Seasonal variation, risk assessment and source estimation of PM 10 and PM10-bound PAHs in the ambient air of Chiang Mai and Lamphun, Thailand

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Received: 27 December 2007 / Accepted: 5 May 2008 / Published online: 8 August 2008 © Springer Science + Business Media B.V. 2008

Abstract Daily PM10 concentrations were measured at four sampling stations located in Chiang Mai and Lamphun provinces, Thailand. The sampling scheme was conducted during June 2005 to June 2006; every 3 days for 24 h in each sampling period. The result revealed that all stations shared the same pattern, in which the PM10 (particulate matters with diameter of less than 10 μ m) concentration increased at the beginning of dry season (December) and reached its peak in March before decreasing by the end of April. The maximum PM10 concentration for each sampling station was in the range of $140-182 \mu g/m^3$ which was 1.1–1.5 times higher than the Thai ambient air quality standard of 120 μ g/m³. This distinctly high concentration of PM10 in the dry season (Dec. 05–Mar. 06) was recognized as a unique seasonal pattern for the northern part of Thailand. PM10 concentration had a medium level of negative correlation ($r = -0.696$ to -0.635) with the

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K. Sopajaree e-mail: fourpppp@gmail.com visibility data. Comparing the maximum PM10 concentration detected at each sampling station to the permitted PM10 level of the national air quality standard, the warning visibility values for the PM10 pollution-watch system were determined as 10 km for Chiang Mai Province and 5 km for Lamphun Province. From the analysis of PM10 constituents, no component exceeded the national air quality standard. The total concentrations of PM10-bond polycyclic aromatic hydrocarbons (PAHs) are calculated in terms of total toxicity equivalent concentrations (TTECs) using the toxicity equivalent factors (TEFs) method. TTECs in Chiang Mai and Lamphun ambient air was found at a level comparable to those observed in Nagasaki, Bangkok and Rome and at a lower level than those reported at Copenhagen. The annual number of lung cancer cases for Chiang Mai and Lamphun Provinces was estimated at two cases/year which was lower than the number of cases in Bangkok (27 cases/year). The principal component analysis/absolute principal component scores (PCA/APCS) model and multiple regression analysis were applied to the PM10 and its constituents data. The results pointed to the vegetative burning as the largest PM10 contributor in Chiang Mai and Lamphun ambient air. Vegetative burning, natural gas burning & coke ovens, and secondary particle accounted for 46–82%, 12– 49%, and 3–19% of the PM10 concentrations, respectively. However, natural gas burning & coke ovens as well as vehicle exhaust also deserved careful attention due to their large contributions to PAHs concentration. In the wet season and transition periods, 42–60% of the total PAHs concentrations originated from vehicle exhaust while 16–37% and 14–38% of them were apportioned to natural gas burning & coke ovens and vegetative burning, respectively. In the dry period, natural gas burning & coke ovens, vehicle exhaust, and vegetative burning accounted for 47– 59%, 20–25%, and 19–28% of total PAHs concentrations. The close agreement between the measured and predicted concentrations data $(R^2 >$ 0.8) assured enough capability of PCA/APCS receptor model to be used for the PM10 and PAHs source apportionment.

Keywords Lung cancer **·** Natural gas burning and coke ovens **·** PAHs **·** PCA/APCS **·** PM10 **·** TEF **·** TTEC **·** Vegetative burning **·** Vehicle exhaust

Introduction

Chiang Mai, with an altitude of approximately 310 m above the sea level, is situated approximately 700 km from Bangkok in the Chiang Mai–Lamphun intermontaigne basin area. It is surrounded by high mountain ranges. The city covers an area of approximately $20,107$ km² and is the country's second largest province. Due to its geographical features, Chiang Mai as well as some provinces in the northern part of Thailand has been annually facing air pollution during the dry season. This pollution has been recorded as a serious problem for well over 10 years. Recently, the problem has become worse than ever. During the first 2 weeks of March 2007, air pollution levels in Chiang Mai and the surrounding provinces rose steadily above the safety limit and produced a yellow-tinged haze that cut visibility down to less than 1,000 m. Respirable particulate matter, having less than 10 μm in diameter (PM10), reached their peak concentration on March 14th 2007 at 383 μg/m³ (PCD [2007b](#page-20-0))—three times higher than Thailand's acceptable safety concentration of $120 \mu g/m³$. Up to 500,000 people were reported by the Public Health Ministry to be affected by the excessive concentration of PM10 this year. Hospitals and clinics across the affected area reported a surge in the number of patients with respiratory problems during the month of March, approximately a 20% increase compared to the same period in 2006.

This study was conducted from June 2005 to June 2006, approximately 9 months before the pollution crisis occurred, in order to characterize spatial and seasonal variations of PM10 and PM10-bond PAHs concentrations as well as their emission sources in the Chiang Mai–Lamphun intermontaigne basin area. The carcinogenic risk assessment of PM10-bond PAHs was also estimated. The result of this study provides interesting information for solving the air pollution problems in the northern area of Thailand.

Materials and methods

PM10 sampling

In this study, three sampling stations located in Chiang Mai province: (1) Yuparaj Witayalai School, (2) Municipality Hospital, (3) Saraphee District and one sampling station in Lamphun province, Kai Kaew Community, were selected (details shown in Table [1](#page-2-0) and Fig. [1](#page-2-0)). PM10 samples were collected every 3 days for 24 h in each sampling period using a High Volume PM10 Air Sampler (Wedding & Associates Inc., USA). The sampling campaign was simultaneously started at 10:00 a.m. for all four sampling sites. Particulate matter was collected by the gravimetric method, in which the surrounding air was sucked through the inlet chamber (at approximately 2 m above the ground level) using an air pump at the flow rate of 1,130 L/min. PM10 in the air was trapped in the quartz fiber filter and weighed using the five digits scale balance (Mettler Toledo (USA) AG285) covered by the temperature and humidity controlling cabinet (DE-300). Particulate concentrations were then determined using the calculated particulate amounts and air flow rate recorded during the sampling time.

Chemical composition of PM10

The post-weighed quartz fiber paper $(8 \times 10 \text{ in.})$ used for PM10 sampling was equally divided into eight pieces (2.5 \times 4 in.) by a stainless steel roller Environ Monit Assess (2009) 154:197–218 199

blade cutter. Two of eight were grouped and analyzed for PAHs, ions, metals and carbon.

Analysis of 16 EPA-PAHs in PM10 samples by GC-MS (U.S. EPA Method TO-13A (USEPA [1999a](#page-21-0)*) and SOP # CH-IN-003*

Two pieces of the quartz filter (one sample) were cut into small pieces by a stainless scissor and put into 60 ml amber-bottle wrapped with aluminium foil and mounted by paraffin film. The sample was 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 into a v-shape flask. The pellet was re-extracted in 30 ml acetronitrile by repeating the previous step. The solution was collected in the same container and evaporated by low-pressure evaporator at 30◦C until nearly dry. The solution

Fig. 1 Location of sampling stations

was transferred into a 1 ml volumetric flask. After that a mixed solution of internal standards (D10-FLU, D10-PYR and D12-BaP) was added and adjusted to 1 ml by acetronitrile. The solution was then analyzed for 16 PAHs (naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IND), dibenz[a,h]anthracene (DBA), benzo[g,h,i]perylene (BPER) by GC-MS (Hewlett Packard, U.S.A., column HPTM-5MS packed with 5% Phenyl methyl polysiloxane, 30 m long, diameter 0.25 mm, 0.25 μm).

Ion analysis of PM10 sample by ion chromatography (Dionex 300, USA and Metrohm, Switzerland)

Dissolved ions including Na^+ , NH⁺, K⁺, Ca²⁺, Mg^{2+} , Cl⁻, NO₃ and SO₄² were analyzed following a method developed from the Technical Document for Filter Pack Method in East Asia (EANE[T](#page-20-0) [2003](#page-20-0)) using Ion Chromatography (EANET [2000](#page-20-0)). Two pieces of the quartz filter (one sample) were cut into small pieces by a stainless scissor and put into 100 ml beakers followed by 50 ml of deionized water. The beakers were then covered by paraffin film and ultrasonicated at 100% ultrasound power (Transsonic Digitals, Elma, Germany) for 30 min. The solution was filtered through a cellulose acetate filter (pore size 45 μm, diameter 13 mm) into a plastic bottle for IC analysis. Analytical columns of Dionex 300 are IonPac® AS4A (4×250 mm) and IonPac® CS12 $(12 \times 250 \text{ mm})$ for anion and cation analysis, respectively, while those of Metrohm are Metrosep A Supp 4 (4×250 mm) and Metrosep C2 100 ($4 \times$ 100 mm), respectively.

Elemental analysis of PM10 sample by inductively couple plasma spectroscopy (ICP-OES PerkinElmer® Optima 3000 operated with WinLab32TM software)

Filters were extracted based on Compendium Method IO-3.1 (USEP[A](#page-21-0) [1999a](#page-21-0)) for metals including Ca, Al, Si, Fe, Mg, K, Zn, Ti, P, Pb, Ba, Sr, Mn, Ni, Cu, V, Cr, Cd, Hg and As were analysed by Inductively Coupled Plasma–Atomic Absorption Spectrometry based on Compendium Method IO-3.4 (USEP[A](#page-21-0) [1999b](#page-21-0)). Two pieces of the quartz filter (one sample) were cut into small pieces by a stainless scissors and put into an Erlenmayer flask followed by 40 ml of mixed acid solution (5.55% HNO3/16.75% HCl). The sample was refluxed on a hot plate for 3 h. It was left until cool and transferred into a 50 ml volumetric flask. The sample was rinsed with deionized water and transferred into the volumetric flask before the volume was adjusted to 50 ml. It was then filtered through a Nylon 0.45 μm syringe filter into a plastic bottle for further analysis.

Carbon analysis of PM10 sample by CHN-S/O elemental analyzer (PE 2400 series II CHNS/O analyzer, Perkin Elmer Cooperation [1991](#page-20-0)*)*

A tin capsule $(1 \times 1$ cm) as a sample container was weighed by six digit analytical balance. The quartz filter that collected the PM10 was punched by a puncher with a diameter of 0.55 cm. The punched filter was put into a prepared capsule, wrapped and weighed. The sample was then analysed for total carbon (TC) by CHN-S/O Elemental Analyzer using L-cystine and BS Slag 2 as standards and an unused filter paper as a blank.

Carcinogenic risk assessment

The relative toxicity of an individual PM10-bond PAH compared to benzo(a)pyrene was calculated based on Eq. 1. The ambient concentrations of 16 PAHs and their toxicity equivalency factor (TEF, unitless) defined by USEP[A \(1993](#page-21-0)) were used in the calculation. The sum of this relative toxicity, namely total toxicity equivalent concentration (TTEC), exhibits the toxicity of PAHs in Chiang Mai and Lamphun ambient air (see Eq. 1).

$$
TTEC = \sum_{n} C_n \cdot TEF_n \tag{1}
$$

Where, TTEC: Total Toxicity Equivalent Concentration, *n*: total number of individual PAH, *Cn*: Concentration of the individual PAH, TEF*n*: Toxic equivalency factor of the individual PAH which is 1.0 for BaP and DBA, 0.1 for BaA, BbF, BkF, 0.01 for CHR and 0 for the other PAHs.

The TTEC, in other words, indicates the total PAHs concentration in terms of BaP equivalent. Regarding the lung cancer risk via inhalation route, WH[O](#page-21-0) [\(1987](#page-21-0)) suggested the unit risk of 8.7×10^{-5} (ng/m³)⁻¹ for a lifetime (70 years) of PAHs exposure, assuming one was exposed to the

average level of one unit BaP concentration (1 ng/m^3) . This study estimated the corresponding lifetime lung cancer risks of Chiang Mai and Lamphun residents using Eq. 2 (Norramit et a[l](#page-20-0). [2005](#page-20-0)). Certain values were assigned to variables in the equation as following, the TTEC $(ng/m³)$ to sum BaP; 2,054,736 (Department of Provincial Administratio[n](#page-19-0) [2005](#page-19-0)) to residents; the 70 years of life expectancy; and the 8.7 × 10⁻⁵ (ng/m³)⁻¹ to unit risk.

Source apportionment of PM10 and PM10-bond PAHs

Source identification and apportionment of PM10 and PM10-bond PAHs were performed by employing the principal component analysis (PCA) and the absolute principal component scores (APCS) followed by the multiple linear regressions. Statistical treatment of data including correlation analysis, PCA/APCS and multiple linear regressions analysis was conducted using the SPSS 11.0 statistical software.

The application of PCA to PM10 composition data is associated with significant setbacks because the outcomes are correlated with but not proportional to source contributions (Anderson et a[l](#page-19-0). [2002](#page-19-0)). However, when it is coupled with the APCS and multi-linear regression, it becomes a powerful tool for source identification and apportionment (Thurston and Spengle[r](#page-21-0) [1985](#page-21-0); Kumar et a[l](#page-20-0). [2001](#page-20-0); Guo et a[l](#page-20-0). [2004](#page-20-0)).

In this study, varimax rotated PCA with eigenvalue over 1.0 followed by APCS was performed as described by Guo et a[l. \(2004](#page-20-0)) on a data matrix (after removing outliers) that consisted of 503 PM10 concentrations and 33 chemical compositions of PM10. The main purpose of the PCA was to reduce the number of inter-correlated variables in the original data to a lesser number of factors (principal component: PC), which are orthogonal to each other. The first factor accounts for the highest variability in the data and subsequent factors account for a progressively less

amount of the data variance. However, as stated earlier, factors are correlated with sources but are not proportional to source contributions. The principal component scores for each sample are obtained from PCA. As the principal component scores are normalized, with a Mean of zero and Standard Deviation equal to unity, the true zero for each factor score was calculated by introducing an artificial sample with concentrations equal to zero for all variables. The absolute principal component scores (APCS) were then calculated by subtracting the principal component scores for this artificial sample from the factor scores of each one of the true samples. Regressing the observed data (PM10 concentration or total PAHs concentration) on these APCS gives estimates of the coefficients which convert the APCS into pollutant source mass contributions from each source for each sample. The source contributions to Ci (PM10 concentration or total PAHs concentration) can be calculated by using a multiple linear regression procedure according to the following equation:

$$
\sum_{p=1}^{n} b_p \text{APCS}_p = C_i \tag{3}
$$

Where $p = 1, 2, \dots, n$ (total number of principal component), is the multiple linear regression coefficient for the source p , is the absolute principal component score of the rotated factor *p* for a sample, represents the contribution of source p to PM10 ($ng/m³$) or total Pm10-bond PAHs concentrations (μ g/m³). If the determination coefficient of Eq. 3 is equal or greater than 0.7, the equation is then considered suitable for the source apportionment. Thus, the percentage contribution of source p to PM10 concentration can be calculated from the ratio $b_p \text{APCS}_p / \sum_{p=1}^n b_p \text{APCS}_p$. Therefore, the seasonal contributions of source p are defined as in Eq. 4 where $i = 1, 2, \dots, m$ (total number of samples in each season).

Seasonal contributions of source

$$
p\left(\% \right) = \frac{\sum\limits_{i=1}^{m} \left[b_p \text{APCS}_p \right]_i \times 100}{\sum\limits_{i=1}^{m} \left[\sum\limits_{p=1}^{n} b_p \text{APCS}_p \right]_i} \tag{4}
$$

Results and discussion

PM 10 concentration

Daily concentrations of PM10 at the four sampling stations varied from 5 to 182 μ g/m³ with the highest concentrations of 149 μ g/m³ at Yuparaj, 140 μg/m³ at Municipality Hospital, 150 μg/m³ at Saraphee and $182 \mu g/m^3$ at Lamphun. As shown in Fig. 2, the PM10 concentration detected at all stations shared the same pattern, in which it increased at the beginning of dry season (December) and reached its peak in March before decreased by the end of April.

The average of daily PM10 concentration compared with the percentages of numbers of days that PM10 concentrations exceeded the 24-h national standard value $(120 \mu g/m^3)$ for each sampling site is summarized in Table [2](#page-6-0). On average, PM10 concentrations and the percentages of numbers of days that PM10 concentrations exceeded the 24-h national standard value at Municipality Hospital (45.3 ± 23.1 μg/m³, 0.8%) and Lamphun $(46.5 \pm 29.2 \,\mu g/m^3, 0.8\%)$ were found to be lower than those at Saraphee (54.5 \pm 31.5 μ g/m³, 5.6%) and Yuparaj $(52.9 \pm 27.3 \,\mu g/m^3, 4.0\%)$. It should be noted here that the average PM10 concentrations at Saraphee and Yuparaj exceeded the Thailand year-round standard value $(50 \mu g/m^3)$. This finding indicates the urgency for the responsible authority in Chiang Mai, especially in the Saraphee and Yuparaj area, as well as Lamphun Provinces to develop more effective strategies to solve the particle pollution during the dry season.

The seasonal patterns of average PM10 concentration observed in this study compared with those measured at different locations by the Pollution Control Department (PCD [2007a](#page-20-0), [b](#page-20-0)) were summarized in Fig. [3](#page-6-0). The distinctly high concentration of PM10 in the dry season (December– March) was found only in the Northern part of Thailand (YP, HP, SP, LP). This unique seasonal pattern of PM10 concentration suggested the presence of temporary source of PM10 as well as the occurrence of an inversion layer in the Chiang Mai–Lamphun intermontaigne basin area during dry period.

The Pearson correlation analysis at a 1% level of significance revealed a significant correlation

Sampling station Number of sample PM10 concentrations exceeded		the 24-h national standard value		Year-round average PM10 concentration \pm standard deviation (μ g/m ³)		
		Number of day ^a	$\%$			
YP	125			4.0 52.9 ± 27.3		
HP	126		0.8	45.3 ± 23.1		
SP	126			5.6 54.5 ± 31.5		
LP	126		0.8	46.5 ± 29.2		

Table 2 PM10 concentration (μg/m³) compared to the 24-h national standard

aDays exceed 24-h national standard were found only in dry season

among PM10 concentrations at three sampling sites in Chiang Mai (correlation coefficient: *r* = 0.819–0.909, $p < 0.01$) in all seasons. A strong correlation ($r = 0.900{\text -}0.988$, $p < 0.01$) was however observed only in the dry season between each couple of PM10 data obtained from all the sampling sites. This result implied that the strategy for controlling PM10 concentration in the dry season needed to be done on the regional level rather than focusing only in an affected province. An increase of PM10 concentration during the dry season in the Chiang Mai–Lamphun intermontaigne basin area was possibly caused by the same sources, such as forest fires or agricultural waste burning. Larger areas burned by forest fire in Chiang Mai during dry season of year 2006 (see Table [3](#page-7-0)) gave evidence to the above interpretation.

The correlation coefficients between meteorological parameters and PM10 concentration at the significant level of 1% are presented in Table [4](#page-8-0). According to the table, PM10 concentration had medium level of negative correlation $(r = -0.696)$ to −0.635) with visibility data. This result implied the possibility of using the visibility as an

indicator for the particle pollution-watch system. Figure [4](#page-8-0) demonstrates a plot of the comparison of PM10 concentrations and the visibility data of which the frequency is equal or greater than 5% of total data. Comparing the maximum PM10 concentration detected at each sampling station with the permitted PM10 level of the national air quality standard, the warning visibility levels for the PM10 pollution-watch system could be determined as 10 km for Chiang Mai and 5 km for Lamphun provinces.

PM10 chemical constituent

The average concentrations and standard deviation of PM10 constituents are summarized in Table [5](#page-9-0). The ambient concentrations of Al, Ca, Fe, K, Si and $\text{Zn } (\mu \text{g/m}^3)$ were higher than the other inorganic elements especially in the dry season. Similarly, the total carbon and the most abundant PAHs in the sample, i.e. CHR, BbF, BkF, BaP, IND, DBA, BPER were also found to be higher in the dry season ($p < 0.05$, *t*-test). This result indicated the priority of total carbon and high molecular weight PAHs to be specially considered

Fig. 3 Seasonal average of 24-hrs PM10 concentrations comparing to those measured by the Pollution Control Department (BKK: Bangkok, KK: Khon Kean, HY: Had Yai)

Period	Parameters		Daily average \pm standard deviation	Area burned by forest fires at
		Chiang Mai Commercial Aviation (2007)	Lamphun Meteorology (2007)	Chiang Mai (Forest Fire Control Division 2007)
Wet (Jun. to Sept. 05)	Rainfall (mm/day)	6.73 ± 10.4	1.79 ± 6.1	$\overline{0}$
	Temperature (C)	26.9 ± 1.4	28.5 ± 5.8	
	Pressure (mmHg)	754.1 ± 1.7	734.6 ± 1.6	
	Relative humidity (%)	85.4 ± 5.7	78.2 ± 7.4	
	Sunlight duration (day)	0.26 ± 0.2	NA	
	Visibility(km)	10.8 ± 0.9	11.0 ± 0.99	
	Wind direction	West and southwest	NA	
	Monthly maximum wind velocity (km/h)	$20.9 - 26.4$	NA	
Transition 1 (Oct. to Nov. 05)	Rainfall (mm/day)	3.4 ± 9.4	0.02 ± 0.1	$\overline{0}$
	Temperature (C)	25.2 ± 1.4	26.2 ± 1.5	
	Pressure (mmHg)	758.5 ± 1.4	738.8 ± 1.3	
	Relative humidity (%)	83.8 ± 4.6	78.9 ± 2.7	
	Sunlight duration (day)	0.5 ± 0.2	NA	
	Visibility(km)	9.6 ± 1.3	6.6 ± 1.98	
	Wind direction	West and northwest	NA	
	Monthly maximum wind velocity (km/h)	$16.3 - 21.8$	NA	
Dry (Dec. 05 to Mar. 06)	Rainfall (mm/day)	0.5 ± 2.2	0.1 ± 0.7	
	Temperature (C)	23.9 ± 3.2	24.4 ± 3.2	
	Pressure (mmHg)	758.4 ± 2.2	738.2 ± 1.8	Jan. 06: 106 Rai
	Relative humidity (%)	69.3 ± 12.0	65.0 ± 11.0	
	Sunlight duration (day)	0.6 ± 0.2	NA	Feb. 06: 1,527 Rai
	Visibility(km)	8.7 ± 1.5	4.0 ± 1.12	
	Wind direction	Not clear	NA	Mar. 06: 4,728 Rai
	Monthly maximum wind velocity (km/h)	$13.5 - 20.6$	NA	
Transition 2 (Apr. to May. 06)	Rainfall (mm/day)	6.9 ± 17.4	0.7 ± 1.2	
	Temperature (C)	27.6 ± 2.6	28.1 ± 2.7	
	Pressure (mmHg)	755.5 ± 1.3	735.9 ± 1.1	
	Relative humidity (%)	74.5 ± 12.3	70.9 ± 12.1	
	Sunlight duration (day)	0.5 ± 0.3		Apri. 06: 937 Rai
	Visibility(km)	10.3 ± 1.3	6.5 ± 2.97	
	Wind direction	West and northwest	NA	May. 06: 8 Rai
	Monthly maximum wind velocity (km/h)	28.1	$\rm NA$	

Table 3 Meteorological parameters and forest fire records

1 Rai = 0.4, acres = $1,600 \text{ m}^2$

NA Not analyzed

Sampling stations	Air quality monitoring stations	Relative humidity and PM_{10}	Visibility and PM_{10}	Precipitation and PM_{10}	Sunlight duration and PM_{10}
YP	Pollution Control Department	-0.436	-0.696	-0.193	$+0.301$
HP	Commercial aviation, Chiang Mai	-0.532	-0.657	-0.291	$+0.235$
LP	Meteorology building, Lamphun	-0.644	-0.685		
SP	Commercial aviation, Chiang Mai	-0.644	-0.644	-0.325	$+0.432 P$

Table 4 Correlation coefficients between meteorological parameters and PM10 concentration at the significant level of 1%

during PM10 pollution crisis. However, no single component exceeded the national air quality standard. Total concentration of low molecular weight PAHs, including NAP, ACY and ACE, represented 0–12% of the total 16 PAHs concentrations. The result agreed well with the general expectation that the components easily volatized and degraded away due to their low evaporation pressures and high degradation rates.

High PAHs concentration in the winter season was also reported in many urban atmospheres as a result of residential heating and the thermal inversion layer (Wild and Jone[s](#page-21-0) [1995](#page-21-0); Panther et a[l](#page-20-0). [1999](#page-20-0); Dimashki et a[l](#page-20-0). [2001](#page-20-0); Vestreng and Klei[n](#page-21-0) [2002](#page-21-0); Guo et a[l](#page-20-0). [2003](#page-20-0)). Since heaters are not normally used in Thailand, vehicle exhaust, vegetative burning such as forest fires and agricultural waste combustion as well as the accumulation of the air pollutants under the formation of a strong inversion layer during the winter period were thought responsible for this increase of PAH concentration during dry season. Total concentrations $(ng/m³)$ of 16 PAHs ranged from 0.05 to 22.4 ng/m³, which are at a level similar to those observed in Bangkok $(35.8-55.5 \text{ ng/m}^3)$ (Thongsan[it](#page-21-0) [2002](#page-21-0)) for annual average concentration) Guangzhou (55.49 ng/m³ for winter and spring), Hong Kong (14.88 ng/m^3) for winter and spring) and Macao (41.70 ng/m^3) for winter and spring) (Qi et a[l](#page-20-0). [2000](#page-20-0)). The correlation coefficients between the total concentrations of 16 PAHs and meteorological parameters (i.e. temperature, relative humidity, visibility, sunlight duration and precipitation) were found to be very low $(r = -0.26 \text{ to } 0.23)$. Contrary to the case of PM10 concentrations, the conventional meteorological parameters were not enough to produce a warning index for PAHs pollution-watch system.

Risk assessment of PM10-bound PAHs

The total of 16 PAHs concentration was calculated in terms of TTEC using the TEF method. As shown in Table [6](#page-11-0), the TTEC of PM10-bond PAHs in Chiang Mai and Lamphun ambient air was found to be at a comparable level with those observed in Nagasaki, Bangkok and Rome and at a lower level than that reported in Copenhagen. Based on the yearly average TTEC, the annual number of lung cancer cases for Chiang Mai and Lamphun provinces was estimated at two cases/ year which was lower than the number of cases in Bangkok (27 cases/year).

Fig. 4 A plot of the comparison of PM10 concentrations and the visibility data of which the frequency is equal or greater than 5%

Table 5 Average concentrations ± standard deviation of PM10 constituents

	YP				HP					
	WET	TRANS1	DRY	TRANS2	WET	TRANS1	DRY	TRANS2		
Al	0.27 ± 0.17	0.287 ± 0.08	0.52 ± 0.24	0.17 ± 0.11	0.24 ± 0.23	0.31 ± 0.1	0.33 ± 0.18	0.13 ± 0.08		
As	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0		
Ba	0.01 ± 0.0	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.0	0.01 ± 0.01	0.01 ± 0.02	0.01 ± 0.01	0.0 ± 0.01		
Ca	1.62 ± 1.49	1.21 ± 0.38	2.22 ± 1.05	0.78 ± 0.5	1.48 ± 1.01	1.47 ± 0.52	1.75 ± 1.46	0.97 ± 0.62		
$\ensuremath{\mathrm{Cd}}$	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0		
Cr	0.0 ± 0.0	0.0 ± 0.01	0.01 ± 0.02	0.0 ± 0.01	0.0 ± 0.01	0 ± 0.0	0.01 ± 0.02	0.01 ± 0.01		
Cu	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.01		
$\rm Fe$	0.14 ± 0.16	0.12 ± 0.18	0.44 ± 0.34	0.21 ± 0.1	0.13 ± 0.19	0.15 ± 0.16	0.31 ± 0.33	0.17 ± 0.1		
Hg	0.01 ± 0.01	0.02 ± 0.04	0.0 ± 0.0	0.0 ± 0.0	0.01 ± 0.01	0.02 ± 0.04	0 ± 0.01	0 ± 0.0		
$\mathbf K$	0.29 ± 0.14	0.53 ± 0.23	1.7 ± 1.09	0.52 ± 0.45	0.26 ± 0.16	0.65 ± 0.72	1.39 ± 0.93	0.37 ± 0.36		
Mg	0.11 ± 0.09	0.1 ± 0.05	0.19 ± 0.13	0.05 ± 0.05	0.09 ± 0.08	0.1 ± 0.06	0.14 ± 0.12	0.06 ± 0.05		
Mn	0.01 ± 0.01	0.02 ± 0.01	0.03 ± 0.02	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.02 ± 0.02	0.01 ± 0.01		
Ni	0.0 ± 0.01	0.0 ± 0.0	0.0 ± 0.01	0.0 ± 0.0	0 ± 0.01	0 ± 0.0	0 ± 0.01	0 ± 0.01		
$\mathbf P$	0.0 ± 0.0	0.0 ± 0.01	0 ± 0.01	0.0 ± 0.01	0.0 ± 0.0	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01		
Pb	0.01 ± 0.01	0.05 ± 0.03	0.05 ± 0.05	0.01 ± 0.01	0.0 ± 0.01	0.07 ± 0.12	0.04 ± 0.05	0.01 ± 0.01		
Si	0.82 ± 0.67	0.92 ± 0.48	1.47 ± 0.88	0.41 ± 0.34	0.6 ± 0.42	1.09 ± 0.51	1.26 ± 0.8	0.42 ± 0.23		
$\rm Sr$	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0 ± 0.0	0 ± 0.0	0.0 ± 0.0		
Ti	0.01 ± 0.0	0.01 ± 0.0	0.01 ± 0.01	0.0 ± 0.0	0.01 ± 0.01	0.01 ± 0.0	0.01 ± 0.01	0.0 ± 0.0		
$\mathbf V$	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0		
Zn	0.29 ± 1.56	0.06 ± 0.04	0.08 ± 0.05	0.03 ± 0.03	0.31 ± 1.56	0.07 ± 0.04	0.1 ± 0.21	0.1 ± 0.25		
SO_4^{2-}	1.29 ± 1.07	6.57 ± 5.92	4.36 ± 2.47	2.97 ± 2.05	1.14 ± 0.96	6.35 ± 5.41	3.58 ± 2.24	1.87 ± 1.38		
NO_3^-	0.49 ± 0.24	0.65 ± 0.3	1.64 ± 1.17	0.68 ± 0.56	0.44 ± 0.24	0.68 ± 0.26	1.37 ± 1.09	0.43 ± 0.41		
Cl^-	0.13 ± 0.11	0.14 ± 0.08	0.17 ± 0.08	0.08 ± 0.06	0.12 ± 0.11	0.18 ± 0.17	0.15 ± 0.08	0.07 ± 0.05		
$NH4+$	0.13 ± 0.14	1.17 ± 1.19	1.48 ± 1.34	0.98 ± 0.9	0.14 ± 0.16	1.1 ± 0.99	1.37 ± 1.26	0.59 ± 0.59		
	SP				LP					
	WET	TRANS1	DRY	TRANS2	WET	TRANS1	DRY	TRANS2		
Al	0.19 ± 0.12	0.20 ± 0.07	0.47 ± 0.27	0.15 ± 0.09	0.15 ± 0.11	0.19 ± 0.06	0.62 ± 0.64	0.17 ± 0.11		
$\mathbf{A}\mathbf{s}$	0 ± 0.0	0.0 ± 0.0	0 ± 0.01	0 ± 0.0	0 ± 0.0	0 ± 0.01	0 ± 0.01	0 ± 0.0		
Ba	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.0	0 ± 0.0	0.01 ± 0.01	0.01 ± 0.01	0 ± 0.0		
Ca	0.83 ± 0.47	0.9 ± 0.36	2.1 ± 1.73	0.59 ± 0.44	0.64 ± 0.33	0.71 ± 0.26	2.11 ± 1.66	0.7 ± 0.36		
Cd	0.0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0		
$\rm Cr$	0.0 ± 0.0	0 ± 0.0	0.01 ± 0.02	0 ± 0.01	0 ± 0.0	0 ± 0.02	0.01 ± 0.023	0 ± 0.0		
Cu	0.02 ± 0.03	0.01 ± 0.02	0.04 ± 0.03	0.02 ± 0.01	0.01 ± 0.01	0 ± 0.0	0.02 ± 0.02	0.01 ± 0.0		
Fe	0.11 ± 0.15	0.09 ± 0.13	0.46 ± 0.44	0.17 ± 0.09	0.11 ± 0.15	0.14 ± 0.2	0.59 ± 0.49	0.19 ± 0.13		
Hg	0.01 ± 0.01	0.02 ± 0.03	0 ± 0.01	0 ± 0.0	0.01 ± 0.01	0.02 ± 0.04	0 ± 0.0	0 ± 0.0		
K	0.37 ± 0.19	0.58 ± 0.19	1.95 ± 1.18	0.55 ± 0.41	0.33 ± 0.17	0.5 ± 0.19	1.66 ± 0.98	0.41 ± 0.36		
Mg	0.09 ± 0.07	0.09 ± 0.05	0.19 ± 0.14	0.06 ± 0.05	0.08 ± 0.07	0.08 ± 0.05	0.18 ± 0.11	0.08 ± 0.06		
Mn	0.01 ± 0.01	0.02 ± 0.01	0.03 ± 0.02	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.03 ± 0.02	0.01 ± 0.01		
Ni	0 ± 0.0	0 ± 0.0	0.01 ± 0.01	0 ± 0.0	0.01 ± 0.02	0 ± 0.0	0.01 ± 0.01	0 ± 0.0		
${\bf P}$	0.01 ± 0.03	0.01 ± 0.02	0.03 ± 0.03	0.02 ± 0.02	0 ± 0.01	0 ± 0.0	0.01 ± 0.02	0 ± 0.01		
Pb	0.01 ± 0.01	0.05 ± 0.03	0.05 ± 0.06	0.01 ± 0.01	0.01 ± 0.01	0.04 ± 0.02	0.04 ± 0.03	0.01 ± 0.01		
Si	0.57 ± 0.43	0.9 ± 0.38	1.45 ± 0.85	0.4 ± 0.31	0.53 ± 0.41	0.88 ± 0.39	1.44 ± 0.8	0.46 ± 0.33		
			0 ± 0.0							
Sr	0 ± 0.0	0 ± 0.0 0.01 ± 0.0	0.01 ± 0.01	0.0 ± 0.0	0.0 ± 0.0	0 ± 0.0	0 ± 0.0	0 ± 0.0		
Ti	0.01 ± 0.0			0.0 ± 0.0	0.0 ± 0.0	0.01 ± 0.0	0.01 ± 0.01	0 ± 0.0		
$\mathbf V$	0 ± 0.01	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0 ± 0.01	0 ± 0.0		
Zn	0.04 ± 0.07	0.06 ± 0.04	0.09 ± 0.08	0.03 ± 0.03	0.3 ± 1.38	0.06 ± 0.05	0.08 ± 0.09	0.04 ± 0.04		
SO_4^{2-}	1.28 ± 0.97	6.76 ± 5.84	4.4 ± 2.36	3 ± 2.37	1.06 ± 0.92	4.43 ± 4.11	3.75 ± 2.47	2.44 ± 1.67		
NO_3^-	0.46 ± 0.26	0.87 ± 0.45	2.18 ± 1.27	0.92 ± 0.76	0.37 ± 0.26	0.66 ± 0.31	2.19 ± 1.53	0.69 ± 0.59		
Cl^-	0.19 ± 0.1	0.3 ± 0.18	0.5 ± 0.44	0.18 ± 0.11	0.16 ± 0.12	0.21 ± 0.13	0.51 ± 0.36	0.11 ± 0.11		
$NH4+$	0.17 ± 0.18	1.4 ± 1.17	1.63 ± 1.16	0.94 ± 0.95	0.15 ± 0.16	0.91 ± 0.91	1.34 ± 1.12	0.66 ± 0.68		

The concentration of inorganic components and TC are in the unit of μg/m3. The concentration of PAHs are in the unit of ng/m³. *TTEC* Total toxicity equivalent concentration (ng/m³) comparing to BaP

Table 6 Yearly average TTEC of Chiang Mai and Lamphun provinces compared with that of other cities

	VР	HP	çр	LP	Bangkok ^a	Nagasaki ^b	Rome ^c	Copenhagen ^d
TTEC	0.78		.60	0.84		20	2.48	
Lung cancer cases per year					-	N	NA	

NA Not ana[l](#page-20-0)yzed, *TTEC* total toxicity equivalent concentration (ng/m³) comparing to BaP ^aNorramit et al. [\(2005](#page-20-0))
^bWada et al. (2001)

^cMenichini et a[l](#page-20-0). [\(1999](#page-20-0))

dNie[l](#page-20-0)sen et al. [\(1996](#page-20-0))

Figure 5 demonstrates the plot of the comparison of TTEC and PM10 concentration in each sample. The result revealed that a higher PM10 level does not always resulted in higher TTEC value. This finding implied the insufficiency of conventional Thai air quality standard. A new criterion for the control of carcinogenic components, i.e., PAHs is needed in order to protect the residents from long-term diseases.

Source apportionment

The principal component analysis with Kaiser– Meyer–Olkin (KMO Measure of Sampling Adequacy) of 0.75 identified 11 possible components (PC1–PC11) explaining 82.5% of the total variance as shown in Table [7](#page-12-0). Elements which had communality values of lower than 0.65 were removed from the calculation. Among the 11 PC, it was observed that the elements that had the highest PC loading (bold) were considered to have a strong influence on that PC. Applying the same comparison among sources on the emis-

Fig. 5 The plot between TTEC and PM10 concentration

sion profiles (Tables $8, 9$ $8, 9$ $8, 9$ and 10), Each PC was named after the source which had highest content of those marker elements (*italics*) in the emission profile.

The first principal component (vehicle exhaust, VEHIC) accounted for 13.1% of the total variance. It was interpreted as vehicle exhaust due to high loading of the dominant four–five rings PAHs, including benzo[b]fluoranthene, benzo[k] fluoranthene, benzo[a]pyrene, indeno[ig1,2,3-cd] pyrene, dibenz[a,h]anthracene and benzo[g,h,i] perylene, which mostly are of vehicle exhaust origin as shown in Table [8](#page-13-0). Especially, benzo $[g,]$ h,i]perylene, which has been identified as a typical tracer of vehicular source of PAHs (Harrison et a[l](#page-20-0). [1996](#page-20-0); Simcik et a[l](#page-21-0). [1999](#page-21-0); Larsen and Bak[er](#page-20-0) [2003](#page-20-0)).

The second principal component was considered as soil particles. It accounted for 9.0% of the variance. The high loading of Al and Si indicated soil particles and Mg represented brake wear that can also be included in soil as shown in Tables [9](#page-14-0) and [10](#page-15-0).

The third principal component (natural gas combustion and coke oven, GAS&COKE) was heavily loaded with PM10, benzo[a]anthacene, chrysene and benzo[k]fluoranthene with a percentage variance of 7.9. Since benzo[a]anthacene and chrysene are the markers of coke ovens and natural gas combustion, and benzo [k]fluoranthene is detected in high amounts in both gasoline vehicle exhaust and natural gas combustion (Table [8](#page-13-0)), GAS&COKE represents the coke oven and natural gas combustion. To support this opinion, types and number of factories located in Chiang Mai and Lamphun provinces as well as the energy consumption data were reviewed. In Chiang Mai province, most of the factories are located in these three districts,

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Table 7 Varimax rotated PC loading (only marker species are demonstrated)

Each PC was named after the sources which had highest (or comparatively high) content of those marker species in the emission profile (*italics*: highly presented in their PAHs profile, *bold*: highly presented in their inorganic species profile). *VEHIC* Vehicle exhaust, *SOIL* soil (mixture of soil and dust from construction), *GAS&COKE* natural gas combustion and coke oven, *ROAD* road dust (mixture of brake dust, tire dust and exhaust ash), *VEGET* vegetative burning, i.e. forest fire, agricultural waste burning, *COOK* fat and oil as well as fuel burning during cooking process, *SECOND* secondary particle (could result from the reaction between vehicle exhaust gas and vegetative burning), *REPAIR* particle emitted during car repairing, *WASTE* solid waste burning, *PAVE* particle emitted from the debris of pavement surface (mixture of brake ware and yellow paint), *HAZWASTE* hazardous waste incineration

Chiang Mai municipality (or Chiang Mai City, 587 factories), San Kamphaeng District (226 factories), and Saraphee District (195 factories). Among the total number (2,126) of factories located in this province, 115 of them are nonmetal industries (i.e. ceramic roof tiles, concrete

PAHs	Coal power plant ^a	Coke ovens ^b	Gasoline vehicle ^c	Diesel vehicle ^c	Natural gas burning ^d	Biomass burninge	Domestic wood burning ^f	Cooking ^g
PHE	15.1	13.2	3.8	17.1	0.2	7.0	11.1	0.8
ANT	1.7	2.6	0.4	2.3		1.0	41.8	NA
FLA	17.0	9.0	8.5	18.2	17.1	13.3	11.8	30.6
PYR	14.9	11.8	10.6	31.6	16.4	17.4	12.5	59.8
BaA	10.5	26.2	8.1	5.1	9.2	6.9	8.6	NA
CHR	20.3	7.4	16.2	13.9	44.5	25.9	6.5	3.0
BbF	7.8	13.9	12.4	4.0	5.3	6.6	2.6	1.3
B _k F	1.6	2.1	8.5	3.7	7.3	7.2	0.9	1.0
BaP	3.7	1.1	8.1	1.9	NA	7.1	2.2	0.1
IND	2.7	2.5	2.0	ND	NA	1.4	0.2	ND
DBA	0.5	0.9	1.4	ND	NA	0.7	0.03	NA
BPER	4.1	9.5	20.0	2.3	NA	5.4	1.7	3.5

Table 8 PAHs source profile (g/100gTotalPAHs)

NA Not ana[l](#page-20-0)yzed, *ND* none detected
^aLi et al. [\(2003](#page-20-0))
^bYang et al. (2002)

 ${}^{\text{c}}$ Rogge et a[l](#page-20-0). [\(1993a](#page-20-0))

 d Rogge et a[l](#page-20-0). [\(1993b](#page-21-0))
 e Rogge et al. [\(1998](#page-20-0))

^fWenborn et a[l](#page-21-0). (1<mark>999</mark>) gZ hao et a[l](#page-21-0). (2007)

block, pottery construction material); 88 of them are metal industries; and 22 factories are chemical industries (Chiang Mai Province Industrial Divisio[n](#page-19-0) [2003](#page-19-0)). According to the presentation document received from the Department of Alternative Energy Development and Efficiency, 14– 22% of total energy consumed by the above three types of industries are obtained from the natural gas (DED[E](#page-19-0) [2005](#page-19-0)). In the case of Lamphun Province, there are 833 factories (The Federation of Thai Industri[es](#page-21-0) [2007](#page-21-0)), 70 of which are the non-metal, metal and chemical industries. These factories could perform industrial combustion consuming considerable amounts of natural gas and coke. The high PC loading of PM10 also implied that the GAS&COKE had a great influence on PM10 level.

Road dust was also identified with 7.8% and explained variance as a source of PM10 due to the high loading of Cd, Mn, Sr and Ti in the fourth principal component (road dust (mixture of brake dust, tire dust and exhaust ash)). Mn, Ti and Sr were considered as a result of the tear of brake pads and tire surfaces while Cd could be emitted from various sources such as diesel

vehicle, aircraft, municipal solid waste combustion and chemical fertilizers (Tables [9](#page-14-0) and [10](#page-15-0)). These elements could accumulate on the road surfaces and be re-suspended into the air due to certain weather conditions.

The fifth principal component (vegetative burning, i.e. forest fire, agricultural waste burning, VEGET) has a high loading of PM10, Ba, K, Pb, K^+ with 7.4% which explained the variance. K and K^+ are well known as the markers of vegetative burning such as forest fires, field crop burning and other agricultural waste combustion (Watson et a[l](#page-21-0). [2001](#page-21-0); Muraleedharan et a[l](#page-20-0). [2000](#page-20-0); Wangkiat et a[l](#page-21-0). [2004](#page-21-0); Cachier et a[l](#page-19-0). [1995](#page-19-0)). VEGET should have a strong influence on PM10 concentrations due to the high PC loading of PM10.

The sixth principal component (fat and oil as well as fuel burning during cooking process) with a 7.4% variance was characterized by fluoranthene and pyrene, which could be heavily emitted during cooking (Table 8). Although the cooking process described in Table 8 is a spicy Sichuan style which may differ from the Thai style but the usage of spices and oil should be similar. This component was named as cooking particle.

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A high loading of SO_4^{2-} and NH_4^+ in the seventh principal component (secondary particle (could result from the reaction between vehicle exhaust gas and vegetative burning)) indicated secondary particulate matters. This source type does not represent any real source of emission but is the result of continuous oxidation of S(IV) to S(VI) taking place during atmospheric transport. The oxidation converts gaseous $SO₂$ to particulate sulphate such as $(NH_4)_2SO_4$. The high correlation coefficient $(r = 0.7 - 0.9)$ between SO₄² and NH₄⁴ concentration (mole/ $m³$) in three seasons excluding the wet season can be seen as the proof of this interpretation.

The eighth principal component (particle emitted during car repairing) with 6.1% variance was identified as the particle emitted from the vehicle repairing and painting. There are some vehicle repairing and painting shops in the vicinity of our four sampling stations. Heavy PC loading of Cu and P probably comes from the cupper– phosphorus alloy which was brushed out from the cars during the repairing and painting. The high correlation coefficient $(r = 0.9)$ between Cu and P indicates this interpretation is reasonable.

The ninth principal component (solid waste burning) was considered as particulate matters originated from solid waste burning. The dominant types, Na⁺ and Cl[−] with high PC loading, did not indicate sea salt because their correlation coefficients for each sampling site were less than 0.5. TC, Na⁺ and Cl[−] could also be emitted into the ambient air from waste burning as shown in Tables [9](#page-14-0) and [10](#page-15-0).

The high loading of Cr and Fe in the tenth principal component (particle emitted from the debris of pavement surface (mixture of brake ware and yellow paint)) implied a mixture of brake wear and pavement painting. This component probably represents pavement debris which was produced during vehicle braking and directly released into the air without accumulation (Tables [9](#page-14-0) and [10](#page-15-0)).

The last principal component (hazardous waste incineration) suggested the particles released from hazardous waste incineration. Major elements in the PC loading are Hg and TC which probably originated from waste incineration (Tables [9](#page-14-0) and [10](#page-15-0)).

Statistical results of regressing APCS on PM10 concentrations and on the total PAHs concentrations are summarized in Table 11. Assigning the value to Eq. 2, seasonal contributions of the possible source were derived as shown in Figs. [6](#page-17-0) and [7](#page-17-0).

From Fig. [6](#page-17-0), at our four sampling sites, vegetative burning accounted for more than 50% of PM10 concentration while 12–45% and 2–12% of the PM10 concentrations were apportioned to natural gas burning & coke ovens and secondary particle. Despite of the fact that the largest forest fires during study period occurred in March (Table [3](#page-7-0)), the year-round highest contributions of vegetative burning were observed in Trans2 period rather than the dry period. This finding can be explained as the result from strong ground-based inversion during dry season by which cold air near the ground surface is blocked under the warmer air layer above. Firstly it is important to notice that the inlets of our PM10 samplers were at 2 m above the ground level. Under the strong groundbased inversion layer, emissions from forest fires on the mountains and tall stacks of industrial

Case (dependent variables)	Source (independent) variables)	Coefficient	sig		R^2		
		Value confidence intervals	Lower boundary of 95% confidence intervals	Upper boundary of 95%			
PM10 $(\mu g/m^3)$	VEGET	0.0167	0.015	0.018		25.588	0.858
	GAS&COKE	0.0174	0.016	0.019		19.656	
	SECOND	0.0053	0.003	0.007	Ω	5.749	
PAHs $(ng/m3)$	GAS&COKE	0.002021	0.002	0.002		29.925	0.918
	VEHIC	0.001277	0.001	0.001		21.625	
	VEGET	0.0006504	0.001	0.001		11.724	

Table 11 Coefficient of multiple regressing models in this study

plants were presumed to take place above the inversion height (Karppinen et a[l](#page-20-0). [2001](#page-20-0)), and these therefore do not substantially influence pollution near the ground level (0–15 m above the ground). This could be the reason for having higher contributions of vegetative burning in Trans2, a period after the ground-based inversion in the dry season had just ended. In the dry season, it is also noticed that natural gas burning & coke oven contribution to PM10 concentrations at Saraphee and Kaikew had almost the same value of those from vegetative burning. This result implied that most of the

Fig. 7 Source contributions of PM10-bound PAHs concentration

natural gas burning & coke ovens in this study occurred at the ground level. During the situation in which the lowest part of the atmosphere is very stable, even average emission volume from the ground-level source is sufficient to maintain an air quality episode (Mäkelä et a[l](#page-20-0). [1998](#page-20-0)). The low stacks of small family-based factories located in Saraphee and Kaikeaw community area, i.e. ceramic ware manufacturing, electroplating, glass manufacturing, which include natural gas burning or coke ovens in the manufacturing process are then postulated as the members of GAS & COKE group.

According to Fig. [7](#page-17-0), no significant spatial variation of PAHs contribution were observed. Here, vegetative burning was ranked as a minor PAHs source due to its being the lowest PAHs contribution. The particulate matter emitted from vegetative burning may have lower PAHs content (μgPAHs/gParticle) than that from the other sources. On the other hand, vehicle exhaust was found to be the major source of the total PAHs concentration during wet, transition1 and transition 2 periods (49–60%) while natural gas burning & coke ovens made the largest contribution to the total PAHs concentration in dry season (47– 59%). These findings also agreed well with the occurrence of a strong ground-based inversion layer in the dry months. Without the formation of a ground-based inversion layer, PAHs from vehicle exhaust of which the releasing point is less than 1 m height from the ground, would possibly remain near the our sampling device in high concentration compared to those emitted from

> \bullet 0.02 0.04 0.06 0.08 0.10 0.12 0.14 0.16 0.18 0.20

observed PM10 concentration (microgram/cubic meter)

observed PM10 concentration (microgram/cubic meter) low stacks of the small factories. However, when the ground-based inversion was formed in the dry period, even the average emission load from GAS & COKE group which are also the ground level sources could strongly result in high PAHs concentration. For this reason, natural gas burning & coke ovens turned out to make the largest contribution to PAHs in the dry season while vehicle exhaust was noted from the results as the major PAHs contributor in the wet season and transition periods. Natural gas burning & coke ovens (15–37%) and vegetative burning (13–37%) were also recognized as the second and third contributors of PAHs during wet months and transition periods. In the dry season, (1) natural gas burning & coke ovens, (2) vehicle exhaust and (3) vegetative burning accounted for 47–59%, 20–25%, and 20–25% of total PAHs concentrations, respectively.

The above findings emphasized the importance of vegetative burning as the major contributor to PM10 concentrations in Chiang Mai and Lamphun ambient air. However, the results revealed that the natural gas burning & coke ovens as well as the vehicle exhaust also deserve careful attention in pollution control strategies because of their largest contributions to the PM10 bond PAHs.

The Q–Q plots of the observed pollutant concentrations and calculated data based on the multiple regression model were shown in Fig. 8. The agreement between the measured and predicted concentrations of PM10 and PM10-bound PAHs $(R^2 > 10.8)$ suggests that the PCA/APCS receptor

Fig. 8 Q-Q plot comparing observed pollutant concentrations and calculated data

0.02

0.02

0.03

 $X = Y$

 $X = Y$

model is applicable for estimation of source contributions to ambient PM10 and PM10-bound PAHs.

Conclusions

PM10 concentration detected at all stations shared the same pattern, in which it increased at the beginning of dry season (December) and reached the peak in March before decreased by the end of April. The average of daily PM10 concentration and percentages of numbers of days that PM10 concentrations exceeded the 24-h national standard value pointed out the urgency for the responsible authority in Chiang Mai and Lamphun provinces to develop more effective strategies to control the particle pollution during the dry season. Medium level of negative correlation between PM10 concentration and visibility data suggested the visibility as a possible indicator for the particle pollution-watch system. The warning visibility values for the PM10 pollution-watch system were determined as 10 km for Chiang Mai and 5 km for Lamphun provinces.

Higher ambient concentrations of Al, Ca, Fe, K, Si and Zn $(\mu g/m^3)$, especially in the dry season, indicated the priority of these metals to be taken as matters of concern during PM10 pollution crisis. No single component exceeded the national air quality standard. Total concentrations $(ng/m³)$ of 16 PAHs ranged from 0.05 to 22.4 ng/m³, which is in the similar level compared to those measured by other researchers. No meteorological parameters could be used as a warning index for the toxic PAHs pollution-watch system. Average TTEC of Chiang Mai and Lamphun ambient air was ranked at a low level compared to that observed in other cities. Based on the yearly average TTEC, the annual number of lung cancer cases for Chiang Mai and Lamphun provinces was estimated at two cases/year which was lower than the those in the cases of Bangkok (27 cases/year). Since TTEC and PM10 concentration are not related to each other, the conventional Thai air quality standard is considered insufficient. A new criterion for the control of carcinogenic component, i.e., PAHs is needed in order to protect the residents from longterm health problems.

The results of source apportionment emphasized the importance of vegetative burning as the major contributor to PM10 concentration in Chiang Mai and Lamphun ambient air. However, careful attention on the natural gas burning & coke ovens as well as the vehicle exhaust is also necessary because of their large contributions to PAHs concentrations. No significant spatial variation of PAHs contribution was observed. The measured concentrations of PM10 and PM10-bound PAHs are in good accord with the predicted concentrations $(R^2 > 0.8)$. Thus, the PCA/APCS receptor model is considered applicable for estimation of source contributions to ambient PM10 and PM10-bound PAHs.

Acknowledgments Our sincere thanks to the following agencies: Thailand Research Fund for providing us with the financial support, the Pollution Control Department for providing the sampling devices and related data, the Chiang Mai and Lamphun Meteorological Center for providing us the Meteorological data, Chiang Mai University and NRC-EWHEM for providing the other research equipment.

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