Atmospheric Pollution Research xxx (2017) 1–12

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Atmospheric Pollution Research

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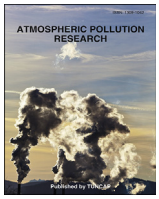

Effect of agricultural waste burning season on $PM_{2.5}$-bound polycyclic aromatic hydrocarbon (PAH) levels in Northern Thailand

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ARTICLE INFO

Article history:
Received 22 January 2017
Received in revised form
10 April 2017
Accepted 17 April 2017
Available online xxx

Keywords:
Agricultural waste burning
Biomass burning
PAHs
Northern Thailand
$PM_{2.5}$

ABSTRACT

Smoke from agricultural waste burning causes hazardous haze pollution in Southeast Asian countries every year. Besides traces of a few toxic gases several persistent organic pollutants, natural, and anthropogenic emissions are released, resulting in adverse health effects. The study focuses on identification of sources of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons (PAHs) during the non-haze and haze seasons and investigates the concentration and toxicity of fine particulate matter, especially in the middle of biomass burning period. Totally, 12 fine particulate PAH congeners were selected and analysed at air quality observatory sites located in nine administrative provinces of Northern Thailand during four different sampling periods from 7th December 2012 to 27th March 2015. Nisbet and Lagoy's toxicity equivalent concentration (*TEQ*) equation revealed that the $B[a]P_{Equivalent}$ value for 2014–2015 roughly surpassed the value of 2012–2013 by a factor of 17. This can be attributed to the significant rise in five-to six-ring PAHs levels in the past two years. Diagnostic binary ratios and linear regression analysis highlight the roles of vehicular exhausts and biomass burning as two major contributors of $PM_{2.5}$-bound PAHs. Interestingly, principal component analysis (PCA) reveals similar loading patterns for PC1 during the non-haze and haze periods, indicating that agricultural waste burning cannot be considered as the sole contributor of particulate PAHs in Northern Thailand.

1. Introduction

Over the past decades, increasing international attention had been paid to fine particles released as a consequence of open biomass burning (agricultural waste, sugarcane, rice straw, and other crops) prior to the harvest season, normally performed to clean land quickly before the next crop cycle (Godoi et al., 2004; Mugica-Alvarez et al., 2015; Pongpiachan, 2016; Pongpiachan et al., 2015a; Vassura et al., 2014). Amongst numerous toxic pollutants emitted during biomass burning, polycyclic aromatic hydrocarbons (PAHs) are of major concern due to their genotoxic effects and adverse impact on human health (Liao et al., 2011; Matsui, 2008; Wickramasinghe et al., 2012). Several studies have investigated the chemical profile of particulate PAHs from various sources such as motorcycle exhausts (Pham et al., 2013; Seggiani et al., 2012), vehicle releases (Chellam et al., 2005; Fraser et al., 1998; He et al., 2006; Phuleria et al., 2006), shipping emissions (Pongpiachan et al., 2015b), and various types of biomass burning (Bari et al., 2011; Hays et al., 2005; Kim Oanh et al., 2015; Lisouza et al., 2011; Park et al., 2013; Rajput et al., 2011; Sanchis et al., 2014; Yang et al., 2006). Additionally, epidemiological studies on agricultural waste burning in Northern part of Thailand have been conducted (Pongpiachan and Paowa, 2014; Pongpiachan, 2016). Pongpiachan (2014b) investigated $PM_{2.5}$ levels from samples collected during haze episodes at three different air quality observatory sites in Chiang-Mai city using Fourier transform infrared spectroscopy (FTIR) technique. Pongpiachan et al. (2015a) were the first to present comprehensive data regarding $PM_{2.5}$-bound PAH samples from the nine northern administrative provinces (NNP) of Thailand during non-haze and haze periods in 2012–2013. Since no statistically significant differences were observed in $PM_{2.5}$-bound

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Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.

http://dx.doi.org/10.1016/j.apr.2017.04.009

PAHs before and after the haze episode, the authors indicated the probable role of vehicular exhausts as a regular source of $PM_{2.5}$-bound PAHs in the NNP. Previous studies have highlighted the impact of vehicular exhausts as major contributors of five- and six-ring PAHs (Chellam et al., 2005; Fraser et al., 1998; Rogge et al., 1993; Zielinska et al., 2004). However, numerous confounding factors such as atmospheric chemical reactions (Wu et al., 2007) and variation in the PAH emission profile itself (Miguel et al., 1998) can alter the chemical compositions of PAHs. Despite numerous studies associated with the fine particles released from biomass burning, only a few studies have focused on identifying sources of $PM_{2.5}$-bound PAHs. Furthermore, limited studies have assessed both the contents of $PM_{2.5}$-bound PAHs and its toxicity (i.e. B[*a*]P equivalent concentrations) particularly during haze episodes when potential risk of human exposure is strongly suspected to be high. Overall, the principle goals of this study are to *i*) investigate the annual variation of chemical compositions of $PM_{2.5}$-bound PAHs before and after a haze episode using dataset from 2012 to 2015 and *ii*) apply diagnostic binary ratios, linear regression analysis (LRA), and principal component analysis (PCA) to identify potential sources of PAHs during agricultural waste burning periods in the NNP.

2. Methods

2.1. Description of sampling campaigns

Sampling method described by the United States – Environment Protection Agency's Quality Assurance Guidance Document (US-EPA, 2002) was used in the study. Twenty-four hour time-integrated $PM_{2.5}$ samples were collected every seven days (from 9:00 a.m. until 9:00 a.m. the next day) on pre-baked (550 °C for 12 h) quartz-fiber filters (QFFs; Whatman 47 mm; Article No. 28418542 (US reference)) using MiniVolTM portable air samplers (Airmetrics) with a flow rate of 5 L min^{-1} through a particle size separator (impactor) and subsequently passed through a 47 mm filter. The weight of fine particles was measured using microbalances (Mettler Toledo, New Classic MF, MS205DU, Switzerland) as per procedure described by the United States Environmental Protection Agency Office of Air Quality Planning and Standards (US-EPA, 1998).

All nine sampling sites are geologically grouped by numerous mountainous belts and are predominantly characterized by tropical savannah vegetation. Four monitoring campaigns were conducted at all the meteorological stations of Pollution Control Department (PCD), Ministry of Natural Resources and Environment (MNRE) listed in Fig. 1. The address and geographical co-ordinates of the stations are available in Appendix A. Sampling equipment were strategically positioned to ensure exposure to winds from all directions, ensuring absence of obstructions in the vicinity.

Air quality observatory programs were categorized into two groups based on the sampling period. Phases I and III were conducted before the agricultural waste burning episode in the winter of 2012 (Phase-I; 7th to 22nd December 2012) and 2014 (Phase-III; 16th December 2014 to 10th January 2015), while air quality observations during Phases II and IV were conducted in 2013 (Phase-II; 23rd February to 20th March 2013) and 2015 (2nd to 27th March 2015). According to the moderate resolution imaging spectroradiometer (MOUDIS) fire images produced by the US NASA Goddard Space Flight Center (available at http://earthobservatory.nasa.gov/NaturalHazards/category.php?cat_id=8), scattered fire spots were detected at distances ranging from a few kilometres around the observatory sites to several kilometres adjacent to the border of Thailand and Myanmar (CMOS and MHOS), Golden Triangle region (CROS), and Thailand and Laos (PYOS and NPOS).

2.2. Analysis of polycyclic aromatic hydrocarbons (PAHs)

The analytical method employed for the determination of $PM_{2.5}$-bound PAHs has been described in detail by Gogou et al. (1996). Briefly, QFFs containing the $PM_{2.5}$ were extracted using Soxhlet extraction with dichloromethane (DCM; Fisher Scientific, HPLC grade). QFFs were spiked with known amounts of two deuterated internal standards (d_{10}-fluorene and d_{12}-perylene) prior to extraction. The extract was rotary evaporated to reduce the volume of DCM and subsequently dried using a nitrogen stream. Finally, it was substituted using hexane (Fisher Scientific, HPLC grade) and evaporated with nitrogen prior to fractionation using flash chromatography with silica gel and solvents of increasing polarity. Details of PAHs fractionation methodology can be found in previous studies by Pongpiachan et al. (2013a, b, 2015a, b) (See Section 2.2 in Supplementary Material).

2.3. Toxicity equivalent concentration (TEQ)

The toxicity equivalent concentration (*TEQ*) equation is widely employed for investigating the risk of exposure to PAHs, and can be computed as below (Pongpiachan et al., 2013a; Pongpiachan, 2016):

$$TEQ = \sum_i [C_i \times TEF_i] \quad (1)$$

where, C_i and TEF_i are concentrations of individual PAHs and the toxic equivalency factors, respectively. By applying *TEF*, the toxicity of a mixture of PAH congeners can be comprehensively calculated as a single number, the toxic equivalency *TEQ*. In this study, the *TEQ* equation proposed by Nisbet and LaGoy (1992) (Eq. (2)), described below was used.

$$TEQ_{Nisbet\ and\ Lagoy} = 0.001(Phe + Fluo + Pyr) + 0.01(An + B[g,h,i]P + Chry) + 0.1(B[a]A + B[b]F + B[k]F + Ind) + B[a]P + D[a,h]A. \quad (2)$$

3. Results and discussion

3.1. Atmospheric concentrations of $PM_{2.5}$-bound PAHs

$PM_{2.5}$-bound PAHs were identified in four different monitoring campaigns described in section 2.1. Table 1 summarizes the statistical analysis of mass concentrations of the selected PAH congeners measured in the 36 samples taken at the nine air quality observatory sites. Σ3,4-rings PAHs and Σ5,6-rings PAHs represent the sum of Phe, An, Fluo, Pyr, B[a]A, Chry + Tri and B[$b + k$]F, B[*e*]P, B[*a*]P, Ind, D[*a,h*]A, B[*g,h,i*]P, respectively.

In the absence of major anthropogenic emission sources such as factories and incinerators in the immediate vicinity of the sampling locations, vehicular exhausts, biomass burning, and soil dust are probably the most significant sources of fine particulate pollutants in these areas (Pongpiachan et al., 2013b, 2015a). Additionally, CMOS is located in the city center of Chiang-Mai province, a tourist spot subject to heavy traffic, where vehicular exhausts can be considered as the main source of air pollutants (Pongpiachan, 2013b). Burning aerosols play an important role in governing air quality of most rural parts of Phayao, Mae Hong Son, Uttaradit, and Phrae provinces, particularly in the middle of haze period (Pongpiachan et al., 2013b, 2015a). Consequently, imperfect

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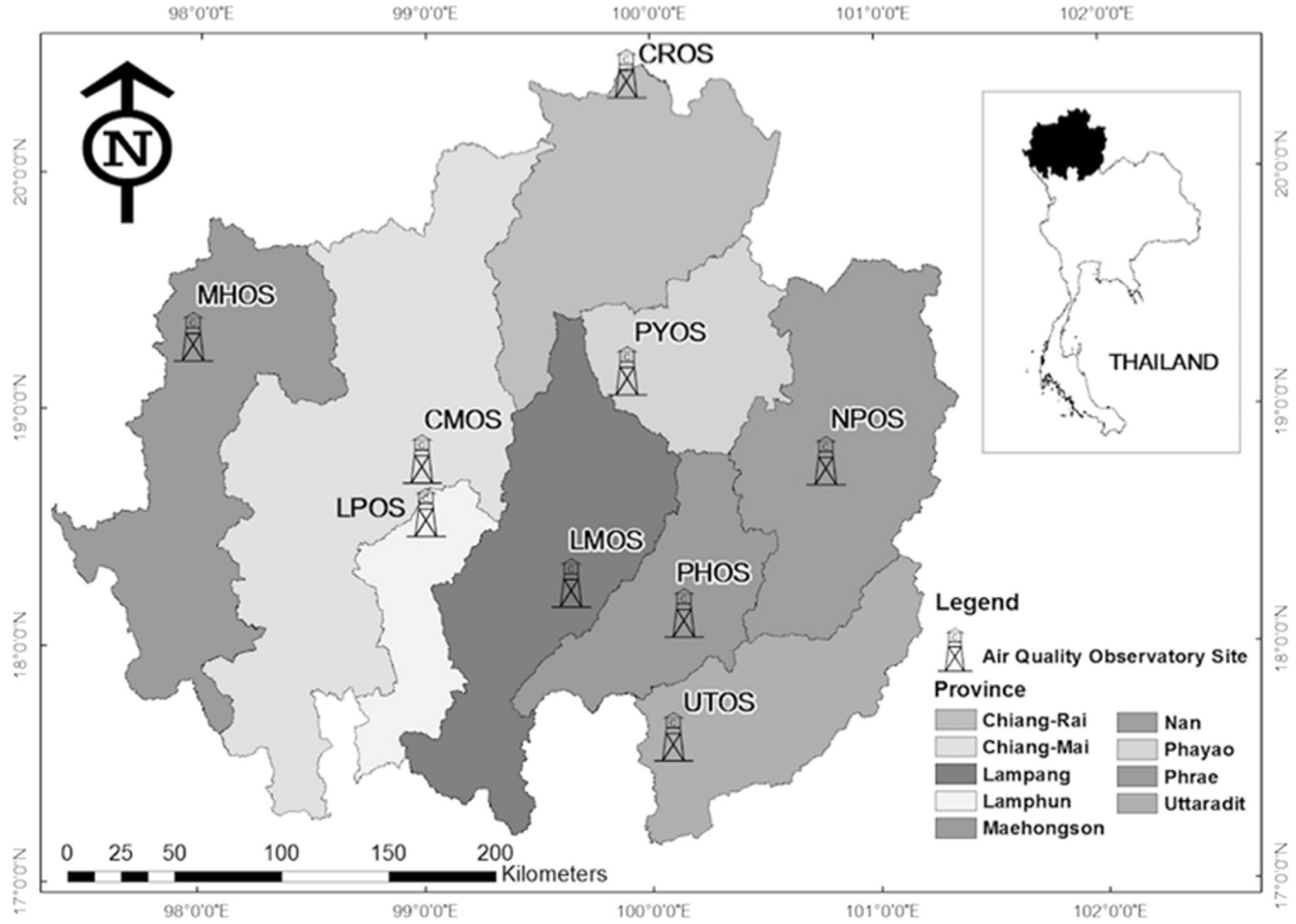

Fig. 1. Map showing location of air quality observatory sites in the nine administrative provinces of Northern Thailand (Table S.1). Please see the following references for more details of air quality observatory sites (Pongpiachan, 2016; Pongpiachan et al., 2015a).

Table 1
Statistical analysis of PM2.5-bounded PAHs [pg m^{-3}] collected from the nine administrative provinces of Northern Thailand.

PAH Congener	Phase-I-2012 Non-haze	Phase-II-2013 Haze	Phase-III-2014 Non-Haze	Phase-IV-2015 Haze	*t*-Test ($p < 0.05$)	ANOVA-Test ($p < 0.05$)	IARC Class[a]
	$n = 9$	$n = 9$	$n = 9$	$n = 9$			
Ph	188 ± 355	185 ± 465	179 ± 516	N.D.		N.S.	3
An	57 ± 68	18 ± 15	45 ± 75	25 ± 42		N.S.	3
Fluo	84 ± 184	81 ± 191	100 ± 195	19 ± 41		N.S.	3
Pyr	182 ± 467	260 ± 696	156 ± 439	36 ± 50		N.S.	3
B[*a*]A	**461 ± 1372**	**337 ± 990**	**183 ± 509**	**11 ± 29**		N.S.	**2A**
Chry	333 ± 986	478 ± 1387	243 ± 313	254 ± 506		N.S.	3
B[*b* + *k*]F	13 ± 25	83 ± 142	4238 ± 5027	3087 ± 3120		S.	2B
B[*e*]P	1767 ± 3246	967 ± 1855	1468 ± 1990	631 ± 770		N.S.	3
B[*a*]P	**17 ± 27**	**7 ± 22**	**775 ± 1647**	**595 ± 1190**		N.S.	**2A**
Ind	5 ± 14	7 ± 21	N.D.	N.D.	N.S.		2B
D[*a*,*h*]A	**N.D.**	**N.D.**	**N.D.**	**N.D.**			**2A**
B[*g*,*h*,*i*]P	7 ± 20	8 ± 24	N.D.	N.D.	S.		3
$B[a]P_{Equivalent}$	69 ± 140	55 ± 103	1220 ± 1723	908 ± 1231		N.S.	
Σ3,4-ring PAHs	1305 ± 1799	1359 ± 1908	906 ± 927	346 ± 512		N.S.	
Σ5,6-ring PAHs	1809 ± 3246	1072 ± 1861	6481 ± 5652	4313 ± 3427		S.	
Fluo/(Fluo + Pyr)	0.32 ± 1.07	0.24 ± 0.85	0.39 ± 1.33	0.35 ± 0.87		N.S.	
B[*a*]A/(B[*a*]A + Chry)	0.58 ± 2.44	0.41 ± 1.71	0.43 ± 1.32	0.043 ± 0.14		N.S.	
B[*a*]P/B[*e*]P	0.010 ± 0.023	0.0072 ± 0.027	0.53 ± 1.33	0.94 ± 2.2		N.S.	
Σ3,4-/5,6-rings PAHs	0.72 ± 1.63	1.27 ± 2.83	0.14 ± 0.19	0.080 ± 0.13		N.S.	

Notes: PAHs probably carcinogenic to humans are highlighted in bold.
[a] Classification for carcinogenic risk by IARC (2010): 1: carcinogenic to humans; 2A: probably carcinogenic to humans; 2B: possibly carcinogenic to humans; 3: not classifiable.

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combustions of biomass can also be regarded as regular emission sources of particulate PAHs in these sites.

For all sampling periods, particulate-phase distributions of PAHs were dominated by high molecular weight (HMW) PAHs such as B[*b*+*k*]F, B[*e*]P, B[*a*]P, and Chry with mean concentrations of 1855 pg m^{-3}, 1208 pg m^{-3}, 349 pg m^{-3}, and 327 pg m^{-3}, respectively. This was followed by relatively high contributions of B[*a*]A, Pyr, and Ph with mean concentrations of 248 pg m^{-3}, 159 pg m^{-3} and 138 pg m^{-3}, respectively. Noticeably, fine particles during the none-haze season, B[*b*+*k*]F with a mean concentration of 2126 ± 5027 pg m^{-3} comprised 41% of the total. HMW PAHs levels were also significant during the haze season with mean concentrations of B[*b* + *k*]F, B[*e*]P, Chry, and B[*a*]P at 1585 ± 3123 pg m^{-3}, 799 ± 2008 pg m^{-3}, 366 ± 1476 pg m^{-3}, and 301 ± 1191 pg m^{-3}, respectively. As displayed in Table 1, only B[*b* + *k*]F showed significantly high concentration ($p < 0.05$) in Phase-III (4238 ± 5027 pg m^{-3}), followed by Phase-IV (3087 ± 3120 pg m^{-3}).

Similarity in distribution patterns of $PM_{2.5}$-bound PAHs' percentage contribution during the periods 2012–2013 and 2014–2015 were observed during the study. B[*e*]P, Chry, B[*a*]A, Pyr, and Ph levels constituted 49%, 16%, 15%, 8.4%, and 6.8%, respectively (Table 1 and Fig. 2) during the 2012–2013. Interestingly, B[*b*+*k*]F and B[*e*]P levels alone constituted 79% of the samples collected from 2014 to 2015. Σ5,6-ring PAHs obtained during 2014–2015 was significantly higher than 2012–2013 levels irrespective of the season. The magnitude of Σ5,6-ring PAHs during the period of 2014–2015 increased by a factor of 3.8 when compared to 2012–2013. Previous studies have highlighted that vehicular exhausts are major contributors of Σ5,6-ring PAHs (Chellam et al., 2005; Fraser et al., 1998; Rogge et al., 1993; Zielinska et al., 2004). However, numerous confounding factors such as atmospheric chemical reactions (Wu et al., 2007), and a variation in the PAH emission profile itself (Miguel et al., 1998) can alter the chemical compositions of PAHs. In spite of the fact that OH radical reactions predominate the fate of semi-volatile organic compounds (SVOCs) in the tropospheric atmosphere, NO_2 and NO_3 radicals coupled with O_3 can also react with organic compounds of which contain an unsaturated C=C bound (Aschmann et al., 2001). These heterogeneous chemical reactions perplexed the clarification of PAH degradation mechanisms particularly in tropical countries. It is also crucial to note that under high NO_x condition, O_3 is the final product as a consequence of CH_4 and CO oxidation cycle, and it is this which generates an OH radical (Pongpiachan, 2006). Previous studies have been highlighted the importance of biomass burnings as a major source of trace gaseous species (Cheng et al., 1998; Cofer et al., 1998; Miranda et al., 1994). Therefore, one can consider that the degradation mechanism of PAHs in northern Thailand during the haze episode should dramatically differ from those of mid- and high latitude areas.

Toxicity of a combination of PAH congeners collected from the NNP during different periods was calculated as a single number using the $B[a]P_{Equivalent}$ (Table 1 and Fig. 3). Due to the comparatively high *TEF* value of B[*b*+*k*]F (0.1), its existence in Phases III and IV significantly surpassed other congeners with the contributions of 91% and 93%, respectively.

$B[a]P_{Equivalent}$ value during the period 2014–2015 roughly increased by a factor of 17, compared to $B[a]P_{Equivalent}$ value of 2012–2013. B[*a*]P concentrations in all phases ranged from N.D. to 5095 pg m^{-3} and with an average of 349 pg m^{-3}. This value is much lower than the guideline limits of annual B[*a*]P concentrations of

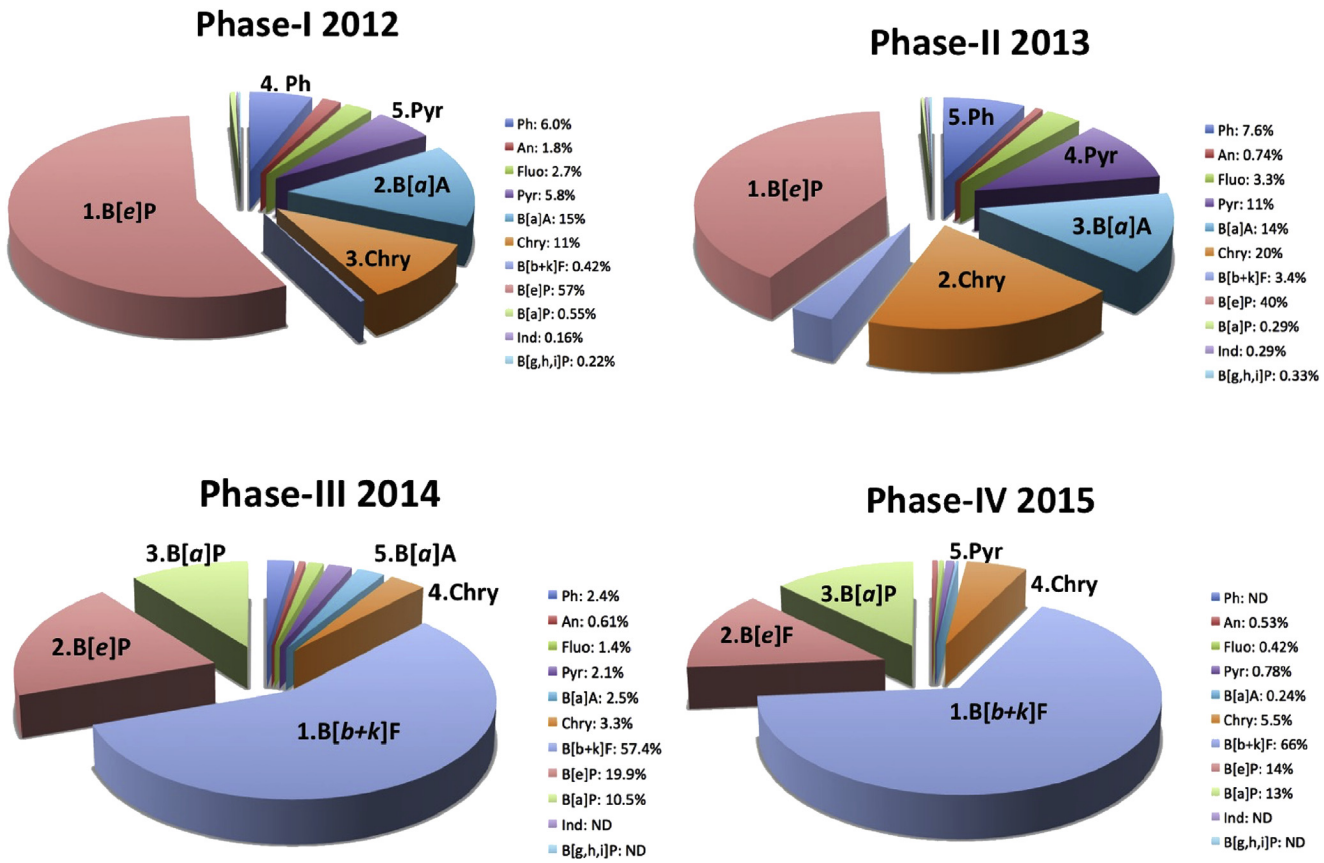

Fig. 2. Percentage contribution of PM2.5-bound PAHs collected at nine administrative provinces of Northern Thailand during four different periods.

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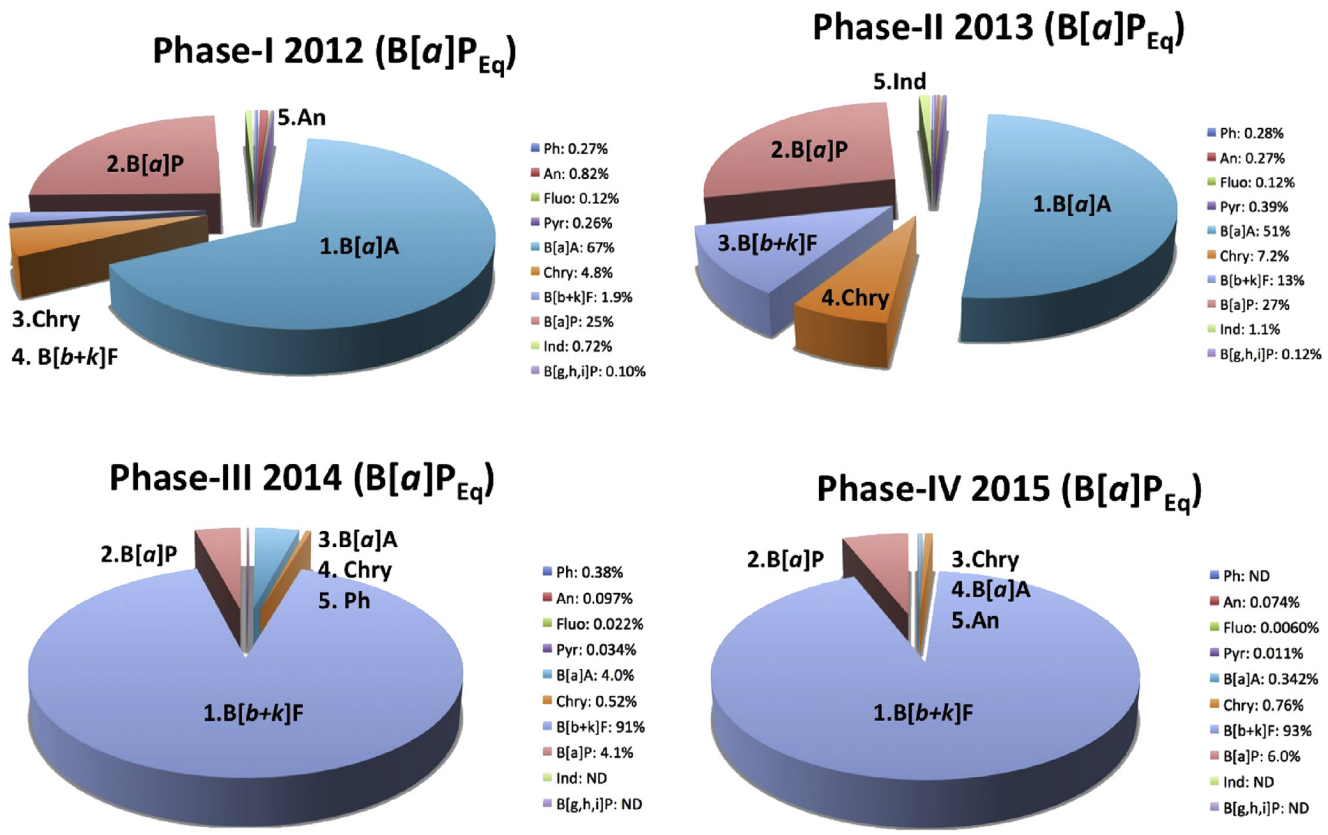

Fig. 3. Percentage contribution of B[a]PEq of $PM_{2.5}$-bound PAHs collected at nine administrative provinces of Northern Thailand during four different periods calculated using Nisbet and Lagoy's equation.

10 ng m^{-3} and 1.0 ng m^{-3} proposed by the German Federal Environmental Agency and the European Union, respectively (Baek et al., 1992). Furthermore, the average values of Phases III and IV (685 pg m^{-3}) was 2.74 times higher than the value of 250 pg m^{-3} proposed by the UK Expert Panel of Air Quality Standard (UK EPAQS, 1998). This indicates that the most recent results from the NNP are still outside acceptable limits despite variable standards across Europe. In addition, this value is comparable to the concentrations reported in Jojutla-Zacatepec Morelos, Mexico (462 pg m^{-3}); Saitama, Japan (520 pg m^{-3}); and Gothenberg, Sweden (540 pg m^{-3}), but much lower than the concentrations reported in Guangzhou, China (1820 pg m^{-3}); Houston, USA (1880 pg m^{-3}); and Augburg, Germany (3270 pg m^{-3}) (Chellam et al., 2005; Gao et al., 2011; Mugica-Alvarez et al., 2015; Naser et al., 2008; Schnelle-Kreis et al., 2005). In addition, it is crucial to note that the $B[a]P_{Equivalent}$ value obtained from Phase-IV-2015 Haze Period (i.e. 908 pg m^{-3}) is almost comparable to previous studies conducted in Chiang-Mai (910 pg m^{-3}) and Bangkok (820 pg m^{-3}), underlining the importance of vehicular exhausts as the main contributors of PAHs irrespective of the season of biomass burnings (Pongpiachan, 2013b,c).

3.2. Diagnostic binary ratios

During the past decades, diagnostic binary ratios of PAHs have been employed for investigating relative discrimination ability (i.e. relative thermodynamic stability) of various types of parent PAHs, uniqueness of different PAH emission sources, and alteration of PAH profiles in sediments (Yunker et al., 2002, 2011; Yunker and Macdonald, 2003). In this study, three types of categories were applied as characteristic chemical proxies to identify probable emission sources (Guo et al., 2003; Pongpiachan et al., 2015a, b; Ravindra et al., 2006). The categories are:

- B[*a*]A/(B[*a*]A + Chry) *vs.* B[*a*]P/(B[*a*]P + B[*e*]P) (Category-I),
- Fluo/(Fluo + Pyr) *vs.* An/(An + Phe) (Category-II), and
- Ind/(Ind + B[*g,h,i*]P) *vs.* B[*b*+*k*]F/Ind (Category-III).

Several types of diagnostic binary ratios have been employed to distinguish petrogenic PAHs from pyrogenic emission sources. For instance, a petrogenic PAH usually possesses a binary ratio of An/(An + Ph) less than 0.1 whilst biomass and coal combustion generally release a binary ratio of Fluo/(Fluo + Pyr) greater than 0.5 (Yunker et al., 2002). Ind/(Ind + B[*g,h,i*]P) and B[*a*]A/(B[*a*]A + Chry) ratios higher than 0.5 indicate that the emission was most likely influenced by the imperfect combustions of coal, grass, and wood (Yunker et al., 2002).

As illustrated in Fig. 4, diagnostic binary ratios of PAHs in fine particles collected from different types of vehicles (e.g. Light Duty Vehicle (LDV), Heavy Duty Vehicle (HDV), motorcycles) with various numbers of fuels (e.g. bioethanol-gasoline fuel blends) and engine types (e.g. four-stroke) as well as numerous biogenic species (e.g. rice straw, sugarcane, crop residues) were plotted and compared with those of all phases (Tables 1 and 2, and Fig. 4).

Noticeable features in all proxy categories were:

(i) Category-I clearly highlights different sources for samples from Phases I and-II and Phases III and IV. While Phases-I and II highly deviate from the group of motorcycle exhausts, LDV/HDV emissions, and rice-straw burning, Phases III and IV are close to B3 (a single cylinder four-stroke motorcycle), C1, C2

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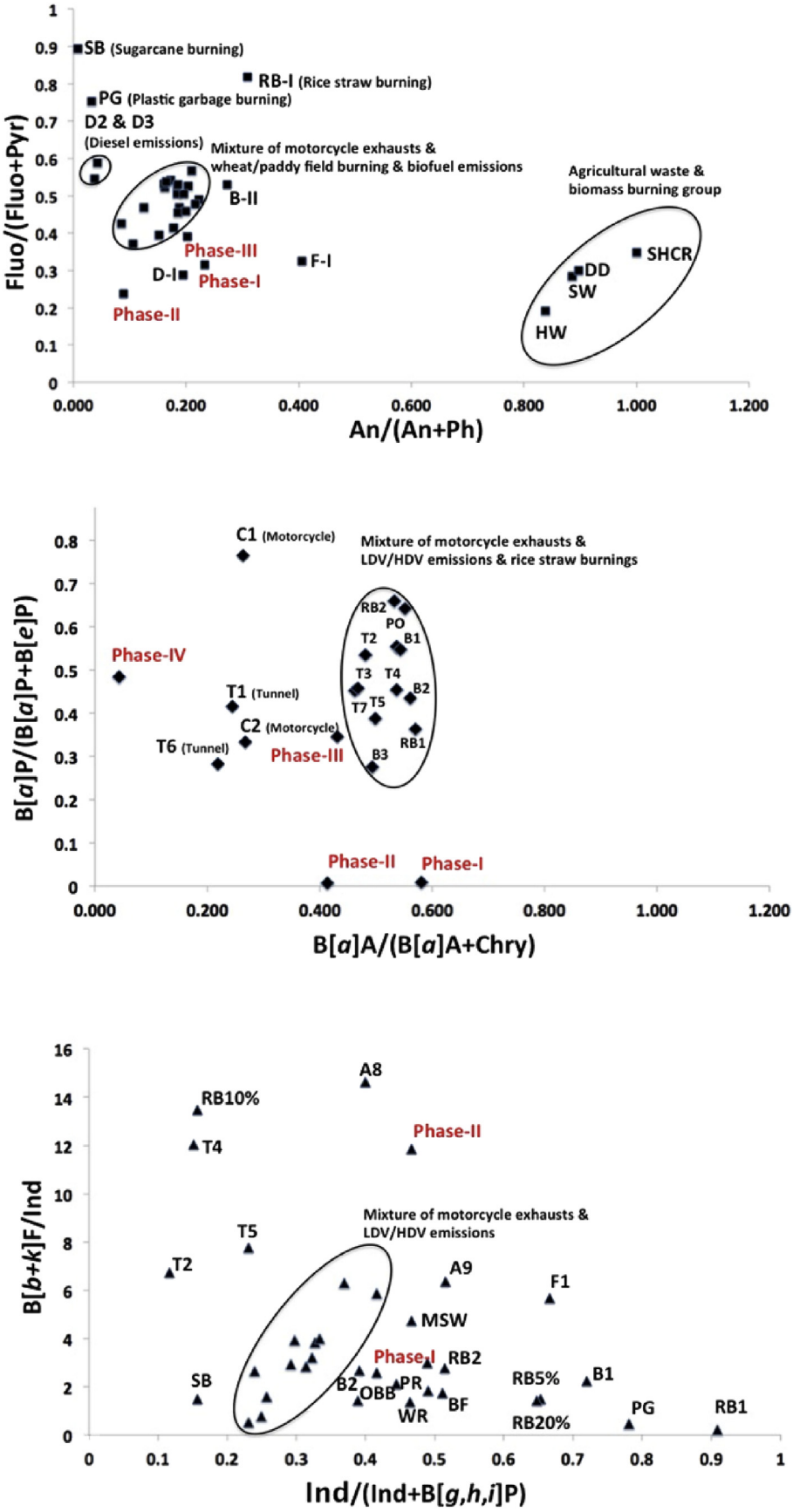

Fig. 4. Diagnostic binary ratios of $PM_{2.5}$-bound PAHs in the NNP.

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Table 2
Slopes, intercepts, and correlation coefficients (R2) of individual PAH congener vs. TPAHs (Total PAHs) for $PM_{2.5}$ collected from urban areas and biomass burning. Values higher than 0.8 have been highlighted in bold.

	Urban Atmosphere				Biomass Burning			
	Slope	Intercept	R^2	n	Slope	Intercept	R^2	n
Ph *vs.* TPAHs	27.4	0.298	0.570	24	5.88	6.14	**0.846**	17
An *vs.* TPAHs	188	−2.10	**0.837**	24	11.4	8.65	0.542	18
Fluo *vs.* TPAHs	11.4	0.372	0.622	31	4.87	7.85	0.712	19
Pyr *vs.* TPAHs	10.4	−0.482	0.660	34	5.56	7.08	0.747	19
B[*a*]A *vs.* TPAHs	11.3	7.76	**0.881**	34	7.26	5.65	0.694	16
Chry *vs.* TPAHs	11.8	−2.65	**0.933**	32	6.69	5.83	0.704	19
B[*b*+*k*]F *vs.* TPAHs	0.458	14.9	0.0134	33	5.82	1.65	0.731	19
B[*e*]P *vs.* TPAHs	11.1	−4.24	**0.970**	23	16.5	−4.71	0.699	15
B[*a*]P *vs.* TPAHs	8.46	5.87	**0.961**	32	11.6	1.37	**0.865**	19
Ind *vs.* TPAHs	6.69	6.19	**0.848**	30	7.66	2.42	0.605	17
B[*g,h,i*]P *vs.* TPAHs	6.61	3.39	**0.932**	29	7.50	3.93	0.646	17

(six catalyst-equipped vehicle), T1, T3,T6, T5, T7 (a mix fleet of LDV and HDV inside tunnels);

(ii) Category-II highlights two-dimensional (2D) plots of agricultural waste and biomass burning group including shrubs and crops residues, dry cow dung, perennial indigenous and fast growing exotic tree species that are highly deviated from other samples in both seasons. Additionally, Phase-I, Phase-II, and Phase-III are adjacent to the group of motorcycle exhausts, wheat/paddy field burning, and bio-fuel emissions;

(iii) Category-III shows comparatively high dispersion of 2D plots for rice straw burning with three different moisture contents (RB5%, RB10%, and RB20%), plastic garbage combustion (PG), and fugitive emissions from fuel (F1). Phase-I is located close to paddy residue burning (PR), rice straw burning (RS), and open burning of bonfires (OBB).

Moreover, Phase-I was located next to the mixture of motorcycle exhausts and LDV/HDV emissions. In spite of some discrepancies in the application of different types of diagnostic binary ratios, it appears that both agricultural waste burnings (i.e. rice straw and paddy field burning) and vehicle exhausts are two major sources of $PM_{2.5}$-bound PAHs in northern of Thailand regardless of the season. Certain diagnostic binary ratios were more useful and sensitive in identifying PAHs from specific sources in the NNP. Fluo/(Fluo + Pyr) and An/(An + Phe) ratios were good indicators in the identification of PAHs originating from agricultural waste and biomass burnings, as seen in Category-II (Fig. 4). Similarly, Ind/(Ind + B[*g,h,i*]P) and B[*b*+*k*]F/Ind ratios successfully identified mixture of particulate PAHs released from motorcycles, LDV, and HDV. However, B[*a*]A/(B[*a*]A + Chry) and B[*a*]P/(B[*a*]P + B[*e*]P) ratios have not been useful in identification of PAHs emitted from vehicular exhausts and rice-straw burning.

3.3. Linear-regression analysis (LRA)

Slopes, intercepts, and correlation coefficients (R^2) of LRA of six different combinations of PAHs (Ph *vs.* An, Fluo *vs.* Pyr, B[*a*]A *vs.* Chry, B[*b*+*k*]F *vs.* B[*e*]P, and B[*a*]P *vs.* Ind) were calculated using data of $PM_{2.5}$-bound PAHs from the present study along with data from earlier studies conducted in urban cities subject to heavy traffic and types of biomass burning, and are presented in Figs. 5 and 6, respectively.

Caution was exercised during data interpretation as several meteorological parameters such as air temperature, wind speed, relative humidity, and the physicochemical properties of aerosol itself could have dramatically alter the LRA variables. Despite these uncertainties, R^2 of six different combinations were comparatively high with average values of 0.87 ± 0.058 and 0.82 ± 0.16 for $PM_{2.5}$-bound PAHs collected from urban cities and agricultural waste burnings, respectively. The relatively strong positive correlation coefficients of two similar structures of PAHs underline the roles of vehicular exhausts and biomass burning as two major sources of particulate PAHs in the NNP. These findings are also in agreement with data interpreted from diagnostic binary ratios in section 3.2. Further attempts to identify major contributors of particulate PAHs in northern Thailand were made by plotting individual PAH congeners against the total quantified PAHs (TPAHs). As illustrated in Table 2, the R^2 of 11 different combinations were considerably high with the average values of 0.75 ± 0.28 and 0.71 ± 0.094 for particulate PAHs obtained from high traffic urban cities and biomass combustions, respectively. It appears reasonable to conclude that An, B[*a*]A, Chry, B[*e*]P, B[*a*]P, Ind, and B[*g,h,i*]P are mainly affected by traffic emissions due to their comparatively high R^2 values (>0.8), while B[*b*+*k*]F showed the lowest R^2 value of 0.013 indicating that it was not affected significantly by vehicular exhausts. On the contrary, R^2 of B[*b*+*k*]F derived from agricultural waste burning showed a moderate positive value of 0.73. Since B[*b*+*k*]F was the dominant PAH congener detected in $PM_{2.5}$ collected during Phase-III (57%) and Phase-IV (66%), it would be safe to consider biomass burning as the major source of B[*b*+*k*]F in the NNP from 2014 to 2015.

3.4. Principal component analysis (PCA)

The multivariate analytical tool PCA was used to extract a small number of latent factors (principal components, PCs) from a set of original variables (detected $PM_{2.5}$-bound PAHs) for investigating correlations between the measured PAHs. Data submitted for the analysis were arranged in a matrix, where each column corresponded to one PAH congener and each row represented the number of the $PM_{2.5}$ sample. Data matrixes were evaluated using PCA allowing the summarized data to be further interpreted. PCA was employed to evaluate the influences of biomass combustions and traffic exhausts on chemical compositions of particulate PAHs (Table 3).

Varimax rotation was used to classify three principal components based on the PAH loadings and accounted for 89.1% and 82.9% of the total variance (i.e. PC1+PC2+PC3) for particulate PAHs collected during the non-haze and haze periods, respectively. Moreover, total variances of PC1 were 2.3 and 3.6 times higher than PC2 during the non-haze and haze periods, respectively. PC1 showed high loading (>0.8) on medium molecular weight (MMW) PAHs (Ph, Fluo, Pyr, B[*a*]A, Chry) observed for both periods. Since there are no major differences on PC1 loading patterns between the two periods, it seems rational to conclude that biomass burning plays a minor role on $PM_{2.5}$-bound PAHs observed at the NNP.

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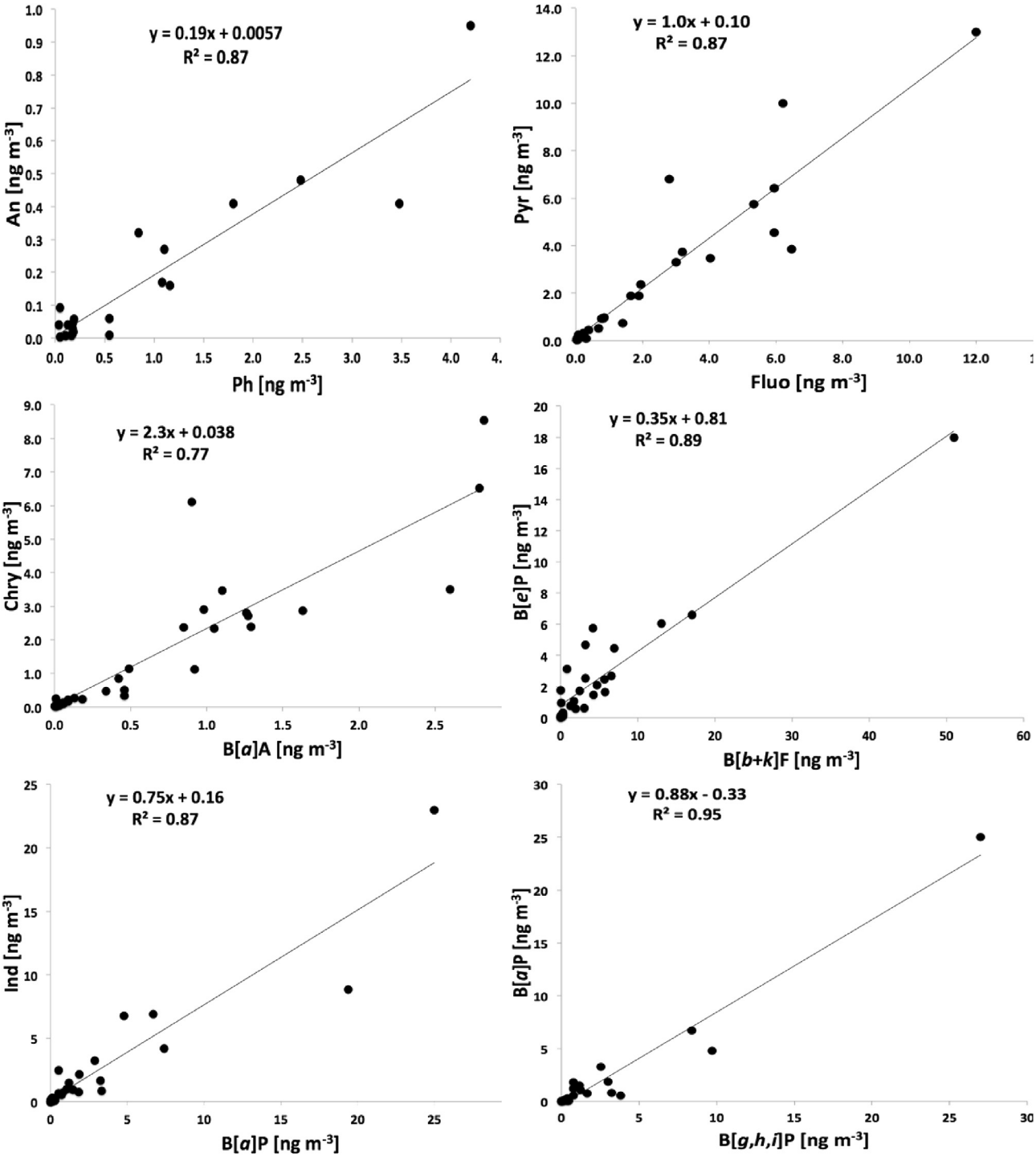

Fig. 5. Linear regression analysis of $PM_{2.5}$-bound PAHs in the NNP (Phases I-IV), in comparison with other urban areas (Afghanistan (Wingfors et al., 2011), Belgium (Ravindra et al., 2006), Brazil (Bourotte et al., 2005; Silva et al., 2010), China (Deng et al., 2006; Gao et al., 2011; Hou et al., 2006; Li et al., 2005; Wang et al., 2006), Germany (Schnelle-Kreis et al., 2005), Hong Kong (Guo et al., 2003), Italy (Vassura et al., 2014), Japan (Naser et al., 2008), Singapore (Karthikeyan et al., 2006), Spain (Barrado et al., 2012; Reche et al., 2012), Sweden (Wingfors et al., 2001, 2011), Taiwan (Fang et al., 2005), USA (Chellam et al., 2005; Poor et al., 2004)).

Previous studies found high levels of Pyr, Fluo, and Ph in emissions from incineration (Smith and Harrison, 1998; Ravindra et al., 2006) and oil combustion (Harrison et al., 1996; Ravindra et al., 2006). Additionally, mobile emissions particularly from diesel trucks are significant sources of MMW PAHs such as Ph, Fluo, and Pyr (Harrison et al., 1996), while rice straw burnings can dramatically increase the contents of particulate Ph, Fluo, B[*a*]A, and Pyr (Korenaga et al., 2001; Sanchis et al., 2014; Yang et al., 2006). Consequently, incineration, rice straw burning and vehicular exhaust are probably three main sources found in PC1 for both periods. Interestingly, PC2 loading patterns show relatively high positive loading (>0.6) of B[*b*+*k*]F and B[*a*]P detected during both seasons with total variances of 22.5% and 15.3% for non-haze and haze periods, respectively (Table 3). Early investigations indicate

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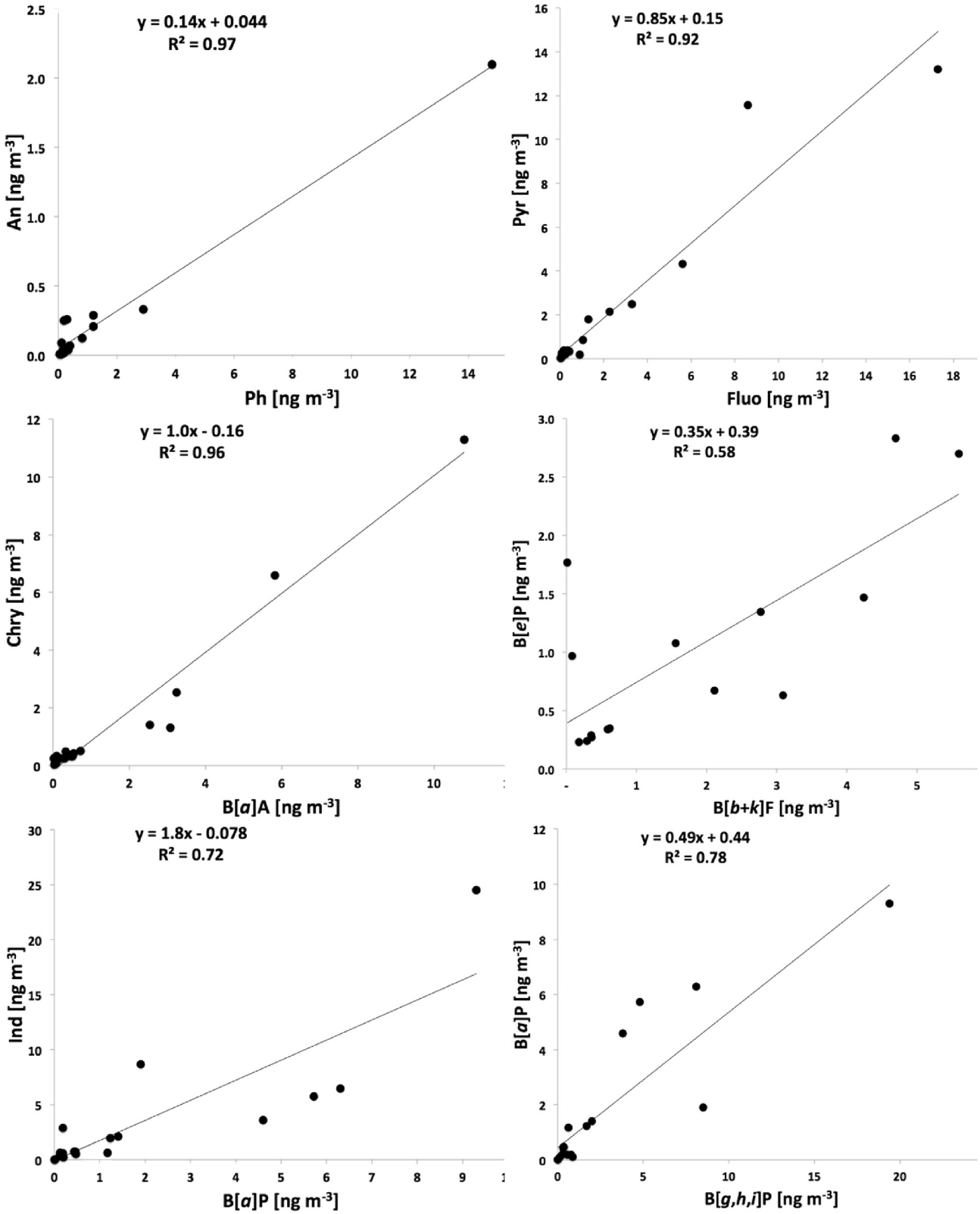

Fig. 6. Linear regression analysis of PM2.5-bound PAHs in the NNP (Phases I-IV), in comparison with other biomass/agricultural waste burnings (agricultural waste/biomass burnings (Pongpiachan, 2014a), bonfire (Pongpiachan, 2013a; Vassura et al., 2014), domestic heating (Ma et al., 2010), and sugarcane open burning (Godoi et al., 2004; Mugica-Alvarez et al., 2015; Silva et al., 2010)).

sugarcane burning as major contributors of particulate B[*b*+*k*]F and B[*a*]P (Mugica-Alvarez et al., 2015; Souza et al., 2014) and thus it would be safe to regard PC2 as resulting from post-harvest agricultural waste burning. It is also well known that diesel emission

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Table 3
Principal components (PC) pattern for Varimax rotated components applied to PM2.5-bound PAHs from the NNP during the haze (Phases II and IV) and non-haze (Phases I and III) periods. Values higher than 0.5 have been highlighted in bold.

	Non-Haze Period			Haze Period		
	Principal Component			Principal Component		
	PC1	PC2	PC3	PC1	PC2	PC3
Ph	**0.897**	−0.092	0.403	**0.983**	−0.074	−0.099
An	0.196	−0.019	**0.917**	−0.159	0.276	0.441
Fluo	**0.940**	−0.025	0.315	**0.992**	−0.092	0.021
Pyr	**0.969**	−0.068	0.204	**0.986**	−0.063	−0.110
B[*a*]A	**0.969**	−0.041	−0.125	**0.989**	−0.055	−0.096
Chry	**0.937**	0.030	−0.278	**0.969**	−0.055	0.007
B[*b*+*k*]F	−0.114	**0.912**	−0.121	−0.044	**0.690**	0.484
B[*e*]P	0.349	0.537	−0.413	0.037	−0.178	**0.860**
B[*a*]P	−0.114	**0.900**	0.134	−0.066	**0.860**	−0.178
% of Variance	52.6	22.5	14.0	55.2	15.3	12.4

has a high factor loading of An (Ho et al., 2002; Omar et al., 2002; Ravindra et al., 2006). Therefore, the strong positive loading of An (0.917) found in PC3 can be attributed to the 14% contribution of diesel emissions during non-haze period.

In conclusion, all data analysis (i.e. diagnostic binary ratios, LRA, and PCA) highlighted the importance of traffic emissions as one of the major contributors of PAH congeners in northern Thailand irrespective of haze period. These findings are in good agreement with the increasing number of vehicle registered in Thailand. According to the reports of Transport Statistics Sub-Division, Planning Division, Department of Land Transport, significant enhancement of vehicles was observed since 2012. For instance, total vehicle number under motor vehicle act was 33,520,175 in 2013 while the number increased up to 36,912,488 in 2016. The number of vehicles has increased considerably not only in the city but also in the rural area in the past five years as a consequence of the so-called first-car buyer scheme in Thailand, which was an incentive introduced by Yingluck Shinawatra's government (see Table. S.2 in Supplementary Material). The scheme offered tax rebates of up to 100,000 baht to customers who purchased passenger vehicles with a maximum engine capacity of 1500 cc or pickup trucks with unlimited engine capacity, but priced within 1 million baht. In addition, wind speed and wind direction profiles of nine air quality observatory sites were obtained from Global Climate Station Summaries reported by National Oceanic and Atmospheric Administration (NOAA) as can be seen in Fig. S.1-S.18 (see Supplementary Material). It is worth mentioning that wind profiles were found to be consistent through out the four different sampling campaigns at nine air quality observatory sites. Therefore, it appears rationale to conclude that vehicle exhausts are one of the major contributors of particulate PAH congeners in northern part of Thailand.

4. Conclusions

Despite some disagreements on applying diagnostic binary ratios, correlations observed during the study support the contention that both vehicular exhausts and biomass combustion are the two major contributors of $PM_{2.5}$-bound PAHs. The PCA results of non-haze period reveal contributions of 52.6% from rice straw burning/vehicular exhausts/incinerations, 22.5% from sugarcane combustion, and 14.0% from diesel emissions. Similarly, the PCA results of haze period constitute 55.2% mobile emissions and 15.3% agricultural waste burning from sugarcane plantations. Since the B[*a*]$P_{Equivalent}$ value for period of 2014–2015 surpassed that of 2012–2013 by a factor of 17, the government policy should target both indirect and direct strategies with greater focus on minimization and elimination of paddy field burning and other agricultural waste combustion. In addition, it is also crucial to focus on finding realistic policy tools for reducing air pollutants from mobile emissions such as increasing vehicle tax, building bicycle lanes, and facilitating utilization of public transportation.

Acknowledgement

This project was financed by National Institute of Development Administration (NIDA) Research Center. The authors acknowledge Dr. Songsirin Ruenvisesh from Department of Nutrition and Food Science, Texas A&M University, College Station, TX USA for her kind assistances on some literature reviews. The authors also thank assistance of Ms. Panatda Kanchai and the local staff from Pollution Control Department, Ministry of Natural Resources and Environment for providing sampling and meteorological data. The authors appreciate Dr. Danai Tipmanee for his work on factor analysis.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.04.009.

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Please cite this article in press as: Pongpiachan, S., et al., Effect of agricultural waste burning season on $PM_{2.5}$-bound polycyclic aromatic hydrocarbon (PAH) levels in Northern Thailand, Atmospheric Pollution Research (2017), http://dx.doi.org/10.1016/j.apr.2017.04.009

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