

## Research Article

# Origin and Distribution of PAHs in Ambient Particulate Samples at High Mountain Region in Southern China

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To understand the deposition and transport of PAHs in southern China, a measurement campaign was conducted at a high-elevation site (the summit of Mount Heng, 1269 m A.S.L.) from April 4 to May 31, 2009, and a total of 39 total suspended particulate samples were collected for measurement of PAH concentrations. The observed particulate-bound PAHs concentrations ranged from 1.63 to 29.83 ng/m<sup>3</sup>, with a mean concentration of 6.03 ng/m<sup>3</sup>. BbF, FLA, and PYR were the predominant compounds. Good correlations were found between individual PAHs and meteorological parameters such as atmospheric pressure, relative humidity, and ambient temperature. The backward trajectory analysis suggested that particulate samples measured at the Mount Heng region were predominantly associated with the air masses from southern China, while the air masses transported over northern and northwestern China had relative higher PAHs concentrations. Based on the diagnostic ratios and factor analysis, vehicular emission, coal combustion, industry emission, and unburned fossil fuels were suggested to be the PAHs sources at Mount Heng site. However, the reactivity and degradation of individual PAHs could influence the results of PAH source profiles, which deserves further investigations in the future.

## 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of widespread environmental contaminants, originating from both natural (e.g., forest fires and volcanoes) and anthropogenic sources (e.g., incomplete combustion and pyrolysis of fossil fuels or organic materials). Between the two sources, human activities (i.e., anthropogenic sources) contribute the most to PAHs emissions. This is particularly the case in urban area or industrial areas, where the PAHs sources can be entirely anthropogenic [1–4]. PAHs are persistent and accumulative in environment, and they can cause carcinogenic and mutagenic effects to living things. Thus, these compounds have received more and more attention by environmental scientists for the last several decades. The emission abatement of PAHs has been listed on the agenda of

international convention processes, such as the 1979 Geneva Convention on Long-Range Transboundary Air Pollution. Since 1990, reduction of PAHs emission has been reported in U.S. and European countries. In contrast to those developed countries, the PAH emissions in China have been continuously increasing due to the rapid industrialization and economic development [5–7]. The total PAH emission in China was estimated to be about 25,300 tons in the year of 2003 and the major sources were suggested to be domestic coal combustion, biofuel burning, and industry emission [5].

Once released into the atmosphere, PAHs can partition between vapor and particle phases, which is dependent on temperature and vapor pressure of the chemicals [8, 9]. PAHs with high molecular weight (4–6 rings) tend to be associated with particle phase, while those with lower molecular weight (2–3 rings) are often concentrated in the vapor phase

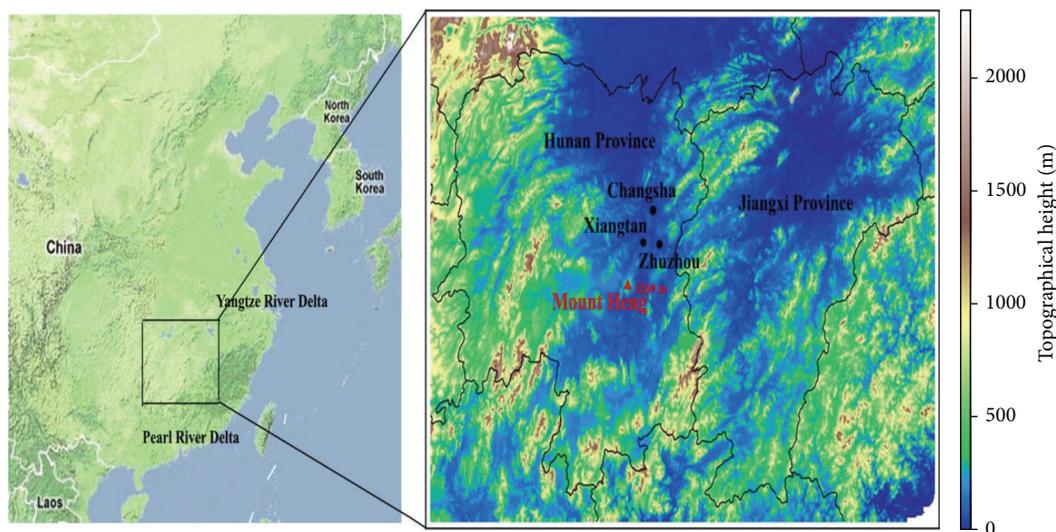


FIGURE 1: Geographical location of Mount Heng (cited from [36]).

[10]. During the course of their transport, PAHs could be removed from the atmosphere due to precipitation and/or dry deposition. Through the grasshopper effect, PAHs can be transported to very long ranges from their original sources [11].

Although a considerable number of studies have been conducted on measurements of PAHs in urban environment, little work has been done in rural sites. Remote mountain regions, particularly with altitude in the free troposphere, are the unique environments for the study of atmospheric pollution load over continental areas, and the study at remote mountain regions can help to get full assessment of the role of the atmosphere in the global distribution of PAHs [12, 13]. In this study, atmospheric particulate samples were collected at the summit of Mt. Heng, which is in the transition region between the free troposphere and boundary layer, and serve as a regionally representative site for studying the long-range transport of pollutants in Southern China. The objectives of the present study were (1) to gain a primary understanding on the distribution of particulate-bound PAHs at Mt. Heng and (2) to examine the possible sources and transport patterns of PAHs in the region.

## 2. Materials and Methods

**2.1. Site Description and Sample Collection.** The Mount Heng is located in Hunan Province, south China, and it is between two major polluted industrial/urban areas—the Pearl River and Yangtze River deltas. There are two big cities, Shanghai and Guangzhou, situated 900 km to the east and 400 km to the south of the Mount Heng, respectively. The sampling site was located at a meteorological station ( $27^{\circ}18'N$ ,  $112^{\circ}42'E$ ), which was set up in the 1937 at the summit of Mount Heng (1269 m A.S.L.) (Figure 1).

The sampling period was carried out from April 4 to May 31, 2009. Total suspended particulate (TSP) samples were collected on glass fibre filters by a middle-volume air

sampler (KC-300, Qingdao Laoshan Electronic Instrument Company) daily (22 hours, from 10 a.m. to 8 a.m.). The sampler was operated at a flow rate of 225 L/min and a total of 39 TSP samples were collected. Prior to sampling, all the glass fibre filters were combusted at  $450^{\circ}C$  in an oven for 6 h to volatilize any organic contaminants. After sampling, the glass fibre filters were wrapped in aluminum foils and frozen at  $-20^{\circ}C$ , and PAHs analysis was then conducted within two weeks after sampling.

**2.2. Sample Preparation and Analysis.** The glass fibre filters were extracted by Accelerated Solvent Extractor (Dionex ASE 300) with acetone/*n*-hexane (1:1, v/v) solvents, and the elute was purified using silica gel columns and condensed to exactly 1 mL with rotary evaporation and nitrogen stream technique. PAHs were then determined using gas chromatography coupled to mass spectrometry (Shimadzu 2010 plus). The details of the extraction and analysis procedure were described elsewhere [1]. Fifteen individual PAHs were detected in this study (Table 1).

**2.3. Quality Control.** Deuterated perylene-d<sub>12</sub> was spiked to all the samples to monitor procedural performance. The mean recoveries based on surrogates in samples were  $98.4 \pm 6.4\%$ .

Field and method blanks were analyzed using the same procedure as particulate samples, and all the PAHs concentrations were corrected for blanks. The control calibration standards were measured regularly to ensure the performance of instrument during sample analysis.

## 3. Results and Discussion

**3.1. Concentrations of PAHs in Particulate Samples.** Descriptive statistics for all valid observations of PAHs concentrations in particulate samples collected at Mount Heng are summarized in Table 2. The concentrations of PAHs varied

TABLE 1: The abbreviations for PAH compounds and their detection limits.

PAHs	Abbreviation	Detection limit (ng)
Acenaphthylene	ACY	4
Acenaphthene	ACE	5
Fluorene	FLU	5
Phenanthrene	PHE	3
Anthracene	ANT	4
Fluoranthene	FLA	7
Pyrene	PYR	4
Benzo[a]anthracene	BaA	2
Chrysene	CHR	7
Benzo[b]fluoranthene	BbF	3
Benzo[k]fluoranthene	BkF	4
Benzo[a]pyrene	BaP	4
Dibenz[a,h]anthracene	DahA	6
Benzo[g,h,i]perylene	BghiP	4
Indeno[1,2,3-cd]pyrene	IcdP	5

from 1.63 to 29.83 ng/m<sup>3</sup>, with a mean concentration of 6.03 ng/m<sup>3</sup>. BbF was the predominant compound, with a mean concentration of 0.86 ng/m<sup>3</sup>, contributing to 14.26% of the total PAH concentration. FLA, PHE, and PYR comprised the second tier of dominant compounds, with mean concentrations of 0.77, 0.74, and 0.60 ng/m<sup>3</sup>, respectively. Similar distribution of individual PAHs has been reported in previous studies in China. For the research done by our group at the Mount Tai (the highest mountain at northern China, 1534 m A.S.L.), BbF, FLA, and PYR were the most abundant PAH compounds in PM<sub>2.5</sub> samples, accounting for 23.55%, 14.21%, and 10.39% of the total PAH concentration, respectively [1]. Wang et al. [14] also suggested that BbF was the predominant compound of PAHs in PM<sub>10</sub> samples in Beijing. In 2012, China Environmental Protection Agency has published the latest ambient air quality standards, in which the BaP daily and annual concentration limits were set at 2.5 ng/m<sup>3</sup> and 1.0 ng/m<sup>3</sup>, respectively. The BaP concentration found in this study met the standard. Together with BaP, several other PAHs compounds, including BaA, BbF, BkF, IcdP, and DahA, were suggested as potential carcinogenic compounds by the International Agency for Research on Cancer (IARC). The total concentration of these six compounds was 2.30 ng/m<sup>3</sup>, accounting for 38.14% of the total PAHs.

The observed PAHs concentrations at the Mount Heng are comparable with our previous studies done at the Mount Tai (fine particles, 6.88 ng/m<sup>3</sup>; Table 3) [1]. When comparing with other areas, the PAHs concentrations at Mount Heng are much lower than the results reported in urban cities or areas in China, including Beijing [14], Guangzhou [15], and Northern Plain [16]. In contrast, the PAHs concentrations at Mount Heng were higher than the observed concentrations at Waliguan (a global standard observation site, 3810 m A.S.L., 2.08 ng/m<sup>3</sup>) [17], high mountain regions of Europe (0.07–1.10 ng/m<sup>3</sup>, 2240–2413 m) [18], Mt. Bachelor Observatory (MBO, <6 ng/m<sup>3</sup>, 2763 m) [19], Far East Asia

TABLE 2: Statistical description of PAH concentrations in particulate samples at Mount Heng.

Species	PAHs concentrations (ng/m <sup>3</sup> )		
	Range	Mean	SD
ACY	Nd-0.11	0.02	0.03
ACE	Nd-0.56	0.10	0.15
FLU	Nd-0.85	0.22	0.23
PHE	0.17–2.83	0.74	0.71
ANT	Nd-0.41	0.12	0.12
FLA	0.15–4.88	0.77	0.97
PYR	0.13–3.62	0.60	0.72
BaA	0.05–1.68	0.27	0.33
CHR	Nd-3.80	0.52	0.80
BbF	Nd-4.24	0.86	0.88
BkF	0.11–1.25	0.36	0.30
BaP	0.08–1.47	0.35	0.31
IcdP	Nd-1.80	0.39	0.39
DahA	Nd-0.53	0.07	0.11
BghiP	0.21–2.60	0.64	0.51
PAHs	1.63–29.83	6.03	5.87

(0.32 ng/m<sup>3</sup>), North Pacific Ocean (0.14 ng/m<sup>3</sup>), and the Arctic area (0.48 ng/m<sup>3</sup>) [20].

**3.2. Correlations of Particulate-Bound PAH Levels with Meteorological Parameters.** The meteorological conditions are important factors that can affect the deposition levels of pollutants [21–23]. In the present study, meteorological parameters including atmospheric pressure (press.), relative humidity (RH), and ambient temperature ( $T$ ) were measured during the sampling period. The correlation coefficients between these meteorological parameters and particulate-bound PAHs were analyzed (Table 4). Good correlation was found between atmospheric pressure and several individual PAHs including BkF, BaP, IcdP, and BghiP, suggesting that high atmospheric pressure enhanced the bound of these compounds to the particulate phase. In the study of Tian et al. [21], significant negative correlation was observed between particulate-bound PAHs and ambient temperature, suggesting the temperature dependence for PAHs. Some other studies also highlighted the important role of temperature in PAHs partitioning between gas and particulate phase [24, 25]. However, in this present study, only five individual PAH compounds including ACY, ACE, BkF, IcdP, and BghiP presented significant negative correlation with temperature. In addition, ACE and FLU had good positive relationships with RH, indicating their RH dependence.

**3.3. Transport Pattern and Sources of Particulate-Bound PAHs.** In previous studies, atmospheric samples always have higher deposition concentrations when they came from the more polluted area [2, 10]. Back trajectory analysis was conducted in this study to characterize the particulate deposition character influenced by different source regions.

TABLE 3: Comparison of particulate-bound PAH levels between Mount Heng and other studies.

Sampling site	Sample type	Altitude (m A.S.L.)	$\sum$ PAHs	PAHs concentrations (ng/m <sup>3</sup> )	Reference
Mount Heng, China	TSP	1269 m	$\sum$ 15	6.03	This research
Mount Tai, China	PM <sub>2.5</sub>	1534 m	$\sum$ 15	6.88	[1]
Waliguan, China	TSP	3810 m	$\sum$ 14	2.08	[17]
High Mountain Regions, Europe	TSP	2240 m–2413 m	$\sum$ 20	0.07–1.10	[18]
MBO, US	TSP	2763 m	$\sum$ 14	<4	[19]
Far East Asia	TSP	NA	$\sum$ 15	0.32	[20]
North Pacific Ocean	TSP	NA	$\sum$ 15	0.14	[20]
Arctic	TSP	NA	$\sum$ 15	0.48	[20]
Beijing, China	PM <sub>10</sub>	NA	$\sum$ 15	44.9	[14]
Northern Plain, China	TSP	NA	$\sum$ 16	346	[16]
Guangzhou, China	TSP	NA	$\sum$ 16	19.5	[15]

TABLE 4: The correlation coefficients between particulate bound-PAH concentrations and meteorological parameters.

	Press. (Pa)	T (°C)	RH (%)
PAHs	0.627	-0.639	0.226
ACY	0.551	-0.764*	0.665
ACE	-0.025	-0.234	0.940**
FLU	-0.078	-0.166	0.938**
PHE	0.389	-0.554	0.700
ANT	0.472	-0.641	0.621
FLA	0.541	-0.571	0.119
PYR	0.555	-0.564	0.076
BaA	0.572	-0.537	0.050
CHR	0.531	-0.475	0.015
BbF	0.688	-0.644	0.174
BkF	0.733*	-0.718*	0.233
BaP	0.722*	-0.706	0.221
IcdP	0.767*	-0.783*	0.336
DahA	0.626	-0.656	0.363
BghiP	0.751*	-0.784*	0.310

Significance level: \*  $P < 0.05$ , \*\*  $P < 0.01$ .

Forty-eight-hour backward trajectories were calculated using the HYSPLIT (hybrid single-particle Lagrangian integrated trajectory) model at a height of 1269 m A.S.L.

Based on the analysis, four major trajectory categories were classified for the samples: N/NW, continental air masses from northern and northwestern China, the most polluted region of China; EA, air masses from eastern China, where Shanghai and its surrounding cities located here; S, air masses from the Southern China, transported through the Pearl River Delta region; and L, samples with looped and short trajectories that stayed within a radius of 500 km of sampling site (Figure 2).

Among the four classified trajectory categories, S happened most frequently, accounting for 42.9% of the total, followed by N/NW (37.1%), EA (14.3%), and L (5.7%). As presented in Figure 3, the order of PAHs concentrations among the trajectory categories was N/NW, L, EA, and S.

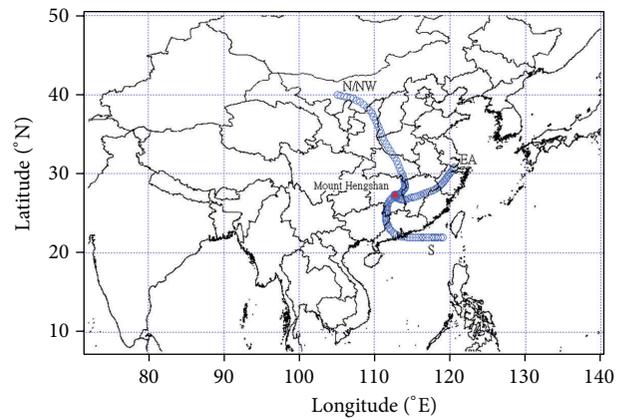


FIGURE 2: Classified pathway of each trajectory category.

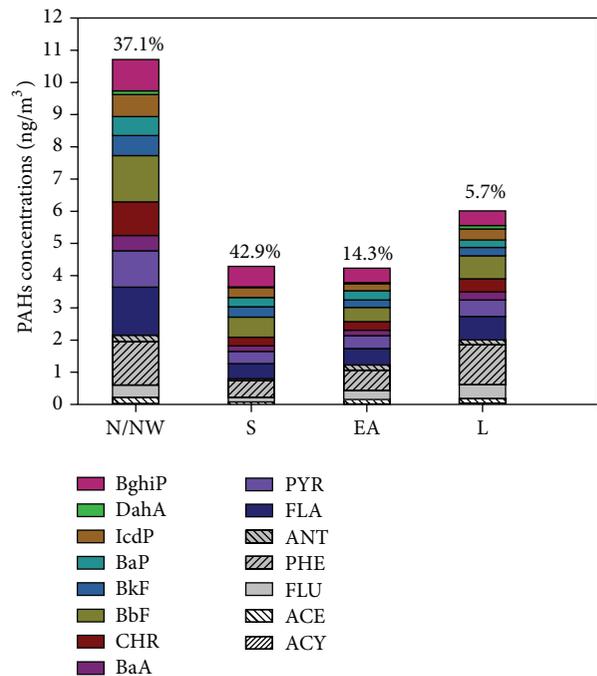


FIGURE 3: PAHs concentrations based on different trajectory categories.

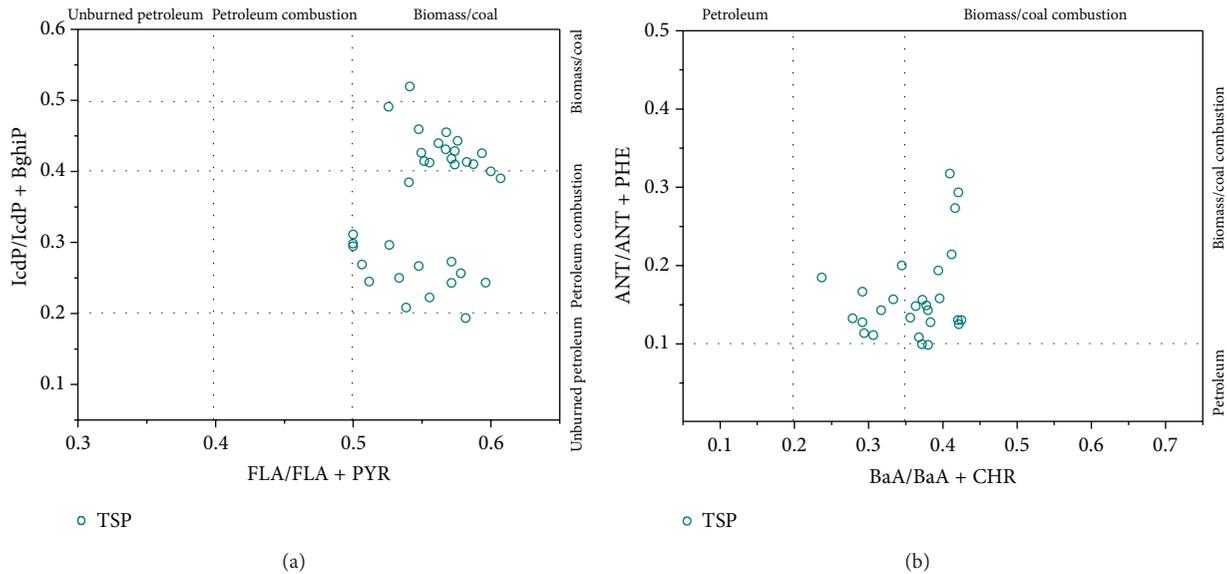


FIGURE 4: Diagnostic PAH ratios for TSP samples collected at Mount Heng.

The air masses of N/NW group originated from the northern and northwestern China, where most industrial activities of China are located and burning of coal and biomass was the dominant energy source. So, particulate samples of this group had the highest PAHs concentrations, reaching  $10.71 \text{ ng/m}^3$ . Unexpectedly, the group of L had higher PAHs concentrations than EA and S groups, and the average concentration reached  $6.01 \text{ ng/m}^3$ . PHE was the predominant compound of L group, contributing over 20% of the total concentration. As presented in the study of Lin et al. [26], PHE is a major component of emissions from incense burning. There are several temples located in the Mount Heng region, and most of them are crowded with visitors. Therefore, incense burning inside or outside the temples is suggested to be an important local PAHs emission source. It should be noted that the relative fewer sample numbers of L group could cause uncertainties to some degree. Air mass from eastern and southern China, the two most developed areas in China, had lower PAHs concentrations, which are  $4.23$  and  $4.29 \text{ ng/m}^3$ , respectively.

**3.4. Diagnostic Ratios of PAH.** As suggested in prior studies, the concentrations and ratios of some tracer PAHs could be used to identify the contribution of different sources. Four common diagnostic ratios including  $FLA/(PYR + FLA)$ ,  $IcdP/(BghiP + IcdP)$ ,  $ANT/(PHE + ANT)$ , and  $BaA/(BaA + CHR)$  were used to analyze the PAHs in particulate samples, and the results are presented in Figure 4.

According to the study of Yunker et al. [27], the ratio of  $FLA/(PYR + FLA)$  lower than 0.4 implies the unburned petrogenic sources and the ratio higher than 0.5 signals the coal and wood combustion, and a ratio between 0.4 and 0.5 indicates the liquid fossil fuel combustion sources. In this study, the ratio was 0.56 for particulate sample, indicating wood and coal combustion was the main source.

For  $IcdP/(BghiP + IcdP)$ , the ratio lower than 0.2 suggests unburned petrogenic sources, the ratio between 0.2 and 0.4 indicates liquid fossil fuel combustion sources, and the ratio higher than 0.5 signals the contribution of coal or wood combustion [6]. The ratio value was 0.38 for the sample, indicating comprehensive contributions from coal and liquid fossil fuel combustion sources. For another common used indicator  $ANT/(PHE + ANT)$ , a ratio lower than 0.1 signals the liquid fossil fuel sources and a ratio higher than 0.1 indicates the combustion sources. As presented in Figure 4, the ratios were all above 0.1 for particulate samples, indicating the combustion source contributions. In previous study, the ratio of  $BaA/(BaA + CHR)$  higher than 0.35 indicates the coal or other biomass combustion sources, while the ratio lower than 0.2 signals the liquid fossil fuel sources [28]. In this study, the ratio was 0.29 for particulate sample, indicating the coal and liquid fossil fuel combustion contribution.

Based on the results of diagnostic ratios analysis, coal combustion and emissions from liquid fossil fuel consumption were suggested to be the dominating sources of particulate-bound PAHs at Mount Heng site. In our previous studies conducted at Mount Tai, the highest mountain in northern China, coal combustion and vehicular emissions were the main sources of PAHs in atmospheric samples as well [1, 2]. The results obtained from the high altitude background sites could reflect an overall picture of PAH possible sources in China. However, special caution should be given when using diagnostic ratios. The ratio values could be altered as PAHs can react with other atmospheric compounds, such as hydroxyl radicals and/or ozone [29, 30]. In addition, degradation during the transport can modify the concentrations and ratios of PAHs, which is another limitation of using diagnostic ratios [31]. For example, BaA and ANT have been proved to degrade more quickly than their isomers-CHR and PHE, respectively [32, 33].

TABLE 5: Factor analysis for particulate-bound PAHs.

PAHs	Factor 1	Factor 2
ACY		0.735
ACE		0.977
FLU		0.962
PHE		0.790
ANT		0.846
FLA	0.857	
PYR	0.882	
BaA	0.940	
CHR	0.922	
BbF	0.954	
BkF	0.709	
BaP	0.810	
IcdP	0.851	
DahA	0.895	
BghiP	0.833	
% of variance	70.77	15.43
Sources	Vehicular and coal combustion emission	Unburned fossil fuels and incense burning

**3.5. Factor Analysis.** As stated above, several constraints exist when using diagnostic ratios to identify PAHs sources. Factor analysis, which served as an exploratory tool, can be used to quantify the major sources of PAHs. In this present study, the total concentrations of 15 PAHs compounds detected in particulate samples were subjected to factor analysis using SPSS version 16 (SPSS Inc.). Factor analysis was conducted with Varimax rotation, and factors with eigenvalue  $>1$  were considered. The results of rotated factors are listed in Table 5.

Two factors were separated, accounting for 86.2% of the total variance in the data. Factor 1, which explained 70.77% of the variance, presented high loading for individual PAHs with higher molecular weight, including FLA, PYR, BaA, CHR, BbF, BkF, BaP, IcdP, DahA, and BghiP. According to previous studies, BbF, BkF, IcdP, and BghiP are the major components of vehicular emission, and the presence of FLA, PYR, BaA, and CHR could point to coal and wood combustion sources [1, 34, 35]. Thus, factor 1 could represent sources from vehicular and coal combustion emission. Factor 2, which explained 15.43% of the variance, is highly loaded on ACY, ACE, FLU, PHE, and ANT. ACY and ACE could signal the contribution of unburned fossil fuels. PHE and ANT have been identified in industry emission in previous studies [1, 6]. PHE is also a major component of emissions from incense burning [26], while Flu could originate from different sources.

Based on the results of factor analysis, vehicular emission, coal combustion, industry emission, and unburned fossil fuels were suggested to be the PAHs sources at Mount Heng site. However, as mentioned earlier, the reactivity and degradation of individual PAHs could influence the results for profiles of PAH sources, and thus much work still needs to be conducted in the future.

## 4. Conclusion

Particulate-bound PAHs concentrations were investigated at high mountain region in Southern China from April to May 2009. The PAH concentrations ranged from 1.63 to 29.83 ng/m<sup>3</sup>, with a mean concentration of 6.03 ng/m<sup>3</sup>. The meteorological parameters including atmospheric pressure, relative humidity, and ambient temperature had good correlations with individual PAHs. The results obtained from air mass back trajectories and PAHs concentrations highlighted the importance of air mass origin. The air masses of N/NW group, transported over the most polluted region of China, had relatively higher PAH concentrations. Both the diagnostic ratio and factor analysis suggested that vehicular emission, coal combustion, industry emission, and unburned fossil fuels were the possible sources of PAHs at Mount Heng site.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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