

## Measurement of Particulate *n*-alkanes and PAHs Inside and Outside a Temple in Xiamen, China

Shui-Ping Wu<sup>1,2</sup>, Xin-Hong Wang<sup>1,2\*</sup>, Hua-Sheng Hong<sup>1,2</sup>, Jing-Ming Yan<sup>1,2</sup>

<sup>1</sup> *State Key Laboratory of Marine Environmental Science, Xiamen University, Xiamen 361005, P. R. China*

<sup>2</sup> *College of Oceanography and Environmental Science, Xiamen University, Xiamen 361005, P. R. China*

---

### Abstract

Total suspended particles samples inside and outside the South Pu-Tuo Temple (SPT), a traditional Buddhist temple in Xiamen, China were collected and further analyzed for *n*-alkanes and polycyclic aromatic hydrocarbons (PAHs) during the periods of worship. It was observed that the concentrations of particulate *n*-alkanes at SPT were abnormally higher compared to the surrounding bus terminus and campus. In addition, benzo[a]pyrene (BaP) equivalent concentrations at SPT (7.1-26.3 ng/m<sup>3</sup>) were significantly higher than those at the bus terminus (5.1-6.9 ng/m<sup>3</sup>) although the PAH concentrations were comparable. The hazard potential of PAHs in terms of the carcinogenicity of BaP inside SPT is not acceptable because the indoor air quality standard of BaP recommended by the State Environmental Protection Administration of China is 1 ng/m<sup>3</sup> (daily mean). Ratios of fluoranthene to fluoranthene plus pyrene, indeno[1,2,3-cd]pyrene to indeno[1,2,3-cd]pyrene plus benzo[g,h,i]perylene and 1,7-DMP (dimethylphenanthrene) to 2,6-DMP plus 1,7-DMP were further calculated; the values of these three together with the ratio of retene to phenanthrene separated the SPT samples from the bus terminus samples, in that SPT samples showed a strong influence of wood burning (such as bamboo sticks, stick coatings, and joss paper).

**Keywords:** Incense burning; Particulate matter; *n*-alkanes; Polycyclic aromatic hydrocarbons; Health risk.

---

### INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs)

are a family of persistent organic pollutants produced mainly by incomplete combustion of fossil fuel and biomass, which are of special interest due to their toxicity, carcinogenicity, and ubiquitous distribution in the environment (IARC, 1991; Khalili *et al.*, 1995). Because of

---

\* Corresponding author. Tel.: 86-592-2187857,  
Fax: 86-592-2180655  
E-mail address: xhwang@xmu.edu.cn

increasing environmental awareness, people are paying more attention to the existence and problems of indoor air pollution since people spend most of their time in either closed or semi-closed environments where carcinogenic PAHs and particulate matter levels are much higher than outdoors, especially those indoor places with activities such as smoking, cooking, and candle and incense burning (Li and Ro, 2000; Lung *et al.*, 2003; de Kok *et al.*, 2004). Incense, which is obtained from certain resinous trees, has been used since ancient times, largely for religious worship or to mask household odors. Incense burning is generally practiced as an important religious rite on a 24-h basis in all Buddhist temples and residential houses. Previous researches have found that the smoke emission from incense burning is full of cancer-causing chemicals and put people who breathe burning incense over long periods at increased risk of lung and bladder cancer as well as occupational skin and scrotal cancers (Koo and Ho, 1996; Reed *et al.*, 2003). Thus, evaluating of the emission of particulate matter and PAHs from incense burning is highly needed. PAHs and particulate matter aside, worshippers and temple staff are also exposed to free radicals and volatile organic compounds emitted from burning incense inside the temple (de Kok *et al.*, 2004; Lee and Wang, 2004).

The South Pu-Tuo temple (SPT) is the biggest and busiest Buddhist temple in Xiamen and attracts a large number of pilgrims and tourists all year round. The activities of incense and candle burning on a 24-h basis would inevitably emit large

amounts of particulate matter and PAHs, and the exposure to contaminated air during religious services may cause severely adverse health effects on the monks and temple staff who spend most of their working time in the temple. The pollutants caused by incense burning in the temple are not regulated by the State Environmental Protection Administration of China (SEPA) and no report concerning indoor air pollution and its impact on human health are found to date. Therefore, the measurement of particulate matter and PAH concentrations inside and outside SPT is necessary to assess the potential effects of incense burning on temple's indoor air quality.

Many studies have reported the emission of particulate matter, PAHs and mutagenic radicals from incense burning. However, there is still very little information regarding the emission of *n*-alkanes from incense burning, although it can cause an increase of lipid peroxidation and the loss of cell respiration (Suleiman, 1987). For chronic exposure, the Voluntary Children's Chemical Evaluation Program (VCCEP) (2005) present the representative and upper margin of exposures (MOE) of the *n*-alkanes (including decane, undecane and dodecane) (6000 and 1900, respectively). The *n*-alkanes provide important source information of organic pollutants in collected aerosols (Azevedo *et al.*, 1999; Kavouras *et al.*, 1999). The present study was aimed at characterizing particulate matter concentrations as well as the profiles of particulate *n*-alkanes and PAHs inside and outside SPT during the periods of worship, and assessing the impact of incense burning

emissions and vehicle exhausts on ambient air quality. The results can provide useful information for assessing health risks and for enhancing health safety self-consciousness in the future.

## EXPERIMENTAL

### *Site Characterization and Sample Collection*

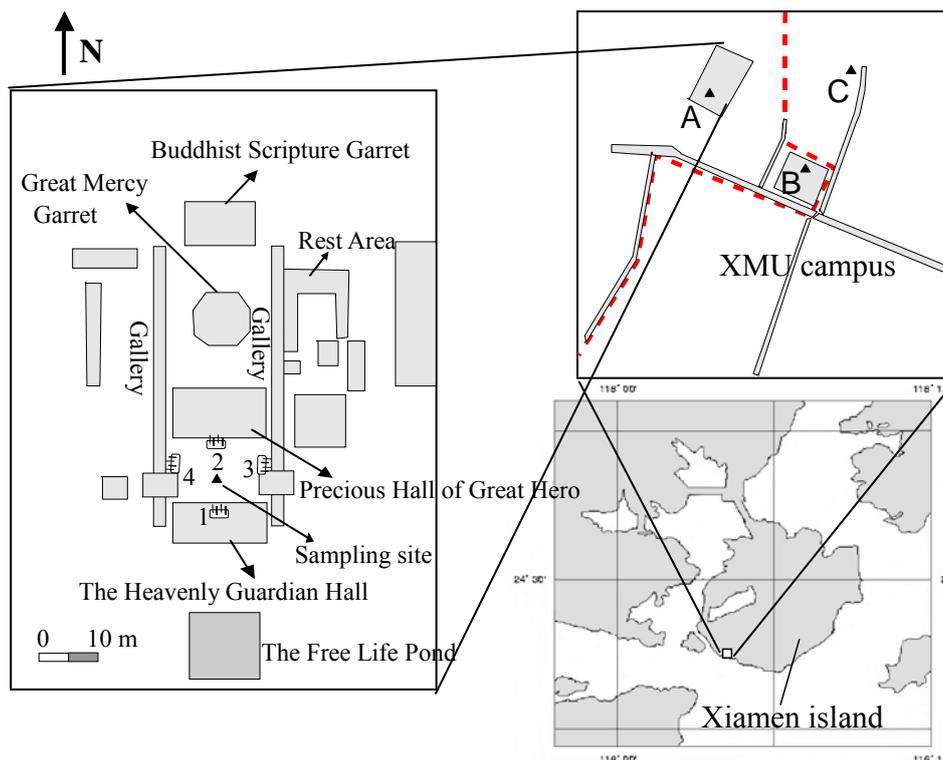
The SPT, located at the southern urban area of Xiamen Island, was chosen for this study to measure particulate matter, *n*-alkanes and PAHs generated from combustion sources, such as the burning of incense and candles. The sampling site was located at the open section of the temple (with no rooftop on the courtyard) at a distance of ~5 m from the nearest censer (Fig. 1). The average number of pilgrims or tourists was around 6000 and 8000 weekday and weekend, respectively, during the sampling period. The number of sticks used was approximately 10 for each pilgrim with a burning rate of 20 min per stick. But not all the sticks were burned out completely because the temple staff would often remove partly burned sticks from the censer in order that other pilgrims or tourists have a place to offer incense. The temple has four large censers in which all the pilgrims' incense was placed. A dispatch building (4 m in height) at the bus terminus and a four-story building in the campus of Xiamen University (12 m in height) were selected to set up samplers to measure outdoor particulate matter (Fig. 1). The bus terminus was a typical site for engine exhaust monitoring. The average traffic

volume of this terminus was approximately 1680 buses per day. The campus site was further departed from general traffic and thus with less impact from vehicle emissions.

Total suspended particles (TSP) were collected on annealed glass fiber filters (Waterman GF/F, 90 mm diameter, baked at 550°C for 4 h) using a medium volume TSP sampler (2030B, Laoshan Instrument and Meter, Qingdao, China) operating at a flow rate of 100 L/min, from 8:30 a.m.-5:30 p.m. daily, from 31 July to 14 August 2006. The sampling height was 1.5 m above ground level to reduce the influence of re-suspended particles and roof dusts. After sampling, the filters were folded in half and put back to their aluminum foil envelopes and transported to the laboratory for chemical analysis as soon as possible. The filters were conditioned at 25°C with a relative humidity of 50% for 24 h. The weight of collected particles was obtained by weighing the filter (Mettler AE240, 0.01 mg, Switzerland) before and after a run. After being weighed, the envelopes were sealed and stored at -20°C in a refrigerator for further analysis. TSP concentrations for each sample and meteorological parameters during the sampling periods are summarized in Table 1.

### *Extraction and Fractionation*

Each sample was extracted in a Soxhlet extractor for 24 h with 150 mL of redistilled dichloromethane (DCM). The extract was concentrated to approximately 15 mL under reduced pressure in a 30°C water-bath using a rotary evaporator. An additional 15 mL



**Fig. 1.** Sampling sites of the TSP samplers in the South Pu Tuo temple (A), bus terminus (B), and XMU campus (C). Location 1, 2, 3 and 4 were the censers in which all the pilgrims' incense was placed.

**Table 1.** Meteorological data and TSP concentrations ( $\mu\text{g}/\text{m}^3$ ) during the sampling period.

Sampling date (2006)	TSP concentration ( $\mu\text{g}/\text{m}^3$ )			Temp ( $^{\circ}\text{C}$ )	Wind speed (m/s)	RH (%)	Weather state
	SPT	Bus Terminus	Campus				
31 Jul (Mon)	245	-	-	26-32	1.6-3.3	50-85	Clear-cloudy
1 Aug (Tue)	188	-	-	26-32	2.4-4.4	55-85	Clear-cloudy
2 Aug (Wed)	-	-	119	26-32	3.3-4.1	70-90	Clear-cloudy
3 Aug (Thu)	-	-	114	26-31	2.0-5.5	70-90	Cloudy-rainy
4 Aug (Fri)	-	57	-	24-29	3.4-5.4	70-95	Rainy
5 Aug (Sat)	336	116	107	24-29	1.6-3.3	75-90	Cloudy
6 Aug (Sun)	570	148	138	25-32	1.6-3.3	50-90	Cloudy
7 Aug (Mon)	-	158	120	25-33	1.6-4.4	55-85	Cloudy-clear
8 Aug (Tue)	-	113	91	26-33	1.6-5.4	55-90	Clear
9 Aug (Wed)	274	161	138	26-33	5.5-9.3	50-90	Cloudy-rainy
10 Aug (Thu)	202	88	67	24-31	5.5-9.3	60-90	Cloudy-rainy
11 Aug (Fri)	168	-	42	26-32	3.5-5.0	50-80	Cloudy
12 Aug (Sat)	429	189	165	26-32	2.5-4.4	55-85	Clear
13 Aug (Sun)	342	152	118	27-34	1.6-3.0	50-80	Clear-cloudy
14 Aug (Mon)	238	126	94	26-34	1.6-5.4	55-85	Clear

*n*-hexane was added to the flask and the contents evaporated again to ~1 mL. The concentrated extract was transferred to the top of a glass column (30 cm × 10 mm i.d.) slurry packed with 10.0 g of silica gel (MACHEREY-NAGEL, 70-230 mesh, activated at 130°C for at least 16 h). Compounds retained on the silica gel columns were eluted sequentially with 25 mL of hexane and then 50 mL of hexane-DCM (6:4) to obtain fractions enriched in *n*-alkanes and PAHs, respectively. Both fractions were further concentrated by nitrogen blow down to a volume of 200 µL, and kept in a Teflon-lined vial insert for gas chromatographic-mass spectrometric (GC-MS) analysis.

### Analysis

Analysis was performed using an Agilent 6890 gas chromatograph with a 5973 mass spectrometry detector operated in an electron impact mode (70 eV). Samples were injected in the splitless mode at 60°C and then the oven temperature was raised by 5°C/min to 280°C (for PAHs) or 300°C (for *n*-alkanes) and held isothermally at each end temperature for 20 min. The GC column used was a 30 m × 0.25 mm i.d. capillary column with 0.25 µm film thickness (HP-5MS, 5% phenyl methyl siloxane). The temperatures of the ion source and the quadrupole mass analyzer were kept at 230°C and 150°C, respectively. Helium, with a purity of 99.999%, was used as a carrier gas at a flow rate of 2.7 mL/min. The injector and transfer line temperatures were held at 250°C and 280°C, respectively. *n*-Alkanes from C<sub>14</sub> to C<sub>35</sub> (Supelco Inc., USA) and PAH

[naphthalene (Nap), acenaphthylene (Acpy), acenaphthene (Acp), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flua), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IP), dibenz[a,h]anthracene (DBA), and benzo[g,h,i]perylene (BghiP), AccuStandard Inc., USA) standard solutions were used to identify the retention times and quantification ions of *n*-alkanes and PAHs in the samples, respectively. The *n*-alkanes were detected in the full-scan mode and ion fragment *m/z*85 was selected for quantitative analysis. The molecular ion fragment of each PAH was selected to identify and quantify the PAH present in the selective ion monitoring mode. In addition, PAHs were also determined in full scan mode to confirm the compounds and to gain more information concerning alkylated homologues. Because of the occasional low resolution of the peaks corresponding to BbF and BkF, these two PAH isomers were reported as their sum, i.e. BbkF. The calibration curves of all the compounds were constructed to determine their concentrations in the samples based on measurement of the above-mentioned standard solutions at different concentrations (Wu *et al.*, 2007).

### Quality Assurance

Field and laboratory blanks were used to check for any background contamination from artificial contaminants, reagents and glassware during the sampling and analysis procedure.

Annealed filters spiked with *n*-alkanes (500 ng~1 µg) and PAH (100 ng~500 ng) mixture standards were also analyzed to determine recovery rates. The average matrix spike recovery rates for PAHs ranged from 79-101%, except for Acpy (59%) and BbkF (111%). Relative standard deviations varied from 8 to 16% (n = 5). For *n*-alkanes, the recovery rates fell between 69% (C<sub>30</sub> alkane) and 108% (C<sub>14</sub> alkane) with an average of 85%. Results are reported as blank and recovery corrected to units of ng/m<sup>3</sup> for each sample.

## RESULTS AND DISCUSSION

### *TSP Concentrations*

TSP concentrations during the monitoring period ranged from 168 to 570 µg/m<sup>3</sup> at the temple, from 57 to 189 µg/m<sup>3</sup> at the bus terminus, and from 42 to 165 µg/m<sup>3</sup> at the campus site (Table 1). The levels measured at SPT were comparable to the range of TSP concentrations (227-348 µg/m<sup>3</sup>) measured in the urban area of Rio de Janeiro city, Brazil (Azevedo *et al.*, 1999), lower than the range (429-991 µg/m<sup>3</sup> and 28-1880 µg/m<sup>3</sup>) reported in the indoor air of the temples investigated in Taiwan (Lin *et al.*, 2001, 2002) and higher than the highest concentration of TSP (119 µg/m<sup>3</sup>) measured in central London (Kendall *et al.*, 2001). The mean value of TSP concentrations inside the temple was 1.5 times greater than the ambient air quality standard of 120 µg/m<sup>3</sup> recommended by WHO and SEPA (1996) for residential areas. However, the lowest concentration of TSP measured on 11

August at the campus site after raining was comparable to the values of TSP (45.6-46.2 µg/m<sup>3</sup>) reported in downtown Hat Yai city, Thailand (Tekasakul *et al.*, 2008), suggesting the importance of precipitation in determining the concentration of TSP in the air.

The levels of TSP at SPT were much higher than the values at the bus terminus and the campus indicating that a large quantity of particulate matter was emitted from incense burning and accumulated in SPT due to the space being not well-ventilated (Fig. 1). The glass fiber filters from SPT turned faint yellow while the filters from the bus terminus and the campus turned gray or black due to different emission sources of airborne particles. The weather conditions were relatively stable (Table 1) and would not significantly influence the distribution of TSP in the ambient air. Thus, the TSP concentration was mainly affected by the amount of particulate matter emitted from local emission sources. It also can be seen from Table 1 that the TSP concentrations on weekends were much higher than those on weekdays. The fact that there were more emissions of particles from incense burning related to the increased number of pilgrims or tourists on weekends than the emissions on weekdays can explain this difference to a great extent. Lung and Kao (2003) report in their research results for two temples in Taiwan that each joss stick in the censer contributed about 0.40 µg/m<sup>3</sup> of particles to the pilgrims' exposure. Burning incense generates a large quantity of fine particles with a mass median diameter

between 0.24 and 0.40  $\mu\text{m}$  (Mannix *et al.*, 1996) or between 0.1 and 0.7  $\mu\text{m}$  (Li and Hopke, 1993). Fine particles in the above size range could deposit in the respiratory tract and cause adverse health effects on the temple staff as well as the pilgrims or tourists to the temple. The amounts of toxic pollutants bound to the inhalable particles are especially important in order to assess the inhalation exposure risk posed.

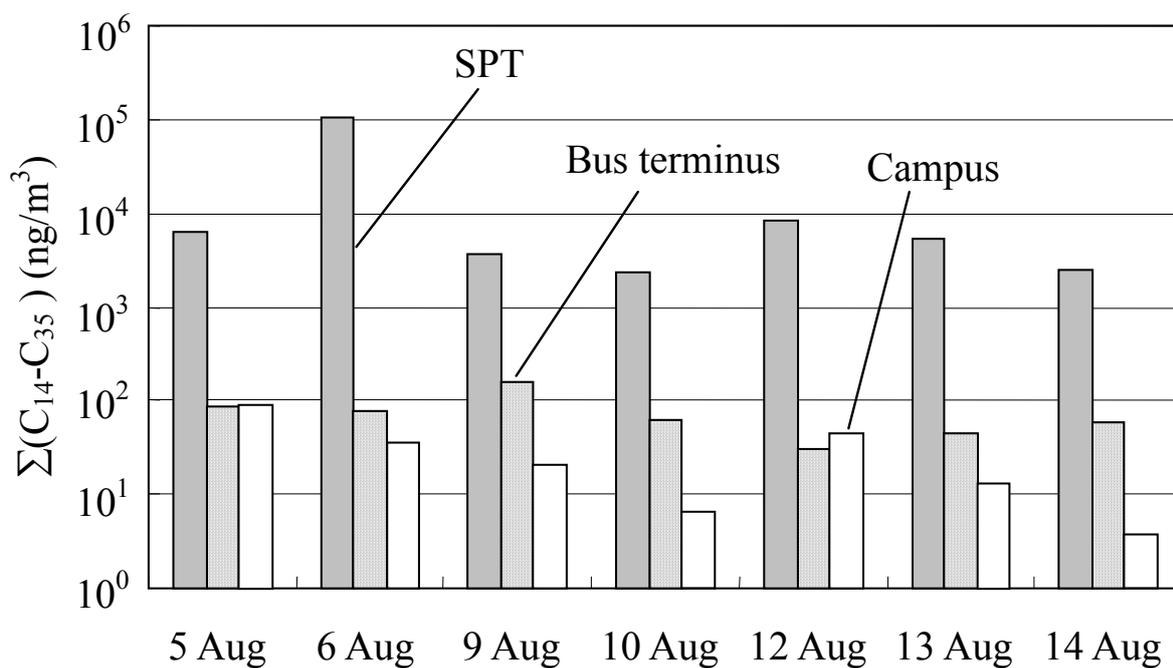
### *n*-Alkanes

A total of 22 *n*-alkanes ( $\text{C}_{14}$ - $\text{C}_{35}$ ) were identified and quantified in the TSP samples. The total concentrations of *n*-alkanes ( $\sum(\text{C}_{14}$ - $\text{C}_{35})$ ) in the SPT samples ranged from  $1.6 \times 10^3$ - $1.0 \times 10^5$   $\text{ng}/\text{m}^3$ , with a mean value of  $1.4 \times 10^4$   $\text{ng}/\text{m}^3$ , while the values of the bus terminus samples ranged from 29.9 to 625.4  $\text{ng}/\text{m}^3$  with a mean value of 129.5  $\text{ng}/\text{m}^3$ , and the levels in the campus samples ranged from 3.7 to 91.2  $\text{ng}/\text{m}^3$  with a mean value of 27.3  $\text{ng}/\text{m}^3$ . Thus the levels of total *n*-alkanes in TSP samples at SPT were 1 to 3 orders of magnitude higher than the levels at the bus terminus and on the campus (Fig. 2). Although the volatilization losses of lower weight molecular compounds are important (20-80% of the total concentrations) (Kavouras *et al.*, 1999), the abnormally high concentrations of particulate *n*-alkanes in the temple samples indicate the importance of incense burning emissions. The compositional differences in *n*-alkanes between the bus terminus and the campus samples were not significant, and both were characterized by strong peaks of  $\text{C}_{24}$ - $\text{C}_{31}$ , whereas the strong peaks in the SPT samples

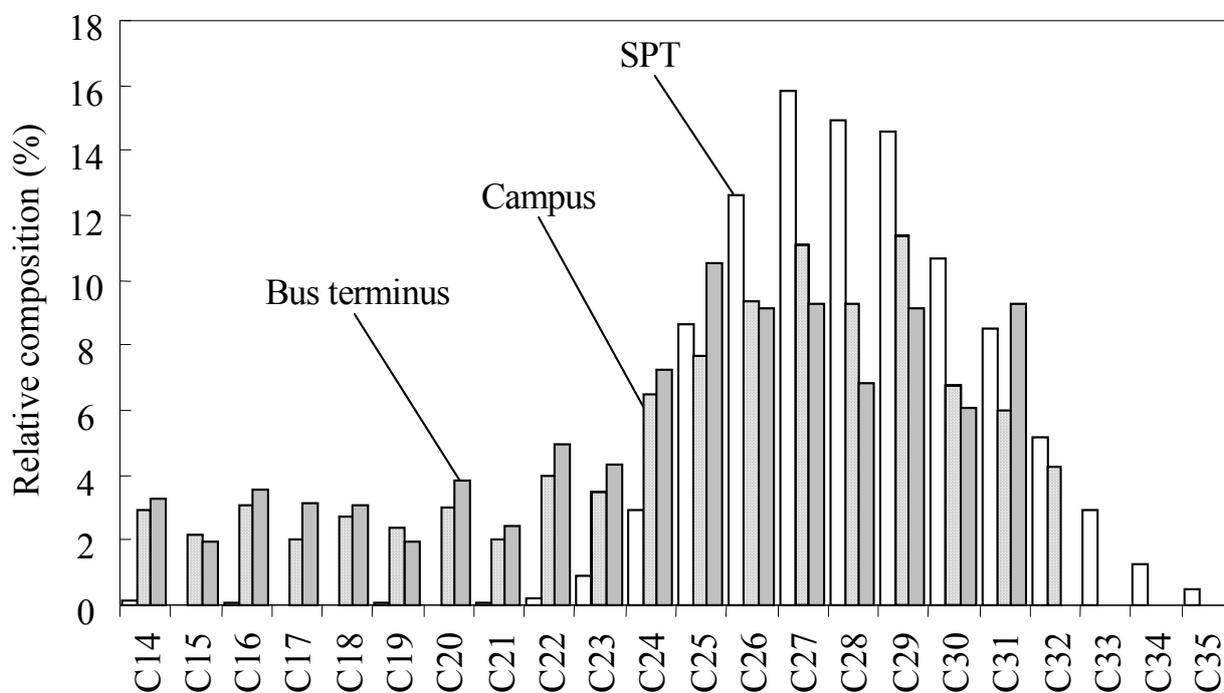
were of  $\text{C}_{25}$ - $\text{C}_{32}$  (Fig. 3). The relative distribution of *n*-alkanes provided some insight into the likely sources that contribute to the ambient samples. The bus terminus and the campus samples showed an *odd-to-even* predominance for the biogenic *n*-alkanes ( $n\text{-C}_{24}$ - $n\text{-C}_{35}$ ), while no *odd-to-even* predominance was observed in the same range for SPT samples. This underscores the contribution of *n*-alkanes emitted directly from epicuticular waxes of *Euphoria Longan* (*Airllus Longan*) leaves near the bus terminus and at the campus sampling sites due to leaf mechanical abrasion (Rogge *et al.*, 1993b). The mean values of the CPI (carbon preference index) for biogenic *n*-alkanes (defined as the sum of odd- over even-carbon number of *n*-alkanes between  $n\text{-C}_{24}$  and  $n\text{-C}_{35}$ ) were  $1.06 \pm 0.03$ ,  $1.12 \pm 0.24$  and  $1.22 \pm 0.29$  for SPT, the bus terminus and the campus samples, respectively. The CPI values near unity signify the minor contribution of *n*-alkanes emitted directly from epicuticular waxes (Bi *et al.*, 2003). The contribution of terrestrial wax *n*-alkanes (wax  $\text{C}_n$ ) was also estimated in order to determine the relative importance of biogenic and petrogenic sources, using the following equation (Simoneit *et al.*, 1991):

$$\text{Wax } \text{C}_n (\%) = \frac{\sum([\text{C}_n] - 0.5([\text{C}_{n-1}] + [\text{C}_{n+1}]])}{\sum[\text{C}_n] \times 100} \quad (1)$$

Negative values of  $\text{C}_n$  were taken as zero. Only  $7.1 \pm 1.8\%$  (SPT),  $31.2 \pm 10.6\%$  (bus terminus) and  $35.5 \pm 9.9\%$  (campus) of biogenic hydrocarbons were observed,



**Fig. 2.** Distribution of total concentration of *n*-alkane between C<sub>14</sub> and C<sub>35</sub> during the simultaneous sampling days.



**Fig. 3.** Average distributions of *n*-alkanes in the TSP samples.

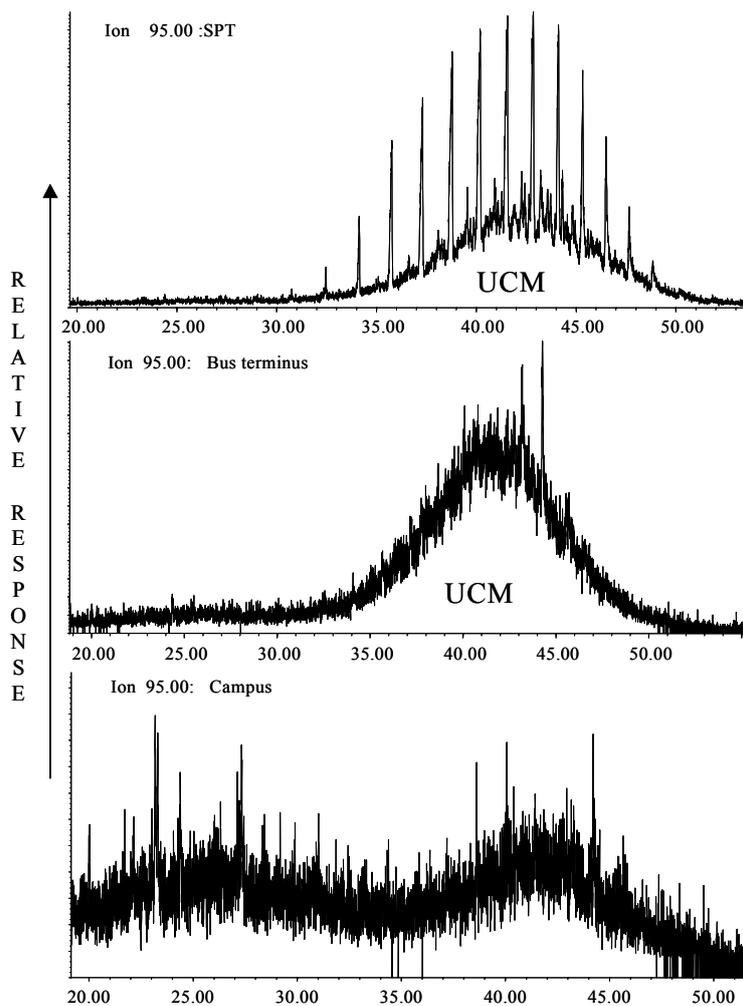
indicating a low contribution of *n*-alkanes from vascular plant wax. This was in agreement with the measurements of CPI values for biogenic *n*-alkanes. Kavouras *et al.* (1999) point out that the source identification of aliphatic hydrocarbons would be wrong if the concentrations of particulate *n*-alkanes are not corrected for volatilization losses. Thus, in future, the vapour phase *n*-alkanes must be quantified as well as the particulate phase in order to accurately identify the source identification of *n*-alkanes in the ambient air.

The unresolved complex mixture (UCM), consists of branched and cyclic hydrocarbons, related to petroleum residues, can be plotted using a *m/z*95 mass chromatogram (Fig. 4). Since unresolved hydrocarbons are emitted from combustion processes, it was possible to deduce whether the particles collected at the campus were less related to the combustion emission than those at SPT and the bus terminus. The diagnostic parameter of U/R (unresolved to resolved hydrocarbon components) can be used to assess the magnitude of anthropogenic sources. The mean value of U/R observed for the bus terminus samples was  $3.60 \pm 0.77$ , falling within the range (1.0-5.9) measured for Hong Kong aerosols (Zheng *et al.*, 2000), whereas the ratio of U/R for the SPT samples was very low ( $0.29 \pm 0.06$ ) and no UCM was detected in the campus samples, implying that the major source of *n*-alkanes at these two sites was not the incomplete combustion of fossil fuel. Although the UCM is seriously underestimated if only glass fiber filters are used for collection, the UCM/*n*-alkanes ratios

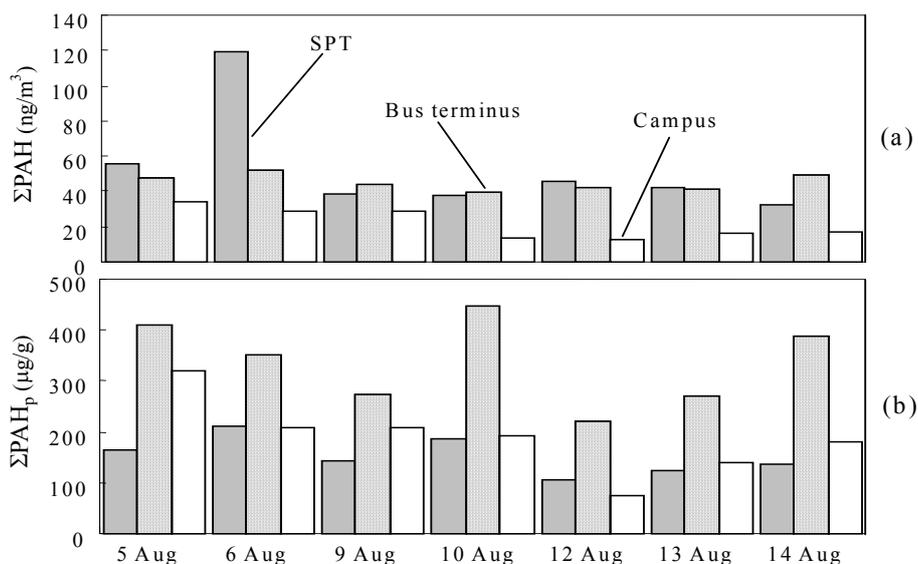
calculated for the particulate phase are very close to those calculated for the vapour phase (Kavouras *et al.*, 1999). Thus, the U:R ratios for particulate *n*-alkanes were still, to some extent, applicable to assess the relative contribution of vehicular emissions and epicuticular wax.

### **Polycyclic Aromatic Hydrocarbons**

The  $\Sigma$ PAH concentrations (defined as the sum of the concentrations of the 16 individual PAHs) ranged from 19.16 to 119.48 ng/m<sup>3</sup> at SPT, from 33.32 to 123.28 ng/m<sup>3</sup> at the bus terminus, and from 8.06 to 34.37 ng/m<sup>3</sup> at the campus. By comparison, Lung *et al.* (2003) report 200-450 ng/m<sup>3</sup> in a closed living room; Lin *et al.* (2002) report 3349-9238 ng/m<sup>3</sup> (particle + gas phase) in a Taiwan temple; Fang *et al.* (1999) report 7 ng/m<sup>3</sup> and 9 ng/m<sup>3</sup> at the urban sites of Malaysia; and Wu *et al.* (2007) report 372-772 ng/m<sup>3</sup> at an urban site in Tianjin, China. The  $\Sigma$ PAH concentrations measured during the 7 simultaneous sampling days at the three sites are presented in Fig. 5(a). The  $\Sigma$ PAH concentrations at SPT were comparable to those measured at the bus terminus and higher than those measured at the campus site. The mean value of  $\Sigma$ PAH on the campus was around 56-57% less than the mean values at the SPT and bus terminus. The results showed that a significant contribution of particulate PAHs was from incense/candle burning at SPT and from bus engine exhausts at the bus terminus. Other human activities such as cigarette smoking and burning joss paper can also contribute to the temple particulate PAH concentrations in such a



**Fig. 4.** Mass chromatogram of  $m/z95$  in the aliphatic hydrocarbon fraction. (indicator for the UCM)



**Fig. 5.** Concentration distributions of  $\Sigma PAH$  (a), and  $\Sigma PAH_p$  (b), at the three sites during the simultaneous sampling days.

semi-closed environment with a high volume of tourists (Yang *et al.*, 2005). The lower concentrations in the campus air may result from the lack of continuous PAH sources, such as bus exhausts and incense burning emission. It also can be seen from Fig. 5(a) that the values of  $\Sigma$ PAH at SPT were higher than those at the bus terminus at weekends (5-6 Aug and 12-13 Aug), while an opposite trend was observed on weekdays (9-10 Aug and 14 Aug). More worshippers and visitors at SPT on weekends produced more PAHs than on weekdays due to the increased amount of incense burning while paying respect to Buddha.

However, the sum of particle-bound concentrations of PAHs ( $\Sigma$ PAHp) (defined as PAH mass adsorbed onto airborne particles and normalized by particle mass,  $\mu\text{g/g}$ ) in the bus terminus air (221.0-738.1  $\mu\text{g/g}$ ) are significantly higher than the values at SPT (14.3-209.7  $\mu\text{g/g}$ ) and in the campus air (8.7-319.7  $\mu\text{g/g}$ ), and are comparable to the range between 271 to 423  $\mu\text{g/g}$  inside a temple in Taiwan (Lin *et al.*, 2001). The comparisons of  $\Sigma$ PAHp among the three sites during the simultaneous sampling days are presented in Fig. 5(b), and are different from the profiles of the atmospheric concentrations (Fig. 5(a)). Similar trends are reported in the research work of Lin *et al.* (2002) who note that the particle-bound PAH concentrations inside a temple (490  $\mu\text{g/g}$ ) are only about 1/30 of the levels of those at the traffic source ( $1.54 \times 10^4$   $\mu\text{g/g}$ ). This can be partly explained by the greater fine particle fraction and elemental carbon content in vehicle exhausts, which

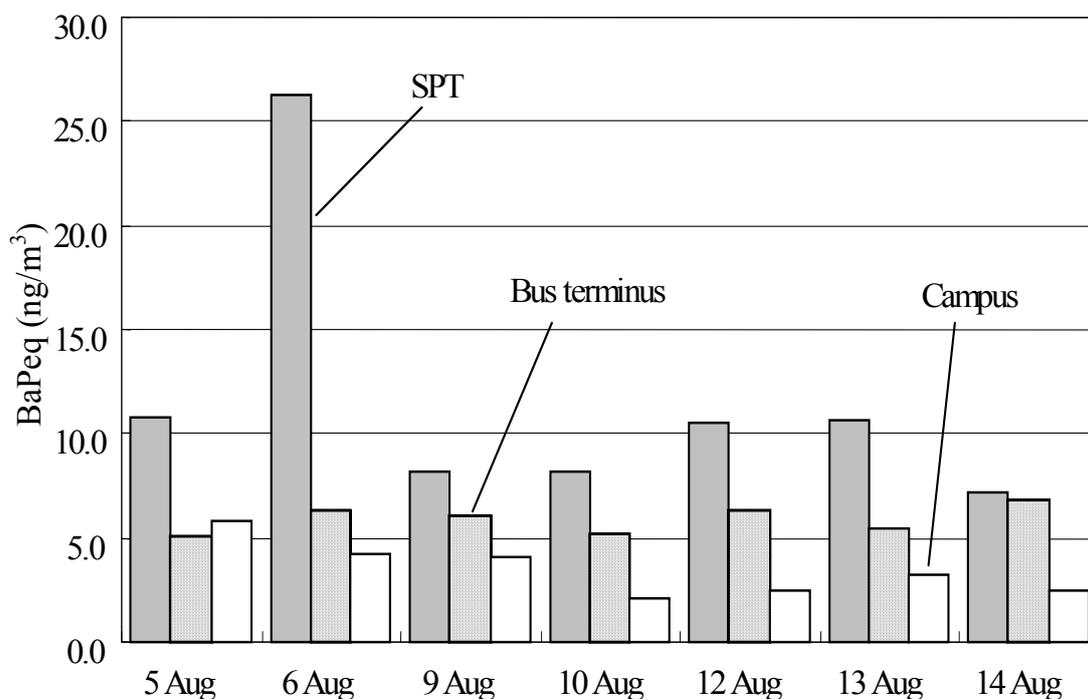
allow more PAH adsorption/absorption per unit of particles (Chen *et al.*, 1997; Marr *et al.*, 2004).

For health risk assessment purposes, the carcinogenic potency of each individual PAH compound should be taken into account. The toxicity equivalent factors (TEFs) of the 16 PAHs given by Nisbet and LaGoy (1992) reflect well the actual state of knowledge on the toxic potency of each individual PAH. The sum of the BaP equivalent concentration of PAHs (BaP<sub>eq</sub>) was calculated by multiplying each concentration by each individual TEF. The calculated BaP<sub>eq</sub> values at SPT (7.1-26.3  $\text{ng/m}^3$ ) were higher than those at the bus terminus (5.1-6.9  $\text{ng/m}^3$ ) and the campus (2.4-5.9  $\text{ng/m}^3$ ) (Fig. 6). This result again clearly indicated that the particle pollution caused by incense burning inside SPT was more serious than that at the bus terminus. BaP concentrations measured inside SPT considerably exceeded the standard for BaP (1  $\text{ng/m}^3$  for 24 h) in indoor air set by SEPA (2002) and the maximum permissible risk level (1  $\text{ng/m}^3$  BaP) for ambient air suggested by Sloff *et al.* (1989) based on the carcinogenic potential of inhaled particle-bound PAHs. It also exceeded the levels of BaP inside residential homes with tobacco smoking (2.2  $\text{ng/m}^3$ ) and homes without specified indoor combustion sources (0.83  $\text{ng/m}^3$ ) (Sheldon *et al.*, 1993). However, the measured BaP concentrations inside the SPT were significantly lower than those measured inside the Taiwanese temple, namely 71.5-124.2  $\text{ng/m}^3$  (Lin *et al.*, 2002) and 57.6-98.2  $\text{ng/m}^3$  (Lin *et al.*, 2001). It is well known that

the high molecular weight PAH compounds (5- and 6-ring PAHs) are mainly associated with fine particles less than 2  $\mu\text{m}$  in diameter (Wu *et al.*, 2006). The high BaPeq values inside the temple might increase the risk of cancer to temple staff due to the fact that, on average, 97.6% of BaPeq were contributed from 5- and 6-ring PAHs in this study. The reported values should be used in the risk assessment with great caution, however, because PAHs are present in both the gas and aerosol phases, with the lower molecular weight PAHs (2- and 3-ring polycyclic compounds) being preferentially in the gas phase and the higher molecular weight (5 and more aromatic rings) associated with particulate matter (Kavouras *et al.*, 1999; Mandalakis *et al.*, 2002). Thus, the risk level of exposure to PAHs would be underestimated

if the volatilization loss of relatively volatile PAHs is not considered even though their TEF values are small (= 0.001). In addition, partitioning of ambient PAHs between the gas and particle phases is temperature dependent (Odabasi *et al.*, 2006). Thus, the amounts of some individual PAHs measured from the filters in this study were lower or higher than the real situation inside or outside air due to the gas-particle transfer process and artifacts during sampling, the transporting process in the air, as well as the ambient temperature increases or decreases.

The relative contribution of individual PAHs to the total PAH mass is shown in Figure 7. The largest contribution in the SPT and bus terminus air was made by BghiP, followed by IP, BaP and Nap while the compound of Nap dominated the PAH profiles



**Fig. 6.** BaP equivalent concentration during the 7 simultaneous sampling days.

in the campus air. The contribution of BghiP to the total PAH mass in the campus air was also significant ( $14.6 \pm 7.2\%$ ) because the campus is located in the urban area of Xiamen city where gasoline exhaust is one of the main sources of urban PAH emissions (Hong *et al.*, 2007). The characteristics of significantly great contribution from BghiP are suggested to be indicative of gasoline vehicles (Rogge *et al.*, 1993a) and incense burning emissions (Huynh *et al.*, 1991). The PAH profiles of low molecular weight PAHs with 2-4 rings (from Nap to Chr) in SPT and bus terminus air were not significantly different. In contrast, significant differences existed for high molecular weight PAHs with 5-6 rings (from benzo[b]fluorene to BghiP) in that the relative contributions of individual PAHs to the total PAH mass in the SPT air were higher than those in the bus terminus air with the exception of BghiP as seen in Fig. 7. Significantly different profiles of 5-6 ring PAHs between SPT and bus terminus air suggested different dominant PAH sources.

Some source-diagnostic ratios between PAH isomers are used to assess their different origins (Simick *et al.*, 1999). But the ratios should be used with caution because their source profiles can be altered due to their reactivity under atmospheric conditions (Zhang *et al.*, 2005). Yunker *et al.* (2002) summarize the literature of PAH ratios for petroleum, single-source combustion and environmental samples and diagnose PAH sources in the Fraser basin using PAH cross plots for the diagnostic ratios. The proportion of 1,7-dimethylphenanthrene (DMP) to 2,6-

DMP is a sensitive indicator to distinguish wood combustion from motor vehicle emission (Benner *et al.*, 1995). Ratios of 1,7-DMP/2,6-DMP + 1,7-DMP (abbreviated as 1,7/2,6 + 1,7-DMP) between 0.70-0.90 indicate wood combustion, while ratios  $< 0.45$  indicate vehicle emissions (Yunker *et al.*, 2002). Ratios of Flua to Flua + Pyr between 0.4 and 0.5 are characteristic of liquid fossil fuel (vehicle and crude oil) combustion whereas ratios  $< 0.4$  and  $> 0.5$  are characteristic of petroleum and grass, wood or coal combustion, respectively. An IP to IP + BghiP ratio less than 0.2 likely implies petroleum, a ratio between 0.2 and 0.5 implies liquid fossil fuel (vehicle and crude oil) combustion, and a ratio  $> 0.5$  indicates grass, wood and coal combustion according to the literature values summarized by Yunker *et al.* (2002). A clear discrimination between the SPT and bus terminus samples was evident in the x-axis distribution of the 1,7/2,6 + 1,7-DMP and Flua/Flua + Pyr ratios and in the y-axis distribution of the IP/IP + BghiP ratio (Fig. 8). The spot distribution of SPT samples in the cross plot is close to the area of wood combustion while the bus terminus samples are close to the transition area between petroleum and mixed sources, suggesting a different origin of particulate PAHs at SPT. The spots of campus samples are relatively scattered, lying between the SPT and bus terminus samples in Fig. 8 and indicating a combination of sources (i.e. fossil and wood combustion) for PAHs.

The ratios of retene (1-methyl-7-isopropylphenanthrene) to Phe (Retene/Phe)

for SPT samples were all greater than those for bus terminus and campus samples during the simultaneous sampling days (Fig. 9),

supporting the above proposal of greater influence from wood combustion (such as bamboo stick, stick coating and joss paper) in

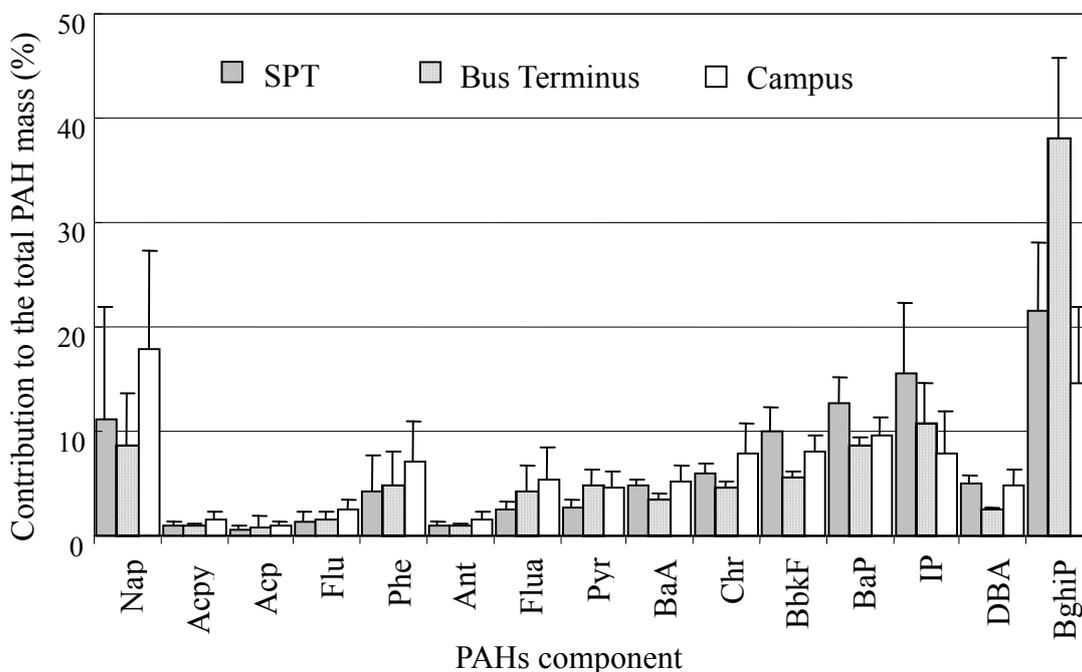


Fig. 7. Mean contribution of individual PAHs to the total PAH mass.

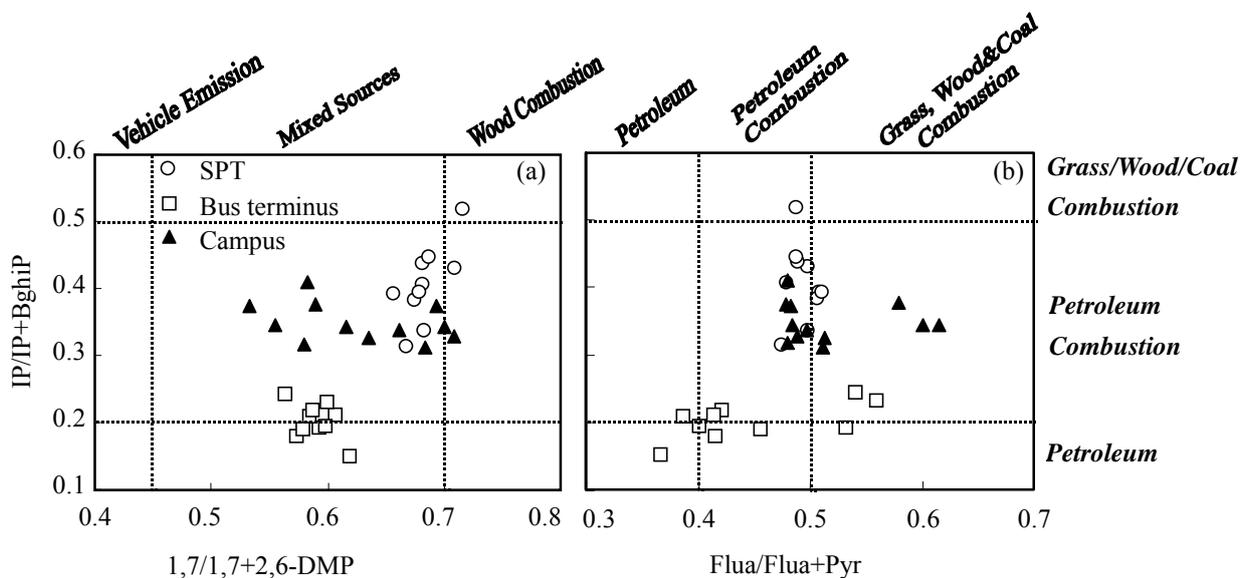
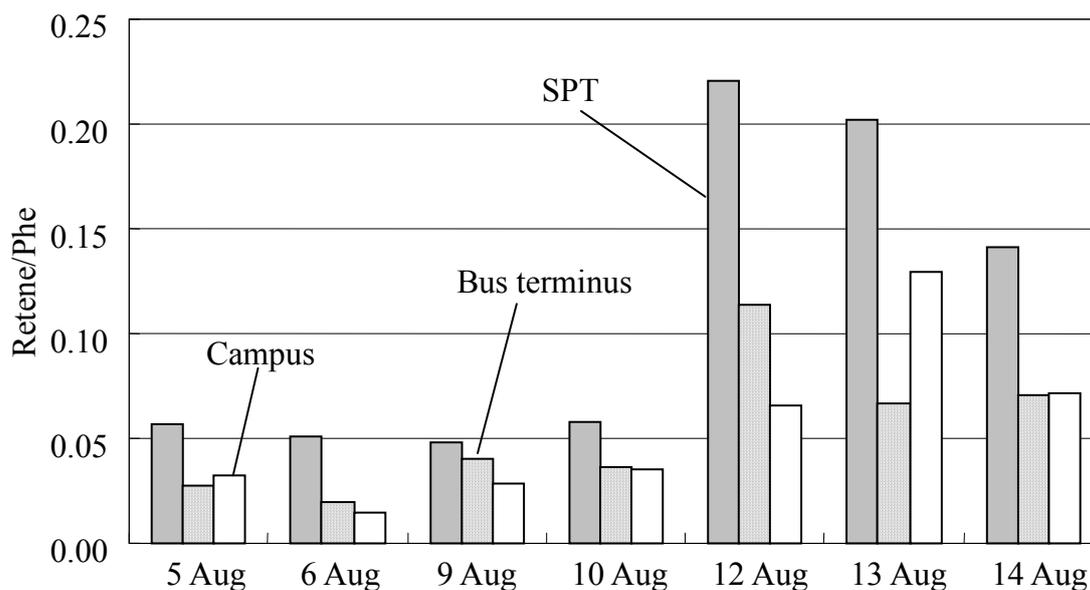


Fig.8. PAH cross plot for the ratio of indeno[1,2,3-cd]pyrene (IP) to IP plus benzo[ghi]perylene (BghiP) versus (a) 1,7-dimethylphenanthrene (DMP) to 2,6-DMP plus 1,7-DMP (1,7/2,6+1,7-DMP), and (b) fluoranthrene (Flua) to Flua plus pyrene (Pyr).



**Fig. 9.** Ratios of retene to phenanthrene during the simultaneous sampling days.

the SPT because the retene is known to be formed from pyrolysis of abietic acid, a component of conifer resin (Thomas, 1983). PAHs produced by incense burning in a temple will be transported to outdoors and deposited on the soil around the temple more or less depending on the construction style of the temple (closed, semi-closed or open). The soil could be contaminated by receiving atmospheric deposition of PAHs emitted from incense burning indoors as well as from the vehicle emissions outdoors. But the contribution of PAHs to the soil from fossil and modern biogenic sources is difficult to distinguish due to the similar profiles between incense burning and vehicle emissions (Fig. 7). In addition, the compound BghiP, which serves as a useful tracer for soot generated by motor vehicles, was also a dominant component in the PAH profiles for the SPT samples. Radiocarbon dating of individual PAHs would allow for quantitative

apportionment of fossil fuel versus modern biomass combustion sources of PAHs (Mandalakis *et al.*, 2004) in the local environment and thus assessment of the influence of incense burning on the environment.

## CONCLUSIONS

Incense burning inside the SPT temple in roughly 100 times (geometric mean) as much particulate *n*-alkanes as that tested at an adjoining busy bus terminus suggesting that the source of particulate *n*-alkanes emitted from incense burning was very strong. Although the levels of PAH concentrations measured inside the temple were comparable to those at the bus terminus, the BaP equivalent concentrations for SPT samples were all higher than those for bus terminus samples, suggesting that incense burning created more carcinogenic pollutants in

comparison with vehicle emissions. In addition, the air inside the temple contained on average 6 times more BaP than the indoor air standard recommended by the State Environmental Protection Administration of China. There is probably not too much to worry about in the case of tourists and pilgrims, but people working long hours in the temple, such as monks, temple staff and cleaners, may face significant exposure. Source analysis of PAHs in airborne particles showed significant difference between samples taken at the temple and bus terminus. Further research is needed to analyze both particulate (inhalable particles, PM<sub>10</sub>) and gas phase PAHs to assess the exposure risk more accurately. The findings so far also suggest the need for future research on the influence of incense burning on the ambient environment using radiocarbon dating of individual PAHs.

## ACKNOWLEDGEMENTS

This work was supported by National Science Foundation of China (No. 20777060). The authors thank the South Pu-Tuo Temple and Xiamen Bus Company for allowing sample collection for this study. Special thanks also go to Professor John Hodgkiss for his kind assistance in English proof reading for this manuscript.

## REFERENCES

- Azevedo, D.A., Moreira, L.S. and Siqueira, D.S. (1999). Composition of Extractable Organic Matter in Aerosols from Urban Areas of Rio De Janeiro City, Brazil. *Atmos. Environ.* 33: 4987-5001.
- Benner, B.A., Wise, S.A., Currie, L.A., Klouda, G.A., Klineidnst, D.B., Zweidinger, R.B., Stevens, R.K. and Lewis, C.W. (1995). Distinguishing the Contributions of Residential Wood Combustion and Mobile Scentrations of Dimethyphenanthrene isomers. *Environ. Sci. Technol.* 29: 2382-2389
- Bi, X., Sheng, G., Peng, P., Chen, Y., Zhang, Z. and Fu, J. (2003). Distribution of Particulate- and Vapor-Phase *n*-alkanes and Polycyclic Aromatic Hydrocarbons in Urban Atmosphere of Guangzhou, China. *Atmos. Environ.* 37: 289-298
- Chen, S.J., Hwang, W.I., Chiu, S.C., Hung, M.C. and Lin, C.C. (1997). Size Distribution of Particle-Bound PAH Composition in the Ambient Air of Bus Station. *J. Environ. Sci. Health A.* 32: 1781-1805.
- de Kok TMCM, Hogervorst J.G.F., Kleinjans, J.C.S. and Briede, J.J. (2004). Radicals in the Church. *Eur. Respir. J.* 24: 1069-1070.
- Fang, M., Zheng, M., Wang, F., To, K.L., Jaafar, A.B. and Tong, S.L. (1999). The Solvent-Extractable Organic Compounds in the Indonesia Bomass Burning Aerosols-Characterization Studies. *Atmos. Environ.* 33: 783-795.
- Hong, H.S., Yin, H.L., Wang, X.H. and Ye, C.X. (2007). Seasonal Variation of PM<sub>10</sub>-Bound PAHs in the Atmosphere of Xiamen, China. *Atmos. Res.* 85: 429-441.

- Huynh, C.K., Savolainen, H., Vu-Duc, T., Guillenmin, M. and Iseln, F. (1991). Impact of Thermal Proofing of a Church on its Indoor Air Quality: the Combustion of Candles and Incense as a Source of Pollution. *Sci. Total Environ.* 102: 241-251.
- IARC (1991). Monographs on the Evaluation of Carcinogenic Risks to Humans, vol. 43-53. International Agency for Research on Cancer, Lyon.
- Kavouras, I.G., Lawrence, J., Koutrakis, P., Stephanou, E.G. and Oyola, P. (1999). Measurement of Particulate Aliphatic and Polynuclear Aromatic Hydrocarbons in Santiago De Chile: Source Reconciliation and Evaluation of Sampling Artifacts. *Atmos. Environ.* 33: 4977-4986.
- Kendall, M., Hamilton, R.S., Watt, J. and Williams, I.D. (2001). Characterization of Selected Speciated Organic Compounds Associated with Particulate Matter in London. *Atmos. Environ.* 35: 2483-2495.
- Khalili, N.R., Scheff, P.A. and Holsen, T.M. (1995). PAH Source Fingerprints for Coke Oven, Diesel and Gasoline Engines, Highway Tunnel, and Wood Combustion Emission. *Atmos. Environ.* 29: 533-42.
- Koo, L.C. and Ho, J.H.C. (1996). Diet as a Confounder of the Association between Air Pollution and Female Lung Cancer: Hong Kong Studies on Exposures to Environmental Tobacco Smoke, Incense, and Cooking Fumes as Examples. *Lung Cancer.* 14: S47-S61.
- Lee, S.C. and Wang, B. (2004). Characteristics of Emissions of Air Pollutants from Burning of Incense in a Large Environmental Chamber. *Atmos. Environ.* 38: 941-951.
- Li, C.S. and Ro, Y.S. (2000). Indoor Characteristics of Polycyclic Aromatic Hydrocarbons in the Urban Atmosphere of Taipei. *Atmos. Environ.* 34: 611-620.
- Li, W. and Hopke, P.K. (1993). Initial Size Distributions and Hygroscopicity of Indoor Combustion Aerosol Particles. *Aerosol Sci. Technol.* 19: 305-316.
- Lin, T.-C., Chang, F.-H., Hsieh, J.-H., Chao, H.-R. and Chao, M.R. (2001). Environmental Exposure to Polycyclic Aromatic Hydrocarbons and Total Suspended Particulates in a Taiwanese Temple. *Bull. Environ. Contam. Toxicol.* 67: 332-338.
- Lin, T.C., Chang, F.H., Hsieh, J.H., Chao, H.R. and Chao, M.R. (2002). Characteristics of Polycyclic Aromatic Hydrocarbons and Total Suspended Particulate in Indoor and Outdoor Atmosphere of Taiwanese Temple. *J. Hazard Mater.* A95: 1-12.
- Lung, S.C. and Kao, M.C. (2003). Worshipper's Exposure to Particulate Matter in Two Temples in Taiwan. *J. Air Waste Manage. Assoc.* 53: 130-135.
- Lung, S.C., Kao, M.C., Hu and S.C. (2003). Contribution of Incense Burning to Indoor PM<sub>10</sub> and Particle-Bound Polycyclic Aromatic Hydrocarbons under Two Ventilation Conditions. *Indoor Air.* 13: 194-199.
- Mandalakis, M., Gustafsson, Ö., Reddy, C.M. and Xu, L. (2004). Radiocarbon

- Apportionment of Fossil Versus Biofuel Combustion Sources of Polycyclic Aromatic Hydrocarbons in the Stockholm Metropolitan Area. *Environ. Sci. Technol.* 38: 5344-5349.
- Mandalakis, M., Tsapakis, M., Tsoga, A. and Stephanou, E.G. (2002). Gas-Particle Concentrations and Distribution of Aliphatic Hydrocarbon, PAHs, PCBs and PCDD/Fs in the Atmosphere of Athens (Greece). *Atmos. Environ.* 36: 4023-4035.
- Mannix, R.C., Nguyen, K.P., Tan, E.W., Ho, E.E. and Phalen, R.F. (1996). Physical Characterization of Incense Aerosols. *Sci. Total Environ.* 193: 149-158.
- Marr, L.C., Grogan, L.A., Wöhrnschimmel, H., Molina, L., Molina, M.J., Smith, T.J. and Garshick, E. (2004). Vehicle Traffic as a Source of Particulate Polycyclic Aromatic Hydrocarbon Exposure in the Mexico City Metropolitan area. *Environ. Sci. Technol.* 38: 2584-2592.
- Nisbet, I.C.T. and LaGoy, P.K. (1992). Toxic Equivalency Factors (TEFs) for Polycyclic Aromatic Hydrocarbons (PAHs). *Regul. Toxicol. Pharm.* 16: 290-300.
- Odabasi, M., Cetin, E. and Sofuoglu, A. (2006). Determination of Octanol-air Partition Coefficients and Supercooled Liquid Vapor Pressures of PAHs as a Function of Temperature: Application to Gas-particle Partitioning in an Urban Atmosphere. *Atmos. Environ.* 40: 6615-6625.
- Reed, M.D., Monske, M.L., Lauer, F.T., Meserole, S.P., Born, J.L. and Burchiel, S.W. (2003). Benzo[a]pyrene Diones are Produced by Photochemical and Enzymatic Oxidation and Induce Concentration-Dependent Decrease in the Proliferation State of Human Pulmonary Epithelial Cells. *J. Toxicol. Environ. Health Part A.* 66: 1189-1205.
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R. and Simoneit, B.R.T. (1993a). Sources of Fine Organic Aerosol 2. Non-catalyst and Catalyst-equipped Automobiles and Heavy-duty Diesel Trucks. *Environ. Sci. Technol.* 27: 636-651.
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R. and Simoneit, B.R.T. (1993b). Sources of Fine Organic Aerosol. 4. Particulate Abrasion Products from Leaf Surfaces of Urban Plants. *Environ. Sci. Technol.* 27: 2700-2711.
- SEPA (State Environmental Protection Administration of China) (1996). Ambient Air Quality Standard. GB3095-1996.
- SEPA (State Environmental Protection Administration of China) (2002). Indoor Air Quality Standard. GB/T18883-2002.
- Sheldon, L., Clayton, A., Keever, J., Perritt, R. and Whitaker, D. (1993). Indoor Concentrations of Polycyclic Aromatic Hydrocarbons in California Residences. Draft Final Report, Contract no. A033-132, Research Triangle Institute.
- Simcik, M.F., Eisenreich, S.J. and Lioy, P.J. (1999). Source Apportionment and Source/Sink Relationships of PAHs in the Coastal Atmosphere of Chicago and Lake

- Michigan. *Atmos. Environ.* 33: 5071-5079.
- Simoneit, B.R.T., Sheng, G., Chen, X., Fu, J., Zhang, J. and Xu, Y. (1991). Molecular Marker Study of Extractable Organic Matter in Aerosols from Urban Areas of China. *Atmos. Environ.* 25: 2111-2129.
- Sloff, W., Janus, J.A., Matthijsen, A.J.C.M. and Montizaan, J.P.M.R. (1989). Integrated Criteria Document PAHs, Report No. 758,474,011, The Netherlands National Institute of Public Health and Environmental Protection, Bilthoven.
- Suleiman, S.A. (1987). Petroleum Hydrocarbon Toxicity in Vitro: Effect of *n*-alkanes, Benzene and Toluene on Pulmonary Alveolar Macrophages and Lysosomal Enzymes of the Lung. *Arch. Toxicol.* 59: 402-407.
- Tekasakul, P., Furuuchi, M., Tekasakul, S., Chomanee, J. and Otani, Y. (2008). Characteristics of PAHs in Particulates in the Atmospheric Environment of Hat Yai City, Thailand, and Relationship with Rubber-wood Burning in Rubber Sheet Production. *Aerosol Air Qual. Res.* 8: 265-278.
- Thomas, R. (1983). Retene-a Molecular Marker of Wood Combustion in Ambient Air. *Nature.* 306: 580-582.
- VCCEP (Voluntary Children's Chemical Evaluation Program) (2005). Peer Consultation Meeting Report on *n*-alkanes. <http://tera.org/peer/vccep>.
- Wu, S.P., Tao, S. and Liu, W.X. (2006). Particle Size Distributions of Polycyclic Aromatic Hydrocarbons in Rural and Urban Atmosphere of Tianjin, China. *Chemosphere.* 62: 357-367.
- Wu, S.P., Tao, S., Zhang, Z.H., Lan, T. and Zuo, Q. (2007). Characterization of TSP-Bound *n*-alkanes and Polycyclic Aromatic Hydrocarbons at Rural and Urban Sites of Tianjin, China. *Environ. Pollut.* 147: 203-210.
- Yang, H.H., Jung, R.C., Wang, Y.F. and Hsieh, L.T. (2005). Polycyclic Aromatic Hydrocarbon Emissions from Joss Paper Furnaces. *Atmos. Environ.* 39: 3305-3312.
- Yunker, M.B., Macdonald, R.W., Vingarzen, R., Mitchell, R.H., Goyette, D. and Sylvestre, S. (2002). PAHs in the Fraser River Basin: a Critical Appraisal of PAH Ratios as Indicators of PAH Source and Composition. *Org. Geochem.* 33: 489-515.
- Zhang, X.L, Tao, S., Liu, W.X., Zuo, Q. and Liu, S.Z. (2005). Source Diagnostics of Polycyclic Aromatic Hydrocarbons Based on Species Ratios: a Multimedia Approach. *Environ. Sci. Technol.* 39: 9109-9114.
- Zheng, M., Fang, M., Wang, F. and To, K.L. (2000). Characterization of the Solvent Extractable Organic Compounds in PM<sub>2.5</sub> Aerosols in Hong Kong. *Atmos. Environ.* 34: 2691-2702.

Received for review, October 24, 2008

Accepted, December 26, 2008