Characteristics and Source Identification of Polycyclic Aromatic Hydrocarbons and *n*-Alkanes in PM$_{2.5}$ in Xiamen

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ABSTRACT

PM$_{2.5}$ samples were collected to characterize the organic compounds of polycyclic aromatic hydrocarbons (PAHs) and *n*-alkanes in Xiamen, China, in 2013. The concentrations of PM$_{2.5}$, PAHs and *n*-alkanes were 55.53 µg m^{-3} and 22.14 µg m^{-3}, 15.73 ± 6.25 ng m^{-3} and 5.65 ± 3.73 ng m^{-3}, and 148.57 ± 36.84 ng m^{-3} and 97.53 ± 67.46 ng m^{-3} in winter and summer, respectively, demonstrating higher pollutant levels in wintertime. Benzo[e]pyrene (BeP) was the most abundant PAH, accounting for 32% and 26% of the total quantified PAHs in winter and summer, respectively, followed by coronene (Cor) and phenanthrene (Phe). 5-ring PAHs were the most dominant group, contributing 50.8% and 44.1% to the total quantified PAHs in winter and summer, respectively. Two peaks were exhibited by the homologue distributions of *n*-alkanes, and the dominant components were enriched in the high molecular weight fraction. Meteorological parameters had a stronger impact on the atmospheric PAH levels in summer than winter, and the most significant parameter was relative humidity (RH), followed by temperature. The diagnostic ratios indicated that PAHs in Xiamen were mainly contributed by petroleum combustion in the two seasons and the concentrations of *n*-alkanes were mainly influenced by anthropogenic sources. The annual values of the benzo[a]pyrene (BaP) equivalent concentration and incremental lifetime cancer risk (ILCR) were 0.83 ± 0.63 ng m^{-3} and 7.17 × 10^{-5} in winter and 1.11 × 10^{-4} ng m^{-3} and 3.29 × 10^{-5} in summer, respectively. The findings illustrated that the overall exposure risk to PM$_{2.5}$-bound PAHs did not trigger an alert in Xiamen, but higher risks in winter were still shown to exist.

Keywords: PM$_{2.5}$; PAHs and *n*-alkanes; Source characterization; Health effects; Xiamen.

INTRODUCTION

Along with the industrial activities, urbanization, and rapid economic expansion, air pollution has become a serious environmental issue in China. Currently, fine particulate matter (PM$_{2.5}$), referring to particle with an aerodynamic diameter less than 2.5 µm, has been identified as the most important primary pollutant in China (www.zhb.gov.cn). In 2013, the mean concentration of PM$_{2.5}$ in 74 Chinese key cities was 72 µg m^{-3}, twice exceed the newly ambient air quality standards in China (GB3095-2012) of 35 µg m^{-3}.

Besides the environmental impacts such as visibility deterioration (Cao *et al.*, 2012a; Wang *et al.*, 2013), PM$_{2.5}$ also affects human health due to particle sizes, carbonaceous contents, and specific chemicals that are bounded to the particles, especially of organic fractions (Duan *et al.*, 2010; Cao *et al.*, 2012b; Huang *et al.*, 2014).

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic substances that are formed during the incomplete combustion processes of fossil fuel and industrial processes in heavily urbanized or industrialized regions (Mostert *et al.*, 2010; Cheruiyot *et al.*, 2015; Lai *et al.*, 2016; Redfern *et al.*, 2017). They can be also emitted from natural processes such as biomass burning (Wang *et al.*, 2007a; Saha *et al.*, 2017). PAHs are ubiquitous environmental pollutants, which widely exist in the atmosphere, water, soil and sediments (Wang *et al.*, 2007a; Mostert *et al.*, 2010) and are toxic to all living organisms. The concentrations of

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PAHs were varied on the size distribution of airborne particulate matters, and about 95% of total PAHs present in the particulate with a diameter less than 3 μm (Baek *et al.*, 1991a, b; Venkataraman *et al.*, 1994). *n*-Alkanes are also ubiquitous which is a group of non-polar and photo-catalytically stable organic compounds. They are generated by biological source and incomplete combustion of fossil fuel, wood and agricultural burning (Simoneit, 1977).

Xiamen is a coastal city located at southeastern China where is in the west coast of the Taiwan Straits. Its climate is dominated by sub-tropic climate influenced by northeastern monsoon from October till April of next year and southwestern monsoon from May till August (Li *et al.*, 2013). The prevailing wind directions in winter are north and northeast, while south and southeast winds are prevailing in summer. As similar as most cities in China, Xiamen has been suffered from air pollution, especially during wintertime (Zhang *et al.*, 2012; Zhang *et al.*, 2016). Most of the previous studies focused on the characterization, seasonal variations and sources apportionment of PM (Zhao *et al.*, 2011a; Zhang *et al.*, 2012; Li *et al.*, 2013; Zhang *et al.*, 2016). The characterization, seasonal variation and spatial and temporal distribution of PAHs in PM$_{2.5}$, PM$_{10}$ and TSP in Xiamen have been studies during 2003–2004 and 2008–2009, respectively (Hong *et al.*, 2007; Wang *et al.*, 2007a; Zhao *et al.*, 2011a, b). Very limited studies were conducted on ambient *n*-alkanes, besides a report on their levels in PM$_{10}$ (All undergraduate of 2003, 2007) and a measurement done inside and outside a temple (Wu *et al.*, 2009). However, none of study focused on both PAHs and *n*-alkanes status. Thus, the results of our work are expected to fully understand the current seasonal variations, characterize their potential sources, evaluate the health risk of PAHs in Xiamen.

MATERIAL AND METHODS

Ambient Samples

All the samples were collected at Huli district in Xiamen (Fig. 1). Huli district was used to be the industry district in Xiamen Island. Recently, in order to protect the environment, most of industries have been moved out of the island. However, there still have port and airport at Huli. Our sampling site is set at a building which is about 8 m above ground with about 300 m to main road, 2 km to port and 4 km to airport. Quartz microfiber filters (47 mm diameter, Whatman) were used for PM$_{2.5}$ sampling which were collected synchronously in January and July representing winter and summer. For each month, three weeks were used for sampling, and each sample was run for 24 h from 10:00 till 10:00 next day.

Analysis

All the samples were analyzed at the Institute of Earth Environment, Chinese Academy of Sciences (Xi'an). The quartz filters were pre-combusted at 800°C for 3 h prior to use. Before sampling, they were maintained at a relative humidity (RH) of 50% and 25°C for over 24 h and then weighted using a Sartorius MC5 electronic microbalance (Sartorius, Göttingen, Germany) with a sensitivity of ± 1 μg. After sampling, the filters were conditioned and weighed again to determine the mass concentration of the filtered particles. One punch (0.526 cm^2) of filter was analyzed using an Agilent 7890A GC/5975MSD system (City, Country, USA) with in-injection port thermal-desorption-gas chromatography/mass spectrometry (TD-GC-MS) method. The details of TD-GC-MS method have been described by Ho and Yu (2004) and Ho *et al.* (2008, 2011). The parameter settings and quality control on the analysis were reported in Chen *et al.* (2014). A total of 17 PAHs (i.e., phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF) benzo [a]fluoranthene (BaF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene (Per), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), benzo[ghi]perylene (BghiP), coronene (Cor), and dibenzo[a,e] pyrene (DaeP)) and 19 *n*-alkanes (i.e., from C$_{17}$ to C$_{35}$) were quantified.
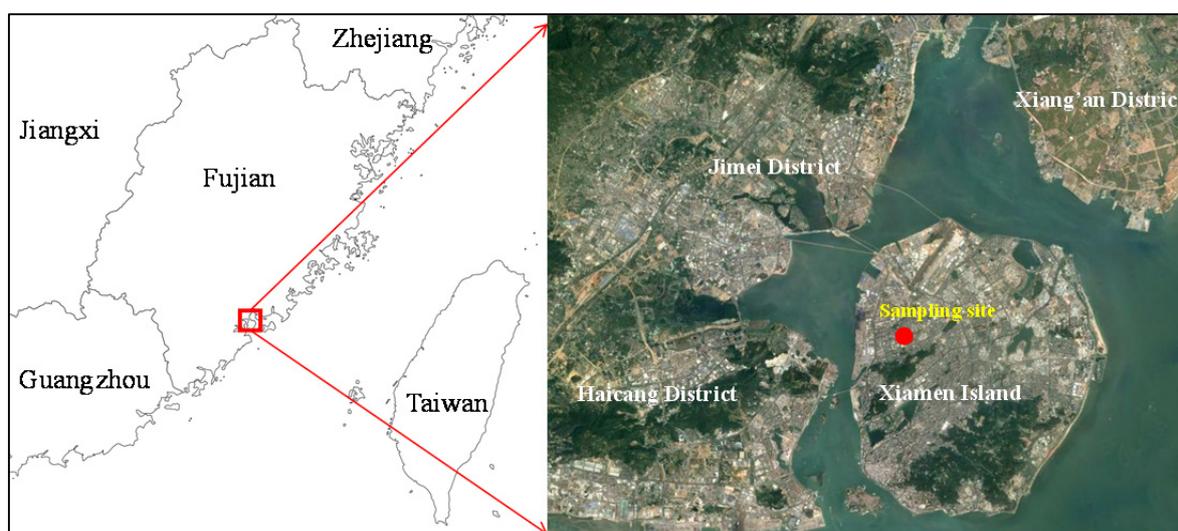

Fig. 1. Location of sampling site.

Meteorological Data and Gases Data

Meteorological parameters including temperature (T), RH, atmospheric pressure (P), wind speed and direction were recorded continuously from Xiamen Meteorological Bureau (Fig. 2). Higher temperature was seen in summer (29.5°C) than winter (14.7°C). However, due to it is located in coastal area, less variations on RHs were seen between winter (57.6%) and summer (66.0%). Affected by continental high pressure system, the pressure was 985.3 hPa in winter, which is higher than 977.2 hPa in summer. A large difference was found for wind direction. The main wind direction is northeast in winter but southwest in summer. The data for other gases including SO_2, NO_2, O_3 and CO were obtained from Xiamen Environmental Monitoring Central Station.

Health Risk Assessment

The health risk of PAHs can be assessed by BaP equivalent concentration (BaP_{eq}). It is calculated by Eq. (1):

$$BaP_{eq} = \sum C_i \times TEF_i \qquad (1)$$

where C_i refers to the mass concentration of specific PAHs components, TEF_i is their corresponding toxic equivalent factor (TEF). According to U.S. EPA (2012), the TEFi values for Phe, Ant, Flu, Pyr, BaA, Chr, BbF, BkF, BeP, BaP, Per, IcdP, BghiP and DahA are 0.001, 0.01, 0.001, 0.001, 0.1, 0.01, 0.1, 0.1, 0.01, 1, 0.001, 0.1, 0.01 and 5 respectively. By defined the UR_{BaP} (unit risk) as the number of people at risk of contracting cancer from inhalation a BaP_{eq} of 1 ng m^{-3} within 70-year lifetime, the
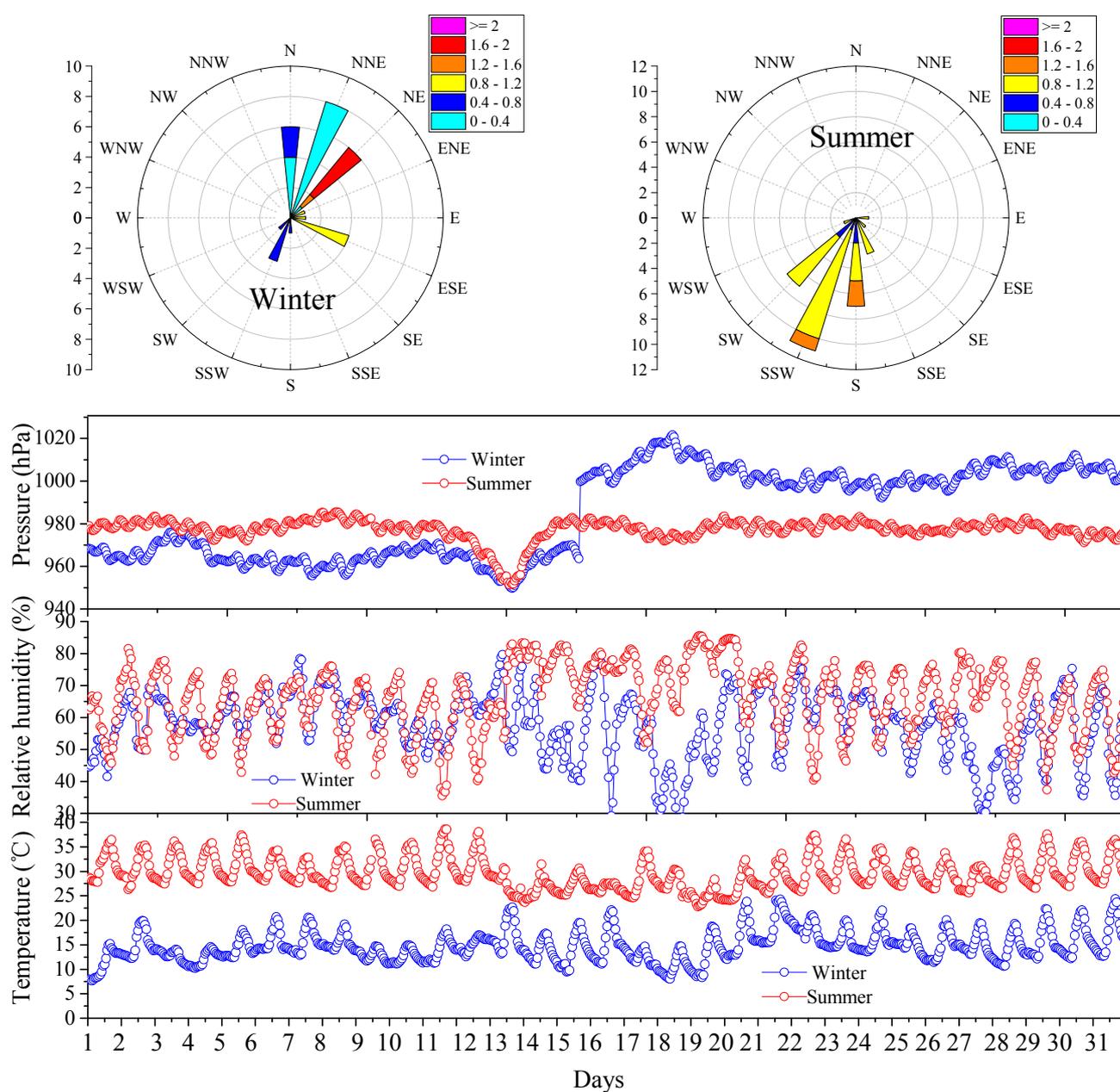

Fig. 2. Variation of meteorological parameters in January (winter) and July (summer).

incremental lifetime cancer risk (ILCR) can be calculated as equation (2), and the value of UR_{BaP} used in this study is 8.7×10^{-5} (WHO, 2000):

$$ILCR = \sum BaP_{eq} \times UR_{eq} \tag{2}$$

RESULTS AND DISCUSSION

Obvious seasonal variations of PM concentrations have been reported in Xiamen by previous works (Zhao *et al.*, 2011a; Li *et al.*, 2013). In this study, the concentrations of $PM_{2.5}$ were 55.53 µg m^{-3} and 22.14 µg m^{-3} in winter and summer, respectively, which were 25% and 12% lower than those measured in winter (74.2 µg m^{-3}) and summer (25.2 µg m^{-3}) in 10 years ago (Cao *et al.*, 2012). Our results indicate that more decrement of $PM_{2.5}$ appeared in winter in the last decade. The annual (average values of winter and summer) $PM_{2.5}$ mass concentration is 38.84 µg m^{-3} which is close to 36 µg m^{-3} issued by Xiamen Environmental Protection Bureau (http://www.xmepb.gov.cn/) and the new ambient quality standard of 35 µg m^{-3} established by Ministry of Environmental Protection of China in 2012 (GB 3095-2012).

PAHs Concentration

The average concentration of total quantified PAHs was 10.10 ± 7.07 ng m^{-3} during the whole sampling period (Table 1), which was lower than 14.78 ng m^{-3} in PM_{10} from 2004 till 2005 and 21.89 ng m^{-3} in TSP from 2008 till 2009 (Hong *et al.*, 2007; Zhao *et al.*, 2011b). The concentration levels in Xiamen are close to Lhasa (i.e., 10 ± 6.6 ng m^{-3} and 20 ± 15 ng m^{-3} at two sites) (Liu *et al.*, 2013), higher than those reported at Hsinchu (1.0–8.4 ng m^{-3}) (Yang *et al.*,

2017), a high mountain site (1.36 ng m^{-3}) (Liu *et al.*, 2017), and some rural areas such as Lulang (0.59 ± 0.52 ng m^{-3}) (Chen *et al.*, 2014) and Mt. Halla (0.40 ± 0.58 ng m^{-3}) (Lee *et al.*, 2008), but much lower than the values of 104 ± 130 ng m^{-3} in Beijing (Ma *et al.*, 2011) and 25.92–90.80 ng m^{-3} in Nanjing. Seasonal differences were seen with the values of 15.73 ± 6.25 ng m^{-3} in winter and 5.65 ± 3.73 ng m^{-3} in summer. The results are consistent to the trend that the PAHs concentrations were higher in winter than summer in most Chinese cities (Zhao *et al.*, 2011b, Bandowe *et al.*, 2014). The ratio of mean winter to mean summer (W/S) is 2.78, which was slight lower than the W/S value (2.82) reported at Xiamen in PM_{10} from 2004 till 2005 (Hong *et al.*, 2007).

As shown in Fig. 3, BeP was the most abundant PAHs, accounting for 32% and 26% of the total quantified PAHs during the winter and summer, followed by Cor and Phe. This result was different from that in a high mount site, where Phe, Flu and IcdP were the top three abundant PAHs species (Liu *et al.*, 2017). Our targeted PAHs can be classified into 3- to 7-aromatic rings, and furtherly defined as low molecular weight (LMW, referred to 3-rings PAHs), middle molecular weight (MMW, referred to 4-rings PAHs), and high molecular weight (HMW, sum of 5-, 6- and 7-rings PAHs) (Table 1). The dominant PAHs in our study were 5-rings in both winter and summer, accounting for 50.8% and 44.1% of the total PAHs respectively (Fig. 3). The contribution of 3-rings PAHs was only 7.8% in winter but as high as 20.4% in summer, explaining by the gas/particulate partitioning effect due to the temperature differences. During the entire sampling period, the ratio of HMW/LMW was 6.51, which is consistent with the ratio of 6.94 reported at Guangzhou, another southern Chinese city (Liu *et al.*, 2015).

Table 1. Mass concentration of PAHs components (ng m^{-3}) in $PM_{2.5}$ at Xiamen.

|	Winter		Summer		Annual		W/S
	Average	SD	Average	SD	Average	SD	
Phe (3 rings)	1.02	0.23	0.98	0.59	1.00	0.46	1.05
Ant (3 rings)	0.21	0.03	0.18	0.11	0.19	0.09	1.16
Flu (4 rings)	0.47	0.20	0.17	0.07	0.30	0.20	2.77
Pyr (4 rings)	0.43	0.15	0.15	0.06	0.27	0.18	2.85
BaA (4 rings)	0.20	0.09	0.11	0.05	0.15	0.08	1.82
Chr (4 rings)	0.73	0.30	0.24	0.12	0.46	0.33	3.05
BbF (5 rings)	0.71	0.28	0.22	0.20	0.44	0.34	3.24
BkF (5 rings)	0.77	0.35	0.25	0.19	0.48	0.38	3.13
BaF (5 rings)	0.64	0.29	0.23	0.16	0.41	0.30	2.80
BeP (5 rings)	5.08	2.40	1.48	1.24	3.07	2.57	3.43
BaP (5 rings)	0.28	0.14	0.10	0.08	0.18	0.14	2.79
Per (5 rings)	0.51	0.29	0.22	0.17	0.35	0.27	2.37
IcdP (6 rings)	0.78	0.37	0.18	0.15	0.45	0.40	4.25
BghiP (6 rings)	0.99	0.50	0.27	0.23	0.59	0.52	3.63
DahA(6 rings)	0.13	0.06	0.03	0.03	0.08	0.07	3.90
Cor (7 rings)	2.48	1.09	0.69	0.60	1.48	1.23	3.58
DaeP (6 rings)	0.29	0.11	0.15	0.20	0.21	0.18	1.90
PAHs	15.73	6.25	5.65	3.73	10.10	7.07	2.78
LMW-PAHs	1.23	0.24	1.15	0.70	1.19	0.55	1.07
MMW-PAHs	1.83	0.67	0.67	0.27	1.18	0.75	2.73
HMW-PAHs	12.67	5.70	3.83	3.17	7.73	6.26	3.31
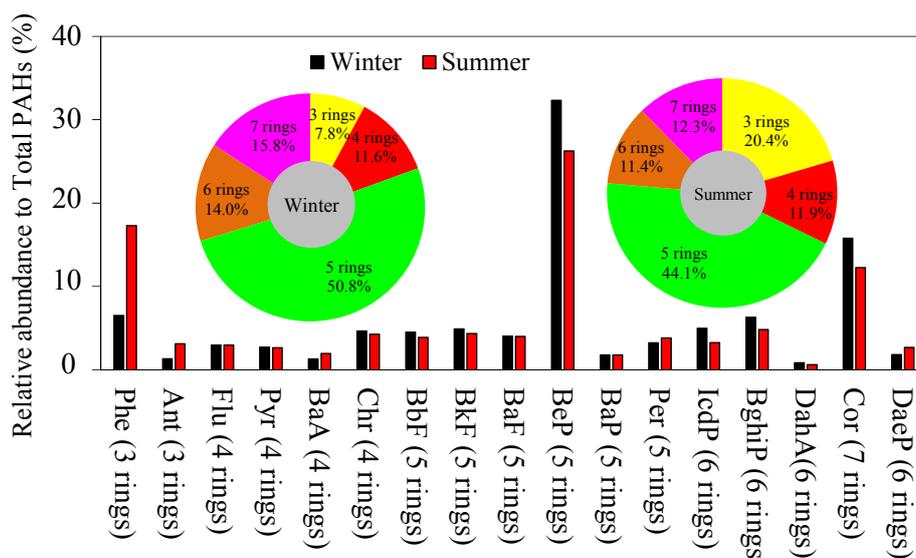
Fig. 3. Relative abundance of detected PAHs components to total PAHs.

Such ratio was 10.30 in winter but as low as 3.31 in summer. It was related to the higher contributions of volatile PAHs in summer, and additional sources for HMW PAHs in winter, such as long-rang transport from northern China (Hong *et al.*, 2007).

n-Alkanes Concentrations

Nineteen *n*-alkanes (C_{17}–C_{35}) were detected in this study with average values of 148.57 ± 36.84 ng m^{-3} and 97.53 ± 67.46 ng m^{-3} during winter and summer times, respectively (Table 2). The mean concentration for the whole sampling period was 120.08 ± 61.09 ng m^{-3}, which is close to 129.5 ng m^{-3}, lower than 1.4×10^{4} ng m^{-3} and higher than 27.3 ng m^{-3} observed at the bus terminus, temple and campus at Xiamen in TSP, respectively (Wu *et al.*, 2009). In comparison, the average concentration level is equal to 163.0 ± 193.5 ng m^{-3} in PM$_{2.5}$ in Beijing (Huang *et al.*, 2006) and 94.2 ± 67.9 ng m^{-3} in TSP at Lahsa (Gong *et al.*, 2011), but lower than the value of 141–392 ng m^{-3} in PM$_{10}$ at Guangzhou (Bi *et al.*, 2002) and 449–1733 ng m^{-3} in PM$_{10}$ at Baoji (Xie *et al.*, 2009). However, our value is higher than an average of 23.5 ng m^{-3} in PM$_{2.5}$ in Hong Kong and some rural areas, such as 1.25 ± 2.28 ng m^{-3} in Lulang (Chen *et al.*, 2014), 7–95 ng m^{-3} in Finland (Rissanen *et al.*, 2006) and 8.2 ng m^{-3} in Sweden (Wingfors *et al.*, 2011). The W/S for *n*-alkanes was 1.52, which was much lower than the value of 6.99 reported in Taiyuan, where strong influence from heating was found during wintertime, indicating the seasonal differences for the levels of *n*-alkanes were relatively small in Xiamen.

The distribution of C_{17}–C_{35} concentration as a function of carbon number had similar pattern during the winter and summer with two peaks obviously divided by C_{25} (Fig. 4). However, the peak values were found at C_{22} and C_{31} during winter while those were at C_{31} and C_{21} during summer. This distribution pattern was as similar as the results found in Tianjin, a coastal city in northern China, where the distributions of *n*-alkanes had two peaks at C_{22}–C_{25} and C_{29}

(Wu *et al.*, 2007). In addition, our result was different with that only one peak at C_{29}–C_{33} was seen in Lulang. The LMW *n*-alkanes (carbon number < 24) are mainly derived from anthropogenic sources, such as incomplete combustion of fossil fuels, while the HMW *n*-alkanes (carbon number ≥ 25) are regarded as originated from strong contributions of plants waxes. The values of LMW/HMW was 1.03 and 0.56 during winter and summer, respectively, indicating that there were more emissions from higher plants in summer.
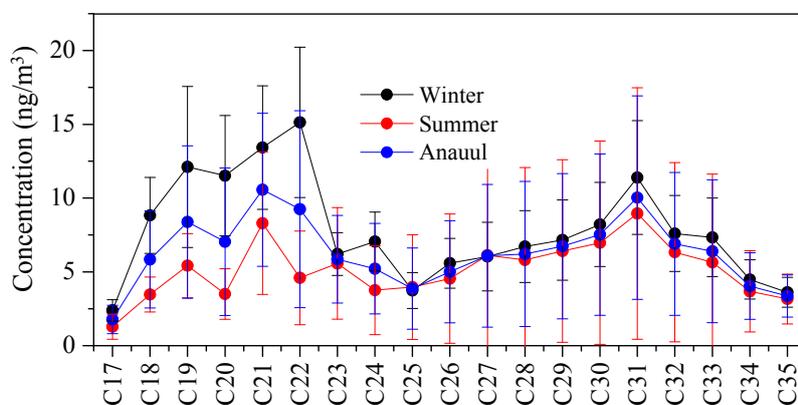
Relations between Meteorological Parameters and Gases with PAHs

Sampling time and period are important factors influenced the relations between PAHs concentration and meteorological parameters. For example, under high temperature and humidity and strong radiation in summer, more PAHs partitioned from particulate phase to vapor phase. Stronger photochemical reactions and thermal circulation, more rainfall and deeper mixing layer can also impact on the PAHs concentration in summer than other seasons. The correlations between PAHs and meteorological parameters have been widely reported in previous studies. Chetwittayachan *et al.* (2002), Hien *et al.* (2007) and Liu *et al.* (2015) have found significant correlations between PAHs and RH; however, no significant correlations were seen by Li *et al.* (2016) and Fang *et al.* (2004), indicating that the relationship between PAHs and meteorological parameters is complex.

In order to investigate the influences of meteorological parameters on the variation of PAHs, Pearson's correlation analysis was applied in this study (Table 3). The relationship between PAHs and meteorological parameters including wind speed, T, RH and P was more significant in summer than in winter, and it may be also due to more influences from long range transport in winter (Zhao *et al.*, 2011b). T has been regarded as the strongest parameter that impact on most of the targeted PAHs. Pehnec *et al.* (2016) and Amarillo and Carreras (2016) have reported that significantly negative correlation was found between T and PAHs. In

Table 2. Mass concentration of *n*-alkanes (ng m^{-3}) components in PM$_{2.5}$ at Xiamen.

	Winter		Summer		Annual		W/S
	Average	SD	Average	SD	Average	SD	
n-Heptadecane (C$_{17}$)	2.40	0.71	1.29	0.85	1.78	0.96	1.86
n-Octadecane (C$_{18}$)	8.83	2.58	3.47	1.18	5.84	3.29	2.54
n-Nonadecane (C$_{19}$)	12.12	5.45	5.42	2.17	8.38	5.16	2.24
n-Icosane (C$_{20}$)	11.52	4.08	3.50	1.72	7.04	5.00	3.29
n-Heneicosane (C$_{21}$)	13.43	4.19	8.30	4.83	10.56	5.19	1.62
n-Docosane(C$_{22}$)	15.13	5.09	4.60	3.17	9.25	6.68	3.29
n-Tricosane (C$_{23}$)	6.21	1.45	5.58	3.78	5.85	2.97	1.11
n-Tetracosane (C$_{24}$)	7.05	1.99	3.76	3.01	5.22	3.07	1.88
n-Pentacosane(C$_{25}$)	3.74	1.22	3.97	3.55	3.87	2.74	0.94
n-Hexacosane (C$_{26}$)	5.59	1.69	4.55	4.38	5.01	3.46	1.23
n-Heptacosane (C$_{27}$)	6.04	2.33	6.13	6.20	6.09	4.84	0.99
n-Octacosane(C$_{28}$)	6.72	2.43	5.81	6.26	6.21	4.92	1.16
n-Nonacosane (C$_{29}$)	7.15	2.72	6.41	6.19	6.74	4.93	1.12
n-Triacontane (C$_{30}$)	8.21	2.85	6.97	6.89	7.52	5.47	1.18
n-Hentriacotane (C$_{31}$)	11.39	3.86	8.96	8.51	10.03	6.90	1.27
n-Dotriacontane(C$_{32}$)	7.60	2.57	6.34	6.07	6.89	4.84	1.20
n-Titriactotane (C$_{33}$)	7.34	2.66	5.64	5.99	6.39	4.84	1.30
n-Tetratriactoane (C$_{34}$)	4.49	1.33	3.68	2.75	4.04	2.25	1.22
n-Pentatriacontane (C$_{35}$)	3.62	1.01	3.16	1.69	3.36	1.43	1.14
n-alkanes	148.57	36.84	97.53	67.46	120.08	61.09	1.52
LMW	76.68	20.62	35.91	18.19	53.92	27.98	2.14
HMW	71.89	23.78	61.62	57.98	66.16	45.93	1.17
CPI 1	0.96	0.17	1.27	0.07	1.13	0.20	0.76
CPI 2	0.86	0.19	1.51	0.15	1.22	0.36	0.57
CPI 3	0.99	0.15	1.11	0.04	1.05	0.12	0.89

Fig. 4. Variation of detected *n*-alkanes concentration.

Table 3. Correlation analysis between differeny rings of PAHs and meteorological parameters and Gases.

	Rings	SO$_2$	NO$_2$	O$_3$	CO	PM$_{10}$	PM$_{2.5}$	WS	T	RH	P
Winter	3	−.070	−.238	.410	.269	.364	.196	.443	−.535[*]	.038	.552[*]
	4	.228	.040	.297	.461[*]	.585[**]	.270	.508[*]	−.378	−.082	.428
	5	.211	.187	−.154	.095	.225	−.225	.081	−.336	.019	.361
	6	.124	.139	−.219	−.012	.094	−.376	.002	−.365	.072	.377
	7	−.045	.015	−.245	−.159	−.095	−.511[*]	−.091	−.419	.140	.410
Summer	3	−.169	−.166	.389	−.212	−.019	−.022	.526[**]	.187	.078	−.130
	4	.228	.326	.106	−.002	.277	.363	−.054	.335	−.441[*]	.338
	5	.378	.352	−.199	−.019	.234	.336	−.402	.472[*]	−.548[**]	.434[*]
	6	.440[*]	.407[*]	−.125	.018	.331	.433[*]	−.364	.461[*]	−.564[**]	.440[*]
	7	.417[*]	.364	−.041	−.045	.334	.407[*]	−.285	.447[*]	−.459[*]	.325

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

our study, the most significant meteorological parameters for PAHs in summer was RH, followed by T (Table 2). RH correlated with 4- to 7-rings PAHs negatively in summer but positive for 3-rings PAHs, and no significant correlation for both PAHs was seen between RH and PAHs in winter. The negative correlations between RH and PAHs were also found at Cordoba, Argentina (Amarillo and Carreras, 2016). Rainfall can wash out particles and lead the decrease of concentrations of PM and its species, while RH increased at the same time. However, positive correlations between PAHs and RH have been reported at Zagreb, Croatia (Pehnec *et al.*, 2016) and Guangzhou, China (Liu *et al.*, 2015).

Due to the influences from traffic emissions, positive correlations between PAHs with CO and NO but negative correlations between PAHs and NO_2 and O_3 were found by Valerio *et al.* (2000) at a heavy traffic street in Genoa, Italy. However, no significant correlation was found between PAHs and gases (SO_2, NO_2, O_3 and CO) in our study, expect the 6-rings PAHs had a significant correlation between SO_2 and NO_2 during summer and the 4-rings had good correlation between CO during winter.

Source Identification

Diagnostic Ratios for PAHs

Diagnostic ratios have been used to identify the possible sources of PAHs (Yunker *et al.*, 2002; Ravindra *et al.*, 2008). Tobiszewski and Namieśnik (2012) have entirely summarized

the PAH diagnostic ratios for the identification of pollution emission in different environmental compartments. Because of their relative stability (Yunker *et al.*, 2002), Ant/(Phe + Ant), Fla/(Fla + Pyr), IcdP/(IcdP + BghiP) and BaA/(BaA + Chr) have been often used to distinguish the possible categories of PAH sources in the environment (Table 4). It has been reported as the petroleum source if the ratio of Ant/(Ant + Phe) is lower than 0.1, and as combustion source if the ratio is higher than 0.1 (Pies *et al.*, 2008; Han *et al.*, 2011). For the ratio of Flu/(Flu + Pyr), its value is lower than 0.4, between 0.4–0.5 and higher than 0.5 considered as petroleum, fossil fuel combustion and grass, wood, coal combustion sources, respectively (Han *et al.*, 2011, Li *et al.*, 2016). As for BaA/(BaA + Chr), it indicates petroleum source, coal combustion source and vehicular emission source when values of the ratios are lower than 0.2, between 0.2–0.35 and higher than 0.35, respectively (Yunker *et al.*, 2002). The IcdP/(IcdP + BghiP) ratios below 0.2 is considered as petroleum, 0.2–0.5 for petroleum combustion and more than 0.5 for biomass and coal combustion (Yunker *et al.*, 2002).

In this study, the mean values of Ant/(Phe + Ant) ratios were 0.17 (ranged from 0.13 to 0.22) and 0.15 (ranged from 0.12 to 0.18) during winter and summer, respectively, suggesting the stable combustion sources both in winter and summer (Fig. 5). For BaA/(BaA + Chr), its ratios ranged from 0.18 and 0.24 in winter and from 0.19 to 0.49 in summer, and

Table 4. Diagnostic ratios of PAHs in previous works and our study.

| Diagnostic ratios	Range	Winter	Summer	Annual
Ant/(Ant + Phe)	< 0.1: Petroleum; > 0.1: combustion	0.17	0.15	0.16
FLA/(FLA + PYR)	< 0.4: Petroleum; 0.4–0.5: fossil fuel combustion; > 0.5: biomass, coal combustion	0.52	0.53	0.52
BaA /(BaA + CHR)	0.2–0.35: petroleum combustion; > 0.35: biomass, coal combustion	0.22	0.31	0.28
IcdP/(IcdP + BghiP)	< 0.2: Petroleum; 0.2–0.5: petroleum combustion; > 0.5: biomass, coal combustion	0.45	0.40	0.42
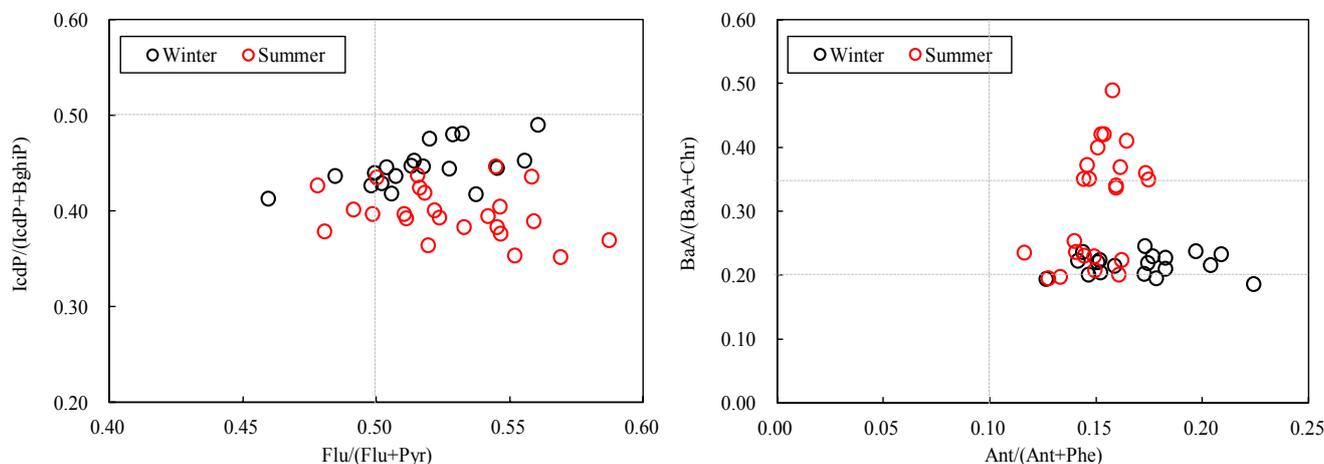
Fig. 5. Diagnostic ratios for determining sources of PAHs.

most of the ratios were in the range of 0.20–0.35, indicating that the main source was petroleum combustion in both seasons. Meanwhile, the ratios exceeded 0.35 in summer should be caused by biomass burning emissions. According to the result showed in Fig. 5, it can be seen that all the values of IcdP/(IcdP + BghiP) ratios were in the range of 0.20–0.50, and most values of Flu/(Flu + Pyr) were higher than 0.5 during summer.

Source Characterization of n-Alkanes

Carbon maximum number (C_{max}) and carbon preference index (CPI) are two common indices to identify the potential sources of *n*-alkanes (Simoneit, 1984; Rogge *et al.*, 1993; Chen *et al.*, 2014). Here, the C_{max} values ranged from C_{19}–C_{22} in winter, suggesting the *n*-alkanes were mainly originated from anthropogenic sources. However, the C_{max} was C_{31} in summer with a dominant peak at C_{21} indicated the mixed sources of plant wax and human activities. The definition and calculation of CPI for the whole range (CPI_1), petrogenic (CPI_2) and biogenic (CPI_3) can be found anywhere (Simoneit, 1984; Rogge *et al.*, 1993; Wu *et al.*, 2007; Chen *et al.*, 2014). Because of the C_{max} at small carbon number (i.e., C < 25), some values of CPI_1 and CPI_2 is below 1, and the range for CPI_1 in winter and summer were 0.74–1.25 and 1.11–1.43 with the mean value of 0.98 and 1.27, respectively. It supported the conclusion above that there were obvious biogenic sources in summer. CPI_2 ranged from 0.61–1.93 during the whole sampling with the average value of 0.94 and 1.59 during winter and summer, respectively. The values were close to unity indicated that anthropogenic sources affected the *n*-alkanes, especially during winter. For CPI3, its mean values were 1.10 ± 0.10 and 1.12 ± 0.04, respectively, during winter and summer, suggesting the possible contribution of biological sources to $PM_{2.5}$-bound *n*-alkanes.

Health Risk Assessment

In our study, the concentration of BaP is 0.28 ± 0.14 ng m^{-3} and 0.10 ± 0.08 ng m^{-3} during winter and summer, respectively, which are both below the annual mean values of 2.5 ng m^{-3} and 1 ng m^{-3} established with the Chinese standard (GB3095-2012) and EU standard (2005/69/EC), respectively. Meanwhile, the mean total BaP_{eq} was calculated, and its values were 1.27 ± 0.60 ng m^{-3} and 0.38 ± 0.29 ng m^{-3} during winter and summer, respectively. The average annual value of BaP_{eq} was 0.77 ± 0.63 ng m^{-3}, lower than the value of 0.92 ng m^{-3} in $PM_{2.5}$ in Xiamen during December in 2004 (Wang *et al.*, 2007a) and 1.36 ng m^{-3} in TSP in Xiamen during 2008–2009 (Zhao *et al.*, 2011b), showing a decrease trend along the years. The value was also lower than those reported in other urban areas, such as 2–64 ng m^{-3} in Xi'an (China) (Bandowe *et al.*, 2014), 4.10 ng m^{-3} in Guangzhou (China) (Li *et al.*, 2004), 7.1 ng m^{-3} in Nanjing (China) (Wang *et al.*, 2006) and 1.91 ng m^{-3} in Fuji (Japan) (Takeshi *et al.*, 2004), but close to 0.84 ng m^{-3} in Nanjing during the period of the 2013 Asian Youth Games (Li *et al.*, 2016) and 0.99 ng m^{-3} at Birmingham (Smith and Harrison, 1996).

It has been reported that ILCR values lower than 1×10^{-6}, between 1×10^{-6} and 1×10^{-4} and higher than 1×10^{-4} can be treated as negligible, acceptable and serious, respectively (Liu *et al.*, 2017, and reference therein). In our study, ILCR values were 1.11×10^{-4} and 3.29×10^{-5} in winter and summer, respectively, and the annual value is 7.17×10^{-5} which was much lower than those in Guangzhou (598) (Liu *et al.*, 2015) and Xi'an (1450) (Bandowe *et al.*, 2014). Although the annual value of ILCR is between 1×10^{-6} and 1×10^{-4} indicated that potential carcinogenic effect of PAHs was not serious in Xiamen, it just exceeds 1×10^{-4} in winter suggesting that risk of carcinogenic exposure increased to a concern level in winter.

CONCLUSIONS

In this study, $PM_{2.5}$ samples were collected in winter and summer to analyze the characterization of PAHs and *n*-alkanes, including their seasonal variation and sources, in Xiamen, and the health risk of PAHs was also assessed. It was found that the mass concentrations of $PM_{2.5}$, PAHs and *n*-alkanes were all higher in winter than in summer, and their concentrations were lower than the values detected by previous work, indicating that the air quality in Xiamen has improved since the last decade. BeP, Cor and Phe were the three most abundant components overall in the PAHs, and 5-rings PAHs accounted for 44.1–50.8% of the total PAHs. For *n*-alkanes, the dominant components were rich in high molecular weight *n*-alkanes, and a bimodal distribution was found for this species. Because of the long transport from other areas in winter and the high RH and T in summer, RH and T were the two most important meteorological parameters influencing the concentration of PAHs in summer. Anthropogenic sources impacted the concentrations of PAHs and *n*-alkanes mostly, and fuel combustion was the main contributor to PAHs and *n*-alkanes both in winter and summer in Xiamen. In total, compared with ambient air quality standards and the values reported in other Chinese cities, the values of BaP, BaP_{eq} and ILCR were much lower, suggesting that the health effect of $PM_{2.5}$-bound PAH pollution is not serious in Xiamen. However, the serious risk of carcinogenic exposure, which increases in winter, should be noticed.

The information from our study will be useful for understanding the recent concentration levels and sources of PAHs and *n*-alkanes, and the health risk of PAHs at Xiamen. Although the concentrations of PAHs and *n*-alkanes, and the health risk of PAHs have not been serious recently, the contributions of PAHs and *n*-alkanes are mainly related to anthropogenic sources, especially fuel combustion. Consequently, the management of industry related to fuel combustion and the improvement of the current air quality, especially in winter, should be considered carefully by the local government for future development.

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