Atmospheric Pollution Research xxx (2017) 1–12

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Atmospheric Pollution Research

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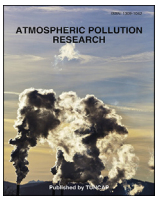

Assessing human exposure to PM_{10}-bound polycyclic aromatic hydrocarbons during fireworks displays

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ARTICLE INFO

Article history:
Received 15 December 2016
Received in revised form
29 January 2017
Accepted 30 January 2017
Available online xxx

Keywords:
PAHs
Firework display
Diagnostic binary ratios
Risk assessment
Incremental lifetime cancer risk

ABSTRACT

The "*Loy Krathong*" festival is a major annual Thai event that includes firework displays. It takes place on the evening of the full moon in the 12th month of the traditional Thai lunar calendar. Since fireworks are widely considered a major source of PAHs, it is considered reasonable to expect a significant increase in PAH levels during this event. The overall PAH profile at the six air quality observatories operated by the Pollution Control Department (PCD), Ministry of Natural Resources and Environment (MNRE), showed that the Kingdom of Thailand's atmosphere was dominated by 5–6-ring PAHs during the firework display period. A significant increase in ΣPAHs (153%) was observed during firework displays. A statistical analysis coupled with the application of diagnostic binary PAH ratios was conducted to determine whether the detected increase in PAH congeners during the festival period was due to firework combustion or whether it was a coincidental effect caused by vehicular exhausts, long-range atmospheric transportation, photolysis and chemical degradation. The average incremental lifetime cancer risk (ILCR) values of adults and children living in Bangkok as estimated by three different TEQs for ingestion, dermal contact, and inhalation exposure pathways were greatly lesser than the US EPA baseline, further highlighting that the cancer risk of bonfire night falls into the "acceptable level" range.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), particularly benzo[*a*]pyrene (B[*a*]P), have been extensively recognized as carcinogenic substances that are also responsible for endocrine disruption, and reproductive and developmental effects (Hoyer, 2001; Liao et al., 2011; Matsui, 2008; Wickramasinghe et al., 2012). As a consequence, several field studies have attempted to elucidate the role of weather conditions in the behaviour of PAHs in cities around the world (Akyüz and Çabuk, 2009; Amodio et al., 2009; Choi et al., 2012; Massei et al., 2003; Pongpiachan, 2013a,b,c; Tham et al., 2008; Zhang and Tao, 2008). It is crucial to emphasize that PAHs arise from various types of emission sources, including traffic emissions, domestic heating, industrial activities, agricultural waste and biomass burnings and forest fires (Li et al., 1999; Lu et al., 2012; Okuda et al., 2002; Rajput et al., 2011; Riva et al., 2011; Slezakova et al., 2011; Yang et al., 2002). Despite numerous studies of the annual and seasonal variations of PAH, their fate as tropical aerosols is still uncertain, particularly in Southeast Asian nations where information on atmospheric PAHs is limited.

Bangkok city, situated in the Chao Phraya River delta in Central Thailand, has an administrative area of 1569 km^2, making it the 73rd largest city in the world. By applying Nielsen's technique, previous studies have highlighted traffic emissions as the most important source of atmospheric PAHs in Bangkok (Pongpiachan et al., 2013a,b). Nielsen (1996) investigated the influence of

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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.2017.01.014

vehicular emission to particulate PAH contents at three air quality observatory sites, namely, a street in Central Copenhagen, a site in a city park and a bus street in Central Aarhus. Since a site in city park positions only a few hundreds of meters from the street, it seems rationale to ignore the impacts of non-traffic sources on PAHs aerosol concentrations coupled with atmospheric chemical and/or physical processes. As a consequence, the only difference in particulate PAH contents between city and park site could be attributed to vehicular exhausts. By adopting the differences in PAH concentrations between two sites, vehicular exhaust profiles of PAHs were computed.

In this study, the average values of the incremental Incremental Lifetime Cancer Risk (*ILCR*) for three different sampling sites fell between 10^{-7} and 10^{-6}, close to the acceptable risk level (10^{-6}) but much lower than the priority risk level (10^{-4}) (Pongpiachan, 2013a). A further investigation was carried out by collecting PM_{10} samples at seven Pollution Control Department (PCD) air quality observatories in Bangkok (Pongpiachan et al., 2013a). Results from Principal Component Analysis (PCA) showed a 47% contribution from both mutagenic and carcinogenic emissions (i.e. PC1 + PC2) at all observatories, emphasizing that traffic emissions are the main contributors to PAHs and other chlorinated dioxins in Bangkok's atmosphere (Pongpiachan et al., 2013a).

Despite numerous studies highlighting the importance of vehicular exhausts in atmospheric PAH contents in Bangkok, no study has focused on the impact of the "*Loy Krathong*" festival (LKF) on the enhancement of particulate PAHs. *Loy Krathong*, which means "to float a basket", is a traditional festival celebrated annually throughout Thailand and certain parts of Southeast Asia (e.g. Malaysia, Laos and Burma). The floating lanterns and fireworks used in the LKF are public concerns causing both environmental damage and adverse health effects. Over the past few years, several studies have underlined the LKF as a major source of PAHs in the urban atmosphere (Pongpiachan, 2013c; Sarkar et al., 2010; Vassura et al., 2014). A previous study highlighted the importance of firework displays as a major contributor to particulate PAHs associated with biomass combustion during the Chinese New Year (CNY) Festival (Shi et al., 2014). Enhanced PAH levels were detected during CNY as a result of cooking emissions on CNY Eve (Li et al., 2009; Feng et al., 2012; Shi et al., 2014), but the fireworks display for the Beijing Olympics opening ceremony rehearsal on the night of August 3, 2008 also dramatically affected PAH source profiles (Wu et al., 2014a,b). Additionally, numerous reports using the source apportionment technique coupled with diagnostic binary PAH ratios have underlined vehicular exhausts as the principle source of PAH emissions, regardless of firework display effects, in the ambient urban atmosphere (Feng et al., 2012; Harrad and Laurie, 2005). It is therefore clear that more information is required to clarify the impact of firework displays on the PAH profile.

Overall, the major aims of this research are to (*i*) study the factors affecting the diagnostic binary PAH ratios for PM_{10} during the LKF, (*ii*) investigate the impact of firework displays on the enhancement of PM_{10}-bound PAH concentrations and (*iii*) conduct a risk assessment regarding particulate PAH levels before and after firework events at the heart of the Bangkok metropolitan area.

2. Materials & methods

2.1. PM_{10} observatories and sample collection

Six Pollution Control Department (PCD) air sample observatories were chosen for the assessment of PM_{10} PAH concentrations (see Fig. 1): the Public Relation Department Observatory Site (PDOS), Bansomdejchaopraya Rajabhat University Observatory Site (BROS), Ramkhamhaeng Conjunction Observatory Site (RCOS), Land Development Department Observatory Site (LDOS), Maboonkrong Conjunction Observatory Site (MCOS) and Victory Monument Observatory Site (VMOS). Intensive monitoring campaigns were performed consecutively before and after the LKF, Father's Day (5th of December), and New Year's Eve celebrations between 2011 and 2014, forming a database of 64 individual air samples. The total numbers of air samples obtained before and after the firework display were 53 and 11 samples, respectively. Graseby-Anderson high volume air samplers, TE-6001, were employed to achieve unmanned 24-h samplings for PM_{10} at the six PCD air quality observatories. A total of 64 air samples were acquired using high volume samples of approximately 1632 m^3 for each 24 h sample. PM_{10} samples were collected on 20 cm × 25 cm Whatman Glass Fibre Filters (GFFs) at a flow rate of about 1.133 m^3 min^{-1} (i.e. 40 cfm). The sample air flow rate was calibrated for standard temperature and pressure conditions. A more comprehensive explanation of the air sampling method is given in "Compendium Method IO-2.2. Sampling of Ambient Air for PM_{10} using an Andersen Dichotomous Sampler" (U.S. EPA, 1998). It is important to note that there are two sampling campaigns namely the Fireworks Display Period (FDP: i.e. the sum of samples collected during LFK, Father's Day, and New Year's Eve celebration) and Non-Fireworks Display Period (NDP: i.e. the sum of samples collected during non-FDP period).

2.2. Polycyclic aromatic hydrocarbons (PAHs)

Half of each GFF was extracted with dichloromethane (Fisher Scientific, HPLC grade) using Soxhlet extraction. The GFFs were spiked with a known amount of deuterated internal standards (including d_{10}-fluorene and d_{12}-perylene) before extraction. The organic extract was vacuum concentrated and evaporated by nitrogen stream to a state of almost complete dryness. This was eventually replaced with hexane and then evaporated again with nitrogen until a final volume of 2 mL was reached prior to the fractionation process. The hexane extract was fractionated by flash chromatography with silica gel, using various solvents of increasing polarity. Details of the PAH fractionation methodology can be found in Pongpiachan, 2006 and Tipmanee et al., 2012. The PAH fraction was eluted with 15 mL of 4:6 (v/v) toluene:hexane, concentrated using nitrogen stream until almost dry, and subsequently altered to cyclohexane and evaporated under gentle nitrogen stream until a final volume of 100 μl was achieved.

All solvents were HPLC grade, purchased from Fisher Scientific. A mix of standard solutions of 15 native PAHs [Norwegian Standard (NS 9815: S-4008-100-T): Phenanthrene (Phe), Anthracene (An), Fluoranthene (Fluo), 11H-Benzo[*a*]Fluorene (11H-B[*a*]F), 11H-Benzo[*b*]Fluorene (11H-B[*b*]F), Pyrene (Pyr), Benz[*a*]Anthracene (B[*a*]A), Chrysene (Chry), Benzo[*b*]Fluoranthene (B[*b*]F), Benzo[*k*]Fluoranthene (B[*k*]F), Benzo[*a*]Pyrene (B[*a*]P), Benzo[*e*]Pyrene (B[*e*]P), Indeno[1,2,3-*c,d*]pyrene (Ind), Dibenz[*a,h*]Anthracene (D[*a,h*]A) and Benzo[*g,h,i*]Perylene (B[*g,h,i*]P)], and a mix of recovery internal standard (IS) PAHs [d_{12}-Perylene (d_{12}-Per) and d_{10}-Fluorene (d_{10}-Fl)] were purchased from Chiron AS (Stiklestadveine 1, N-7041 Trondheim, Norway). Standard stock solutions of deuterated and native PAHs were prepared in nonane. Working solutions were obtained through appropriate dilution in cyclohexane. All 15 PAHs were isolated on a 60 m length × 0.25 mm i.d. capillary column coated with a film of 0.25 m thickness (phase composition: cross-linked/surface bonded 5% phenyl, 95% methylpolysiloxane with Agilent JW Scientific DB- 5 GC columns). Helium (99.999%) was used as the carrier gas at a constant column flow rate of 1.0 mL min^{-1} with a pressure pulse of 25 psi over 0.50 min. All injections (1 L) were conducted using a universal injector in splitless mode and standards were introduced using a 10 L Hamilton

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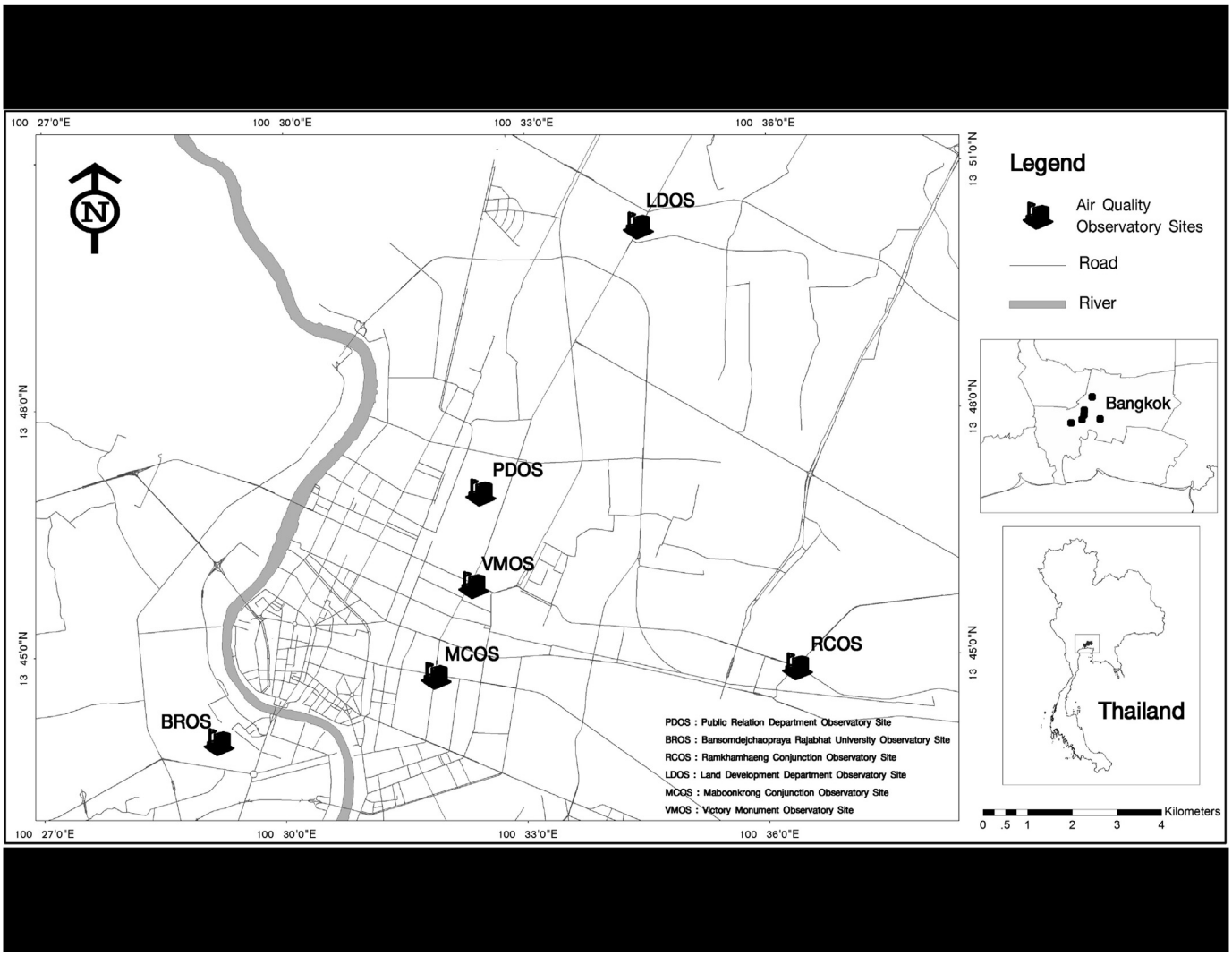

Fig. 1. Map of Pollution Control Department (PCD) air quality observatory sites used in this study.

syringe. The GC oven temperature was set for 1 min at 40 °C, then heated at 8 °C min^{-1} to 300 °C and then held at this temperature for 45 min. A Shimadzu GCMS-QP2010 Ultra system was used, comprising a high-speed performance system with ASSP (Advanced Scanning Speed Protocol) function (i.e. achieving a maximum scan speed of 20,000 u sec^{-1}) and ultra-fast data acquisition for comprehensive two-dimensional gas chromatography (GC × GC). A more comprehensive explanation of the GC/MS method is given in Pongpiachan et al., 2009, 2011. Both precision and accuracy were tested by employing standard reference material (SRM, 1941b) provided by the National Institute of Standard and Technology (NIST). The precision of the procedure was calculated based on the relative standard deviation of duplicate samples, which was less than 10% for 13 PAH congeners (i.e. Phe, An, Fluo, Pyr, B[*a*]A, Chry, B[*b*]F, B[k]F, B[*a*]P, B[e]P, Ind, D[a,h]A and B[g,h,i]P). All sample concentrations were calculated using standardized Relative Response Factors (RRFs) run with each batch (Pongpiachan et al., 2009, 2011).

2.3. Health risk assessment of PAHs

2.3.1. Toxicity equivalent concentration (TEQ)

Since B[*a*]P has been recognized as one of the most toxic PAH congeners, the carcinogenicity of individual obtained PM_{10} air samples was also quantified in terms of its B[*a*]P equivalent concentration ($B[a]P_{eq}$). The total carcinogenic potency of PAH exposure was subsequently calculated as the sum of each individual $B[a]P_{eq}$. To estimate the B[*a*]Peq for each individual PAH congener, the toxicity equivalent concentration (*TEQ*) equation, which is widely adopted for assessing the risk of exposure to PAHs, must be applied. It is calculated as described below (Yang et al., 2007; Yu et al., 2008; Pongpiachan et al., 2013b):

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

In this study, C_i and TEF_i represent the concentrations of individual PAHs and toxic equivalency factors, respectively. It is worth mentioning that the toxicity of a single PAH compound can vary by orders of magnitude. As a consequence, it is crucial to normalize the carcinogenicity of PAHs in terms of their most toxic form, B[*a*]P. By adopting *TEF*, the toxicity of a mixture of PAH compounds can be recalculated as a single number, typically referred to as toxic equivalency, *TEQ*. *TEQ* is a specific number resulting from the product of the concentrations and individual *TEF* values of each PAH, as described in Eq. (1). In this research, three *TEQ* equations were used, based on Nibset and LaGoy (1992) (Eq. (2)), U.S. EPA

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(1993) (Eq. (3)) and Cecinato (1997) (Eq. (4)). In these three *TEQ* formulas, the abbreviations for PAH congeners represent their concentrations (pg m^{-3}).

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

$$TEQ_{US-EPA} = 0.06(B[a]A) + 0.07(B[b]F + B[k]F) + B[a]P + 0.08(Ind) + 0.6(D[a,h]A) \quad (3)$$

$$TEQ_{Cecinato} = 0.01(Chry) + 0.1(B[a]A + B[b]F + B[k]F + Ind) + B[a]P + D[a,h]A \quad (4)$$

Additionally Σ3,4-ring PAHs and Σ5,6-ring PAHs stand for the sum of Phe, An, Fluo, Pyr, B[*a*]A, Chry + Tri and B[*b* + *k*]F, and of B[*e*]P, B[*a*]P, Ind, D[*a*,*h*]A and B[*g*,*h*,*i*]P, respectively.

2.3.2. Risk assessment of PM_{10}-bound PAH intake in preschool children during the Loy Krathong festival

The cancer risks related to non-dietary ingestion of PM_{10}-bound PAHs were estimated for preschool children living adjacent to PCD air quality observatories using Eq. (5) (Maertens et al., 2008; U.S. EPA, 1997).

$$Cancer\ risk = \sum_{i=1}^{n}\left(\frac{(TEQ_i) \times IR \times EF \times SF \times AF}{BW} \times CF\right) \quad (5)$$

where *TEQ* stands for toxicity equivalent concentrations based on the procedures explained by Nibset and LaGoy (1992) (Eq. (2)), U.S. EPA (1998) (Eq. (3)), and Cecinato (1997) (Eq. (4)). Two ingestion rates (*IR*) for indoor PM_{10} (mg day^{-1}) were used in this study: 50 mg day^{-1} and 100 mg day^{-1} (Maertens et al., 2008). *EF*, *SF*, *AF* and *BW* respectively represent the exposure factor (with a mean fraction taken over the 70-year period during which preschool children are believed to be exposed to PM_{10} via non-dietary ingestion), the slope factor (with employment of an oral slope factor for B[*a*]P of 7.3 mg kg^{-1} day^{-1} (U.S. EPA, 2005)), an adjustment factor (an adjustment factor of 5.8 was employed for exposure occurring during the early life stages of preschool children, as proposed by Maertens et al., 2008) and body weight (this study assumes an average body weight for preschool children of 15 kg (U.S. EPA, 1997)). In addition, *CF* stands for the conversion factor (10^{-6}).

2.3.3. Daily exposure to PM_{10}-bound PAHs

Calculations of daily exposure to PM_{10}-bound PAHs were conducted using three different *TEQ* values, as described in section 2.3.1.

$$M_{PDI-ingestion} = \sum_{13}\frac{TEQ_{PM10} \times M_{DID}}{1,000} \quad (6)$$

M_{DID} (mg day^{-1}) represents the daily intake of dust by either adults or children. In this study, M_{DID} values of 4.16–100 mg day^{-1} and 55–200 mg day^{-1} were applied for adults and children, respectively (U.S. EPA, 1997). 4.16 mg day^{-1} and 55 mg day^{-1} were assumed to represent a comparatively low exposure situation for adults and children, respectively, while the corresponding high exposure limits were set at 100 mg day^{-1} and 200 mg day^{-1}. A similar concept was applied in the calculation of daily exposure via inhalation to PM_{10}-bound *TEQs* ($M_{PDI\text{-}inhalation}$; ng day^{-1}) using Eq. (7):

$$M_{PDI-inhalation} = IR_{inhalation}\sum_{13} TEQ_{PM10} \quad (7)$$

where (ng m^{-3}) is the sum of the *TEQ* contents of the 13 PAHs measured in PM_{10}. The inhalation rate ($IR_{inhalation}$) established for the purposes of this research was 10 m^3 day^{-1} and 20 m^3 day^{-1} for children and adults, respectively (SFT, 1999). All statistical analyses (i.e. *t*-Test, Analysis of Variance (ANOVA), Principal Component Analysis (PCA)) were carried out using the software Statistical Package for the Social Sciences (SPSS Inc. Version 13).

2.3.4. Incremental lifetime cancer risk from PM_{10}-bound PAHs

Assuming (*i*) that the only three types of contact with dust particles are ingestion, inhalation and dermal contact, which represent the three main routes for human exposure; (*ii*) that the total carcinogenic risk can be estimated by applying *TEQ* values, as formerly described in section 2.3.1; and (*iii*) that the intake rates and particle emissions can be evaluated by applying procedures revised for soil dusts, the risk of exposure to environmental PAHs can be numerically estimated by applying the idea of incremental lifetime cancer risk (*ILCR*) (Chen and Liao, 2006; Peng et al., 2011; U.S. EPA, 1991) as clearly described in Supplementary Material (Eqs. S1–S3).

3. Results & discussion

Table 1 demonstrates the atmospheric contents of PM_{10}-bound PAH congeners, as well as the ΣPAHs in PM_{10} collected during FDP and NDP as previously described in section 2.1. The daily average PM_{10}-bound PAH concentrations ranged from 2.64 ± 3.70 pg m^{-3} (An) to 3540 ± 4141 pg m^{-3} (B[g,h,i]P) with ΣPAHs of 8301 ± 4910 pg m^{-3} and 3540 ± 4141 pg m^{-3} for the FDP and NDP, respectively (see Table 1). The average contents of particulate PAHs for the FDP and NDP are compared in Table 1. As demonstrated in Table 1, the majority of particulate PAH levels measured in the FDP were significantly ($p < 0.05$) higher than those measured in the NDP, which can be attributed to the high variability and complexity

Table 1
Statistical description of PM_{10}-bound PAHs collected during fireworks display period (FDP) in comparison with those collected during the non-fireworks display period (NDP).

Conc. (pg m^{-3})	Fireworks Display Period (FDP)		Non-Fireworks Display Period (NDP)		*t*-Test ($p < 0.05$)
	n = 11		n = 53		
	Aver.	Range	Aver.	Range	
Phe	123	44–274	137	34–406	NS[a]
An	2.64	N.D. ~8	11.2	N.D. ~55	S[b]
Fluo	104	44–225	127	30–402	S
Pyr	121	64–230	161	36–407	S
11H-B[*a*]F	17.0	8–28	27.7	5–94	S
11H-B[*b*]F	6.00	2–13	14.9	2–70	S
B[*a*]A	42.5	14–95	62.5	10–193	S
Chry	104	50–188	127	26–324	S
B[*b*]F	1114	175–3789	344	N.D. ~1152	S
B[k]F	580	119–1747	197	85–794	S
B[e]P	882	155–2875	279	99–1037	S
B[*a*]P	139	N.D. ~264	153	N.D. ~344	NS
Ind	1526	301–4946	416	181–1699	S
D[a,h]A	N.D.		N.D.		
B[g,h,i]P	3540	717–11,471	1226	537–3917	S
ΣPAHs	8301	1930–24,419	3283	1503–8932	S

[a] NS: Non Significant.
[b] S: Significant.

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of emissions sources in the different sampling periods. The atmospheric concentrations of HMW (High Molecular Weight) PAHs such as B[*b*]F, B[k]F, B[e]P, Ind and B[g,h,i]P were significantly higher in the FDP, which can be explained by the particle additions from fireworks. The fact that there are no official requirements to conduct an approved fireworks display in Thailand supports this interpretation.

As displayed in Fig. 2, similar distribution patterns of PM_{10}-bound PAHs were observed in the NDP and FDP in this study. The main compositions of particulate phase PAHs during the FDP were 6-ring PAHs (i.e. B[g,h,i]P and Ind) and 5-ring PAHs (i.e. B[*b*]F, B[k]F, B[e]P, and B[*a*]P), comprising 61% and 33% of the total, respectively (see Fig. 2). The HMW PAHs, such as B[g,h,i]P and Ind, all of which are widely recognized as vehicular markers, were predominantly found in the particulate phase, making up 43% (B[g,h,i]P) and 18% (Ind), and 37% (B[g,h,i]P) and 13% (Ind), of the total during the FDP and NDP, respectively (see Fig. 2).

Numerous reports have highlighted the importance of fireworks as major contributors to 5–6-ring PAHs in ambient air (Harrad and Laurie, 2005; Kong et al., 2015; Pongpiachan, 2013c). For instance, PAH mass contents clearly increased, particularly for HMW PAHs, by 13.8 times during the CNY intensive fireworks displays in comparison with the previous day (Kong et al., 2015). Significant increases in HMW PAH levels were also observed in Birmingham on Bonfire Night (Pongpiachan, 2013c). Interestingly, only Phe and B[*a*]P showed no statistically significant differences between the FDP and NDP, as displayed in Table 1. This can be ascribed to the fact that the overwhelming majority of these two congeners are released from vehicle exhausts, particularly diesel emissions in the case of Phe, which affects the air quality of observation sites. Particulate PAHs measured in the FDP and NDP both followed the trend B[g,h,i]P > Ind > B[*b*]F > B[e]P > B[k]F, suggesting that fireworks are probably not the main source of PAHs. This is trend is associated with traffic emissions. This supports our assumption that vehicular exhaust was the major contributor to PAH levels in Bangkok, regardless of the fact that firework displays occurred during the LKF.

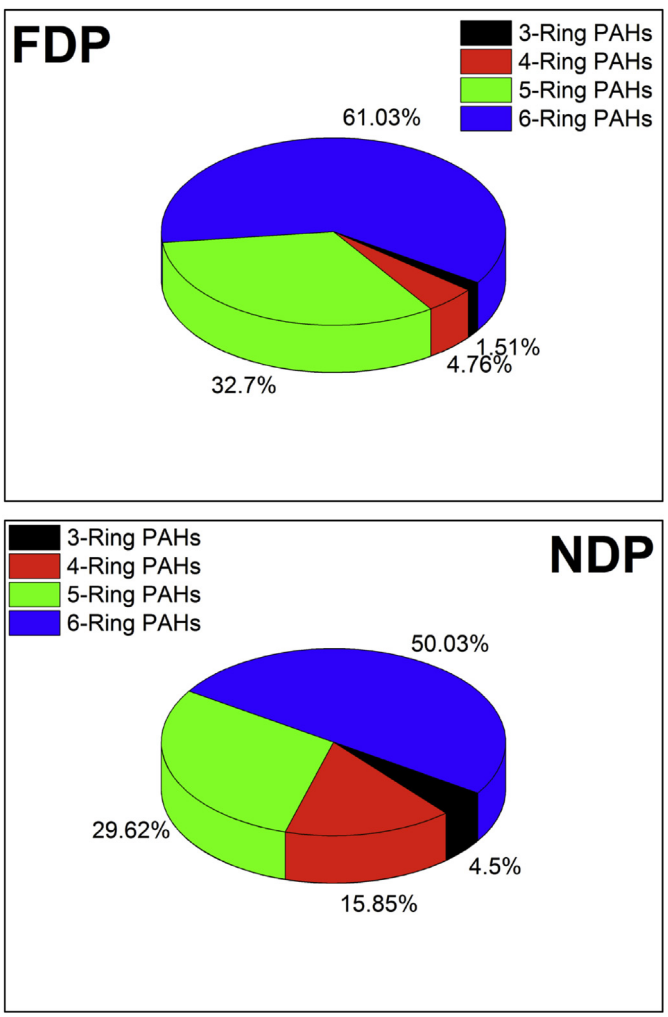

Fig. 2. Percentage contributions of PM_{10}-bound PAHs collected during the two different sampling campaigns.

3.1. Diagnostic binary PAH ratios

PAHs are generally classified based on their volatilities into Σ3,4-ring PAHs (Light Molecular Weight (LMW), MW ≤ 228) and Σ5,6-ring PAHs (HMW, MW ≥ 252). The Σ3,4-ring PAHs/Σ5,6-ring PAHs ratio can be used to investigate the source characteristics observed in the two different sampling campaigns. The two-sample *t*-test showed significant differences in the Σ3,4-ring PAHs/Σ5,6-ring PAHs ratios for the FDP and NDP, with values of 0.067 and 0.26, respectively. This finding indicates that HMW PAHs are major contributors during the LKF, which is in good agreement with earlier studies (Harrad and Laurie, 2005; Kong et al., 2015; Pongpiachan, 2013c). As displayed in Table 2, five different diagnostic binary PAH ratios (i.e. An/(An + Phe), Fl/(Fl + Pyr), B[*a*]A/(B[*a*]A + Chry), Ind/(Ind + B[g,h,i]P), and B[*a*]P/B[g,h,i]P) were calculated in this study and compared with previous reports. A previous study criticized the validity of applying diagnostic binary ratios of PAH congeners since this concept is based on many assumptions (Watson, 1984), two of which may be especially problematic for PAHs (Galarneau, 2008). Firstly, PAH source profiles are not unique by source type. Despite the fact that vehicular exhausts, diesel/gasoline emissions, low smoke sparklers, whistling sparklers, candle, combustions of petroleum, coal, and wood have been employed for source identification studies (Betha and Balasubramanian, 2014; Gschwend and Hites, 1981; Khalili et al., 1995; Li and Kamens, 1993; Orecchio, 2011; Rogge et al., 1993; Sicre et al., 1987; Tobiszewski and Namiesnik, 2012; Yassaa et al., 2001; Yunker et al., 2002), PAH diagnostic binary ratios demonstrate both substantial intrasource variability and intersource similarity (Galarneau, 2008). These problems are particularly severe for coarse categories, which are difficult to discriminate in the case of using B[*a*]A/(B[*a*]A + Chry) (Yunker et al., 2002). However, these limitations are of minor importance since the main contributor of particulate PAHs in Bangkok is vehicular exhausts not coarse combustions (Pongpiachan et al., 2013a). Secondly, it is crucial to underline that relative PAH contents are not conserved in the atmospheric environment (Galarneau, 2008). Several meteorological factors such as temperature, relative humidity, solar radiation coupled with both heterogeneous and homogeneous reaction with trace gaseous species (e.g. NO_x and OH-radical) can dramatically alter particulate PAH contents. For purposes of simplicity, it is assumed that meteorological parameters as well as gas-particle partitioning effects only play a minor role on variation of PAH concentrations due to the comparatively stable weather conditions during the observation period.

A Fl/(Fl + Pyr) ratio of 0.4 has been reported for traffic exhaust, >0.5 for coal/wood combustion, 0.44 for whistling sparklers and 0.49 for candles (Betha and Balasubramanian, 2014; Kong et al., 2013; Orecchio, 2011; Simcik et al., 1999; Yunker et al., 2002). The Fl/(Fl + Pyr) ratios in this study were 0.44 and 0.46 for the NDP and FDP, respectively, indicating that the PAHs were probably derived

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Table 2
Diagnostic binary ratios of PM_{10}-bound PAHs obtained from this study in comparison with other reports.

Source type	An(An + Phe)	Fl/(Fl + Pyr)	B[*a*]A/(B[*a*]A + Chry)	Ind/(Ind + B[g,h,i]P)	B[*a*]P/B[g,h,i]P	Reference
Emission Studies						
Non-Fireworks Display Period (NDP)	0.076	0.44	0.33	0.25	0.12	This Study
Fireworks Display Period (FDP)	0.021	0.46	0.29	0.30	0.039	This Study
Bonfire Night in Birmingham, UK	0.12	0.56	0.38	0.68	0.88	Harrad and Laurie, 2005
Fireworks during Chinese New Year			0.41	0.62	1.09	Shi et al., 2014
Fireworks during Diwali Festival		0.83		0.23		Sarkar et al., 2010
Vehicular Emission-1			>0.35			Tobiszewski and Namiesnik, 2012
Vehicular Emission-2					0.3–0.78	Simcik et al., 1999
Diesel Emission-1			0.38–0.64	0.35–0.7	0.46–0.81	Sicre et al., 1987 Rogge et al., 1993 Simcik et al., 1999 Khalili et al., 1995
Diesel Emission-2				0.5		Yassaa et al., 2001
Gasoline Emission-1			0.22–0.55	0.4	0.3–0.4	Sicre et al., 1987 Rogge et al., 1993 Khalili et al., 1995
Gasoline Emission-2		0.4		0.22		Simcik et al., 1999 Gogou et al., 1996
Cooking Emission			0.62	0.63	0.74	Li et al., 2003
Immission Studies						
Low smoke sparklers	0.32	0.49	0.64	0.76		Betha and Balasubramanian, 2014
Whistling Sparklers	0.14	0.44	0.86	0.78		Betha and Balasubramanian, 2014
Candle	0.28	0.49	0.35	0.71		Orecchio, 2011
Petroleum Combustion-1	<0.1			<0.4		Tobiszewski and Namiesnik, 2012
Coal Combustion-1			0.2–0.35			Tobiszewski and Namiesnik, 2012
Coal Combustion-2			0.5–0.55		0.9–6.6	Simcik et al., 1999 Gschwend and Hites, 1981 Masclet et al., 1987
Wood Combustion-1			0.43	0.62		Li and Kamens, 1993
Coal/Wood Combustion-2		>0.5				Yunker et al., 2002

from mixed sources with the major contributors being firework combustion and vehicular emissions. It should be noted that although the diagnostic ratios of Medium Molecular Weight (MMW) PAHs measured between the two sampling periods are similar, there may still be significant differences between the two periods owing to various source factors including atmospheric processing (Alam et al., 2013).

It is also interesting to note that the PAH source profile in the FDP has unique characteristics in comparison with other studies. For instance, an Ind/(Ind + B[g,h,i]P) ratio of 0.68 was found for Bonfire Night in Birmingham, UK, which is twice that observed during the FDP (0.30) (Harrad and Laurie, 2005). Similarly, a Fl/(Fl + Pyr) ratio of 0.83 was observed during the Diwali Festival, or the "Festival of lights", which is twice that observed during the FDP (0.46) (Sarkar et al., 2010). It is crucial to underline that the average sum of 16 PAH levels declined from background (72.8 ng m^{-3}) to pre-Diwali (102.6 ng m^{-3}) and subsequently drop sharply on Diwali (44.3 ng m^{-3}) as a consequence of the dearth of traffic during the event (Sarkar et al., 2010). It is also notable that the B[*a*]P/B[g,h,i]P ratio for the FDP (0.039) was respectively 23 times and 28 times lower than the ratios observed during Bonfire Night in Birmingham (Harrad and Laurie, 2005) and the Chinese New Year fireworks display (Shi et al., 2014). These inconsistencies reflect some unique characteristics of PAH fingerprints that arise from firework displays, which may be explained by differences in (*i*) the chemical formulas used in the gunpowder, (*ii*) the combustion temperature coupled with other meteorological parameters (Amarillo and Carreras, 2016) and (*iii*) chemical degradations via oxidation processes with hydroxyl radicals, ozone and NO_3 radicals (Arey et al., 1986; Kwok and Atkinson, 1995; Zhang et al., 2011).

Examination of Pearson correlations revealed that the following PAH congeners had strong positive correlations (i.e. $0.80 < R < 0.89$) during the FDP: Phe *vs*. An, Phe *vs*. Fluo, Phe *vs*. B[*a*]A, Pyr *vs*. Fluo and Pyr *vs*. B[*a*]A (see Table 3). Additionally, extremely strong positive correlation coefficients (i.e. $R > 0.90$) were found for Phe *vs*. Pyr, Phe *vs*. Chry, Fluo *vs*. Pyr, Fluo *vs*. Chry, Pyr *vs*. Chry, B[*b*]F *vs*. B[e]P, B[k]F *vs*. B[e]P, Ind *vs*. B[e]P, B[g,h,i]P *vs*. B[e]P and Ind *vs*. B[g,h,i]P. Interestingly, moderate positive correlations (i.e. $0.70 < R < 0.79$) were observed for An *vs*. Pyr, An *vs*. 11H-B[*a*]F, An *vs*. 11H-B[*b*]F, An *vs*. B[*a*]A, An *vs*. Chry, 11H-B[*a*]F *vs*. 11H-B[*b*]F and Fluo *vs*. B[*a*]A during the NDP, as displayed in Table 4. In contrast to the correlations observed in the FDP, strong positive correlations were observed in the NDP for HMW PAHs including B[*b*]F *vs*. B[e]P, B[k]F *vs*. B[e]P, Ind *vs*. B[e]P and B[g,h,i]P *vs*. B[e]P (see Table 4). Overall, the relatively high *R*-values for HMW PAHs illustrated in Table 3 suggest that firework displays can be considered major contributors to B[*b*]F, B[e]P, B[k]F, Ind and B[g,h,i]P during the LKF. In addition, the moderate positive correlations observed in the NDP suggest that 11H-B[*a*]F and 11H-B[*b*]F have similar emission sources to MMW PAHs (i.e. Pyr, An, Fluo and B[*a*]A), which can be attributed to vehicular exhausts.

3.2. Hierarchical cluster analysis (HCA)

HCA is a technique that can be used to discriminate between PAHs from vehicular exhaust and from firework displays. To achieve greater insight into the origins of PAHs in Bangkok, HCA was conducted for the 64 variables (i.e. the database of 64 individual air samples) and the 14 parameters (i.e. the total number of PAH congeners). The HCA successfully revealed the existence of two distinct groups by using the 64 individual air samples. The dendrogram in Fig. S1 consists of two main groups: the first cluster ($n = 3$) consists of air samples collected on 04/12/12, 05/12/12 and 05/12/13, and the second cluster ($n = 61$) contains the remaining air samples (Please see Supplementary Material). His Majesty King Bhumibol Adulyadej's birthday on December 5th is a national holiday and the most celebrated civil festival in Thailand. During this period, countless varieties of fireworks were launched between

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Table 3
Pearson correlation coefficients for PM_{10}-bound PAHs collected during the FDP sampling campaign.

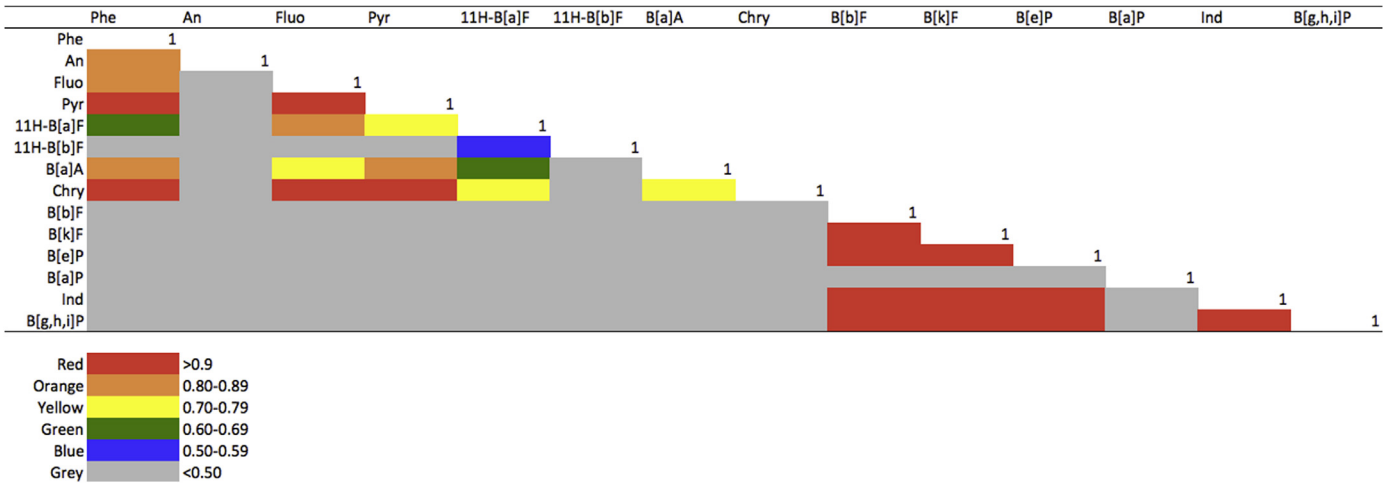
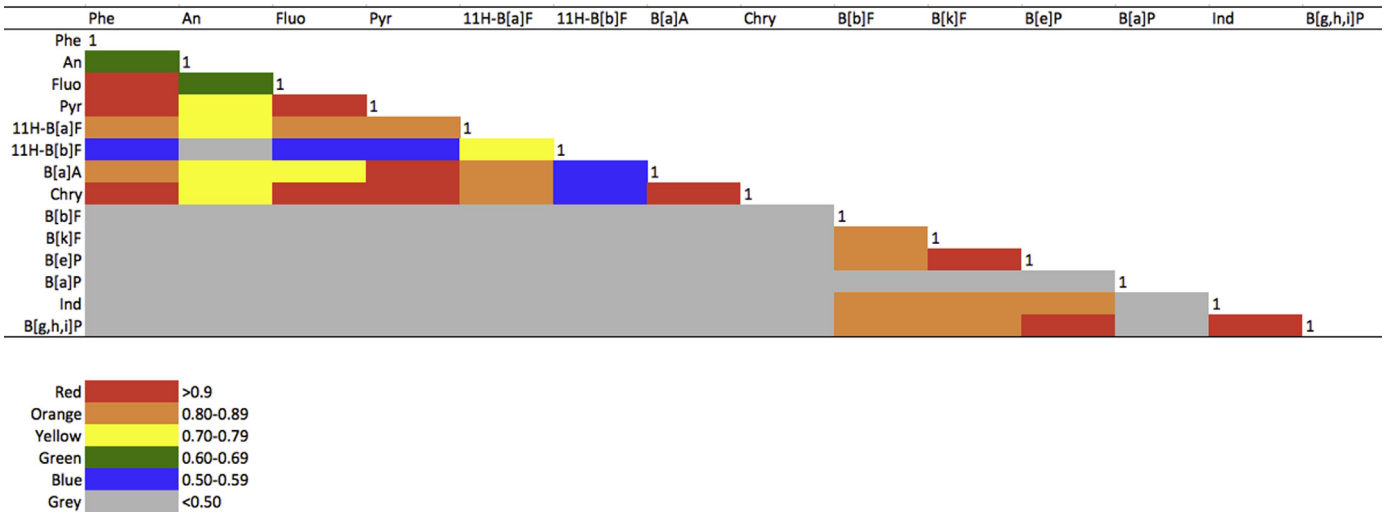


Table 4
Pearson correlation coefficients for PM_{10}-bound PAHs collected during the NDP sampling campaign.



the 4th and 6th December. The air samples present in the first cluster were generally of firework origin. However, it is important to note that sub-groups of firework origin (i.e. 27/11/12 (LKF), 28/11/12 (LKF), 06/12/12 (King's Birthday), 02/01/13 (New Year)) can also be detected in the second cluster, highlighting the importance of firework displays as major contributors of PAHs in ambient air.

Further attempts to clarify the impact of firework displays on the proximity matrix for carcinogenic compounds were conducted by analysing the 14 PAH congener contents of the 64 air samples collected during the FDP and NDP, as displayed in Figs. S2 and S3, respectively. These two dendrograms are very similar and can be divided into three subgroups: the first subgroup contains B[g,h,i]P, the second subgroup is composed of B[*b*]F, B[k]F, B[e]P and Ind, and the third consists of Phe, An, Fluo, Pyr, 11H-B[*a*]F, 11H-B[*b*]F, B[*a*]A, Chry and B[*a*]P. According to literature findings, the PAH congeners present in the first, second and third subgroups are predominantly of vehicular exhaust origin, firework display origin and diesel emission origin, respectively (Harrad and Laurie, 2005; Keyte et al., 2016; Kong et al., 2015; Olson et al., 2012; Pongpiachan, 2013c). These findings can be attributed to the fact that all the air quality observatories are located adjacent to heavily trafficked roads, as illustrated in Fig. 1, and thus traffic emissions can be considered the main contributors to PAHs at these observatories irrespective of firework display events.

3.3. Principal Component Analysis (PCA)

Table 5 displays the principal component patterns for varimax rotated components of the PAH dataset obtained from this study. To enable further interpretation of potential PAH sources throughout the observation period (i.e. NDP + FDP), a PCA model was computed with five significant PCs, respectively representing 40.1%, 35.4%, 7.96%, 7.26% and 5.53% of the variance, and thus accounting for 96.2% of the total variation in the data. The first component (PC1) displays high loading (i.e. >0.8) for Phe, Fluo, Pyr, 11H-B[*a*]F, B[*a*]A and Chry. These findings confirm that these MMW PAHs have vehicular exhausts particularly diesel emissions as a common source (Keyte et al., 2016; Olson et al., 2012). The second

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Table 5
Principal components (PC) pattern for varimax rotated components applied to the PAH congener dataset (NDP + FDP) collected from the PCD air quality observatories in Bangkok.

	PC1	PC2	PC3	PC4	PC5
Phe	**0.950**	0.011	−0.010	0.115	−0.033
An	0.582	−0.104	0.229	0.059	**0.761**
Fluo	**0.969**	−0.033	−0.044	0.086	0.016
Pyr	**0.972**	−0.042	0.081	0.098	0.131
11H-B[*a*]F	**0.818**	−0.101	0.025	0.427	0.297
11H-B[*b*]F	0.456	−0.156	0.005	**0.871**	0.029
B[*a*]A	**0.867**	−0.119	0.265	0.131	0.234
Chry	**0.972**	0.012	0.085	0.115	0.131
B[*b*]F	−0.035	**0.985**	0.027	−0.042	−0.035
B[k]F	−0.060	**0.990**	−0.012	−0.063	−0.038
B[e]P	−0.032	**0.995**	0.018	−0.042	−0.012
B[*a*]P	0.128	0.010	**0.983**	0.003	0.109
Ind	−0.035	**0.984**	−0.087	−0.040	−0.039
B[g,h,i]P	−0.037	**0.994**	0.041	−0.054	−0.014
% of Variance	40.878	35.415	7.962	7.262	5.534

* The bold numbers represent correlation coefficients higher than 0.7.

component (PC2) has higher loadings (i.e. >0.9) for B[*b*]F, B[k]F, B[e]P, Ind and B[g,h,i]P, which could plausibly be interpreted as source markers for firework displays (Harrad and Laurie, 2005; Kong et al., 2015; Pongpiachan, 2013c). PC3, PC4 and PC5 show comparatively high loadings for B[*a*]P (0.983), 11H-B[*b*]F (0.871) and An (0.761), respectively. These four PCs (i.e. PC1+PC3+PC4+PC5) explained 60.9% of total variances, indicating the importance of traffic emissions in PAH contents in the ambient air of Bangkok.

3.4. Health risk assessments

3.4.1. Excess cancer risk and human exposure assessment

The excess cancer risk per million people (ECR) was collected during the NDP and FDP, based on UR-B[*a*]P_{eq} of California Environmental Protection Agency (CalEPA) and World Health Organization (WHO), including the sum of the 12 priority PAHs using Nisbet and Lagoy's calculation ($\sum$PAH12-B[*a*]P_{eq}), the sum of the three MW 178 PAHs (Σ178PAH3-B[*a*]P_{eq}), the sum of the four MW 202 PAHs (Σ202PAH4-B[*a*]P_{eq}), the sum of the four MW 228 PAHs (Σ228PAH4-B[*a*]P_{eq}), the sum of the five MW 252 PAHs (Σ252PAH5-B[*a*]P_{eq}) and the sum of the six MW 276,278 PAHs (Σ276,278PAH6-B[*a*]P_{eq}). Results are illustrated in Table 6. The ECR-$\sum$PAH12-B[*a*]P_{eq} level observed in the NDP, based on the calculations of both CalEPA and WHO, were approximately one order of magnitude lower than those of World Cities (WCs). However, the ECR-$\sum$PAH12-B[*a*]P_{eq} level detected in the FDP was comparable to values for WCs, highlighting the substantial contribution of firework displays to public health risks, particularly during traditional festivals and New Year celebrations. Similar trends were also observed in ECR-Σ252PAH5-B[*a*]P_{eq}, and ECR-Σ276,278PAH6-B[*a*]P_{eq}, indicating higher excess cancer risks through HMW PAH exposure during the LKF. The average ECR-$\sum$PAH12-B[*a*]P_{eq} exposure for a lifetime of 70 years varied from 8.6×10^{-7} (NDP-CalEPA) to 3.2×10^{-4} (WCs-WHO), as illustrated in Table 6. As a consequence, an estimated mean excess of cancer cases for a lifetime of 70 years of 3.246 per million people (CalEPA) or 256.7 per million people (WHO) is attributable to inhalation of particulate $\sum$PAH12 for the residents during the FDP. These numbers are only 1.3 times lower than those of WCs using both CalEPA (i.e. 4.079 per million people) and WHO (i.e. 322.6 per million people), underlining that firework display sources may play a rather minor role in cancer risks in comparison with the role played by fireworks in other world cities. It is important to note that ECR values of WCs have generated much higher results than those predicted for the FDP and NDP. Comparatively low ECR values in Bangkok are apparently related to lower factory exhausts and traffic emissions, lower household fuel combustion owing to overall warm climate conditions and higher rainfalls (i.e. wet deposition effect) in comparison with other cities around the world. Interestingly, this study revealed higher percentage contributions of ECR-$\sum$276,278PAH6-B[*a*]P_{eq} for the FDP (64%) and NDP (57%), which were almost twice as those of WCs (33%). In contrast, the percentage contribution of ECR-$\sum$252PAH5-B[*a*]P_{eq} of WCs (63%) was 1.8 times and 1.5 times

Table 6
Estimated excess inhalation cancer risk attributed to measured concentrations of PM_{10}-bound PAHs for the FDP, NDP, and WCs. Two types of excess cancer risk were calculated according to the UR-B[*a*]P[a] from the WHO[b] (8.7×10^{-5} per ng m^{-3}) and CalEPA[c] (1.1×10^{-6} per ng m^{-3}) procedures.

	Excess cancer risk per million people (based on UR-B[*a*]P of CalEPA)						Excess cancer risk per million people (based on UR-B[*a*]P of WHO)					
	FDP[d] (n = 11)		NDP[e] (n = 53)		WCs[f] (n = 39)		FDP (n = 11)		NDP (n = 53)		WCs (n = 39)	
	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.
Ph-B[*a*]P_{eq}	1.4E-10	8.2E-11	1.5E-10	1.0E-10	1.6E-09	3.8E-09	1.1E-08	6.5E-09	1.2E-08	8.0E-09	1.3E-07	3.0E-07
An-B[*a*]P_{eq}	2.9E-11	4.1E-11	1.2E-10	1.4E-10	4.4E-09	1.2E-08	2.3E-09	3.2E-09	9.7E-09	1.1E-08	3.4E-07	9.3E-07
Fluo-B[*a*]P_{eq}	1.1E-10	6.5E-11	1.4E-10	1.0E-10	2.1E-09	2.6E-09	9.1E-09	5.1E-09	1.1E-08	8.0E-09	1.7E-07	2.1E-07
Pyr-B[*a*]P_{eq}	1.3E-10	6.8E-11	1.8E-10	1.2E-10	3.2E-09	3.1E-09	1.1E-08	5.4E-09	1.4E-08	9.2E-09	2.5E-07	2.5E-07
B[*a*]A-B[*a*]P_{eq}	4.7E-09	2.9E-09	6.9E-09	4.9E-09	1.2E-07	1.3E-07	3.7E-07	2.3E-07	5.4E-07	3.9E-07	9.5E-06	1.1E-05
Chry-B[*a*]P_{eq}	1.1E-09	5.3E-10	1.4E-09	9.5E-10	2.4E-08	2.9E-08	9.0E-08	4.2E-08	1.1E-07	7.5E-08	1.9E-06	2.3E-06
B[*b*+*k*]F-B[*a*]P_{eq}	1.9E-07	1.6E-07	5.9E-08	2.4E-08	5.9E-07	9.3E-07	1.5E-05	1.3E-05	4.7E-06	1.9E-06	4.7E-05	7.4E-05
B[*a*]P-B[*a*]P_{eq}	9.7E-07	1.2E-06	3.1E-07	1.7E-07	2.0E-06	2.6E-06	7.7E-05	9.6E-05	2.4E-05	1.3E-05	1.6E-04	2.1E-04
Ind-B[*a*]P_{eq}	1.5E-08	8.4E-09	1.7E-08	7.7E-09	3.3E-07	5.6E-07	1.2E-06	6.7E-07	1.3E-06	6.1E-07	2.6E-05	4.4E-05
D[*a*,*h*]A-B[*a*]P_{eq}	1.7E-06	2.1E-06	4.6E-07	2.5E-07	10E-07	1.4E-06	1.3E-04	1.6E-04	3.6E-05	2.0E-05	7.9E-05	1.1E-04
B[*g*,*h*,*i*]P-B[*a*]P_{eq}	3.9E-07	4.6E-08	1.3E-08	6.4E-09	3.4E-08	5.3E-08	3.1E-05	3.6E-06	1.1E-06	5.1E-07	2.7E-06	4.2E-06
ΣPAH12-B[*a*]P_{eq}	3.2E-06	2.4E-06	8.6E-07	3.0E-07	4.1E-06	3.2E-06	2.6E-04	1.9E-04	6.8E-05	2.4E-05	3.2E-04	2.5E-04
Σ178PAH3-B[*a*]P_{eq}	1.6E-10	9.1E-11	2.7E-10	1.7E-10	6.0E-09	1.2E-08	1.3E-08	7.2E-09	2.2E-08	1.4E-08	4.7E-07	9.8E-07
Σ202PAH4-B[*a*]P_{eq}	2.5E-10	9.4E-11	3.2E-10	1.5E-10	5.3E-09	4.1E-09	2.0E-08	7.4E-09	2.5E-08	1.2E-08	4.2E-07	3.2E-07
Σ228PAH4-B[*a*]P_{eq}	5.8E-09	3.0E-09	8.3E-09	5.0E-09	1.4E-07	1.4E-07	4.6E-07	2.4E-07	6.5E-07	4.0E-07	1.1E-05	1.1E-05
Σ252PAH5-B[*a*]P_{eq}	1.2E-06	1.2E-06	3.7E-07	1.7E-07	2.6E-06	2.8E-06	9.1E-05	9.7E-05	2.9E-05	1.3E-05	2.0E-04	2.2E-04
Σ276,278PAH6-B[*a*]P_{eq}	2.1E-06	2.1E-06	4.9E-07	2.5E-07	1.4E-06	1.5E-06	1.6E-04	1.6E-04	3.9E-05	2.0E-05	1.1E-04	1.2E-04

[a] UR: Unit Risk.
[b] WHO: World Health Organization.
[c] CalEPA: California Environmental Protection Agency.
[d] FDP: Firework Display Period.
[e] NDP: Non-firework Display Period.
[f] WCs: World Cities.

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higher than those for the FDP (36%) and NDP (42%), respectively. These findings suggest that firework displays are not only responsible for the enhancement of particulate 6-ring PAH contents but also for increasing the excess cancer risks of inhaling ∑276,278PAHs.

Cancer risks associated with non-dietary ingestion of PAHs in house dust for preschool children, as estimated using Eq. (5), are displayed in Table 7 based on the application of three different formulas (U.S. EPA, 1997; Maertens et al., 2008). It should be noted that "non-dietary exposure" can be defined as human exposure to $PM_{2.5}$-bound PAHs via both household air and dust (Wilford et al., 2005). Table 7 shows cancer risks related to non-dietary ingestion of PAHs in house dust for preschool children obtained for the FDP, NDP and WCs, with risks ranging from 4.5×10^{-3} (for the NDP with an ingestion rate of 50 mg day^{-1} estimated using the US-EPA method) to 14.5 (for WCs with an ingestion rate of 100 mg day^{-1} predicted by adopting the Nisbet-Lagoy method). The average cancer risks of both FDP and NDP were 7.3×10^{-3} and 1.5×10^{-2} for ingestion rates of 50 mg day^{-1} and 100 mg day^{-1}, respectively. The cancer risks in WCs for both ingestion rates were significantly greater than those obtained in this study. In spite of the fact that the cancer risks for the FDP and NDP were much smaller than those detected in WCs, with the mean values of the former varying from 10^{-3} to 10^{-2} (i.e. one cancer incidence case per million), they are still regarded as unacceptable (Maertens et al., 2008). These values are higher than the occupational exposure of foundry workers (9.1×10^{-4} and 1.1×10^{-3}) (Liu et al., 2010), sinter metal workers (3.2×10^{-5} and 5.0×10^{-5}) (Lin et al., 2008; Chiang et al., 2009), and the Canadian maximum acceptable level of risk (1×10^{-5}) (Maertens et al., 2008). As a consequence, this research highlights some causes for concern associated with the potential adverse human health effects of low-level home environmental exposure to $PM_{2.5}$-bound PAHs in preschool children.

Human exposure assessments, for both ingestion and inhalation routes, regarding the daily intake of PM_{10}-bound PAHs were computed by applying three different TEQ values (i.e. Nisbet-Lagoy, US-EPA and Cecinato); results for the two exposure routes are displayed in Tables 8 and 9, respectively. In this study, the $M_{PDI\text{-ingestion-}}$

Table 9
Human exposure assessments for inhalation routes from daily intake of PM_{10}-bound PAHs (M-PDI-inhalation: ng day^{-1}) calculated using three different TEQ values, namely Nisbet-Lagoy (Nibset and LaGoy, 1992), US-EPA (U.S. EPA, 1991), and Cecinato (1997), as displayed in Eq (7).

Inhalation Rate [m^3 day^{-1}]	Adult ($n = 11$)		Children ($n = 11$)	
	Aver.	St. dev.	Aver.	St. dev.
	20	20	10	10
FDP[a]				
M_{PDI}-inhalation-Nisbet-Lagoy	1.4E+00	2.9E+00	7.0E-01	1.4E+00
M_{PDI}-inhalation-US-EPA	9.2E-01	1.6E+00	4.6E-01	8.0E-01
M_{PDI}-inhalation-Cecinato	1.4E+00	2.9E+00	6.9E-01	1.4E+00
NDP[b]				
M_{PDI}-inhalation-Nisbet-Lagoy	1.1E+00	2.2E+00	5.5E-01	1.1E+00
M_{PDI}-inhalation-US-EPA	6.8E-01	1.2E+00	3.4E-01	6.1E-01
M_{PDI}-inhalation-Cecinato	1.1E+00	2.2E+00	5.5E-01	1.1E+00
WCs[c]				
M_{PDI}-inhalation-Nisbet-Lagoy	1.5E+02	1.8E+02	7.3E+01	8.8E+01
M_{PDI}-inhalation-US-EPA	6.0E+01	7.5E+01	3.0E+01	3.8E+01
M_{PDI}-inhalation-Cecinato	7.3E+01	8.9E+01	3.7E+01	4.5E+01

[a] FDP: Firework Display Period.
[b] NDP: Non-firework Display Period.
[c] WCs: World Cities.

Table 7
Cancer risks associated with non-dietary ingestion of PAHs in house dust for preschool children, as estimated using Eq. (5) (U.S. EPA, 1997; Maertens et al., 2008).

PAH Congener	FDP[a] ($n = 11$)		FDP ($n = 11$)		NDP[b] ($n = 53$)		NDP ($n = 53$)		WCs[c] ($n = 39$)		WCs ($n = 39$)	
Ingestion Rate [mg/day]	50	50	100	100	50	50	100	100	50	50	100	100
	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.
Cancer Risk-Nisbet-Lagoy (Nibset and LaGoy, 1992)	9.3E-03	1.9E-01	1.9E-02	3.8E-01	7.3E-03	1.5E-01	1.5E-02	3.0E-01	7.3E+00	8.7E+00	1.5E+01	1.7E+01
Cancer Risk-US-EPA (U.S. EPA, 1991)	6.1E-03	1.1E-01	1.2E-02	2.1E-01	4.5E-03	8.0E-02	9.1E-03	1.6E-01	3.0E+00	3.7E+00	6.0E+00	7.4E+00
Cancer Risk-Cecinato (Cecinato, 1997)	9.2E-03	1.9E-01	1.8E-02	3.8E-01	7.3E-03	1.5E-01	1.5E-02	2.9E-01	3.6E+00	4.4E+00	7.2E+00	8.8E+00

[a] FDP: Firework Display Period.
[b] NDP: Non-firework Display Period.
[c] WCs: World Cities.

Table 8
Human exposure assessments for ingestion routes from daily intake of PM_{10}-bound PAHs (M_{PDI}- ingestion: ng day^{-1}) calculated using three different TEQ values, namely Nisbet-Lagoy (Nibset and LaGoy, 1992), US-EPA (U.S. EPA, 1991), and Cecinato (1997), as displayed in Eq (6).

M_{DID} (mg day^{-1})	Adult ($n = 11$)				Children ($n = 11$)			
	55	55	200	200	4.16	4.16	100	100
	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.
FDP[a]								
M_{PDI}- ingestion-Nisbet-Lagoy	5.2E+01	1.1E+03	1.9E+02	3.8E+03	3.9E+00	8.0E+01	9.5E+01	1.9E+03
M_{PDI}- ingestion-US-EPA	3.4E+01	5.9E+02	1.2E+02	2.1E+03	2.6E+00	4.4E+01	6.2E+01	1.1E+03
M_{PDI}-ingestion-Cecinato	5.1E+01	1.0E+03	1.9E+02	3.8E+03	3.9E+00	7.9E+01	9.3E+01	1.9E+03
NDP[b]								
M_{PDI}- ingestion-Nisbet-Lagoy	4.1E+01	8.2E+02	1.5E+02	3.0E+03	3.1E+00	6.2E+01	7.4E+01	1.5E+03
M_{PDI}- ingestion-US-EPA	2.5E+01	4.5E+02	9.2E+01	1.6E+03	1.9E+00	3.4E+01	4.6E+01	8.1E+02
M_{PDI}-ingestion-Cecinato	4.1E+01	8.1E+02	1.5E+02	3.0E+03	3.1E+00	6.2E+01	7.4E+01	1.5E+03
WCs[c]								
M_{PDI}- ingestion-Nisbet-Lagoy	2.0E+04	2.4E+04	7.3E+04	8.8E+04	1.5E+03	1.8E+03	3.7E+04	4.4E+04
M_{PDI}- ingestion-US-EPA	8.3E+03	1.0E+04	3.0E+04	3.8E+04	6.3E+02	7.8E+02	1.5E+04	1.9E+04
M_{PDI}-ingestion-Cecinato	1.0E+04	1.2E+04	3.7E+04	4.5E+04	7.6E+02	9.3E+02	1.8E+04	2.2E+04

[a] FDP: Firework Display Period.
[b] NDP: Non-firework Display Period.
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Please cite this article in press as: Pongpiachan, S., et al., Assessing human exposure to PM_{10}-bound polycyclic aromatic hydrocarbons during fireworks displays, Atmospheric Pollution Research (2017), http://dx.doi.org/10.1016/j.apr.2017.01.014

Table 10
Incremental lifetime cancer risk from PM_{10}-bound PAHs assuming that the only forms of contact with dust particles are ingestion, inhalation, and dermal contact.

	FDP[a]				NDP[b]				WCs[c]			
	Adult ($n = 11$)		Children ($n = 11$)		Adult ($n = 11$)		Children ($n = 11$)		Adult ($n = 11$)		Children ($n = 11$)	
	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.	Aver.	St. dev.
ILCRs-Ingestion	4.9E-09	9.6E-08	2.5E-09	4.8E-08	3.8E-09	7.4E-08	1.9E-09	3.7E-08	1.4E-06	1.7E-06	6.9E-07	8.3E-07
ILCRs-Dermal	8.7E-10	1.7E-08	3.0E-10	6.0E-09	6.8E-10	1.3E-08	2.4E-10	4.6E-09	2.4E-07	3.0E-07	8.6E-08	1.0E-07
ILCRs-Inhalation	3.8E-14	7.4E-13	4.7E-15	9.2E-14	3.0E-14	5.8E-13	3.7E-15	7.2E-14	1.1E-11	1.3E-11	1.3E-12	1.6E-12
Compare to Superfund 10^{-6}												
ILCRs-Ingestion	204	10.4	408	20.9	262	13.5	525	27.0	0.728	0.599	1.46	1.20
ILCRs-Dermal	1149	58.7	3276	167	1477	75.9	4208	216	4.10	3.37	11.7	9.6
ILCRs-Inhalation	26,324,242	1,344,903	210,593,939	10,759,227	33,819,339	1,737,956	27,0554,714	13,903,649	93,814	77,288	750,513	618,303

[a] FDP: Firework Display Period.
[b] NDP: Non-firework Display Period.
[c] WCs: World Cities.

$_{\text{Nisbet-Lagoy}}$ values for the FDP (52 for adults with M_{DID} of 55 mg day^{-1}) and the NDP (41 ng day^{-1} for adults with M_{DID} of 55 mg day^{-1}) were almost 388 times and 495 times lower than those for WCs, respectively. It is crucial to underline that the values of $M_{PDI\text{-}ingestion}$ were two to three orders of magnitude greater than those of $M_{PDI\text{-}inhalation}$ for all three calculations. This pattern was identical to that found for nine administrative provinces in the northern part of Thailand (Pongpiachan et al., 2015) and Guangzhou (Wang et al., 2013), indicating that dust ingestion is extremely important in non-dietary PAH exposure in comparison with $PM_{2.5}$ respiration. Previous studies noted that 50–70% of household dust is believed to come from outdoor soils. Thus, more recent studies have attempted to report results as ingestion of "soil/dust", rather than ingestion of household dust. Furthermore, it is widely known that incidental soil ingestion among children must be considered owing to the prevalence of floor-based play (i.e. relevant for nearly 87% of children aged 1–4 years) and the fact that almost 50% of children play on grass or dirt. In the case of children, other activities can also lead to soil ingestion, such as eating food dropped on the ground, and putting soil-contaminated fingers in their mouths (Akinwunmi et al., 2017; Bacigalupo and Hale, 2012; Juhasz et al., 2016).

3.4.2. Incremental lifetime cancer risk (ILCR)

This is the only research study to specifically focus on the potential cancer risk of human exposure to PM_{10}-bound PAHs through different exposure routes before and after the firework display episodes using three different *TEQ* methods. Since the *ILCR* is generally defined as the average daily dose averaged over a lifetime multiplied by the cancer slope factor, this concept can assess the probability of developing cancer during a lifetime, with scores varying between zero and one. The potential cancer risks for adults and children can be evaluated by applying the *ILCR* for different exposure routes (i.e. ingestion, dermal contact and inhalation) as set out in Eqs. (S1)–(S3), respectively (Table 10) (Peng et al., 2011; Chen and Liao, 2006; Wang et al., 2011). We highlight that $ILCR_{Dermal}$ and $ILCR_{Ingestion}$ ranged from 10^{-15} to 10^{-9} in both the FDP and NDP samples, while those for WCs ranged from 10^{-12} to 10^{-6}, roughly three orders of magnitude higher than the FDP and NDP results. This can be interpreted as a consequence of the comparatively high anthropogenic emissions in urban areas, as these emissions are major contributors governing the potential cancer risk from human exposure to PM_{10}-bound PAHs. As displayed in Table 10, both $ILCR_{Dermal}$ and $ILCR_{Ingestion}$ were clearly larger than $ILCR_{Inhalation}$, emphasizing that inhalation risk of particulate PAHs was essentially negligible when compared with the dermal contact and ingestion exposure pathways.

These results are consistent with previous reports of exposure to heavy metals in street dust in Angola (Ferreira-Baptista and De Miguel, 2005), aerosol samples obtained from Guangzhou (Wang et al., 2011) and $PM_{2.5}$ collected from nine administrative provinces in Northern Thailand before and after a haze episode (Pongpiachan et al., 2015). According to the current international guidelines for carcinogen risk assessment, a potential cancer risk is defined as values of *ILCR* between 10^{-6} and 10^{-4}, and a high potential cancer risk and a practical safety level can be defined as values higher than 10^{-4} and 10^{-6}, respectively (Chen and Liao, 2006; Wang et al., 2011). In this study, the mean values of *ILCR* for children during the FDP were $2.4 \times 10^{-9} \pm 4.7 \times 10^{-8}$, $3.1 \times 10^{-10} \pm 6.0 \times 10^{-9}$ and $4.7 \times 10^{-15} \pm 9.2 \times 10^{-14}$ for ingestion, dermal contact and inhalation, respectively; these risk levels are respectively 408, 3276 and 210,593,939 times lower than the U.S. EPA (1991) baseline. In addition, the mean *ILCRs* for adults during the FDP, estimated for the three exposure pathways, were 281 and 520 times lower than the WCs and U.S. EPA baseline, further suggesting that the cancer risk from firework displays falls into the "acceptable level" range.

4. Conclusions

Significant decreases in Σ3,4-ring PAHs/Σ5,6-ring PAHs ratios observed during the FDP highlight that HMW PAHs with the exception of B[*a*]P are the main compositions present during the bonfire night episode, which is consistent with previous investigations. Cluster analysis revealed the importance of traffic emissions as major contributors to PAHs, independent of the LKF. Principal Component Analysis highlighted the importance of both traffic emissions and firework displays as representing 61% (i.e. PC1 + PC3 + PC4 + PC5) and 35% (i.e. PC2) of the total variances of eigen values, respectively. This study also focused on the incidental ingestion of soil and dust that results from children's typical ingestion behaviours. In the case of children, the $ILCR_{Ingestion}$ was significantly higher than $ILCR_{Dermal}$ for both the FDP and NDP, indicating the importance of hand-to-mouth activity as a key contributor to the cancer risk for children. These findings are also in good agreement with previous assessments of the potential risk from children's exposure to dust in Hermosillo, Sonora, Mexico (Meza-Figueroa et al., 2007) and Guangzhou, China (Wang et al., 2011). The U.S. EPA Risk Assessment Guidance for Superfund Programs adopts a cancer risk level of 10^{-6} as the point at which risk management decisions should be taken. Applying this guidance, the chance of adverse cancer-causing effects during the LKF was significantly lower, particularly considering the incremental lifetime cancer risk from PM_{10}-bound PAHs by inhalation.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.01.014.

Please cite this article in press as: Pongpiachan, S., et al., Assessing human exposure to PM_{10}-bound polycyclic aromatic hydrocarbons during fireworks displays, Atmospheric Pollution Research (2017), http://dx.doi.org/10.1016/j.apr.2017.01.014

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Please cite this article in press as: Pongpiachan, S., et al., Assessing human exposure to PM_{10}-bound polycyclic aromatic hydrocarbons during fireworks displays, Atmospheric Pollution Research (2017), http://dx.doi.org/10.1016/j.apr.2017.01.014