcinogenic potential in humans. Statistical analysis showed that total PAHs and carcinogenic PAHs are strongly correlated with the mass of PM10 but weakly correlated with the traffic volume of the area. Results of the finding underlines and confirms the importance of BaP as a prominent carcinogenic compound for PAH mixtures in air of the study area. The annual number of lung cancer case of Bangkok residents caused by these carcinogenic PAH compounds was estimated as 27 persons per million. However, the concentration of carcinogenic PAHs found in Bangkok city in 2002/2003 is comparable to those previous studies in Nagasaki and Rome.

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