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Original article

Determination of $PM_{2.5}$ and polycyclic aromatic hydrocarbons from incense burning emission at shrine for health risk assessment

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ABSTRACT

This study aims to determine fine particles ($PM_{2.5}$) and polycyclic aromatic hydrocarbons (PAHs) bounded with $PM_{2.5}$ emitted from incense burning at shrine to assess human health risk. The $PM_{2.5}$ samples were collected by a mini volume air sampler during special occasions and normal period at a shrine located in the city center of Chiang Mai, Thailand. The samples were analyzed for 16-PAHs by GC–MS. The descending order of 8- and 24-hrs $PM_{2.5}$ concentrations ($\mu g/m^3$) were 625 ± 147 and 406 ± 159 (Chinese New Year) > 184 ± 85 and 133 ± 71 (other special occasions) > 100 ± 35 and 50 ± 20 (normal period). Their concentrations in each occasion were significantly different due to number of visitors and amount of incense being burned. Total PAHs concentrations (ng/m^3) for 8- and 24-hrs in descending order were 90 ± 41 and 45 ± 29 (Chinese New Year), 71 ± 30 and 30 ± 12 (other special occasions) and 25 ± 15 and 14 ± 9 (normal periods). Correlation between $PM_{2.5}$ and total PAHs concentrations was relatively strong. In addition, $PM_{2.5}$ concentrations were highly correlated ($r = 0.618$) with carcinogenic PAHs (c-PAHs) indicated that carcinogenic compounds were dominant in particulate PAHs and generated from incense burning. The values of toxicity equivalent concentration (TEQ) indicate human health risk from PAHs inhalation. According to European guideline, it should be less than1 ng/m^3 in ambient air. During Chinese New Year, they were relatively high (32 ± 27 ng/m^3 (8 h) and 10 ± 4 ng/m3 (24 h)). Moreover, the isometric ratio analysis revealed that emission from incense burning was the main source of $PM_{2.5}$ and $PM_{2.5}$-bound PAHs.

1. Introduction

The polycyclic aromatic hydrocarbons (PAHs) are comprised of a group of semi-volatile organic pollutants containing at least two fused aromatic rings. PAHs are toxic chemical group, which some of them are mutagenic or carcinogenic. PAHs are produced from incomplete combustion of fossil fuels and organic materials. Although some natural sources (forest fire) can contribute to the PAHs burden (Wiriya et al., 2013; Pongpiachan, 2015), human activities contribute most to PAHs emissions and the sources are entirely anthropogenic in urban and industrial atmospheres (Orecchio, 2010; Masih et al., 2012c; Wu et al., 2015). The study from Billet et al. (2007) found that about 92% of PM was $PM_{2.5}$, which collected from ambient in heavy industrial city. Furthermore, PAHs associated with particulate matter (PM). Evagelopoulos et al. (2010) collected $PM_{2.5}$ sample from urban area surrounded by opencast coal mining emission and found that $PM_{2.5}$-bound PAHs concentrations were four times higher than PM_{10}-bound PAHs. The total PAHs on PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ was collected from indoor and outdoor at the retirement home and a school dormitory in Tehran. The predominant of PM-bound total PAHs found in $PM_{2.5}$ was 83–88% (Hassanvand et al., 2015). Therefore, PAH concentrations associated with particulate matters are highly dependent on fine particles (Duan et al., 2005). Inhalation of PAHs in particulates is a potentially serious health risk linked to an excess risk of lung cancer, which some PAHs are considered carcinogens.

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Some previous studies (Wu et al., 2015) studied the emission of $PM_{2.5}$ and PM_{10} during the peak tourist season at Mount Wutai Buddhism, Shanxi province, China. $PM_{2.5}$ and PM_{10} concentrations were 1.43–59.20 μg/m^3 and 17.40–161.45 μg/m^3, respectively. The main source was released from incense and candle burning. Lin et al. (2002) revealed that total suspended particle (TSP) – bound PAHs concentrations were 6258 ng/m^3 (indoor) and 231 ng/m^3 (outdoor). The concentration indoor PAHs was 27 times higher than outdoors. The highest top five PAH concentrations (particulate + gas phase) were identified as 3583 ng/m^3 acenaphthylene, 1264 ng/m^3 naphthalene, 349 ng/m^3 acenaphthene, 243 ng/m^3 fluoranthene and 181 ng/m^3 phenanthrene. Liao and Chiang (2006) reported the emission from incense burning in Taiwanese temples. The median PAH concentrations found in the inside and outside air of the temples were 478.4 and 232.5 ng/m^3, respectively. Moreover, benzo[a]pyrene (BaP) value of inside was 10 times higher than outside in temples (Chiang et al., 2009). The total toxic of the BaP equivalent concentration of the solid-phase PAHs (S-PAHs) was over 40 times higher than that of the corresponding gas-phase PAHs, indicating that the S-PAHs in incense smoke may pose potential health risk (Yang et al., 2007). In Thailand, Nonthakanok (2013) investigated emission of $PM_{2.5}$ and $PM_{2.5}$-bound PAHs from incense burning at temples in Bangkok. The results showed the 8-hrs $PM_{2.5}$ and total PAHs concentrations found in personal modular impact (PMI) were 54.64 ± 6.16 to 982.20 ± 196.54 μg/m^3 and 2.43 ± 2.77 to 121.73 ± 22.73 ng/m^3, respectively. Moreover, the values were 2–9 and 2–24 times higher than ambient air, which was collected by minivol tactical air sampler, respectively. However, Several studies assessed human exposure to airborne PMs and PAHs from heavy incense burning in temples may be associated with many adverse health effects including cancer, respiratory morbidity, central nervous system damage and lung irritation (Chiang and Liao, 2006; Lin et al., 2008; Navasumrit et al., 2008; Pan et al., 2014). Incense burning is a serious issue in concerning human health effect. Consequently, the main objective of the research is to determine $PM_{2.5}$ and $PM_{2.5}$-bound PAHs emitted from incense burning to assess health risks.

2. Materials and methods

2.1. $PM_{2.5}$ sampling

The sampling shrine is located in Waroros market, which is one of the most crowded areas in the city of Chiang Mai (Fig. 1). The shrine was established in 1897, which is famous and popular in Chiang Mai City. This sampling site shrine is situated near a narrow road with high traffic density, which included motorcycles and small diesel trucks (pick-ups), on a daily basis. The shrine has an area of approximately 960 m^3. It has quite a good ventilation system, which has 4 turbine ventilators on the roof of the worship room and its ceiling is open. A visitor to this sampling site shrine would purchase a package of 28 small incense sticks. The stick has a diameter of ~2.4 mm with a length of ~32 cm and ~21 cm coated with combustible part. Twenty six of them were burned in the worship room and the rest were burned outdoors. A number of incense burned each day depends on number of visitors, which was counted during the sampling period. Apart from incenses, some candles and lanterns filled with vegetable oil were also burned inside the shrine. However, the amount of incense used was much higher that number of candles and lanterns.

Indoor $PM_{2.5}$ samples were collected on Teflon fiber filters (2 μm PTFE, 46.2 mm diameter, Whatman's filter paper) using Minivol Air Samplers (Air metric, USA) at a flow rate of 5 L/min for 8 h (8 am–4 pm) and 24 h (8 am–8 am) in the same day. The filters were pre-weighed by a microbalance in a controlled room (25.4 ± 2.8 °C, 41.3 ± 5.4% RH). After the sampling, the filters were kept in aluminium foil plates inside a desiccator for 48 h before being re-weighed and stored in a freezer (−4.0 °C) until analysis. The indoor temperature and humidity were measured for the whole sampling period using a hygrometer (Sato keiryoki MEG. CO., LTD.). Their ranges were 27.0–35.5 °C and 51.5–75.7%, respectively.

The sampling was carried out during special occasions and over normal periods (background value). The special occasions in this study were those associated with the Chinese lunar calendar including Chinese New Year (22-24/01/2012 and 9-11/02/2013), the Chinese Ghost festival (31/08/2012), the moon festival (30/09/2012), a vegetarian festival (15-16 and 19-20/10/2012) and the celebration of establishment's anniversary (30-31/12/2012 and 01/01/2013). The sampling in the normal periods was randomly carried out during non-event for the whole year (2012). The amount of incense being burned in each occasion depends on number of visitors. During special occasions, more visitors come and high number of incenses is used.

2.2. Extraction and analysis of PAHs

The samples were extracted in 25 mL dichloromethane (DCM) and n-hexane mixture (1:1, v/v) for 45 min by an ultrasonicator. The extracted solutions were than purified using a 0.45 μm nylon syringe filter (Agela Technologies, USA) and were dried using a rotary evaporator. The solution was added with a mixture of internal standards (acenaphthene-d12 and perylene-d10) and was adjusted to 2 mL volumetric flask with solvent mixture. 16-PAHs were analyzed by gas chromatography – mass spectrometer (GC–MS, Agilent, USA) equipped with a 30 m HP-5MS capillary column. The heating program was set for column oven from 60 °C to 290 °C at 6 °C/min, then hold for 20 min (Wiriya et al., 2013). The MS was operated in selective ion monitoring mode (SIM).

The total PAHs (t-PAHs) are all 16 EPA-PAHs, which was identified and quantified including naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IND), dibenz[a,h]anthracene (DbA), benzo[g,h,i]perylene (BPER). The quality control of PAHs was studied using 50 mg of standard reference material (SRM 1649b; urban dust, NIST, USA) and mixed PAHs standard solution (Restex, USA) was spiked on a Teflon fiber filter to obtain 0.02 mg/L (n = 3) for the extraction and analysis method. For SRM method, recovery efficiency of 12 PAHs (n = 3) ranged 61% (CHR) to 126% (ANT) and averaged 84%. The measure values of spiking method are presented in Table 1. The detection limit (n = 7) in this study were 0.08–0.66 ng/m^3.

2.3. Data analysis

Concentration of $PM_{2.5}$ and $PM_{2.5}$-bound PAHs inside the shrine in each occasion was compared by One-Way ANOVA test. The correlation coefficient was assessed by the spearman's rank correlation (r_s), which were calculated correlations between the various indoor $PM_{2.5}$ and total PAHs bounded with $PM_{2.5}$ (t-PAHs, c-PAHs and nc-PAHs) in order to identify their relationship. The isomeric ratios were used to indicate the possible sources of PAH in each occasion.

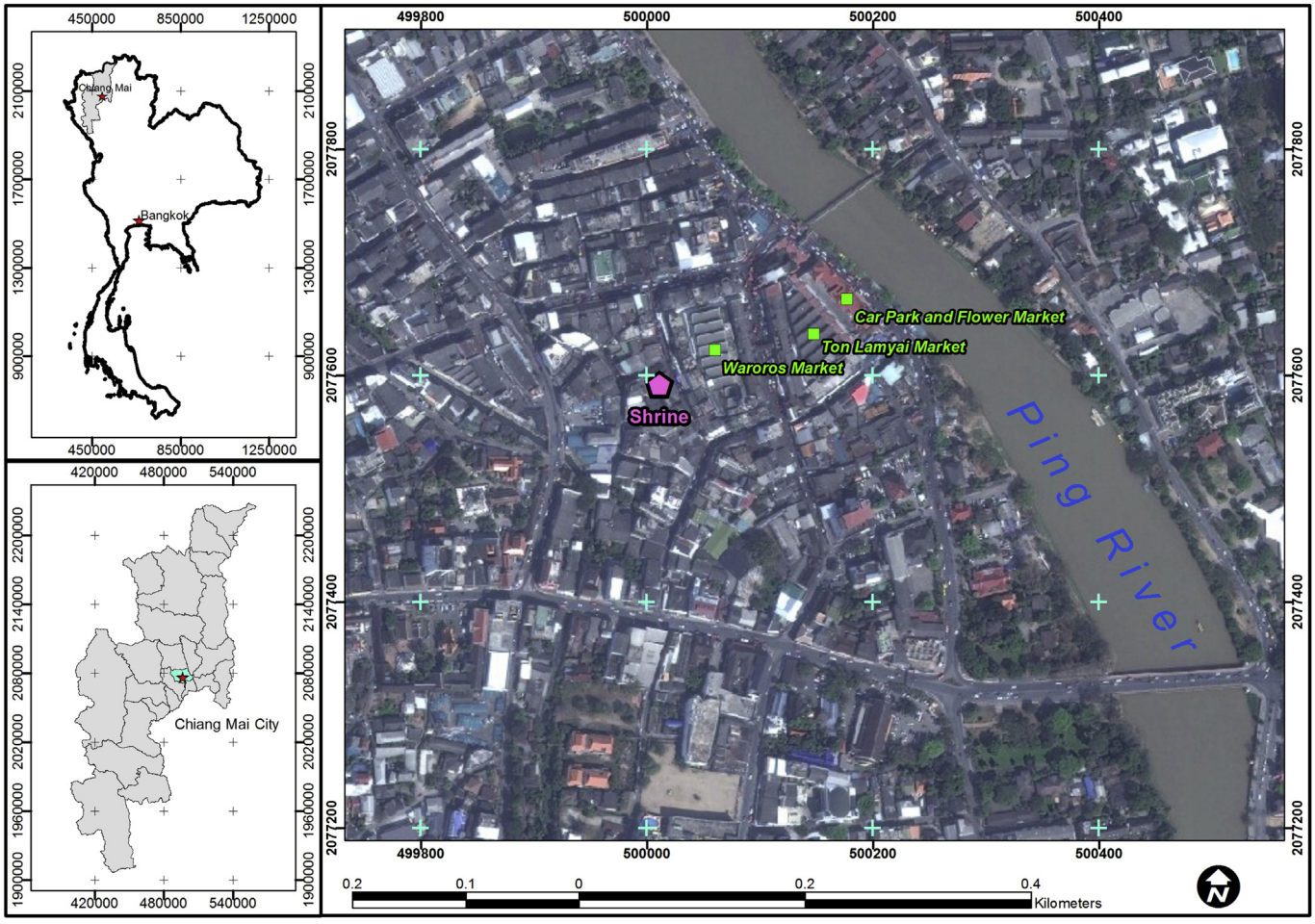

Fig. 1. Sampling site.

Table 1
Limit of detection (LOD) of GC–MS for individual PAH, and efficiency of the extraction and analysis of PAHs.

PAHs	Abbreviation	Ring number	Quantification ion (m/z)	LOD* (ng/m^3)	% recoveries from spiking method	Certified value (mg/kg)	Measured value (mg/kg)
Naphthalene	NAP	2	128	0.16	87	–	–
Acenaphthylene	ACY	3	152	0.10	90	–	–
Acenaphthene	ACE	3	154	0.50	98	–	–
Fluorene	FLU	3	166	0.17	95	–	–
Phenanthrene	PHE	3	178	0.13	93	3.94 ± 0.05	3.00 ± 0.62
Anthracene	ANT	3	178	0.08	103	0.40 ± 0.00	0.54 ± 0.11
Fluoranthene	FLA	4	202	0.17	90	6.14 ± 0.12	5.96 ± 0.95
Pyrene	PYR	4	202	0.17	91	4.78 ± 0.03	3.67 ± 0.63
Benzo[a]anthracene	BaA**	4	228	0.10	84	2.09 ± 0.05	1.42 ± 0.18
Chrysene	CHR**	4	228	0.20	82	3.01 ± 0.04	1.71 ± 0.41
Benzo[b]fluoranthene	BbF**	5	252	0.25	85	5.99 ± 0.20	5.32 ± 1.01
Benzo[k]fluoranthene	BkF**	5	252	0.20	92	1.75 ± 0.08	1.28 ± 0.11
Benzo[a]pyrene	BaP**	5	252	0.66	80	2.47 ± 0.17	1.87 ± 0.60
Indeno[1,2,3-cd]pyrene	IND**	6	276	0.49	90	2.96 ± 0.17	1.89 ± 0.38
Dibenz[a,h]anthracene	DbA**	5	278	0.57	94	0.29 ± 0.00	0.32 ± 0.15
Benzo[g,h,i]perylene	BPER	6	276	0.46	89	3.94 ± 0.05	±0.57

Note; *LOD (ng/m^3) = [(LOD (ng/mL)) × (final volume of solution (2 mL))]/(average volume of air (7.1 m^3)).
**Carcinogenic PAHs (c-PAHs).

3. Results and discussion

3.1. Concentrations of $PM_{2.5}$ and $PM_{2.5}$-bound PAHs

Concentrations of indoor $PM_{2.5}$ and total PAHs bounded with $PM_{2.5}$ measured on special occasions (with the exclusion of Chinese New Year), Chinese New Year and the normal period (background value) at the sampling site shrine are shown in Table 2. One-Way ANOVA was used to determine the differences of mean $PM_{2.5}$ and $PM_{2.5}$-bound PAHs concentrations between occasions. The $PM_{2.5}$ concentrations were 1/square root − transformed to achieve normal distribution, while concentrations of t-PAHs, c-PAHs and nc-PAHs were square root-transformed to achieve normal distribution. It was found that the average $PM_{2.5}$ concentrations measured during normal period were significantly less than those of other special occasions and Chinese New Year ($p < 0.05$). The $PM_{2.5}$ concentrations were associated with number of visitors, which were strongly depended on the amount of incense being burned. Numbers of visitors (8-hrs and 24-hrs sampling period) visiting the sampling site shine during Chinese New Year (390 ± 291 and 631 ± 319 persons) were about 15 times higher than those in other special occasions (23 ± 13 and 42 ± 23 persons). Moreover, they were ~40–50 times higher than normal periods (9 ± 5 and 12 ± 6 persons) (Bootdee and Chantara, 2014).

The $PM_{2.5}$ concentrations were significantly different in both the other special occasions and normal period. The highest value of indoor $PM_{2.5}$ concentrations ($\mu g/m^3$) was obtained in Chinese New Year for both of 8- and 24-hrs periods (625 ± 147 and 406 ± 159, respectively). The 8-hrs $PM_{2.5}$ value was about two times higher than that collected during Chinese New Year at the temple in Hong Kong (360 ± 23 $\mu g/m^3$) (Wang et al., 2007). The 8-hrs $PM_{2.5}$ concentrations collected at temples in Bangkok, Thailand using personal modular impactors (PMI) were 54.6 ± 6.1 to 982 ± 196 $\mu g/m^3$ (Nonthakanok, 2013), which were slightly higher than those found in this study.

In order to compare indoor and outdoor $PM_{2.5}$ at the same period, $PM_{2.5}$ concentrations obtained from the air quality monitoring (AQM) station (Pollution Control Department; PCD) located in Chiang Mai City (approximately 2 km from the sampling site shrine) from the same sampling period were compared. The mean outdoor $PM_{2.5}$ concentrations ($\mu g/m^3$) measuring for 8 and 24 h during Chinese New Year, other special occasions and normal periods were 54.92 ± 2.67 and 35.05 ± 24.25, 23.38 ± 10.90 and 29.96 ± 13.96, and 18.82 ± 7.94 and 21.12 ± 8.38, respectively. It was obvious that the outdoor $PM_{2.5}$ concentrations during Chinese New Year were ~2 times higher than other occasions, but the others were not much different. Moreover, the indoor values measured in the sampling site shrine during the same occasions were much higher than outdoors.

Total PAHs (t-PAHs) concentrations in Chinese New Year were higher than those in the other special occasions and normal period. The values were significantly different between the other special occasions and normal period, while the values of the other special occasions were not significantly different from Chinese New Year ($p > 0.05$) (Table 2). The values of 24-hrs total PAH concentrations during Chinese New Year, other special occasions and normal period were 45 ± 29, 30 ± 12 and 14 ± 9 ng/m^3, respectively. The values were much lower than those found in India (Masih et al., 2012a; Masih et al., 2012b), where the particle-bound 18 PAHs inside roadside homes with oil fumes for cooking and smoking & incense burning during winter and summer were 487 ng/m^3 and 160 ng/m^3, respectively. Moreover, concentrations of 23 PAHs in gas and particle emitted from biomass burning and oil fumes for cooking and heating at rural area were 2312 ng/m^3 (winter), 1489 ng/m^3 (summer) and 879 ng/m^3 (rainy). The 24-hrs total PAHs values in indoor TSP emitted from incense burning in Taiwanese temple were 478 ng/m^3 (Liao and Chiang, 2006) and 6258 ng/m^3 (Lin et al., 2002). Moreover, the 8-hrs total PAHs concentrations in $PM_{2.5}$ emitted from incense burning at Thai temple in Bangkok (1.01 ± 0.98 to 47.7 ± 72.8 ng/m^3) were lower than in this study (25 ± 15 to 90 ± 41 ng/m^3) (Nonthakanok, 2013). Fig. 2 shows PAH composition extracted from $PM_{2.5}$ samples collected for 8 and 24 h inside the sampling site shrine (Table 2). The dominant PAHs species found in this study were BbF (6.2 ± 6.8 to 26.9 ± 18.2 ng/m^3), BaP (1.4 ± 0.9 to 10.0 ± 2.3 ng/m^3), IND (0.4 ± 1.0 to 12.8 ± 9.6 ng/m^3) and BPER (1.0 ± 0.8 to 9.7 ± 7.0 ng/m^3). The result was found to be similar with Navasumrit et al. (2008) and Lin et al. (2002), which indicated that BaA, BbF, BaP and DbA were the dominant carcinogenic PAHs in Thai temples, while the dominant PAHs in Taiwanese temple were BaP, BPER, DbA and BbF. Moreover, Orecchio (2011) revealed that the highest mean of carcinogenic BbF (0.62 μg/kg) was observed from candle burning in a chamber. Derudi et al. (2014) reported that the heavy slack wax candle emitted BbF (3.46 ± 3.45 ng/g), BkF (3.50 ± 3.49 ng/g) and BaP (3.44 ± 3.40 ng/g). Furthermore, Ciecierska and Obiedzinski (2013) investigated the 13 samples of vegetable unconventional oils and found that they were contaminated with 4 heavy PAHs including BaP (1.02 ± 0.09 to 15.74 ± 0.66 μg/kg), CHR (0.89 ± 0.08 to 9.05 ± 0.85 μg/kg), BaA (0.32 ± 0.01 to 8.60 ± 0.53 μg/kg) and BbF (0.64 ± 0.07 to 4.99 ± 0.19 μg/kg). Chiang et al. (1999) investigated vegetable oil fumes and found carcinogenic BaP (21.6 ± 1.3 $\mu g/m^3$), DBA (3.2 ± 0.1 $\mu g/m^3$), BbF (2.6 ± 0.2 $\mu g/m^3$) and BaA (2.1 ± 0.4 $\mu g/m^3$). Those findings supported the result of high concentration of BbF being emitted because burning of candles and incenses were common behavior of the visitors during praying. However, the 8-hrs indoor BaP concentrations measured from special occasions including Chinese New Year (6.8 ± 4.9 to 10.0 ± 2.2 ng/m^3) in this study were much lower than the values found in Taiwanese temple (57.6–98.2 ng/m^3 (Lin et al., 2001) and 125 ± 88 ng/m^3 (Lin et al., 2002)). However, it was higher than the concentration found in Thai temples (2.52 ± 0.83 ng/m^3) reported in the previous study (Navasumrit et al., 2008). Moreover, The 24-hrs BaP concentrations collected indoor of incense burning homes in Taipei, Taiwan (2.4 ± 2.7 ng/m^3) (Li and Ro, 2000) was lower than the result from other special occasions and Chinese New Year in this study (3.38 ± 1.48 and 6.26 ± 2.53 ng/m^3, respectively). This is probably due to lower amount of incense being burnt during the sampling period.

Table 2
Indoor $PM_{2.5}$ and $PM_{2.5}$-bound PAHs concentrations (mean ± SD) in the shrine in various occasions.

Sampling periods	$PM_{2.5}$ concentrations ($\mu g/m^3$)		Total − PAHs concentrations (ng/m^3)	
	8 h	24 h	8 h	24 h
Chinese New Year (n = 3* and n = 6**)	625 ± 147[a]	406 ± 159[a]	90 ± 41[a]	45 ± 29[a]
Other special occasions (n = 9* and n = 8**)	184 ± 85[b]	133 ± 71[b]	71 ± 30[a]	30 ± 12[a]
Normal period (n = 9* and n = 13**)	100 ± 35[c]	50 ± 20[c]	25 ± 15[b]	14 ± 9[b]

[a, b, c] = significant differences ($p < 0.05$) among groups of sampling periods.
*The amount of samples (8 h) and **the amount of samples (24 h).

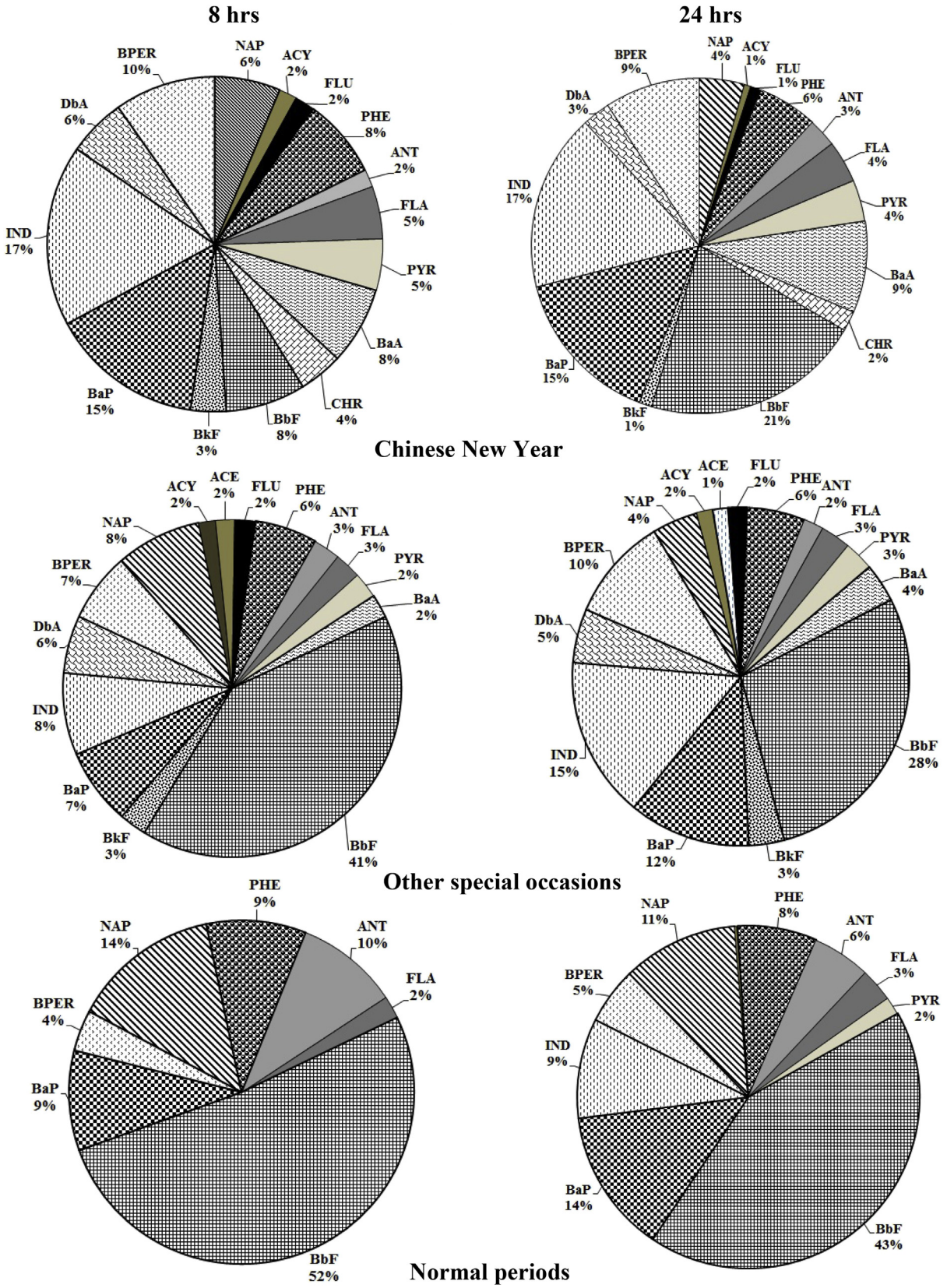

Fig. 2. Variation of PAHs-bound $PM_{2.5}$ at shrine.

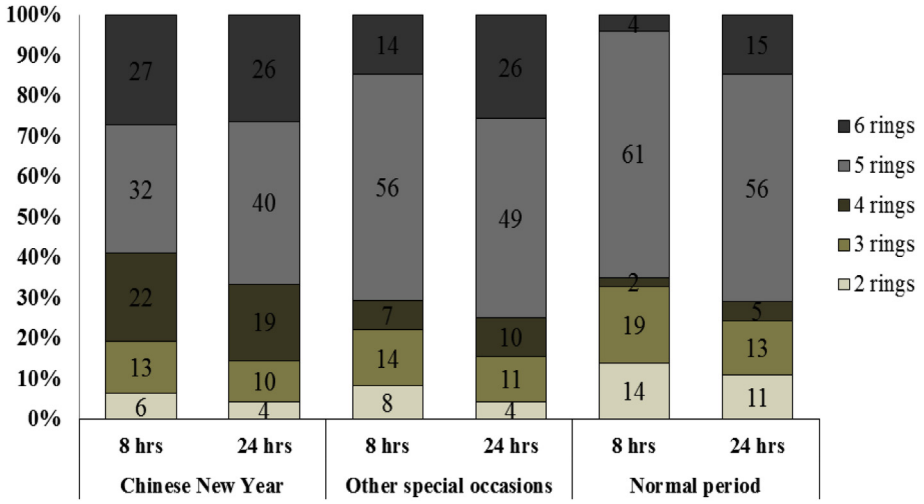

Fig. 3. Ratio of PAHs containing 2–6 rings in $PM_{2.5}$ collected inside shrine.

The 16 PAHs were clustered based on the number of aromatic rings in their structure (2, 3, 4, 5 and 6 rings) (Fig. 3). It was found that 5 rings PAHs were dominated in all occasions. The major component of indoor $PM_{2.5}$-bound PAHs emitted from incense burning was 5-ring PAHs (32–61%). Yang et al. (2013) reported that high molecular weight PAHs with high carcinogenic potency are dominant in the particulate phase from incense burning. Dewangan et al. (2014) also mentioned that the smoldering conditions of incense burning was considered to emit high molecular weight (4–5 rings) of PAHs adsorbed to particle phase, in which 35–37% for 4 rings and 31–35% for 5 rings on particle phase were observed. Wu et al. (2006) mentioned that high molecular weight of PAHs (5 and 6 rings) were mainly associated with diameter of particles less than 2 μm. The results in this study were well agreed with those previous studies.

$PM_{2.5}$ samples were collected for 8 h (8 am–4 pm) and 24 h (8 am–8 am) at the sampling site shrine. The mean values of t-PAHs of the 8-hrs samples were greater than those of the 24-hrs samples in all occasions (Table 2). Moreover, the trend of t-PAH concentrations in every occasion were well related with the trend of $PM_{2.5}$ concentrations. The reason might be due to an opening period of the shrine, which is regularly open in the daytime (8 am–5 pm) except only for Chinese New Year, when the shrine is all day open.

The average concentrations of carcinogenic PAHs (c-PAHs) and non-carcinogenic PAHs (nc-PAHs) are shown in Table 3. The c-PAHs were BaA, CHR, BkF, BbF, BaP, IND, and DbA while nc-PAHs were NAP, ACY, ACE, FLA, PHE, ANT, FLU, PYR and BPER (US-EPA, 2008). The average concentrations (ng/m^3) of c-PAHs and nc-PAHs for 8 and 24 h samplings were highest in Chinese New Year following with other special occasions and normal period. However, they were not significantly different ($p > 0.05$) between Chinese New Year and other special occasions but significantly higher than normal period. The exception was the average concentrations of c-PAHs (24 h sampling), which was significantly higher in Chinese New Year than the other special occasion. Moreover, the c-PAHs concentrations of both 8 and 24 h samplings were about 1–2 times higher than those of nc-PAHs in every occasion including normal period. The c-PAHs were found in high concentrations because high molecular weight PAHs (>200) could be more easily adsorbed to particulate phase (Orecchio, 2011).

3.2. Correlations between $PM_{2.5}$ and PAHs

The correlations of concentration of $PM_{2.5}$, t-PAHs, c-PAHs and nc-PAHs obtained in all occasions are shown in Table 4. The Spearman's correlations can be classified into strong, moderate and weak correlation with correlation coefficient (r) in the range value of 1.000–0.500, 0.499–0.300 and 0.299–0.000, respectively, while negative correlation is presented in negative value (Xie et al., 2015). Therefore, relatively strong correlations ($p < 0.01$) between $PM_{2.5}$ and all type of PAHs were found in 24-hrs sampling ($r \sim 0.6$–0.7, $p < 0.01$). Comparing with a study of Masiol et al. (2012) on gaseous air pollutant in Italy, the correlation coefficient of 24-hrs $PM_{2.5}$ and t-PAHs concentrations from the inside of a shrine in this study was lower than that in spring ($r = 0.85$) but higher than that in autumn ($r = 0.40$). The obtained results indicated that PAHs concentrations associated with fine particles are highly dependent on the incense burning at shrine (Bootdee and Chantara, 2014).

3.3. Source of $PM_{2.5}$-bound PAHs

Besides incense burning, the location of the sampling shrine has been influenced by traffic. In order to confirm that the PAHs concentrations measured in this study were mainly from incense

Table 3
Concentrations (mean ± SD) of c-PAHs and nc-PAHs (ng/m^3) in the shrine in various occasions.

Sampling periods	c-PAHs		nc-PAH	
	8 h	24 h	8 h	24 h
Chinese New Year (n = 3* and n = 6**)	63.8 ± 34.1^a	27.4 ± 14.3^a	26.4 ± 9.4^a	17.8 ± 15.6^a
Other special occasions (n = 9* and n = 8**)	48.5 ± 24.5^a	20.5 ± 10.3^a	22.2 ± 8.4^{ab}	9.6 ± 3.6^{ab}
Normal period (n = 9* and n = 13**)	15.4 ± 12.0^b	9.6 ± 7.5^b	9.8 ± 7.0^b	4.6 ± 2.6^b

$^{a, b}$ = Significant differences ($p < 0.05$) among groups of sampling periods (vertical direction).
*The amount of samples (8 h) and **the amount of samples (24 h).

Table 4
Correlations of $PM_{2.5}$ and PAHs concentrations for 8-hrs and 24-hrs sampling in all occasions.

	8 h (n = 21)				24 h (n = 27)			
	$PM_{2.5}$	t-PAHs	c-PAHs	nc-PAHs	$PM_{2.5}$	t-PAHs	c-PAHs	nc-PAHs
$PM_{2.5}$	1.000				1.000			
t-PAHs	0.475[b]	1.000			0.674[a]	1.000		
c-PAHs	0.451[b]	0.982[a]	1.000		0.618[a]	0.980[a]	1.000	
nc-PAHs	0.448[b]	0.795[a]	0.700[a]	1.000	0.738[a]	0.850[a]	0.757[a]	1.000

[a] Correlation is significant at the 0.01 level (2-tailed).
[b] Correlation is significant at the 0.05 level (2-tailed).

burning, sources of the PAHs pollution in the matrix were investigated and estimated by isomeric ratios. The isomeric ratios are concentration ratios of some PAHs considering as fingerprint of an emission source (Orecchio, 2011; Tobiszewski and Namienik, 2012; Wiriya et al., 2013). PAHs isomer pair ratios have extensively been used to explain the characteristics of the specific source. Table 5 shows isomeric ratios from various studies. The FLA/(FLA + PYR) and IND/(IND + BPER) ratios have been used to specify sources of PAHs. Value of FLA/(FLA + PYR) and IND/(IND + BPER) higher than 0.50 indicated grass, wood and coal combustion, while value lower than 0.50 indicated fossil fuel combustion or petroleum input (Hischenhuber and Stijve, 1987; Yunker et al., 2002). Ratio of BaA/(BaA + CHR) implied petroleum (<0.20), petroleum or combustion (0.20–0.35) and combustion (>0.35) (Hischenhuber and Stijve, 1987). The ratio value of ANT/(ANT + PHE) < 0.10 was referred to petroleum, while a ratio > 0.10 indicated high temperature source (combustion) (Yunker et al., 2002).

When the investigated samples calculated by isomeric ratios are not in agreement with each other, a total index was used to confirm emission of PAHs. Mannino and Orecchio (2008) suggested the total index equation (Eq. (1)), which defined the total index as the sum of the above mentioned isomeric ratios. The total index higher than 4.0 indicated PAHs from high temperature processes (combustion), while the value less than 4.0 indicated low temperature (petroleum products).

$$\text{Total index} = \frac{\text{FLA}/(\text{FLA}+\text{PYR})}{0.4} + \frac{\text{ANT}/(\text{ANT}+\text{PHE})}{0.1} + \frac{\text{BaA}/(\text{BaA}+\text{CHR})}{0.2} + \frac{\text{IND}/(\text{IND}+\text{BPER})}{0.5} \quad (1)$$

The median values of isomeric ratios in 8- and 24-hrs sampling in this study were applied to identify sources of PAHs. Our results revealed that FLA/(FLA + PYA) and IND/(IND + BPER) ratios were higher than 0.50, which indicated grass, wood and coal combustion. The value of BaA/(BaA + CHR) ratio was 0.64–1.00, which implied combustion, while ratio of ANT/(ANT + PHE) was higher than 0.10 indicating high temperature source (combustion). Furthermore, the total index is 7.1–10.8, which PAHs assumed that ratios >4.0 indicates high temperature processes (combustion). All results of the isomeric ratios were in good agreement with the reference values for emission from grass, wood and coal combustion and high temperature source (combustion) as shown in Table 5. Therefore, a significant source of PAHs measured from indoor air of the shrine ensured incense burning as a major combustion source in the sampling site shrine.

3.4. Heath risk assessment of PAHs-bound $PM_{2.5}$

3.4.1. Toxicity equivalent (TEQ) and mutagenic equivalent (MEQ) concentration

The toxicity equivalent concentration (TEQ) is widely used to assess risk of carcinogenic potency of each individual PAH. This parameter is calculated from total of individual concentration of each PAH multiply by their toxic equivalent factor (TEF) relative to the carcinogenic potency of BaP (Eq. (2)), which was used as a reference carcinogenic compound. The toxicity equivalent concentration (TEQ) was calculated based on Nisbet and LaGoy (1992). The mutagenic equivalent concentration (MEQ) was calculated by multiplying the concentration of each PAH compound with the mutagenic equivalent factor (MEF) relative to the mutagenic potency of BaP (Eq. (3)) (Durant et al., 1999; Jung et al., 2010). The

Table 5
Isomeric ratios of some PAHs.

Isomeric ratios	Sampling hours	Ratio values			Reference values	Sources	References
		Chinese New Year	Other special occasions	Normal periods			
FLA/(FLA + PYR)	8	0.51	0.50	1.00	<0.40	Petroleum input	Yunker et al. (2002)
	24	0.51	0.51	0.65	0.40–0.50	Fossil fuel combustion	
					>0.50	**Grass, wood and coal combustion**	
IND/	8	0.64	0.54	ND	0.20	Petroleum	Hischenhuber and Stijve (1987)
(IND + BPER)	24	0.65	0.61	0.64	0.20–0.50	Liquid fossil fuel combustion	
					>0.50	**Grass, wood and coal combustion**	
BaA/	8	0.64	1.00	ND	<0.20	Petroleum	Hischenhuber and Stijve (1987)
(BaA + CHR)	24	1.00	1.00	ND	0.20–0.35	Petroleum or combustion	
					>0.35	**Combustion**	
ANT/	8	0.18	0.30	0.51	<0.10	Low temperature source (petroleum)	Yunker et al. (2002)
(ANT + PHE)	24	0.32	0.25	0.42	**>0.10**	**High temperature source (combustion)**	
Total index	8	7.5	10.3	7.6	<4.0	Low temperature source (petroleum)	Mannino and Orecchio (2008)
	24	10.8	10.0	7.1	**>4.0**	**High temperature source (combustion)**	

ND = not detected. Bolds represent values and their meanings obtained from this study.

mutagenic activity is not uniformly implicated for cancer (Zeiger, 2001) but it is associated to some adverse health effect such as pulmonary diseases (DeMarini et al., 2004). However, the carcinogenic potency is more worldwide using than mutagenic potency.

$$\begin{aligned}TEQ = {} & 0.001(NAP + ACY + ACE + FLU + PHE + FLA + PYR) \\ & + 0.01(ANT + BPER + CHR) \\ & + 0.1(BaA + BbF + BkF + IND) + BaP + DbA\end{aligned} \quad (2)$$

$$\begin{aligned}MEQ = {} & 0.082(BaA) + 0.017(CHR) + 0.25(BbF) + 0.11(BkF) \\ & + 0.31(IND) + 0.29(DbA) + 0.19(BPER) + BaP\end{aligned} \quad (3)$$

The average TEQ and MEQ values are shown in Table 6. The 8-hrs TEQ and MEQ values were higher than 24-hrs sampling. The TEQ values are mainly calculated based on c-PAHs concentrations. Therefore their numbers were well associated. The highest TEQ values were found in Chinese New Year, which were 31.8 ± 27.4 ng/m^3 (8 h) and 9.8 ± 4.3 ng/m^3 (24 h). Comparing the 8-hrs TEQ value of this study with the previous study conducted in a temple by collecting of 8-hrs particle samples using personal samplers. It was found that the values of TEQ in our study (3.2 ± 1.8 to 31.8 ± 27.4 ng/m^3) were obviously higher than the previous work (0.29 ± 0.12 to 4.60 ± 1.35 ng/m^3) (Navasumrit et al., 2008). Comparing with the study in Taiwan (Kuo et al., 2008), where PM_{10} samples were collected for 8-hrs in a shrine, the TEQ value (36.6 ng/m^3) was almost the same with the value obtained during Chinese New Year in this study. The TEQ values found in this study were much higher than the European guideline (1 ng/m^3). The average MEQ values (8- and 24-hrs) in Chinese New Year, other special occasions and normal periods were 25.3 ± 12.2 ng/m^3 and 11.9 ± 5.4 ng/m^3, 18.0 ± 8.0 ng/m^3 and 8.0 ± 3.1 ng/m^3 and 5.2 ± 3.3 ng/m^3 and 3.6 ± 2.2 ng/m^3, respectively. Most of MEQ values were higher than TEQ values for all occasions because MEF values of some PAHs i.e. BbF, INP and BPER were almost 2–3 times higher than TEF values (Eqs. (1) and (2)). Nevertheless, both of TEQ and MEQ values presented the similar trends. The result was well agreed with TEQ and MEQ values for 8-hrs $PM_{2.5}$ sampling from indoor and outdoor residents of young children in New York City (Jung et al., 2010). They found that the average TEQ values (0.478 ± 0.709 ng/m^3) were lower than MEQ values (0.590 ± 1.320 ng/m^3) from indoor sampling due to the dominant of BbF, INP and BPER. Moreover, MEQ values (0.81–14.56 ng/m^3) of 8-hrs $PM_{2.5}$ collected from indoor of primary school in Lithuania (Krugly et al., 2014) were similar to the results found in our study (special occasions and normal period). However, the TEQ and MEQ values from the studied of Jung et al. (2010) and Krugly et al. (2014), were lower than those values obtained in Chinese New Year in this study. The main factors should be ventilation system and the amount of incense being burned. When compared with a study in ambient air of Chiang Mai (Peangchai et al., 2008; Chantara and Sangchan, 2009; Wiriya et al., 2013), where 24-hrs PM_{10} samples were collected in urban and sub-urban areas. It was found that the ranges of TEQ values were 0.92–1.76 ng/m^3 and 0.75–1.60 ng/m^3 in urban area and 0.25–3.70 ng/m^3 in sub-urban. It can be concluded that TEQ values calculated from ambient samples were much lower than the values reported in this study in every occasion.

3.4.2. The inhalation cancer risk assessment

The inhalation cancer risk (ICR) was used to estimate the value of cancer risk from PAHs exposure during the different periods and can be calculated using Eq. (4) (Sarkar and Khillare, 2012; Wiriya et al., 2013). Several studies (Jung et al., 2010; Jia et al., 2011; Sarkar and Khillare, 2012; Wiriya et al., 2013) used this equation to estimate the cancer risk.

$$ICR = TEQ \times IUR_{BaP} \quad (4)$$

where, IUR_{BaP} is the inhalation unit risk. Two different IUR_{BaP} values were used in this study to estimate the inhalation cancer risk. The recommended IUR_{BaP} of the World Health Organization (WHO) is 8.7×10^{-5} m^3/μg (WHO, 2000), while the IUR_{BaP} of the Office of Environmental Health Hazard Assessment (OEHHA) of California Environmental Protection Agency (CalEPA) is 1.1×10^{-6} m^3/μg (Collins et al., 1998; OEHHA, 2003) for lifetime (70 years) PAH exposure. The mean TEQ value was obtained from calculation of each period (Eq. (1)). The inhalation cancer risk was calculated and compared among different periods as shown in Table 6. The 24-hrs of lifetime inhalation cancer risks calculated based on WHO guideline for Chinese New Year, other special occasions and the normal periods were 85×10^{-5}, 60×10^{-5} and 23×10^{-5}, respectively, while those based on CalEPA were 11×10^{-6}, 75×10^{-7} and 29×10^{-7}, respectively. Hence, if a million people were exposed to 10 ng/m^3 TEQ (Chinese New Year) for 70 years, 850 persons may have a chance of cancer development. Following the same calculation, 11 persons based on CalEPA may develop cancer. The ICR values between 10^{-6} to10^{-4} are potential risk and ICR of 10^{-6} represents a lower-bound zero risk, while the upper 10^{-4} of ICR indicates high potential health risk (Chiang and Liao, 2006; Sarkar and Khillare, 2012). The calculated ICR values were found to be in the high potential health risk based on WHO, while those based on CalEPA were in a lower-bound zero risk. The societal inhalation cancer risk was obtained by multiplication of ICR values with million people (Jung et al., 2010). The 24-hrs average value of ICR in normal period based on WHO guideline were 230 cases/million people or 3.30 cases/year, while those based on CalEPA was

Table 6
TEQ and MEQ concentrations (mean ± SD) and inhalation cancer risk (ICR) of $PM_{2.5}$-bound PAHs at the shrine in various occasions.

Risk assessment of indoor		Sampling hours	Chinese New Year	Other special occasions	Normal periods
TEQ concentration (ng/m^3)		8	31.8 ± 27.4 (n = 3)	15.1 ± 7.6 (n = 9)	3.2 ± 1.8 (n = 10)
		24	9.8 ± 4.3 (n = 6)	6.8 ± 2.6 (n = 8)	2.6 ± 1.5 (n = 13)
MEQ concentration (ng/m^3)		8	25.3 ± 12.2 (n = 3)	18.0 ± 8.0 (n = 9)	5.2 ± 3.3 (n = 10)
		24	11.9 ± 5.4 (n = 6)	8.0 ± 3.1 (n = 8)	3.6 ± 2.2 (n = 13)
ICR	WHO (8.7×10^{-5} m^3/ng)	8	28×10^{-4}	13×10^{-4}	28×10^{-5}
		24	85×10^{-5}	60×10^{-5}	23×10^{-5}
	CalEPA (1.1×10^{-6} m^3/ng)	8	35×10^{-6}	17×10^{-6}	35×10^{-7}
		24	11×10^{-6}	75×10^{-7}	29×10^{-7}
Risk (ICR × 10^6)	WHO	8	2800	1300	280
		24	850	600	230
	CalEPA	8	35	17	4
		24	11	8	3

3 cases/million people or 0.04 cases/year. Comparing with a previous studies, the inhalation cancer risk of PM_{10} in ambient air of Chiang Mai, based on WHO was lower (2 cases/year (Pengchai et al., 2008) and 1.3 cases/year, (Wiriya et al., 2013)) than the values found in this study (3.3 cases/year). However, these results should be noted that the values are only a crude estimation of cancer risk from the $PM_{2.5}$-bound PAHs inhalation.

4. Conclusion

The concentrations of total PAHs found in the sampling site shrine were associated with indoor $PM_{2.5}$ samples. It was clearly seen that levels of those pollutants were significantly higher than on special occasions than normal periods. During special occasions, with high amount of incense burned, emission of $PM_{2.5}$ and c-PAHs could be high and can cause a potential risk to human health. The inhalation cancer risk assessment was calculated based on the toxicity equivalent concentrations. The results revealed that the equivalent values during Chinese New Year were much higher than other occasions, which was related with concentrations of PAHs.

Conflict of interest statement

The authors declare that there are no conflicts of interest.

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