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Spatial and temporal variations of ambient PM_{10}-bound polycyclic aromatic hydrocarbons in Chiang Mai and Lamphun Provinces, Thailand

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ABSTRACT

Respirable airborne particulate matters (PM_{10}) were collected using high volume air samplers for 24 h every three days from four sampling stations in Chiang Mai and Lamphun Provinces, Thailand. The maximum PM_{10} concentration for each sampling station ranged from 140 to 182 $\mu g/m^3$. PM_{10}-bound polycyclic aromatic hydrocarbons (PAHs) were extracted from quartz fiber filters and analysed by gas chromatography-mass spectrometry (GC-MS). Mean concentration of total PAHs was in a range from 1.7 to 12.2 ng/m^3. Ratio of non-carcinogenic and carcinogenic PAHs found was approximately 1:3 or 1:4 depending on sampling sites. Concentrations of most of carcinogenic PAHs were found to be positively correlated to PM_{10} concentrations. Seasonal variation of PM_{10}-bound PAHs was obviously seen in all sites. The highest concentration was found in dry season (December–March). Spatial variation was observed in one sampling site, where PAHs content was significantly higher than other sites. Apart from traffic congestion, this was probably due to specific local activities i.e. biomass burning.

Keywords: Air pollution; Biomass burning; GC-MS; PAHs; PM_{10}

1. Introduction

Air pollution is now mostly seen in developing countries with mega-cities. Atmospheric aerosol particles originate from a wide variety of natural and anthropogenic sources. Primary particles are directly emitted from sources such as biomass burning, incomplete combustion of fossil fuels and traffic-related suspension of road, soil, dust, sea salt and biological materials. Secondary particles are formed by gas-to-particle conversion in the atmosphere [1]. Concentration, composition and size distribution of atmospheric aerosol particles are temporally and spatially highly variable. The effects of aerosols on the atmosphere, climate and public health are among the central topics in current environmental research. An airborne particulate matter contains several of inorganic and organic species, many of which can adversely affect human health [1–3]. Of these constituents, the classes of polycyclic aromatic hydrocarbons (PAHs) are widespread environmental pollutants which are formed during incomplete combustion process of organic material emitted from a large variety of industrial processes, motor vehicles, domestic waste burning and regenerated burning in agriculture [4]. They show adverse effects on health (carcinogenic and/or mutagenic activity) and the ecosystems. Therefore, the air polluted by airborne PAHs is of significant concern.

In attempt to identify major emission sources responsible for adverse health effects, several comprehensive surveys of atmospheric contaminants from a variety

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of sources have been performed worldwide [5–8]. In Thailand, only some studies have been conducted to determine atmospheric PAHs in big cities such as Bangkok and Chiang Mai. In particular, the northern part of Thailand has been faced with air pollution in the dry season almost every year. It is important to know the levels of air pollutants, including PAHs and their variation, throughout the year. Moreover, sources of air pollutants are also needed to be identified. This study aims to assess spatial and temporal variation of PM_{10}-bound PAHs in different areas of Chiang Mai and Lamphun Basin and to identify their possible sources based on the diagnostic ratio. The results are expected to be used for the regional environmental plan focusing on air quality.

2. Methods

2.1. PM_{10} sampling

Three study sites were in Chiang Mai Province: Yuparaj Witayalai School (YP), Municipality Hospital (HP), Saraphee District (SP) and one site was in Muang District, Lamphun Province (LP). Map of study is illustrated in Fig. 1. Twenty-four hours PM_{10} samples were collected on quartz fiber filters every three days at each sampling site using a High Volume PM_{10} Air Sampler (Wedding & Associates Inc., USA) at the flow rate of 1,130 L/min. Sampling had been done for one year during June 2005–June 2006 to cover three seasonal periods, which were wet season (June–September), dry season (December–March) and transition periods (October–November and April–May). PM_{10} samples were weighed using a five-digit scale balance (Mettler Toledo (USA) AG285) positioned in a temperature and humidity controlled cabinet (DE-300). More detail of the PM_{10} Sampling can be found in Ref. [9].

2.2. Validation of analysis method for PM_{10} bound PAHs by GC-MS

2.2.1. Determination of GC-MS performance

GC-MS (Hewlett Packard, USA) was used for investigation of PAHs in this study. One micro liter of

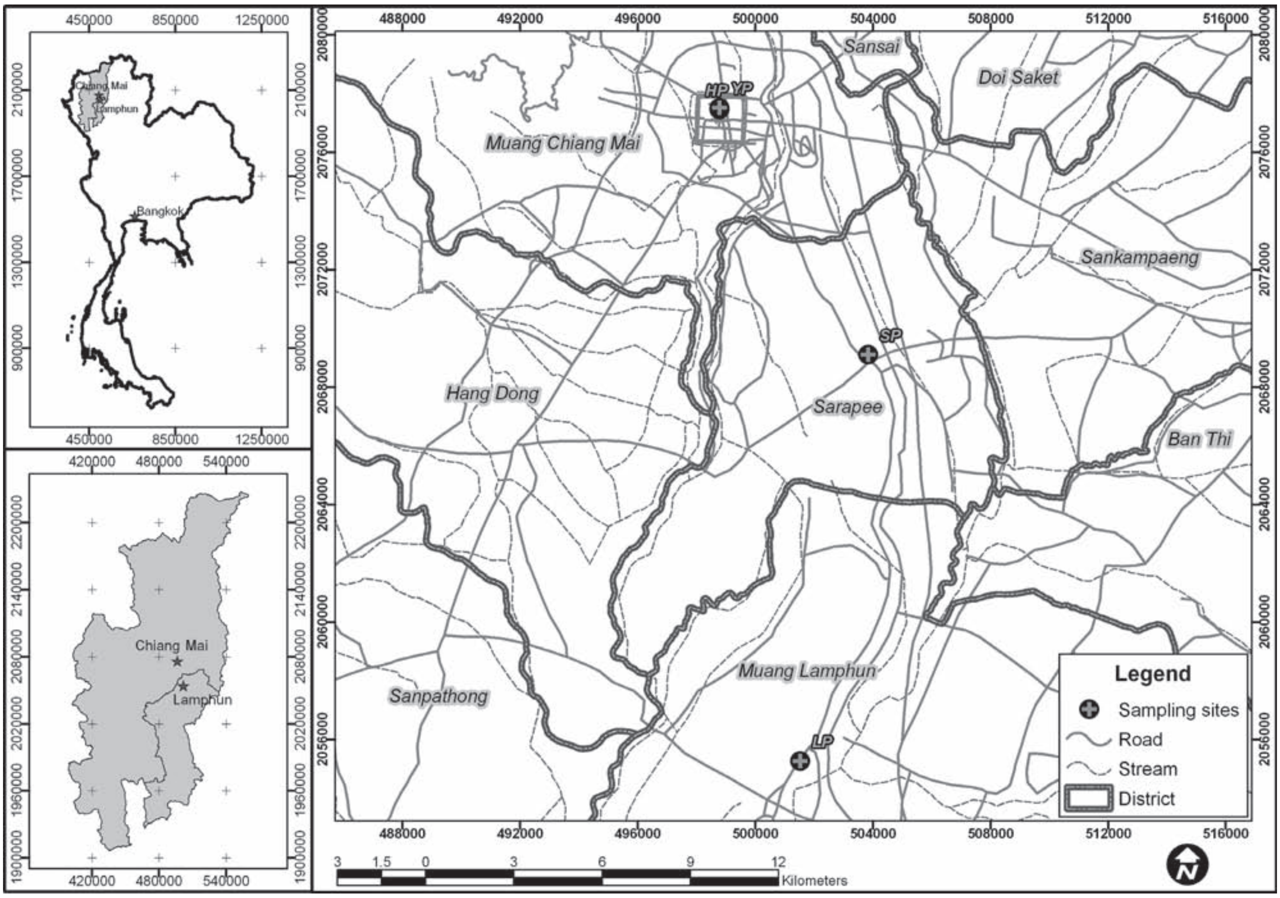

Fig. 1. Map of sampling sites.

a mixed solution of 16-PAH standards (2,000 μg/ml in methylenechloride, Restek, USA) and internal standards (D_{10}-ACE, D_{10}-PHE and D_{12}-PYR from Supelco, USA) was injected in splitless mode onto an HP-5MS GC capillary column of 30 m length, 0.25 mm diameter and 0.25 μm thickness internally coated with 5% phenyl methyl polysiloxane stationary phase. Standard solutions of the 16 PAHs with concentration ranging from 0.005 to 0.50 μg/ml added with 0.1 μg/ml internal standards were injected onto the same GC capillary column of the GC-MS employed under an optimum condition in order to obtain a suitable calibration curve in which peak area ratios were plotted against concentrations. The GC condition was as follows. The injector temperature was 275°C. The GC oven was programmed with an initial temperature of 70°C, held for 2 min, then increased to 290°C at 8°C/min. The MS was operated in SIM mode. Characteristic ions of selected PAHs were used for quantification. Quantification was based on the peak area of the PAH standard relative to the deuterated internal standard closest in molecular weight to the analyte.

2.2.2. Optimization of ultrasonic extraction

Optimization for the extraction procedure of PAHs in PM_{10} samples was conducted using ultrasonication to obtain an appropriate extraction volume of acetonitrile (HPLC grade, Lab-Scan Analytical Science) and evaporation conditions. The quartz filter was cut into small pieces using a pair of stainless scissors and the cut filter was put into a 60-ml amber-bottle wrapped with aluminium foil and mounted by paraffin film. The 0.3 μg/ml of mixed 16-PAH standards was spiked onto the paper, covered and left for 10 min. The spiked samples were extracted in 35 ml acetonitrile by ultrasonicator (T710DH, Elma, Germany) at 100% ultrasound power for 30 min under controlled temperature (approximately 20°C). The solution was filtered through a 0.45 μm nylon filter and evaporated by low-pressure evaporator at 30°C until it became nearly dry. The pellet was re-extracted in 30 ml acetronitrile by repeating the previous step. The solution was transferred into a 1 ml volumetric flask and a mixed solution of internal standards was added with the final volume adjusted to 1 ml with acetronitrile. The solution was then analyzed for 16 PAHs by GC-MS. Percent recoveries of individual PAHs were obtained.

2.2.3. Quality control of the PAH analysis

Standard reference material (SRM) 1649a (urban dust) from the National Institute of Standards and Technology, USA, which is an atmospheric particulate material collected in an urban area, was used in evaluating analytical methods for the determination of selected PAHs in atmospheric particulate material and similar matrices. Total 12 PAHs (Table 1) of the SRM were matched with the 16 EPA-PAHs in this study. Approximately 70 mg urban dust was accurately weighed to six decimal points using an appropriate analytical balance.

Table 1
Detection limits (DL) of GC-MS for PAHs and recoveries of PAHs from extraction in comparison with SRM (urban dust) values.

Compound	Number of ring	DL (ng/m^3)	Recovery ± RSD (%) of spiked samples	Mean analyzed concentration (mg/kg)	Concentration of SRM (mg/kg)
Naphthalene (NAP)	2	0.006	64 ± 5	0.38	–
Acenaphthylene (ACY)	3	0.005	76 ± 6	0.18	–
Acenaphthene (ACE)	3	0.006	66 ± 6	0.05	–
Fluorene (FLU)	3	0.004	80 ± 7	0.07	–
Phenanthrene (PHE)	3	0.006	60 ± 1	2.85	4.14 ± 0.37
Anthracene (ANT)	3	0.004	77 ± 2	0.53	0.432 ± 0.082
Fluoranthene (FLA)	4	0.003	76 ± 2	5.13	6.45 ± 0.18
Pyrene (PYR)	4	0.003	72 ± 1	4.21	5.29 ± 0.25
Benzo(a)anthracene[a] (BaA)	4	0.007	120 ± 4	2.29	2.208 ± 0.073
Chrysene[a] (CHR)	4	0.005	78 ± 2	3.16	3.049 ± 0.060
Benzo(b)fluoranthene[a] (BbF)	5	0.006	112 ± 7	6.46	6.45 ± 0.64
Benzo(k)fluoranthene[a] (BkF)	5	0.007	84 ± 4	1.79	1.913 ± 0.031
Benzo(a)pyrene[a] (BaP)	5	0.005	81 ± 12	1.88	2.509 ± 0.087
Indeno(1,2,3-cd)pyrene[a] (IND)	5	0.004	73 ± 14	2.22	3.18 ± 0.72
Dibenzo(a,h)anthracene[a] (DBA)	6	0.005	77 ± 16	0.31	0.288 ± 0.023
Benzo(g,h,i)perylene (BPER)	6	0.007	58 ± 15	2.41	4.01 ± 0.91

[a] Carcinogen suspects.

The collected dust was extracted by ultrasonication and the resultant solution volume was reduced by evaporation under the same conditions described above. Following the ultrasonication extraction, a mixture of two chosen internal standards was spiked to the extracted standards. The solution was then analysed by GC-MS. The concentrations obtained were calculated for recoveries of the PAHs.

2.3. Analysis method of 16 EPA-PAHs in PM_{10} samples by GC-MS

After sampling, the weighed quartz filter was cut into small pieces by a pair of stainless scissors and put into a 60-ml amber-bottle wrapped with aluminium foil and mounted with paraffin film. The samples were extracted using the same conditions as the standards described above. Each of the sample solutions was then analyzed for 16 PAHs by GC-MS.

3. Results and discussions

3.1. GC-MS performance for analysis of PAHs

Detection limit (DL) of GC-MS for an individual PAH was calculated as three times of the standard deviation of blank concentration [10]. Detection limits for 16-PAHs obtained were ranged from 0.003 to 0.007 ng/m^3 (Table 1). Recoveries of PAHs from the extraction of spiked filters ranged from 58% to 120% (Table 1). Of all the 16 PAHs investigated, 12 PAHs were found to yield the recovery higher than 70%.

3.2. Analysis method validation

The measured concentrations of the 12 PAHs were compared with the actual concentrations available in the SRM (Table 1). The results revealed that 5 of the PAHs were measured with a low percentage error (0.2%–7.6%), while four of them had an error of 20%–25%. The rest was in error by more than 30%.

3.3. PM_{10} concentrations

Spatial and temporal variation of mean PM_{10} concentrations is shown in Table 2. Concentration of PM_{10} collected in the dry season (December–March) was significantly higher than other seasons, while the lowest concentration was found in the rainy season (June–September). Average PM_{10} concentration in each sampling site was very much similar. However, the mean concentration of the Sarapee (SP) site was higher than those of other sites. This is probably due to local activities, i.e. traffic density and open burning.

3.4. Analysis of 16 PAHs in PM_{10} samples by GC-MS

Figure 2 illustrates spatial and temporal variation of PAHs concentrations. The highest concentrations of an individual and total PAHs as well as carcinogenic PAHs were found in the dry season and higher than those in other seasons.

Seasonal variation of PM_{10}-bound PAHs can be observed in Table 3. Mean concentration of various groups of PAHs including those with 2–3 rings, 4–6 rings, carcinogenic and non-carcinogenic PAHs and sum of 16 PAHs were statistically tested based on the season basis. The results revealed that all PAH groups presented a similar pattern. The highest concentration was found in the dry season (December–March) and significantly higher ($p < 0.05$) than the other seasons, in which PAHs concentrations were not significantly different. In terms of sampling site (Table 4), high molecular weight (4–6 rings) and carcinogenic PAHs were significantly higher at the SP site than other sites.

Table 5 illustrates the ratios of non-carcinogenic and carcinogenic PAHs found in PM_{10} samples from each sampling site, which were 1.0:3.7 (LP), 1.0:3.8 (SP), 1.0:2.7 (YP) and 1.0:2.8 (HP). SP and LP ratios revealed that carcinogenic PAHs were obviously higher than non-carcinogenic ones. The highest carcinogenic PAHs content was found at the SP site and was significantly higher than other sites (Table 4). The carcinogenic ratios

Table 2
Mean concentrations (μg/m^3) ± SD of PM_{10} for 4 seasons and 4 sampling sites [11].

Season^{a,b}	PM_{10} concentration	Sampling site^{a,b}	PM_{10} concentration
December–March (*n* = 160)	73.64 ± 25.34$^{a/7}$	LP (*n* = 127)	46.53 ± 29.30$^{a/1}$
April–May (*n* = 84)	***40.44 ± 25.33***$^{a/2}$	SP (*n* = 127)	**55.94 ± 36.64**$^{a/2}$
June–September (*n* = 184)	*33.17 ± 21.39*$^{a/1}$	HP (*n* = 127)	44.08 ± 20.60$^{a/3}$
October–November (*n* = 80)	**47.19 ± 12.36**$^{a/2}$	YP (*n* = 127)	**50.30 ± 23.26**$^{a/6}$

$^{a/n}$Number of non detected or incomplete data.
^{b}Groups of number in *italic* and/or **bold** are not significant different (p > 0.05).

of all sites found in this study were higher than those found in roadsides of the inner part of Bangkok (1.0:2.5) during November 2002–April 2003 [12]. This reveals that our study sites presented higher concentrations of carcinogenic PAHs but lower PM$_{10}$ contents in comparison to the Bangkok area. This fact should be noted and brought into the attention of relevant authorities for subsequent air quality management.

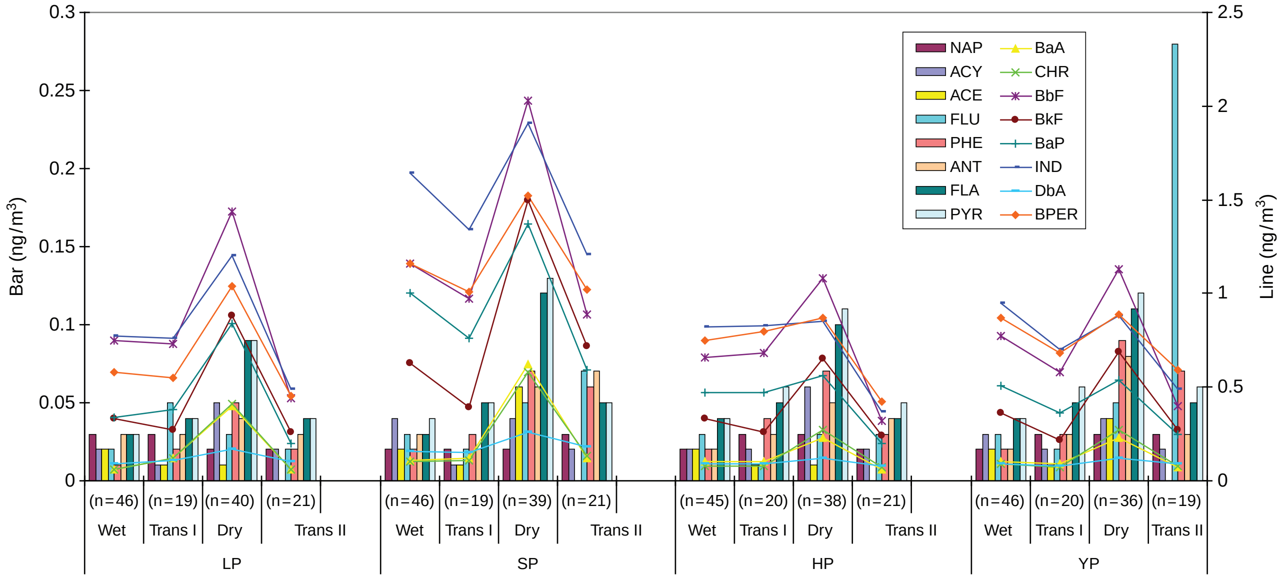

Fig. 2. Spatial and temporal variation of PAHs concentrations.

Table 3
Mean concentrations (ng/m^3) ± SD of PM$_{10}$-bound PAHs in the four seasons.

PAHs^{a,b}	December–March (n = 160)	April–May (n = 84)	June–September (n = 184)	October–November (n = 80)
2–3 rings	**0.15 ± 0.13**$^{a/1}$	0.10 ± 0.16$^{a/2}$	0.07 ± 0.04$^{a/5}$	0.08 ± 0.06$^{a/4}$
4–6 rings	**6.58 ± 3.20**$^{a/1}$	$\underline{2.76 \pm 2.05}^{a/2}$	*4.06 ± 2.96*$^{a/1}$	$\underline{\mathit{3.59 \pm 1.69}}^{a/2}$
Carcinogenic^{c}	**5.29 ± 2.72**$^{a/1}$	$\underline{2.05 \pm 1.65}^{a/2}$	*3.15 ± 2.42*$^{a/1}$	$\underline{\mathit{2.73 \pm 1.41}}^{a/2}$
Non carcinogenic	**0.36 ± 0.20**$^{a/1}$	0.20 ± 0.18$^{a/2}$	0.13 ± 0.06$^{a/1}$	0.18 ± 0.09$^{a/2}$
Total 16 PAHs	**6.72 ± 3.20**$^{a/1}$	$\underline{2.87 \pm 2.08}^{a/2}$	*4.13 ± 2.97*$^{a/1}$	$\underline{\mathit{3.67 \pm 1.71}}^{a/2}$

$^{a/n}$Number of non detected or incomplete data.
^{b}Groups of number in *italic*, underline and/or **bold** are not significant different (p > 0.05).
^{c}Carcinogenic PAHs are BaA, CHR, BbF, BkF, BaP, DBA and IND.

Table 4
Mean concentrations (ng/m^3) ± SD of PM$_{10}$-bound PAHs from four sampling sites.

PAHs^{a,b}	LP (n = 127)	SP (n = 127)	HP (n = 127)	YP (n = 127)
2–3 rings	0.07 ± 0.06$^{a/5}$	0.10 ± 0.09$^{a/3}$	0.10 ± 0.08$^{a/2}$	**0.14 ± 0.17**$^{a/2}$
4–6 rings	3.97 ± 2.53$^{a/1}$	**6.93 ± 4.16**$^{a/2}$	3.58 ± 2.03$^{a/1}$	3.82 ± 1.81$^{a/2}$
Carcinogenic^{c}	3.18 ± 2.12$^{a/1}$	**5.57 ± 3.51**$^{a/2}$	2.71 ± 1.61$^{a/1}$	2.88 ± 1.46$^{a/2}$
Non carcinogenic	0.17 ± 0.11$^{a/1}$	***0.23 ± 0.16***$^{a/2}$	**0.22 ± 0.15**$^{a/1}$	*0.28 ± 0.22*$^{a/2}$
Total 16 PAHs	4.04 ± 2.55$^{a/1}$	**7.03 ± 4.18**$^{a/2}$	3.68 ± 2.06$^{a/1}$	3.96 ± 1.84$^{a/2}$

$^{a/n}$Number of non detected or incomplete data.
^{b}Groups of number in *italic* and/or **bold** are not significant different (p > 0.05).
^{c}Carcinogenic PAHs are BaA, CHR, BbF, BkF, BaP, DBA and IND.

Table 5
Ratios of non-carcinogenic to carcinogenic PM_{10}-bound PAHs.

Sampling site	Percentage of non–carcinogenic PAHs	Percentage of Carcinogenic PAHs	Non-carcinogenic: carcinogenic PAHs
LP	21.3	78.7	1.0:3.7
SP	20.8	79.2	1.0:3.8
HP	26.4	73.6	1.0:2.8
YP	27.3	72.7	1.0:2.7
Bangkok [12]	28.6	71.4	1.0:2.5

Table 6
Pearson correlation of PM_{10} and PAHs.

Season	LP	SP	HP	YP
Wet (June–September)	0.530	0.549	0.703	0.595
Dry (December–March)	0.704	0.572	0.601	0.590
Transition period (April–May, October–November)	0.257	0.650	0.690	0.365
All season	0.731	0.686	0.659	0.618

3.5. Correlation of PM_{10} and PAHs

Correlations between PM_{10} contents and total PAH concentrations for each sampling site in each season are shown in Table 6 and Fig. 3. Correlations between PM_{10} contents and PAH concentrations for all seasons were found to be in the range 0.618–0.731. The highest correlation was found in the LP site, while the lowest one was in the YP site. The correlations for all sites were relatively high in the dry low in the wet season, respectively. The correlations for the SP and HP sites were high throughout the year; they were also high in transition periods and obviously higher than those for the LP and YP sites. This is probably due to local sources, which generated PM_{10}-bound PAHs into the atmosphere during the sampling time period. In terms of PAH source, both SP and HP sampling sites are surrounded by markets with high traffic volume and human activities. The difference between these two places is that the SP site is a place where small scale factories are situated. The fuels used in such factories could have contributed to the level of pollutants emitted to the air.

In comparison to previous studies, the correlation found in this study is almost the same as that reported by Ref. [13] in which the mass of PM_{10} collected by a Minivol air sampler in traffic areas was found to be slightly correlated ($R^2 = 0.45$) with the total PAH concentration of the solution obtained from extraction of the collected particulate. The afore-mentioned correlation was higher than the value in the study by Ref. [14], in which airborne particulate samples were collected from commercial and residential sites of Chiang Mai from July to December 1989 and their results revealed that there was not a large differnce in PAHs concentrations between different sampling sites. However, the airborne particulates and large amounts of rough particulates encountered in their study prompted them to suggest wide spread air pollution over the city.

3.6. The BaP concentration and benzo(a)pyrene-equivalent carcinogenic power

The limit value of BaP recommended by WHO is 1 ng/m^3 due to its high carcinogenic property. Other PAHs such as BaA, BbF, BkF and DBA also have carcinogenic potential. To combine these potential carcinogenic compounds, the benzo(a)pyrene-equivalent carcinogenic power (BaPE) is calculated. The BaPE is an index that has been introduced for better denoting aerosol carcinogenicity related to the whole PAH fraction instead of the BaP [5]. The BaPE was calculated in this study using the following equation.

$$\mathrm{BaPE} = (\mathrm{BaA} \times 0.06) + ((\mathrm{BbF} + \mathrm{BkF}) \times 0.07) + \mathrm{BaP} + (\mathrm{DBA} \times 0.6) + (\mathrm{IND} \times 0.08) \tag{1}$$

Values of monthly mean BaPE and PM_{10} concentrations for each sampling site are illustrated in Fig. 4. It can be obviously seen that the BaPE values of the SP site are the highest, while those of the other sites are approximately two times lower. It can therefore be concluded that the air quality of the SP site was worse than the other sampling sites.

Patterns of the BaPE values from June 2005 to June 2006 were found to be similar for all sites. Fig. 4 shows two peaks of BaPE values. The first peak was obtained in the rainy season (August 2005) while the second one in the dry season (January–March 2006). Based on the fact that traffic is the main PAHs source in the urban area, it presents almost the same level of pollutants to the air

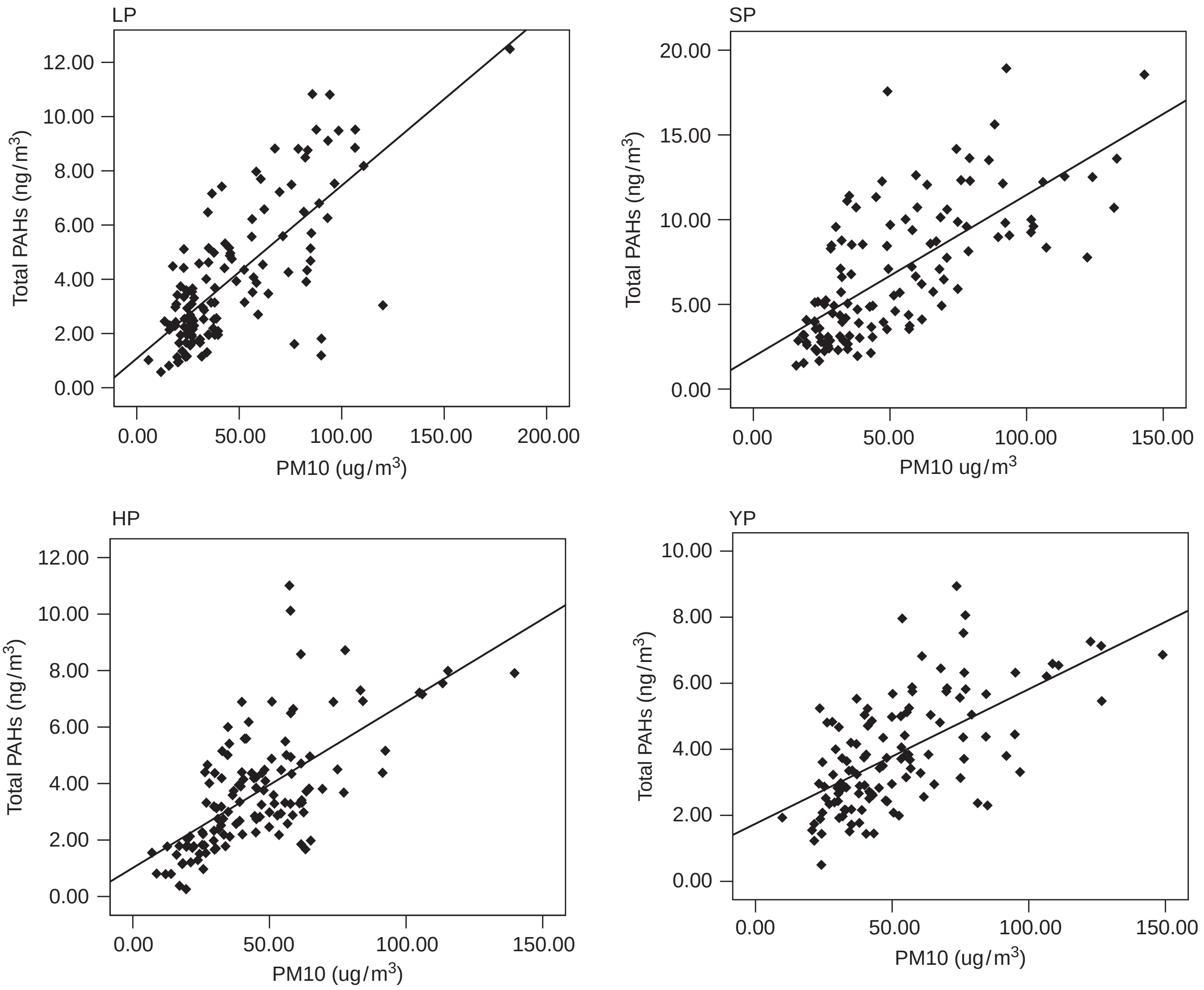

Fig. 3. Correlation between PM_{10} and PM_{10}-bound PAHs for all seasons in each sampling site.

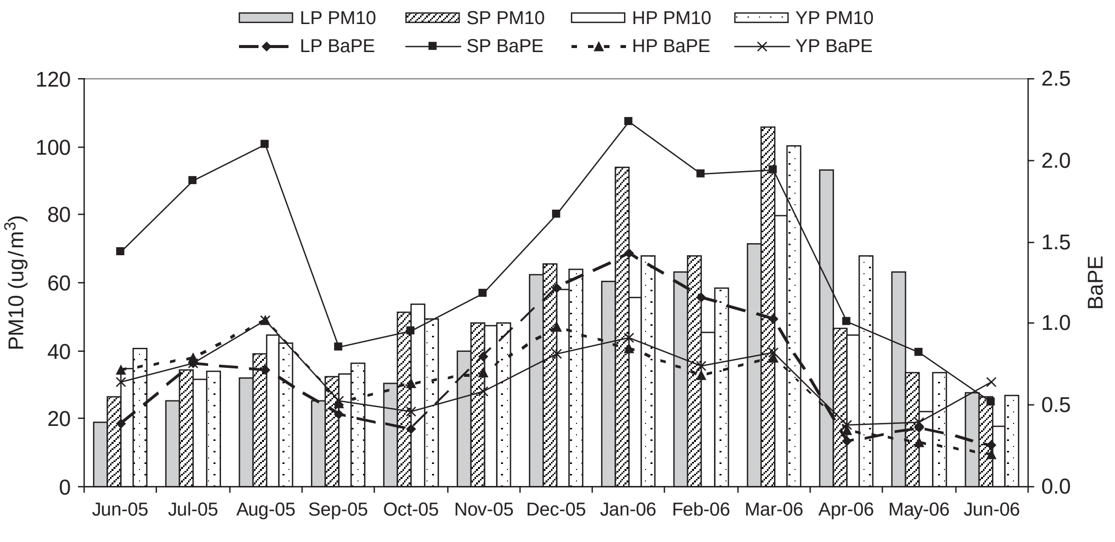

Fig. 4. Monthly mean BaPE and PM_{10} concentrations from all sampling sites.

Table 7
Diagnostic ratios of PAHs for four sampling sites in different months.

	IND/(IND + BPER)				BaP/BPER				BaP/(BaP + CHR)			
	LP	SP	HP	YP	LP	SP	HP	YP	LP	SP	HP	YP
June 2005	0.54	0.57	0.50	0.48	***0.61***	***0.87***	***0.63***	0.50	0.66	0.57	0.58	0.60
July 2005	0.53	0.55	0.49	0.48	***0.70***	***1.06***	***0.66***	***0.61***	0.86	0.79	0.83	0.83
August 2005	0.61	0.62	0.56	0.55	***0.61***	***0.95***	***0.70***	***0.67***	0.90	0.83	0.86	0.86
September 2005	0.57	0.60	0.52	0.52	0.46	0.59	0.48	0.46	0.77	0.71	0.71	0.72
October 2005	0.60	0.58	0.52	0.52	0.48	***0.62***	0.51	0.45	0.83	0.78	0.79	0.80
November 2005	0.56	0.55	0.50	0.50	***0.81***	***0.94***	***0.69***	***0.61***	0.75	0.77	0.83	0.86
December 2005	0.52	0.54	0.47	0.48	***0.83***	***1.03***	***0.64***	***0.63***	0.68	0.66	0.73	0.76
January 2006	0.52	0.56	0.45	0.46	***0.86***	***0.86***	***0.66***	***0.64***	0.67	0.56	0.81	0.78
February 2006	0.57	0.58	0.53	0.52	***0.89***	***1.01***	***0.71***	***0.63***	0.65	0.62	0.81	0.78
March 2006	0.55	0.55	0.54	0.54	***0.65***	***0.79***	0.59	0.54	0.68	0.55	0.67	0.67
April 2006	0.52	0.55	0.48	0.48	0.53	***0.66***	0.50	0.48	0.72	0.49	0.65	0.64
May 2006	0.52	0.53	0.45	0.43	0.40	0.50	0.45	0.38	0.79	0.73	0.81	0.77
June 2006	0.52	0.52	0.47	0.54	0.44	0.56	0.47	***0.61***	0.88	0.80	0.88	0.80
Average	**0.55**	**0.56**	**0.50**	**0.50**	**0.64**	**0.80**	**0.59**	**0.56**	**0.76**	**0.68**	**0.77**	**0.76**
SD	0.03	0.03	0.03	0.03	0.17	0.20	0.10	0.09	0.09	0.11	0.09	0.08

throughout the year. Therefore, the BaPE values contributed from the traffic should be a background value for all sites. It is quite clear that in the dry season open burning, i.e. burning of agricultural waste, community garbage as well as forest fire, mainly contributed pollutants to the atmosphere. In the rainy season, the high BaPE values might have come from other sources. The air pollutants in this season could be from various local activities apart from open burning. One of the possibilities is from small scale factories in the area that whereby seasonal agriculture products are processed.

3.7. Diagnostic ratios

Dianostic ratio can be used for identification of possible emission sources. The diagnostic ratios of this study were calculated by separating sampling sites and months as shown in Table 7. The IND/(IND + BPER) ratio can be used to identify traffic sources. The value in a range from 0.35 to 0.70 indicates that the PAHs were emitted from diesel engines [5–6]. In this study, the average ratios of all sites were 0.50–0.56. Therefore, the PAHs found in this study could have originated from diesel engine emission. The BaP/BPER ratio was also used for characterization of the PAHs sources. The low BaP/BPER ratio (<0.60) is an evidence of greater emission of BPER from traffic sources [15]. The mean ratios of the four sampling sites were 0.56–0.80. The mean ratios of HP and YP sites were fitted with the low ratio value, therefore traffic might be the main source for these two sites. However, the ratio values in some months, especially in the dry season were greater than 0.60, which indicated some other PAH sources. The SP and LP sites had a higher mean ratio value (0.80 and 0.64, respectively) as well as mothly values, depicted as *italic* values in Table 7. This could indicate that the PAHs in these two sites were not only from the traffic but were also from some other sources on a similar basis described for the BaPE above. Apart from these two ratios, the BaP/(BaP + CHR) ratio was also considered. The ratio values of 0.57 and 0.89 could be related to diesel and gasoline exhaust, respectively [8]. The BaP/(BaP + CHR) ratio values obtained in this work were 0.68–0.77, which could be reckoned as somewhat between 0.57 and 0.89. Based on this ratio alone, it could not be concluded what was the major source of PAHs in the areas studied in this work. However, the IND/(IND + BPER) ratio of 0.50–0.56 obtained in this study fits within the range of 0.35–0.70 which suggests that the diesel engine source plays a more important role than the gasoline source.

4. Conclusions

PM_{10} concentrations were found to be significantly higher in the dry season than those in other seasons. PAHs concentrations were positively correlated with the PM_{10} weight. While there was no significant difference of the composition of PM_{10} found among the sampling sites, PM_{10}-bound PAHs were significantly higher at the SP site than the other sites. This is confirmed by the BaPE value, which indicates air quality of the area, since the BaPE value for the SP site was double that of the other sites. The diagnostic ratio indicates that diesel combustion is the main source of PAHs from the traffic sector. However, other sources of PAHs from local activities such as open burning of biomass could not be ruled out. It can be concluded that temporal variation plays a more important role than spatial variation in this study.

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References

[1] U. Pöschl, Atmospheric aerosols: Composition, transformation, climate and health effects, Angewandte Chemie International Edition, 44 (2005) 7520–7540.

[2] C.A. Alves, Characterisation of solvent extractable organic constituents in atmospheric particulate matter: An overview, Annals of the Brazilian Academy of Sciences, 80(1) (2008) 21–82.

[3] J.L. Mauderly and J.C. Chow, Health effects of organic aerosols, Inhalation Toxicology, 20 (2008) 257–288.

[4] W.J. Lee, Y.F. Wang, T.C. Lin and Y.Y. Chen, PAH Characteristics in the ambient air of traffic-source, Science of the Total Environment, 159 (1995) 185–200.

[5] L.B. Liu, Y. Hashi, M. Liu, Y. Wei and J.M. Lin, Determination of particle-associated polycyclic aromatic hydrocabens in urban air of Beijing by GC/MS, Analytical Science, 23 (2007) 667–671.

[6] I.G. Kavouras, P. Koutrakis, M. Tsapakis, E. Lagoudaki, E. Stephanou, D. Von Baer and P. Oyola, Source apportionment of urban particulate aliphatic and polynuclear aromatic hydrocarbons (PAHs) using multivariate methods, Environmental Science and Technology, 25 (2001) 2288–2294.

[7] C.A. Pio, C.A. Alves and A.C. Duarte, Identification, abundance and origin of atmospheric organic particulate matter in a Portuguese rural area, Atmospheric Environment, 35 (2001) 1365–1375.

[8] N.R. Khalili, P.A. Scheff and T.M. Holsen, PAH source fingerprints for coke ovens, diesel and gasoline engines, highway tunnels, and wood combustion emissions, Atmospheric Environment, 29 (1995) 533–542.

[9] P. Pengchai, S. Chantara, K. Sopajaree, S. Wangkarn, U. Tengcharoenkul and M. Rayanakorn, Seasonal variation, risk assessment and source estimation of PM_{10} and PM_{10}-Bound PAHs in the ambient air of Chiang Mai and Lamphun, Thailand, Environmental Monitoring and Assessment, 154 (2009) 197–218.

[10] J.C. Miller and J.N. Miller, Statistics for Analytical Chemistry, 2nd edn., Ellis Horwood, New York, NY, 1988, p. 115.

[11] S. Chantara, S. Wangkarn, U. Tengcharoenkul, W. Sangchan and M. Rayanakorn, Chemical analysis of airborne particulates for air pollutants in Chiang Mai and Lamphun Provinces, Thailand, Chiang Mai Journal of Science, 36(2) (2009) 123–135.

[12] P. Norramit, V. Cheevaporn, N. Itoh and K. Tanaka, Characterization and carcinogenic risk assessment of polycyclic aromatic hydrocarbons in the respirable fraction of airborne particles in the Bangkok metropolitan area, Health Science, 51 (2005) 437–446.

[13] S. Chantara and W. Sangchan, Sensitive analytical method for particle-bound polycyclic aromatic hydrocarbons: A case study in Chiang Mai, Thailand. ScienceAsia, 35 (2009) 42–48.

[14] M. Koyano, O. Endo, S. Goto, K. Tanabe, S. Koottatep and H. Matsushita, Carcinogenic polynuclear aromatic hydrocarbons in the atmosphere in Chiang Mai, Thailand, Japan Journal of Toxicology and Environmental Health, 44(3) (1998) 214–225.

[15] P.K. Pandey, K.S. Patel and Lenicek J. Polycyclic aromatic hydrocarbons: Need for assessment of health risks in India? Environmental Monitoring and Assessment, 59 (1999) 287–319.