

Characterizing PAH emission concentrations in ambient air during a large-scale joss paper open-burning event

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Abstract

Large-scale open burning of joss paper is an important ritual practice for deity worshipping during Buddhist and Taoist festivals. Since Buddhism and Taoism are two of the most popular religions in Chinese societies and some Asian countries, the impact of joss paper burning on the air quality needs further investigation. This study explores the concentrations of polycyclic aromatic hydrocarbons (PAHs) in ambient air during one of the most important festivals, in which large-scale burning of joss paper occurs in temples and in people's houses. The PAH concentrations were measured simultaneously at a temple site and a background site during both the festival and non-festival (ordinary) periods. Each ambient sample was extracted by the Soxhlet analytical method (for both particle-bound and gas-phase) and analyzed with gas chromatography. Experimental results indicate that the total PAH concentration during the festival period is approximately 4.2 times higher than that during the ordinary period (5384 ng m^{-3} vs. 1275 ng m^{-3}). This study also employed statistical methods including diagnostic ratios and principal component analysis (PCA) to identify the possible PAH emission sources. Joss paper burning and vehicular emissions are identified as the principal sources of airborne PAHs during the large-scale open-burning event. The results of this work provide useful information for public awareness concerning PAH emission from the open burning of joss paper.

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1. Introduction

The burning of joss paper and incense is a sacred ritual practice for deity worshipping in Buddhism and Taoism. It is widely practiced in Asia since Buddhism and Taoism are the two most prevalent religions in Taiwan as well as some other Asian countries. Moreover, Chinese and other Asian immigrants in western countries also perform these ritualistic practices. The ritual is generally practiced on the 1st and 15th (or 2nd and 16th) day of the Chinese lunar month. Joss paper is mainly composed of recycled paper and bamboo. The typical chemical composition of joss paper is around 45% oxygen, 40% carbon, 5% hydrocarbon, and small amounts of nitrogen, sul-

fur, and chlorine. Among these compositions, the percentages of combustible materials, moisture and ash content are about 88%, 7%, and 5%, respectively [1]. During important festivals, joss paper is usually burned in the open or in temple furnaces, as well as in the home. However, some investigation indicated that incense and joss paper burning is a significant source of particulate matter (PM) [2–4], metal elements [5,6], and polycyclic aromatic hydrocarbons (PAHs) [7–9] in ambient air. Epidemiological studies performed in several countries show that consistent exposure to ambient particles has adverse health effects, such as increased mortality, respiratory or cardiovascular diseases, respiratory symptoms, and decreased lung function. Epidemiological time-series studies have identified dose–response functions between an increase in PM and adverse health effects [10].

During religious festivals there can be an increase in air pollutants due to incense and joss paper burning. A study by

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Yang et al. [1] revealed that PAHs can be generated by the burning of joss paper. Chao et al. [11] investigated incense burning. Their research indicates that virtually all PAHs generated by incense burning are released into the air because the incense ash remained after burning is very low in PAH content. Because incense burning creates a large number of particles in the air, the habitual use of incense raises the exposure to particles of respirable size. However, literature does not provide epidemiological links between respiratory diseases and incense usage [12]. Furthermore, the influence of PAHs, particularly their impacts on human health, is currently not regulated by the Taiwan EPA.

Previous studies mainly concentrated on investigating the indoor/outdoor air quality of temples and emissions of joss paper burned in joss paper furnaces [1,8,9,13]. However, the emission of air pollutants from the open burning of joss paper has not been well characterized. An in-depth review of literature reveals that no attempts have yet been made to quantify the emissions of larger scale joss paper burnings at an open site. Therefore, this study characterized the concentrations of PAH emissions in the ambient air of Chu-Shan, a town located in Central Taiwan, during the Zhong Yuan Jie festival held on 8th August 2006. Zhong Yuan Jie is one of the most important Chinese festivals, involving large-scale open burning of joss paper in temples, as well as joss paper burning in the home. Concentrations of ambient air particulates, and of sixteen major PAHs, were examined and characterized both during the open-burning event and the ordinary period. This study may provide useful information to raise public awareness of PAH emission resulting from the open burning of joss paper.

2. Experiment set-up

2.1. Description of sampling sites

The sampling area of this study was the eastern region of the town of Chu-Shan, which is one of the most prosperous areas in Central Taiwan. For comparisons, samples were collected from a temple site (TS) and a background site (BS). TS is located in open ground about 20 m beside the Ling-Sing Temple, where tons of joss paper was burned in the open. In contrast, BS is located in an area where there are no temples in the neighborhood. Fig. 1 illustrates the geographical information of the sampling sites. The samples were accumulated during the Zhong Yuan Jie festival from the 8th to the 10th of August 2006, and during an ordinary (non-festive) period on the 27th of October. There are no significant industrial emissions around this area, meaning that vehicles are the major emission sources of air pollutants. The traffic density ranged from 10 (around midnight) to 250 vehicles per hour during the sampling period.

During the festival, most of the inhabitants gathered at the temple to worship the souls of the deceased. Some joss sticks (incense) were burned during the worship ceremony. When the ceremony finished, massive amounts of joss paper were piled up on the open ground by the temple and burned. Since joss paper is the major material burned at the open-burning site, the impacts of incense burning is ignored in this study.

2.2. PAHs sample collection

Consecutive samplings of PAHs (gaseous and particulate) in ambient air were performed 3 times per day for 1–3 h each time at TS and BS simultaneously. Meteorological conditions including temperature, relative humidity, wind speed and wind direction were also gathered using a Watch Dog Model 550 weather station. The samples were obtained by a PAH sampling system (Anderson-Graseby Auto5) modified by Li-Teh Co., Taiwan, in accordance with the U.S. EPA Modified Method 5 (40CFR60). This system has been widely adopted for PAH sampling [1,13–16]. Airborne samples were accumulated using two PAH sampling systems with a pump that drew air through a tissue quartz filter (2500 QAT-UP, dimension: 47 mm) to collect airborne particulate PAHs (p-PAHs). The glass cartridge was packed with a XAD-4 resin and supported by a polyurethane foam (PUF) plug to collect the gaseous PAHs (g-PAHs). The sampling train was rinsed with *n*-hexane following each sampling cycle. The PUF plug and resin were stored in a clean jar wrapped with aluminum foil during sample transportation and storage, and the quartz filters were placed in a sealed 60 mm plate. The quartz filters were weighed before and after sampling to determine the amount of particulate accumulated. They were baked in an oven at 450 °C for 8 h before sampling to eliminate organic impurities. Glass cartridges were cleaned by sequential Soxhlet extractions with distilled deionized water, methanol, dichloromethane and a mixture of dichloromethane for 24 h, and finally dried in a contamination-free oven at 45 °C to eliminate the residual solvent.

2.3. Chemical analysis of the PAH samples

To analyze the PAH samples, all quartz filters and glass cartridges were placed separately in the appropriate Soxhlet extractors after being weighed, and the PAHs were extracted with dichloromethane for 24 h. The extract was then concentrated using a rotary evaporator, cleaned, and re-concentrated with ultra-pure nitrogen to exactly 1 mL using a procedure previously described by Lee et al. [17]. All extracts were analyzed by a gas chromatograph (GC, Agilent 5890) with a GC capillary column (Agilent Ultra 2: 50 m × 0.32 mm × 0.17 mm). The volume of the injected sample was 1 mL and was not split. The temperatures of the injector and transfer line were maintained at 310 and 300 °C, respectively. The analytical method was based on the US EPA Method TO-13 [18].

2.4. Quality control

A standard PAH mixture purchased from Supelco Co., USA containing 16 PAH compounds (concentration: 100–2000 µg mL⁻¹) was used to create calibration curves. Sixteen PAHs, including naphthalene (Nap), acenaphthylene (AcPy), acenaphthene (AcP), fluorine (Flu), phenanthrene (PA), anthracene (Ant), fluoranthene (FL), pyrene (Pyr), cyclopenta[c,d]pyrene (CYC), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene

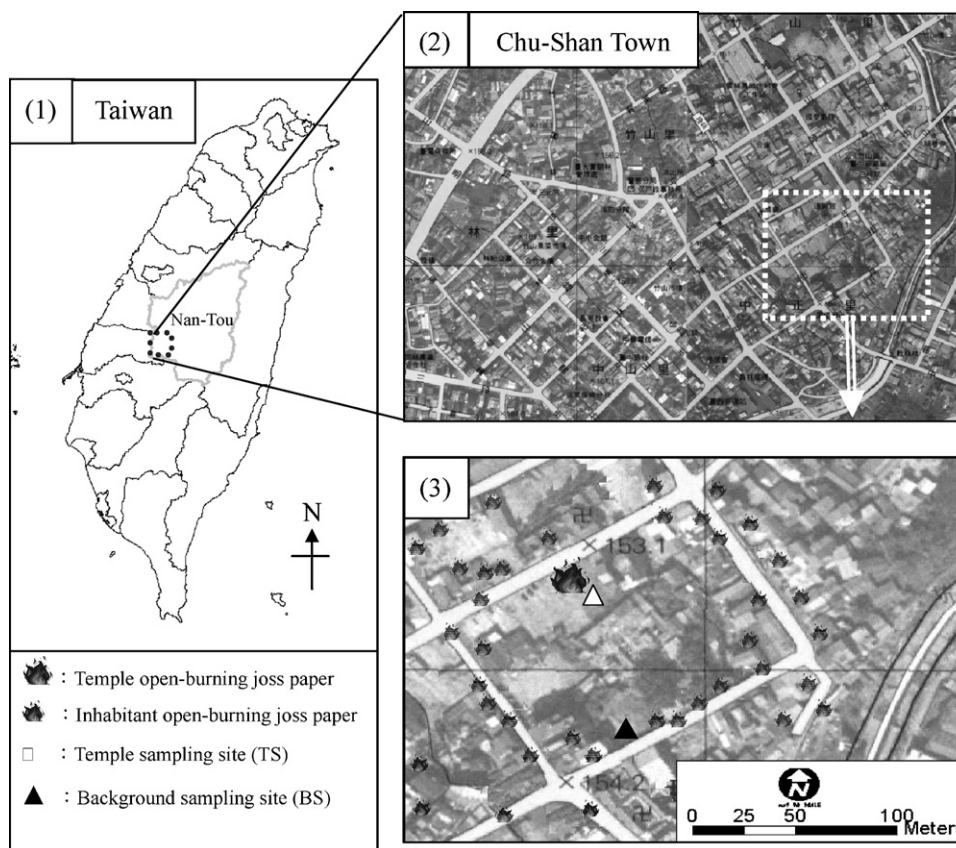


Fig. 1. The sampling positions of this study.

(PER), were identified and quantified for each collected sample. Analysis of serial dilutions of PAH standards showed the method detection limit (MDL) to be between 0.17 (BkF) and 0.27 (BaP) $\mu\text{g mL}^{-1}$. The recovery efficiencies of 16 individual PAHs were determined by processing a solution containing known PAH concentrations following the same experimental procedure used for the samples. A NIST SRM-1597 was used in this study to check the recovery efficiency of PAHs, and the results showed that the recovery efficiency varied between 77% and 108% and averaged 86%. Field and laboratory blank samples were routinely analyzed to evaluate analytic bias and precision. Blank levels of individual PAHs were normally very low, and in most cases, the GC integrated area was less than the detection limit.

3. Results and discussion

3.1. Particulate mass concentration in ambient air

The meteorological conditions during the sampling period are listed in Table 1. The temperature was between 24 and 38 °C, and the relative humidity was between 28% and 92%. The maximum wind speed was 2.7 m s^{-1} , and the atmospheric pressure was between 1006.7 and 1011.2 hpa. The predominant wind directions were south-west and south-east during the festival and ordinary sampling periods, respectively.

The particulate concentrations during the festival period at TS and BS, as demonstrated in Table 1, were 176–582 and

96–346 $\mu\text{g m}^{-3}$, respectively. The maximum particulate concentrations of both TS (582 $\mu\text{g m}^{-3}$) and BS (346 $\mu\text{g m}^{-3}$) exceeded the regulatory limits set by the World Health Organization (150–230 $\mu\text{g m}^{-3}$) [19]. The maximum particulate concentration at TS was higher than the BS sampling site by about 1.7 times during the festival period (582 $\mu\text{g m}^{-3}$ vs. 346 $\mu\text{g m}^{-3}$). During the ordinary period (27th October), the particulate concentrations were 142 and 86 $\mu\text{g m}^{-3}$ at TS and BS, respectively. The maximum concentrations at TS and BS during the festival period were about 4.0 times higher than the concentrations during the ordinary period (582 $\mu\text{g m}^{-3}$ vs. 142 $\mu\text{g m}^{-3}$, and 346 $\mu\text{g m}^{-3}$ vs. 86 $\mu\text{g m}^{-3}$), indicating that the open burning of joss paper significantly raised the particulate concentrations in the ambient air.

3.2. PAH concentrations in ambient air

Table 2 presents the average concentrations of the g-PAHs and p-PAHs at TS and BS during the festival period. The table illustrates that, for the gas-phase, Nap, AcPy, and AcP were the three most abundant g-PAHs for both TS and BS, followed by Flu, PA, and Ant. In the particle phase, BeP, BbF, and BkF were the three most abundant p-PAHs for both TS and BS, followed by BaP, PER, BaA, and CHR.

Table 2 also demonstrates that almost all of the total concentrations of individual PAH at TS were higher than those at BS. The average concentrations of the total PAHs at TS and BS were 2684 and 1885 ng m^{-3} , respectively, indicating that

Table 1
Basic information of the sampling sites

Sampling date (month-date-year)	Sampling period	TS ^a ($\mu\text{g m}^{-3}$) ^h	BS ^b ($\mu\text{g m}^{-3}$) ^h	T ^c ($^{\circ}\text{C}$)	RH ^d (%)	WS ^e (m s^{-1})	WD ^f	P ^g (hPa)
08-08-2006	14:00–21:00	352 ± 145	346 ± 101	27–38	28–41	0.4–1.9	SE	1006.7
08-08 to 09-2006	21:00–02:00	582 ± 226	225 ± 89	24–27	51–75	0.2–2.1	S	1007.1
08-09-2006	02:00–09:00	214 ± 125	96 ± 60	25–35	62–92	0.0–2.0	SE	1007.2
08-09-2006	09:00–21:00	242 ± 116	137 ± 62	27–37	45–69	0.1–1.2	SE	1006.9
08-09 to 10-2006	21:00–06:00	176 ± 85	112 ± 67	24–28	43–70	0.4–2.7	SE	1007
08-10-2006	06:00–12:00	184 ± 67	121 ± 24	26–37	62–87	0.2–1.4	SEE	1006.8
10-27-2006	09:00–16:00	142 ± 70	86 ± 31	28–35	42–63	0.4–1.7	SW	1011.2

^a Temple-burning site.

^b Background site.

^c Temperature.

^d Relative humidity.

^e Wind speed.

^f Wind direction.

^g Atmospheric pressure.

^h Particulate concentration.

Table 2
Average concentrations (ng m^{-3}) of gaseous PAHs (g-PAHs) and particulate PAHs (p-PAHs) in ambient air at TS and BS sampling sites during open-burning period

PAHs	TS			BS		
	g-PAH	p-PAH	Total	g-PAH	p-PAH	Total
Nap	627 ± 361	5 ± 3	632 ± 375	437 ± 312	8 ± 3	445 ± 341
AcPy	374 ± 273	13 ± 4	387 ± 281	262 ± 193	18 ± 5	280 ± 17
AcP	285 ± 188	15 ± 11	300 ± 192	189 ± 85	12 ± 6	201 ± 92
Flu	179 ± 124	12 ± 5	191 ± 125	167 ± 86	10 ± 5	177 ± 89
PA	156 ± 90	23 ± 12	179 ± 112	186 ± 107	13 ± 6	199 ± 108
Ant	140 ± 81	24 ± 17	164 ± 86	105 ± 89	17 ± 5	122 ± 96
FL	38 ± 15	17 ± 9	55 ± 19	31 ± 18	17 ± 6	48 ± 19
Pyr	43 ± 29	24 ± 12	67 ± 47	39 ± 26	10 ± 2	49 ± 27
CYC	33 ± 14	31 ± 17	64 ± 27	27 ± 12	24 ± 16	51 ± 24
BaA	24 ± 12	52 ± 24	76 ± 29	32 ± 12	27 ± 16	59 ± 27
CHR	18 ± 6	49 ± 23	67 ± 30	23 ± 5	16 ± 13	39 ± 16
BbF	24 ± 2	82 ± 63	106 ± 69	11 ± 3	39 ± 22	50 ± 24
BkF	10 ± 2	75 ± 50	85 ± 57	9 ± 6	37 ± 25	46 ± 26
BeP	11 ± 4	149 ± 95	160 ± 103	8 ± 3	52 ± 30	60 ± 31
BaP	10 ± 6	72 ± 57	82 ± 59	7 ± 5	19 ± 15	26 ± 17
PER	7 ± 3	62 ± 24	69 ± 32	6 ± 3	27 ± 12	33 ± 14
Total	1979 ± 1065	705 ± 262	2684 ± 1952	1539 ± 867	346 ± 172	1885 ± 1076

the concentration at TS is approximately 50% higher than the concentration at BS. Moreover, the total g-PAH concentrations are much higher than p-PAH, revealing that most of the PAH emissions of joss paper burning were in gaseous phase.

Fig. 2 displays the fluctuation of PAH concentrations at TS and BS during the sampling periods. Since incomplete combustion occurred in the open burning of joss paper and persisted for a long time, all average PAH concentrations at TS were higher than those at BS. Furthermore, for the ordinary period (stage G), the average total PAH concentrations at the TS and BS sampling sites were 1275 and 524 ng m^{-3} , respectively. The highest concentration at TS, about 4.2 times that for ordinary periods (5384 ng m^{-3} vs. 1275 ng m^{-3}), was observed during stage B, which is the period in which open burning of a large amount of joss paper was just beginning. The concentration at BS during this stage also remained at a high level extended from stage A. However, the concentrations of TS and BS both fell significantly

when a light rainfall began in stage C, leading to the lowest TS value. Nevertheless, the PAH concentration rose by 70% in stage D (from 1309 to 2420 ng m^{-3}) after the rainfall stopped.

The PAHs contain a different number of aromatic rings. Table 3 summarizes the number of aromatic rings of the 16 PAHs analyzed in this study. In general, PAHs having more aromatic rings possess higher molecular weights. Therefore, PAHs can be classified into low molecular

Table 3
Aromatic ring number contained in different PAHs

Number of aromatic rings	PAHs
2	Nap
3	AcPy, AcP, Flu, PA, Ant
4	FL, Pyr, BaA, CHR
5	CYC, BbF, BkF, BeP, BaP, PER

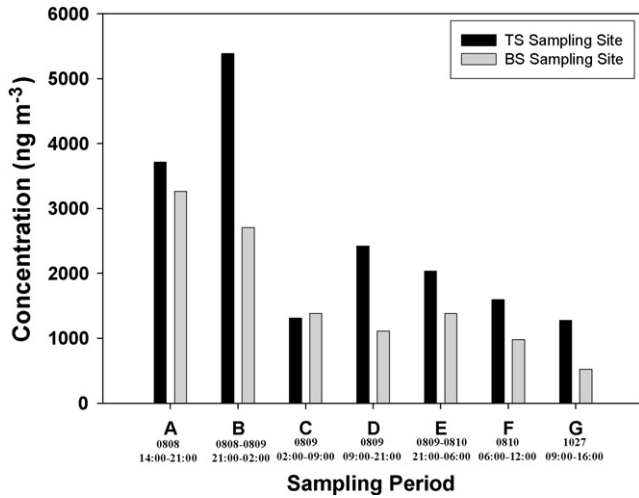


Fig. 2. Total PAH concentrations in ambient air at TS and BS sampling sites during sampling period.

weight (LMW, containing 2- and 3-ringed PAHs), medium molecular weight (MMW, containing 4-ringed PAHs) and high molecular weight PAHs (HMW, containing 5-ringed PAHs) [20].

Graphical statistics and the ratios of the concentration of the igniting stage to the final-smoldering stage (I/F concentration

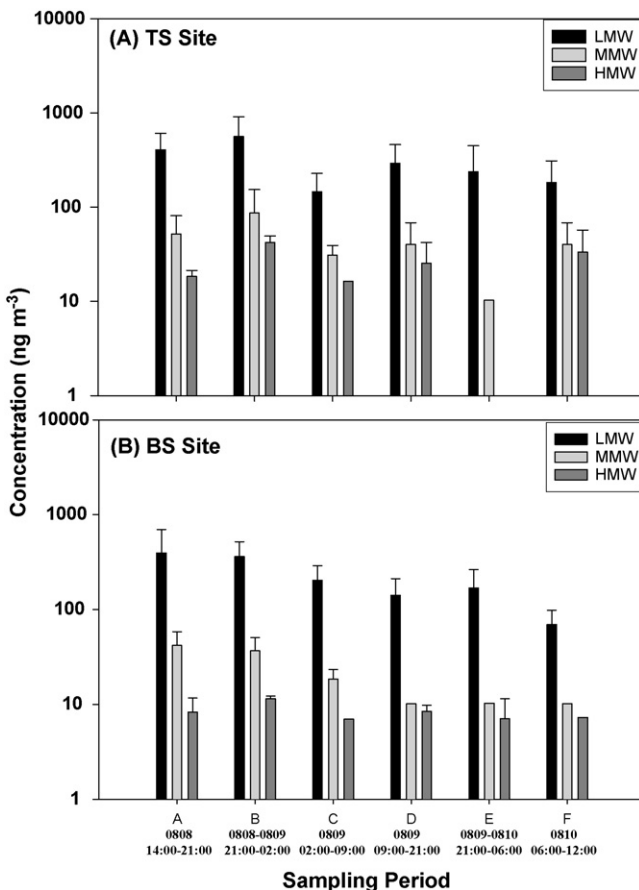


Fig. 3. Average gaseous PAH concentrations in ambient air at TS (A) and BS (B) sampling sites during festival period.

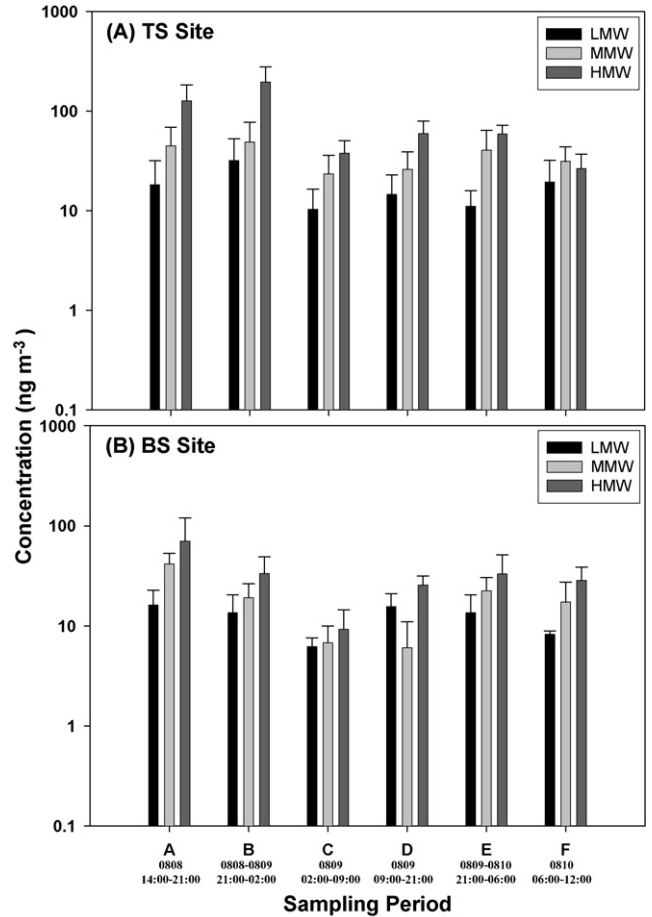


Fig. 4. Average particulate PAH concentrations in ambient air at TS (A) and BS (B) sampling sites during festival period.

ratio) were adopted in this study to investigate the emission characteristics of the three types of PAHs. Figs. 3 and 4 depict the profiles of the average concentrations of g-PAHs and p-PAHs in ambient air at TS and BS, respectively. The highest concentrations of LMW g-PAHs and HMW p-PAHs among all 16 PAHs at the TS and BS sampling sites were observed during the igniting stage (stages A and B) of the open-burning event. LMW g-PAHs were the dominant components for gaseous phase PAHs, while HMW p-PAHs were the dominant components for particle phase PAHs. I/F concentration ratios are summarized in Table 4. In the gaseous phase, it was observed that LMW g-PAHs had the highest I/F ratio at TS and BS, followed by MMW and HMW g-PAHs. However, no obvi-

Table 4
Summary of I/F concentration ratios for gaseous and particulate PAHs during open-burning period

	LMW	MMW	HMW
Temple site (TS)			
g-PAHs	3.1	2.0	1.3
p-PAHs	1.6	1.5	7.5
Background site (BS)			
g-PAHs	5.2	3.6	1.6
p-PAHs	1.5	1.1	1.2

Table 5
Diagnostic rates between total PAH at TS and BS sampling sites

Diagnostic ratio	TS	BS
CHR/BeP	0.51	1.44
Flu/Pyr	2.87	3.66
BaA/BaP	0.94	1.88
Pyr/BaP	0.80	2.10
BaA/CHR	1.08	1.31
BaP/CHR	1.12	0.70

ous trend was observed for the particle phase except that the I/F ratio of HMW p-PAHs at TS was significantly higher than others.

3.3. Statistical analysis for pollution sources identification

An increasing number of recent studies have applied diagnostic ratios to help identify possible PAH emission sources [7,20–24]. Table 5 presents six diagnostic ratios of total PAH concentrations at TS and BS during the festival period. At TS, the ratios of CHR/BeP and Pyr/BaP were 0.51 and 0.8, respectively. These were similar to the ratios measured for joss-paper burning in a previous study [1], which found CHR/BeP and Pyr/BaP ratios of 0.26–0.54 and 0.75–1.55, respectively. Ratios of Flu/Pyr at TS and BS were 2.87 and 3.66, respectively, and they were within the range from 1.7 to 4.2 for diesel engines reported by previous studies [20–22], implying that diesel emission might be a potential emission source at the sampling sites. With regard to other diagnostic ratios such as BaA/BaP, BaA/CHR, and BaP/CHR, no obvious range of values corresponding to this study were available in literature. Therefore, statistical methods, as described in the next paragraph, were adopted in this study to help identify the possible PAHs emission sources.

This study employed principal component analysis (PCA), which is the most widely used multivariate statistical technique in atmospheric sciences, to further identify the major sources of pollutant emissions. By grouping variables with similar characteristics into factors, PCA transforms the original set of variables into a smaller set of linear combinations that retain the original information as much as possible [25]. Factor analysis in this study was carried out using the statistical analysis SPSS® 10.0 software package. Only principal components with eigenvalues >1.0 are discussed. The results of the PCA analyses for TS and BS are presented in Table 6, where the factors along with their factor loadings for individual PAHs are given. The PCA results show that three factors can explain a major part of the data variance. At TS, factor 1 explained 37.9% of the total variance of the data and had high loadings of Flu (0.73), PA (0.86), Ant (0.75), Pyr (0.82), CYC (0.74), and BkF (0.82). Flu, PA, and Ant are the indicator PAHs associated with diesel powered vehicles, [7,20,22–23,25–28], and Pyr, CYC, and BkF are associated with gasoline powered vehicles [7,20,28–30]. Since these six PAHs are all indicators of vehicular emissions, factor 1 was characteristic of vehicular emission. Factor 2 explained 17.5% of the total variance and had high loadings of NaP (0.79), AcP (0.77), and CHR (0.92). From previous studies [31–33],

Table 6
Factor analysis of total PAH concentrations data set at TS and BS sampling sites

PAH	TS			BS	
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2
Nap	0.54	0.79	–	–	0.76
AcPy	0.28	0.42	–	–	0.62
AcP	0.42	0.77	–	–	0.42
Flu	0.73	–	0.21	0.75	0.15
PA	0.86	–	0.35	0.74	–
Ant	0.75	0.25	0.12	0.65	–
FL	–	0.41	0.55	–	0.31
Pyr	0.82	–	0.27	–	0.64
CYC	0.74	–	–	0.87	0.10
BaA	–	0.22	0.78	0.37	0.81
CHR	0.41	0.92	0.14	0.55	0.42
BbF	–	0.43	0.81	0.86	0.15
BkF	0.82	–	0.25	0.92	–
BeP	0.21	–	0.74	0.61	0.48
BaP	0.24	0.15	–	0.28	0.62
PER	0.62	–	0.14	0.42	0.51
Eigen value	7.42	4.31	3.36	8.38	6.73
Variance (%)	37.89	17.45	15.32	37.47	21.25
Cumulative (%)	37.89	55.34	70.66	37.47	58.72
Origin	Diesel and gasoline	Paper burning	Stationary source	Diesel and gasoline	–

Note: Only factor loading values with moduli large than 0.1 are presented and the factor loading values with moduli higher than 0.7 are in bold.

NaP, AcP, and CHR are the indicator PAHs of wood combustion emission, indicating that factor 2 was characteristic of joss paper burning emission. Factor 3 is responsible for 15.3% of the total variance, and is related to a stationary emission source with high loadings of BaA (0.78), BbF (0.81), and BeP (0.74) [7,20,22]. At BS, 58.7% of the variance was explained by two factors, while the remaining 41.3% of the variance was unique to individual variables. Factor 1 accounted for 37.5% of the total variance with high loadings of Flu (0.75), PA (0.74), CYC (0.87), BbF (0.86), and BkF (0.92), revealing that vehicular emissions also affected the air quality at the BS sampling site. The diagnostic ratios and PCA analyses indicate that joss paper burning and vehicular emissions were the principal sources of airborne PAHs during the large-scale open-burning event.

4. Conclusions

This study contributes to literature on the characterization of PAH emissions by analyzing air pollutants emitted on large-scale joss paper open-burning event.

Experimental results show that the particulate concentrations exceeded the regulatory limits set by the World Health Organization. The average PAH concentrations in ambient air at the open-burning site (TS) and background site (BS) during the festival period were found to be 2684 and 1885 ng m⁻³, respectively. Most of the individual PAH concentrations at TS were higher than those at BS. The collected PAH concentrations during the festival period were approximately 4 times higher than

those during the ordinary period. The PAH profiles observed during the open-burning event reveal that Nap (31.7%) dominates the gaseous PAH emission, followed by AcPy (18.9%) and AcP (14.4%). Meanwhile, BeP (21.1%) dominates the particulate PAH emission, followed by BbF (11.6%), BkF (10.6%), and BaP (10.2%). Most of the gaseous PAHs with low molecular weights and particulate PAHs with high molecular weights were found to be emitted at the igniting stage of open burning. A statistical analysis shows that the principal sources of pollution at TS and BS during the sampling period were the burning of joss paper and vehicle emissions.

Analytical results indicate joss paper burning is a significant source of air pollutants. These results suggest that joss paper should be incinerated in incinerator or joss paper furnaces that are equipped with pollutant removal facilities, which provide better combustion and pollution control. This can help reduce PAHs in the ambient air, which can make strive to help reduce the sources of air pollution that cause common respiratory disease. This study also provides useful information for public awareness concerning PAH emission from the open burning of joss paper.

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