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Analysis of the Effect of Meteorological Factors on $PM_{2.5}$-Associated PAHs during Autumn-Winter in Urban Nanchang

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ABSTRACT

The objectives of this study were to examine the association between $PM_{2.5}$-associated polycyclic aromatic hydrocarbons (PAHs) and their meteorological factors, and the relationship between $PM_{2.5}$-associated PAH concentrations and temperature, air pressure and air humidity. Data collected by Center for Disease Control (CDC) in Nanchang urban areas during the fall and winter seasons of 2014–2015 were analyzed by using high performance liquid chromatography (HPLC). Our study showed that the $PM_{2.5}$ mass concentration had a mean of 0.088 µg m^{-3}. Our results showed that the total concentration of PAHs in Nanchang was 22.54 ng m^{-3} (RSD: 8.50) and 15 different types of PAHs examined in this study all exceeded the China national standard. Multiple regression analysis revealed that the daily average concentration of total PAH was significantly associated with the temperature and daily minimum humidity ($p < 0.05$), but not daily wind speed and rainfall. Principal component analysis and characteristic ratio study indicated that the source of PAHs in $PM_{2.5}$ were mainly from vehicle exhaust and coal and gas combustion in Nanchang.

Keywords: $PM_{2.5}$; Meteorological factors; Polycyclic Aromatic Hydrocarbons (PAHs); HPLC.

INTRODUCTION

Haze weather is a state of atmosphere and is defined as excess level of all kinds of suspended particles in the atmosphere due to the formation of core substance floating in the air, smoke, dust and other particles (Li *et al.*, 2014). Haze is different from fog since the latter is a naturally occurred phenomenon due to more condensed water in the air. Haze weather is mainly the formation of man-made pollution of the environment, combined with low temperature, low air velocity and other natural conditions leading to very limited spread of pollutants. In recent years, haze has occurred frequently in most cities in China (Kong *et al.*, 2015; Li *et al.*, 2016). As a major component of haze, particulate matter $PM_{2.5}$ is known to cause serious illness to human body such as the respiratory and cardiovascular systems (Samat *et al.*, 2015; Anyenda *et al.*, 2016). Different profiles and distribution patterns of individual PAHs in $PM_{2.5}$ are characteristic of different sources of air pollution (Wang *et al.*, 2007). According to the "2012 Chinese cancer registry annual report", the total cancer cases in China reached 3.12 millions in 2009, indicating an average of 8,550 new cases per day, and six people diagnosed with malignant tumor per minute. National cancer mortality rate in China is 180.5/100,000 with estimated death of 2.7 millions every year (about 7,300 people die of cancer every day). Currently, about 20.0%–30.0% of cancer patients can survive for five years and the national cancer cases went up to 3.64 million in 2015. Among all reported cancer cases in China, lung cancer has the highest fatality rate and the current haze whether in China has been directly linked to the high incidence of lung cancer. Recent reports have indicated the culprit of haze, particulate matter $PM_{2.5}$, to be a serious health risk to human body, the polycyclic aromatic hydrocarbons adhered on the surface of the $PM_{2.5}$ is a persistent organic pollutant with carcinogenicity effect to humans (Choi *et al.*, 2015; Dauner *et al.*, 2016; Pongpiachan, 2016). Currently, there have been some reports emphasizing on the content and source of polycyclic aromatic hydrocarbons in $PM_{2.5}$ (Geng *et al.*, 2014; Cvetkovic *et al.*, 2015; Zhao *et al.*, 2015; Wang *et al.*, 2016). However, little is currently known regarding

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how meteorological factors influence the concentration and variation of $PM_{2.5}$-associated PAHs (Li *et al.*, 2012, Dan *et al.*, 2004). In particular, recent studies have shown that meteorological factors in different climatic conditions could have different effects on $PM_{2.5}$ (He *et al.*, 2014; Liu *et al.*, 2015; Zhang *et al.*, 2015b). Therefore, there is a need to sexamine meteorological factors and to understand concentration and distribution of $PM_{2.5}$-associated PAHs. Currently, rapid development of economics and the building booms has resulted in serious pollution of the air in many major cities in China, like Nanchang (SBJX, 2014a; Wang *et al*., 2015), thus it become important and necessary to study those factors that are associated with PAHs in $PM_{2.5}$.

METHODS

Sampling of Atmospheric $PM_{2.5}$

The Qing Yun Pu division of Jiangxi Center for Disease Control (CDC) collected the data. The $PM_{2.5}$ samples were collected from several monitoring locations of Qing Yun Pu district between 10th to 16th of each month from September 2014 to February 2015. These sampling locations had continuous airflow and were free of major pollution sources or obstacles within one-kilometer radius. With airflow velocities of 100 L min^{-1}, traffic $PM_{2.5}$ samplers containing glass fiber membranes were chosen and vertically placed 10–15 meters above the ground. Samples were collected at every other 10 minutes interval and each sampling time was 10 minutes and a total of ≥ 20 hours per sampling day.

Meteorology and National Monitoring Point Data

Data was collected every day for the test period from the environmental air quality monitoring station in Nanchang and collected samples were measured for SO_2, NO_2, PM_{10}, CO, O_3 and $PM_{2.5.}$ Other factors such as the daily temperature, relative humidity, atmospheric pressure (maximum, minimum and average), daily precipitation, and wind speed.

Analytical Methods

HPLC method was used to measure the concentration of 15 different types of $PM_{2.5}$-accociated PAHs (Olsson *et al.*, 2014; Jiang *et al.*, 2016). The 10/90 (v/v) ether/hexane mixed solvent was adopted to extract total $PM_{2.5}$ collected in the membrane. Resultant extracts were concentrated and purified through the standard protocols published by the Chinese Environmental Protection Department (NSPRC, 2012), then measured for $PM_{2.5}$ concentration using a HPLC separation apparatus with fluorescence and ultraviolet detectors. In addition, concentration of 15 different categories of PAHs were also analyzed including naphthalene, fluorene, acenaphthene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, 1,2-benzanthracene, benzo(a) fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, indene benzene (1, 2, 3 - CD) pyrene, and benzo (g, h, I) pyrene.

Statistical Analysis

Descriptive statistics was used to summarize the air pollution indexes. Independent two-sample t-tests were conducted to compare the difference of PAHs in $PM_{2.5}$ between fall and winter times. Pearson's correlation coefficients were used to report the association between PAHs categories and meteorological factors. In addition, stepwise multiple linear regression analysis was conducted to investigate the most meteorological significant factors associated with total PAHs in $PM_{2.5}$. Statistical software SPSS20 was used for data management and analyses. All statistical inferences were conducted at 5% alpha level.

Quality Control

Sample collection, preservation and storage, along with laboratory procedures for this study were carried out in highly strict accordance with the "Ambient air and stationary source emissions-determination of gas and particle-phase polycyclic aromatic hydrocarbons with gas chromatography/mass spectrometry" released by the Ministry of Environmental Protection of the People's Republic of China (MEPPRC). The sampling time was not less than 22 hours, which covered the rush hours. Qualified and trained graduate students from medical school of Nanchang universities, and researchers from Chinese Center for Disease Control and Prevention and Qing Yun Pu division of Jiangxi Center for Disease Control conducted the study, and collected and analyzed the data. Data were entered twice separately by two researchers into two different files and cross-checked to ensure no error.

RESULTS AND DISCUSSION

The Concentration of Atmospheric $PM_{2.5}$ and Polycyclic Aromatic Hydrocarbons

As shown in Table 1, the average daily concentration of $PM_{2.5}$ in Nanchang city during the monitoring time was 0.088 mg m^{-3} (SD: 0.03), which is 0.013 mg m^{-3} higher than the Chinese standard of 0.075 mg m^{-3} (GB3095-1996) Thirty days among the total of 40-day monitoring time (> 71%), the monitored concentration of $PM_{2.5}$ is over the air quality standard. This is consistent with the 70.91% reported by the Environmental Protection Bureau of Nanchang city, indicating that $PM_{2.5}$ pollution is serious in Nanchang. In addition, total PAH concentration was 22.54 ng m^{-3} (SD: 8.47), which is very close to the value (21.48 ng m^{-3}) reported from other rapid developing cities in China (Ray *et al.*, 2012). Fifteen categories of PAHs tested in this study all exceeded the allowable concentrations of the national air quality standards (HJ646-2013). The concentrations of pyrene, fluoranthene benzo (b) and chrysene were highest. The concentrations of fluorine, acenaphthylene, dibenzo (a,h), anthracene and anthracene were the lowest; all the others below 1.00 ng m^{-3} (Table 1).

PAHs belong to a larger group of aromatic carbons consisting of 2 to 13 cyclic rings that are fused together. The PAHs with 2 to 3 fused benzene rings are classified as low molecular weight PAHs (LMW-PAHs), while the 4-ringed PAHs are classified as middle molecular weight (MMW-PAHs), and the those with 5 to 7 rings are classified as higher molecular weight PAHs (HMW-PAHs) (Cheruyiot

Table 1. Concentrations of 15 categories of PAHs in $PM_{2.5}$ in Nanchang (ng m^{-3}) observed over 30 days from September 2014 to February 2015.

Category of PAH	Abb*	# of benzene rings	Mean	SD	Min	Max	Relevance ratio	Ambient air quality standard
$PM_{2.5}$ (ng m^{-3})	—	—	0.088	0.031	0.026	0.156	100	0.075
Total PAHs	∑PAHs	—	22.54	8.47	5.33	73.49	100	—
Naphthalene	Nap	2	1.16	0.92	0.26	3.86	100	0.4
Fluorene	Flu	2	0.21	0.22	0.13	0.58	100	0.3
Acenaphthene	Any	2	0.43	0.16	0.20	0.89	73	0.3
Phenanthrene	Phe	3	1.37	1.29	0.24	4.59	97	0.2
Anthracene	Ant	3	0.32	0.28	0.14	0.87	97	0.2
Fluoranthene	Flt	3	2.24	2.04	0.37	6.93	100	0.2
Pyrene	Pyr	4	3.27	2.23	1.10	8.65	100	0.2
Chrysene	Chr	4	2.56	1.74	0.52	6.51	100	0.3
1,2-benzanthracene	BaA	4	1.15	1.38	0.12	5.85	100	0.4
Benzo (a)fluoranthene	BbF	4	3.05	2.64	0.48	11.09	100	0.4
Benzo (k)fluoranthene	BkF	4	1.07	0.86	0.19	3.56	100	0.3
Benzo (a) pyrene	BaP	5	1.85	1.81	0.30	7.94	100	0.4
Dibenz (a,h)anthracene	DBA	5	0.32	0.27	0.09	1.11	100	0.3
Indene and (1, 2, 3 - CD) pyrene	IcdP	5	1.61	1.31	0.25	5.79	100	0.4
Benzo (g, h, I) pyrene	BghiP	6	2.07	1.74	0.34	7.30	100	0.3

*Abb – abbreviation.

et al., 2015). The results showed that different numbers of benzene ring for PAHs are distributed differently on the surface of $PM_{2.5}$ showing 4 rings > 2 rings > 3 rings > 5 rings > 6 rings (Fig. 1). The PAHs with number of 4 rings are most abundant and this may be largely due to the number of 4 ring PAHs are relatively less volatile. Due to dry weather in autumn and winter, less volatile and larger relative molecular mass PAHs enhanced their concentration on $PM_{2.5}$ particles surface (Zhu *et al.*, 2011; Shi *et al.*, 2015).

Table 2 shows the mean and SD's of the 15 different categories of PAHs. The mean of total PAH concentration level in the winter of 2015 (mean: 45. 53, SD: 15.15) was significantly higher than that determined in the fall of 2014 (mean: 13.91, SD: $p < 0.05$). In addition, the mean concentration for each type of 15 PAHs in winter was higher than that observed in the fall with the exception of acenaphthylene, which was detected only once in the fall and twice during the winter.

The correlation between the measured level of PAHs and meteorological factors that includes daily air pressure, temperature, humidity, precipitation, average wind speed, and sunshine time are presented in Table 3. The acenaphthylene level was excluded for this analysis due to its small sample size. As shown in Table 3, PAHs were negatively correlated with temperature and humidity. The multiple regression analysis showed that the total PAHs were significantly associated with daily temperature and daily minimum humidity (Table 4). These results are very similar to the findings recently reported by Liu and his co-workers (Liu *et al.*, 2015). Monitoring deposition characteristics of air contaminant in different geographic sites and areas is of great importance in understanding the fate and distribution of these pollutants in the environment as well as the estimating potential exposure to the populations living in these locations (Suryani R. *et al.*, 2015). However the effect of daily temperature and humidity were not found in some other studies conducted in Wuhan, Shenzhen and other southern areas in China (Sun *et al.*, 2015; Zhang *et al.*, 2015b). A very recent study have indicated that since there are no heating devices in south part of China, thus the impact of temperature, precipitation and relative humidity on the concentration of total PAH should be very limited (Zhang *et al.*, 2015a). These findings suggest that different regional climate characteristics (Ray *et al.*, 2012) could have different effects on PAHs and there is a need for more in-depth research to clarify linkage of daily temperature and humidity to PAH concentration. Guangzhou is a subtropical monsoon city with high humidity, high temperature and more rain weather, while Nanchang is a subtropical monsoon city with moist climate, especially in the fall and winter with low level of water precipitation (Huang *et al.*, 2012). Thus, it is not impossible to conclude that the rainfall has very limited effect on the concentration of total PAH in atmospheric $PM_{2.5}$ in Nanchang, although the influence of temperature and humidity on the concentration of total PAH in atmospheric $PM_{2.5}$ is consistent with other studies (Zhang *et al.*, 2015b).

The method used for factor analysis in this study is a commonly used one for the determination of the source of polycyclic aromatic hydrocarbons (Akhbarizadeh *et al.*, 2016; Ma *et al.*, 2016). Fifteen types of PAHs were extracted for two common factors of eigenvalues greater than 1, and the cumulative variance contribution rate was as high as 92.20%. The 4-ring, 5-ring, and 6-ring PAHs in principal component factor 1 are loaded on high side as shown in the factor loading/matrix results (Table 5), which indicates vehicle emissions to be the primary source of the pollution (Cai *et al.*, 2014). The fluorene and acenaphthene in common

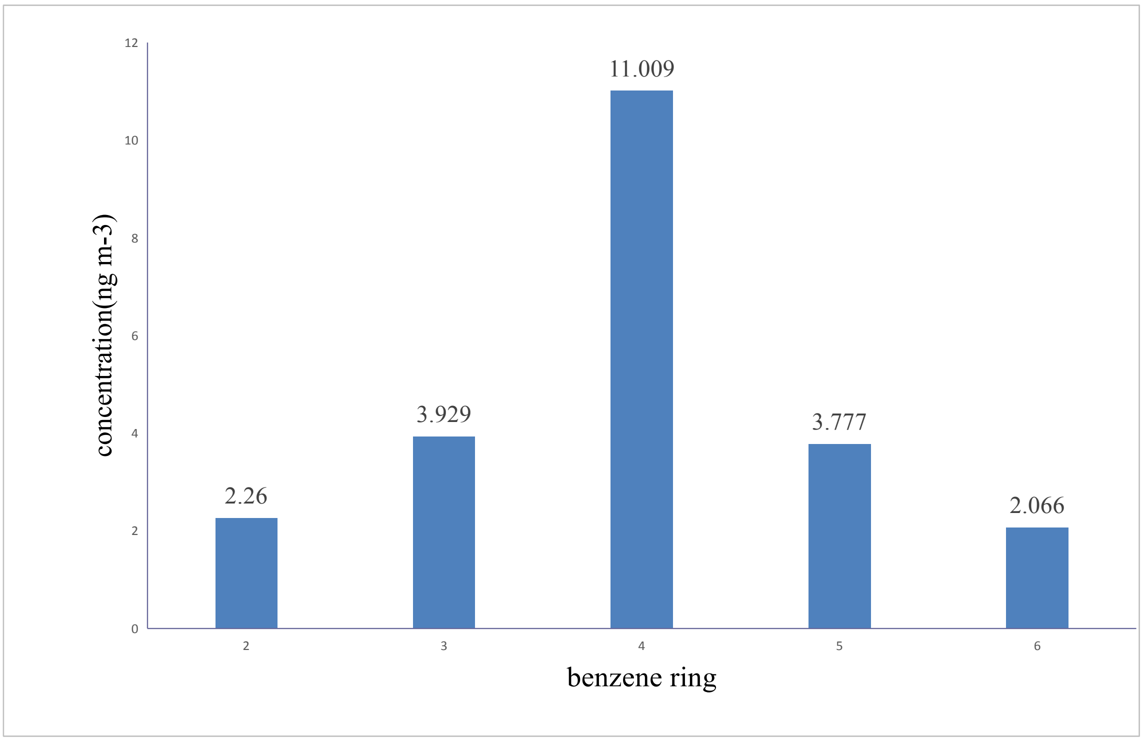

Fig. 1. Distribution of 16 kinds of PAHs in $PM_{2.5}$ in Nanchang city observed over 30 days from September 2014–February 2015.

Table 2. *t* means and SDs of different types of PAHs during the fall and winter in Nanchang.

Category of PAHs	Fall		Winter		*t*-value	*p*-value
	Mean	SD	Mean	SD		
Nap (ng m^{-3})	0.74	0.39	2.26	1.03	–4.32	0.002
Flu (ng m^{-3})	0.09	0.14	0.50	0.04	–12.93	< .001
Any (ng m^{-3})	0.36	0.12	0.57	0.15	–3.53	0.002
Phe (ng m^{-3})	0.63	0.39	3.26	0.67	–13.94	< .001
Ant (ng m^{-3})	0.16	0.13	0.72	0.13	–11.08	< .001
Flt (ng m^{-3})	1.10	0.68	5.29	0.97	–14.00	< .001
Pyr (ng m^{-3})	2.04	0.71	6.53	1.40	–9.20	< .001
Chr (ng m^{-3})	2.20	1.71	3.51	1.52	–2.01	0.054
BaA (ng m^{-3})	0.51	0.38	2.87	1.63	–4.30	0.002
BbF (ng m^{-3})	1.85	1.36	6.27	2.55	–4.94	0.001
BkF (ng m^{-3})	0.71	0.52	2.03	0.86	–5.40	< .001
BaP (ng m^{-3})	1.04	0.73	4.02	2.08	–4.20	0.002
DBA (ng m^{-3})	0.22	0.18	0.59	0.30	–4.38	< .001
IcdP (ng m^{-3})	1.33	1.00	4.03	1.82	–4.22	0.002
BghiP (ng m^{-3})	1.08	0.78	3.03	1.43	–3.88	0.003
∑PAHs	13.91	7.76	45.53	15.15	–5.97	< .001

factors 2 are loaded on higher side belongs to low ring PAHs, which suggests that the sources of these PAHs are mainly from coal combustion (Jung *et al.*, 2010; Samburova *et al.*, 2016). These findings are similar to the study about polycyclic aromatic hydrocarbons in the atmosphere of two subtropical cities in southeast China conducted by Wu *et al.* (2014) and the study about polycyclic aromatic hydrocarbons in $PM_{2.5}$ and $PM_{2.5–10}$ during the autumn and winter time in Urumqi of China conducted by Yi *et al.* (2013). Both these studies employed the principal component analysis (PCA) as an exploratory tool to identify possible sources of particulate PAHs. Qing Yun Pu district belongs to the industrial zone of Nanchang city that administers a number of large industrial enterprises that account for nearly 50% of the economic contribution to the entire Nanchang industrial area (SBJX, 2014b). Moreover, the Qing Yun Pu district is at the center of Nanchang city, where the traffic is usually heavy. This result allows to conclude that air pollution in Qing Yun Pu district is largely due to automobile exhaust emissions and industrial pollution.

Table 3. Correlations between PAHs and meteorological factors. Bolded numbers indicate significant correlation at 5% level.

PAHs (ng m^{-3})	average pressure (hpa)	maximum pressure (hpa)	minimum pressure (hpa)	average temperature (°C)	maximum air temperature (°C)	minimum air temperature (°C)	average relative humidity (%)	minimal humidity (%)	precipitation (mm)	daily average wind speed (m s^{-1})	sunshine duration (hours day^{-1})
∑PAHs	0.20	0.21	0.20	–0.85	–0.78	–0.79	–0.68	–0.71	–0.18	–0.23	0.25
Nap	0.16	0.16	0.16	–0.67	–0.61	–0.62	–0.59	–0.61	–0.10	–0.05	0.28
Flu	–0.08	–0.07	–0.08	–0.85	–0.82	–0.88	–0.49	–0.48	–0.12	–0.19	–0.07
Any	–0.08	–0.08	–0.09	–0.41	–0.39	–0.45	–0.45	–0.41	–0.22	–0.03	0.03
Phe	0.22	0.22	0.22	–0.90	–0.86	–0.81	–0.55	–0.59	–0.13	–0.16	0.12
Ant	0.23	0.24	0.23	–0.91	–0.85	–0.83	–0.64	–0.66	–0.16	–0.27	0.16
Flt	0.19	0.20	0.19	–0.93	–0.87	–0.85	–0.57	–0.62	–0.14	–0.21	0.13
Pyr	0.21	0.21	0.21	–0.90	–0.83	–0.84	–0.57	–0.63	–0.14	–0.25	0.18
Chr	0.22	0.22	0.22	–0.51	–0.43	–0.44	–0.57	–0.62	–0.23	–0.26	0.31
BaA	0.17	0.17	0.16	–0.77	–0.71	–0.70	–0.69	–0.65	–0.14	–0.11	0.25
BbF	0.21	0.21	0.21	–0.81	–0.74	–0.74	–0.67	–0.69	–0.18	–0.24	0.26
BkF	0.22	0.22	0.22	–0.77	–0.69	–0.71	–0.69	–0.71	–0.19	–0.28	0.29
BaP	0.19	0.19	0.18	–0.78	–0.70	–0.71	–0.70	–0.68	–0.17	–0.19	0.27
DBA	–0.23	–0.23	–0.23	–0.66	–0.57	–0.79	–0.69	–0.68	–0.18	–0.24	0.19
IcdP	0.21	0.21	0.20	–0.75	–0.66	–0.69	–0.67	–0.69	–0.20	–0.28	0.29
BghiP	0.21	0.21	0.20	–0.76	–0.67	–0.69	–0.68	–0.69	–0.19	–0.26	0.29

Table 4. Most significant meteorological factors that are associated with PAHs*.

	Parameter Estimate	Standard Error	*t*-value	*P*-value
Intercept	67.63	4.05	16.68	< .001
Average temperature (°C)	–1.62	0.21	–7.82	< .001
Minimal humidity (%)	–0.39	0.09	–4.36	< .001

* Results were obtained from multiple stepwise linear regression analysis and R^2 of the regression model is 0.835.

Table 5. The biggest factor analysis of variance factor loading matrix table.

Element	Factor 1	Factor 2
Fluorene	0.818	0.527
Acenaphthene	0.620	0.574
Phenanthrene	0.890	0.377
Anthracene	0.950	0.228
Fluoranthene	0.899	0.371
Pyrene	0.909	0.280
Chrysene	0.750	–0.570
1,2-benzanthracene	0.950	–0.103
Benzo b) fluoranthene	0.985	–0.147
Benzo (k) fluoranthene	0.957	–0.261
Benzo (a) pyrene	0.971	–0.171
Dibenz (a,h) anthracene	0.951	–0.241
Benzo (g, h, I) pyrene	0.958	–0.254
Indene and (1, 2, 3 - CD) pyrene	0.947	–0.286
Naphthalene	0.865	–0.113
The variance contribution rate	80.944	11.255

CONCLUSIONS

The $PM_{2.5}$ mass concentration in fall and winter of Nanchang city was 0.088 ± 0.031 µg m^{-3}, ranged from 0.026 to 0.156 µg m^{-3}. 71.43% of monitoring days exceeding the air quality standard in China. The total PAH concentration in $PM_{2.5}$ was 22.536 ± 8.469 ng m^{-3}, and 15 types of PAHs were detected to be over the allowable values of the national standard. The atmospheric $PM_{2.5}$ mass concentration and the concentrations of 15 kinds of PAHs were higher in winter than in autumn in Nanchang. Statistical analysis revealed that the total PAH concentration of atmospheric $PM_{2.5}$ to have a significantly negative correlation to daily average temperature and minimum humidity ($P < 0.05$), but effect from the daily average wind speed and average rainfall was not significant ($P > 0.05$). The polycyclic aromatic hydrocarbons in atmospheric $PM_{2.5}$ mainly come from vehicle exhaust, coal combustion and factory gas sources. These findings allow to suggest that industrial pollution and automobile exhaust pollution be the major source of air pollution in Nanchang. In addition, with the rapid industrialization in Nanchang city, the toxicity of PAHs in $PM_{2.5}$ could pose serious health risks to the public even in the suburban areas. Moreover, the carcinogenic risk of PAHs to the public can be more serious due to increasing traffic activity in the area. Therefore, more stringent measurement on the traffic control should be implemented in order to meet the new criterion on ambient air quality and to reduce the health risk to the public. In particular, more efficient emission control measures are desperately needed for controlling current air pollution in urban and suburban atmosphere of China.

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