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## Changes in concentration, composition and source contribution of atmospheric organic aerosols by shifting coal to natural gas in Urumqi

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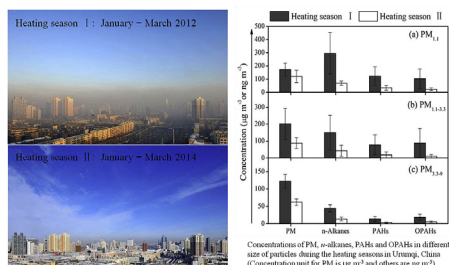
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### HIGHLIGHTS

- *n*-Alkanes, PAHs and OPAHs decreased by 70–90%.
- Coal combustion contribution decreased from 21–75% to 4–21%.
- Number of lung cancer related to PAHs exposure decreased by 73%.
- Replacement of coal by natural gas mitigated air pollution significantly.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Size-segregated aerosols were collected in Urumqi, a megacity in northwest China, during two heating seasons, i.e., before (heating season I: January–March 2012) and after (heating season II: January–March 2014) the project “shifting coal to natural gas”, and determined for *n*-alkanes, PAHs and oxygenated PAHs to investigate the impact of replacement of coal by natural gas on organic aerosols in the urban atmosphere. Our results showed that compared to those in heating season I concentrations of *n*-alkanes, PAHs and OPAHs decreased by 74%, 74% and 82% in heating season II, respectively. Source apportionment analysis suggested that coal combustion, traffic emission and biomass burning are the major sources of the determined organics during the heating seasons in Urumqi. Traffic emission is the main source for *n*-alkanes in the city. Coal combustion is the dominant source of PAHs and OPAHs in heating season I, but traffic emission becomes their major source in heating season II. Relative contributions of coal combustion to *n*-alkanes, PAHs and OPAHs in Urumqi decreased from 21 to 75% in heating season I to 4.0–21% in heating season II due to the replacement of coal with natural gas for house heating. Health risk assessment further indicated that compared with that in heating season I the number of lung cancer related to PAHs exposure in Urumqi decreased by 73% during heating season II due to the project implementation. Our results suggest that replacing coal by clean energy sources for house heating will significantly mitigate air pollution and improve human health in China.

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## 1. Introduction

Coal combustion not only releases inorganic pollutants such as SO<sub>2</sub>, NO<sub>x</sub> and fly ash but also emits carbonaceous material, some of which are toxic such as polycyclic aromatic hydrocarbons (PAHs) (Mittal and Van Grieken, 2001). Burning coal for house heating has been a wide spread activity/practice in north China and is regarded as an important air pollution source in the country. China is the biggest user of coal in the world with 28 billion tons of coal burned in 2015 ([http://www.stats.gov.cn/tjsj/zxfb/201602/t20160229\\_1323991.html](http://www.stats.gov.cn/tjsj/zxfb/201602/t20160229_1323991.html)). A substantial fraction of the combusted coal was used for house heating without efficient emission control, resulting in high PM levels and organic matter concentrations in winter (Bandowe et al., 2014; Wei et al., 2015; Zhang et al., 2015; Zhang and Tao, 2008). A recent study confirmed that air pollution derived from house heating activities have caused human life expectancies to decrease by ~5.5 years in north China because of an increased incidence of cardiorespiratory mortality (Chen et al., 2013).

In order to mitigate the air pollution problems, Chinese government has replaced coal with clean energy to reduce the pollutant emissions from house heating system in many mega-cities. Urumqi (43.9°N and 87.5°E) is the capital city of the Xinjiang Uygur Autonomous Region in northwest China with an area of 14 216 km<sup>2</sup> and a population of 3.5 million (<http://www.wlmtj.gov.cn/tjgb/2014.htm>), which had been one of the cities with the worst air quality during winter in the country (<http://finance.chinanews.com/ny/2014/01-10/5724345.shtml>) (Table S1). Since 2012 a great reform of the energy consumption structure has been performed by “shifting coal to natural gas” for house heating. Over two years Urumqi has raised RMB 25.8 billion to control air pollution and became the first city with all the heating systems based on natural gas and also one of the cities with the most significant improvement of air quality in China (Fig. S1). The project “shifting coal to natural gas” in Urumqi is currently the largest domestic clean energy replacement operation with scale and speed of the construction unprecedented in the country. For example, the project of “shifting coal to natural gas” in Beijing has lasted for 15 years and totally transformed 10 thousands of coal-burning boilers to natural gas-burning ones. In contrast, it only took six months to transform 12.9 thousands boilers into natural-gas burning type in Urumqi. Such a rapid transform from coal to natural gas in the city is an opportunity to assess the effectiveness of coal consumption reduction for air quality improvement. However, up to now only one study conducted such an assessment (Song et al., 2015), which found a significant decline in concentrations of PM<sub>2.5</sub>, inorganic ions and metals in 2013 and 2014 compared with in 2012 and 2011 in Urumqi. To investigate the impact of replacement of coal by natural gas on organic aerosols in the urban atmosphere of Urumqi, here we collected size-segregated samples before and after the “shifting coal to natural gas” project, performed a detailed characterization of organic aerosols and examined the changes in concentrations, composition, and sources of atmospheric aerosols in the city due to the project implementation.

## 2. Methodology

### 2.1. Descriptions for sampling sites and sampling protocol

Size-segregated samples were collected on the rooftop of a three-story building (about 10 m above the ground) on the campus of the Institute of Desert Meteorology at the urban center of Urumqi by using an Anderson 9-stage air sampler (Thermo-electronic Company, USA) with the cutoff points of 0.43, 0.65, 1.1, 2.1,

3.3, 4.7, 5.8 and 9.0 μm at an airflow rate of 28.3 L min<sup>-1</sup> (Fig. S2) (Aggarwal et al., 2013). The samples were collected for about 72 h in each set and twice a month from 1st to 4th and from 15th to 18th during two heating seasons, respectively (January, February and March in 2012 and 2014, and thereafter referred to as heating season I and II, respectively). A total of 6 sets of size-segregated filter samples were collected in each season. Field blank filters were also collected in each season at the beginning and the end of the sampling campaign by mounting blank filters onto the sampler for about 10 min without pumping air. All the samples were collected on pre-baked (450 °C for 8 h) quartz fiber filters (Whatman 42). After sampling, the filter samples were sealed in an aluminum foil bag individually and stored in a freezer at -18 °C prior to analysis.

Because of the project implementation of “shifting coal to natural gas”, natural gas consumption in Urumqi increased rapidly from  $5 \times 10^8$  m<sup>3</sup> in the wintertime of 2011–2012 to  $2.8 \times 10^9$  m<sup>3</sup> in the wintertime of 2013–2014 (Fig. S3), while the coal consumption for house heating decreased by 2.3 million tons in October 2012–April 2013 (after the project) in comparison with that in October 2010–April 2011 (before the project), resulting in the emission reductions of SO<sub>2</sub> and soot by 11.7 and 5.3 thousand tons, respectively ([http://www.360doc.com/content/14/0217/00/15659173\\_353094278.shtml](http://www.360doc.com/content/14/0217/00/15659173_353094278.shtml)).

### 2.2. Sample analysis

In order to reduce the labor-consuming, we grouped the size-segregated filter samples into three size ranges: i.e., <PM<sub>1.1</sub> (the sum of <0.43, 0.43–0.65 and 0.65–1.1 μm), PM<sub>1.1–3.3</sub> (the sum of 1.1–2.1 and 2.1–3.3 μm) and PM<sub>3.3–9.0</sub> (the sum of 3.3–4.7, 4.7–5.8, and 5.8–9.0 μm). In this work PM<sub>9</sub> is defined as the sum of PM<sub>1.1</sub>, PM<sub>1.1–3.3</sub> and PM<sub>3.3–9.0</sub>. Details of sample extraction and derivatization were documented elsewhere (Wang et al., 2006, 2009b). Briefly, a quarter of the filter was extracted with a mixture of dichloromethane/methanol (2:1, v/v) and concentrated to dryness. Then the extracts were reacted with N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) and analyzed by using gas chromatography (HP 7890A, Agilent Co., USA) coupled with mass spectroscopy detector (GC/MS) (HP 5975, Agilent Co., USA). It should be noted that both polar and non-polar compounds in the samples were simultaneously measured, based on the above extraction and derivatization technique (Wang et al., 2006, 2009b). Here we only focused on non-polar organics, i.e., *n*-alkanes, PAHs and OPAHs, and other polar organics will be presented in another paper.

### 2.3. Quality assurance and quality control (QA/QC)

All pre-baked (450 °C for 8 h) glassware were rinsed with methanol, dichloromethane and hexane immediately before using to avoid any contamination. Limit of detection (LOD) and quantification (LOQ) of the target compounds were calculated with signal-to-noise ratios of 3:1 and 10:1, according to the method reported by previous studies (Bandowe et al., 2014; Li et al., 2016). In this work LOD of *n*-alkanes, PAHs and OPAHs are in the range of 0.004–0.049, 0.003–0.008 and 0.01–0.37 ng μL<sup>-1</sup>, respectively. LOQ of them are in the range of 0.0146–0.1629, 0.0095–0.0266 and 0.0242–1.2276 ng μL<sup>-1</sup>, respectively (Table S2). The accuracy of the methodology is 98.2%, which is determined by the error obtained between the mean values of triplicates of a 2 ng μL<sup>-1</sup> standard solution. The precision of the methodology is 3.1%, calculated as the relative standard deviation (%RSD). Target compounds in the field blanks are less than 5% of those in real samples, indicating no serious contamination. Recovery experiment was done by spiking

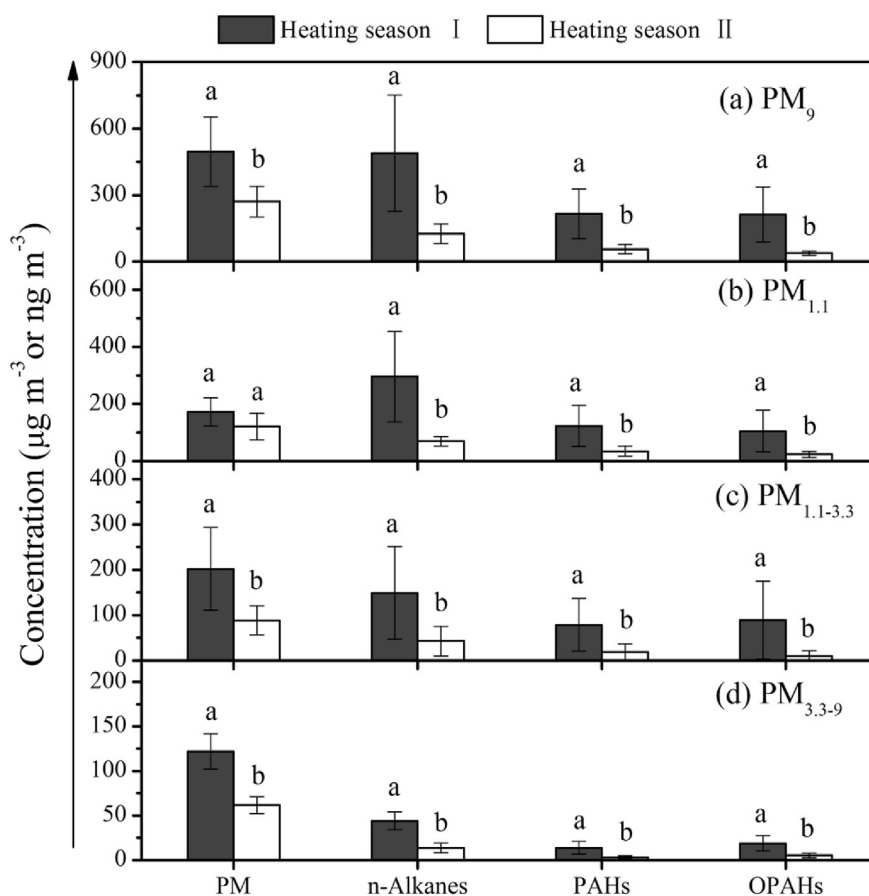
the standard solution onto blank filters ( $n = 3$ ). Amounts of target compounds in the standard solution including *n*-alkanes, PAHs and OPAHs are similar to those in real samples. After being evaporated to dryness, the spiked filters were analyzed in a manner same as the real samples. Recoveries of all the target compounds are 80–120%. Data reported here are all corrected for the blanks but not corrected for the recoveries.

### 3. Results and discussion

#### 3.1. Particle mass (PM) concentrations

Concentrations of PM and organic compounds in the three grouped size ranges are listed in Fig. 1 and Table S3.  $PM_{10}$  (the sum of the three grouped size ranges, i.e., <1.1, 1.1–3.1 and 3.3–9.0  $\mu m$ ) are  $496 \pm 157 \mu g m^{-3}$  (range: 262–685  $\mu g m^{-3}$ ) in heating season I and

$271 \pm 69 \mu g m^{-3}$  (166–364  $\mu g m^{-3}$ ) in heating season II, respectively. Compared with that in heating season I,  $PM_{10}$  concentration in heating season II decreased by 45%, showing a significant decline ( $p < 0.05$ ) (Fig. 1a). We also found similar decrease pattern for the mass concentrations of  $PM_{1.1}$ ,  $PM_{1.1-3.3}$  and  $PM_{3.3-9}$  in heating season II. As seen in Table 1, the annual average concentration of  $SO_2$  significantly decreased by more than 50% from 58  $\mu g m^{-3}$  in 2012 to 25  $\mu g m^{-3}$  in 2014. Meteorological parameters are similar during the two heating seasons. Thus, the significant decreases in PM and  $SO_2$  levels in heating season II can be ascribed to the reduction of coal combustion, which is in agreement with the results from the previous study by Song et al. (2015), who found that meteorological conditions were not the main factors responsible for the significant decrease in  $PM_{2.5}$  concentrations and local pollutant emissions dominated the pollution evolution (Song et al., 2015).



**Fig. 1.** Concentrations of PM, *n*-alkanes, PAHs and OPAHs in different size of particles during the heating seasons in Urumqi, China (Concentration unit for PM is  $\mu g m^{-3}$  and others are  $ng m^{-3}$ ;  $PM_{10}$ : the sum of the three grouped size ranges; The mean values with statistic difference ( $p < 0.05$ ) are labeled with a and b, while those with no statistic difference are labeled with a and a).

**Table 1**

A Summary of average concentrations of  $SO_2$  and meteorological conditions<sup>a</sup> during sampling in Urumqi.

	Heating season I (January–March 2012)	Heating season II (January–March 2014)
$SO_2$ ( $\mu g m^{-3}$ ) <sup>b</sup>	58	25
T ( $^{\circ}C$ ) <sup>c</sup>	$-7.4 \pm 6.3$ (–16–6)	$-10 \pm 7.9$ (–18–12)
RH (%) <sup>c</sup>	$78 \pm 5.9$ (67–90)	$77 \pm 6$ (60–85)
WS (km/h) <sup>c</sup>	$5.0 \pm 2.1$ (3.0–11)	$5.0 \pm 1.6$ (3.0–10)

<sup>a</sup> Meteorological data from <http://www.wunderground.com>.

<sup>b</sup> Data as annual average concentrations and from <http://www.wlmqhb.gov.cn>.

<sup>c</sup> T: Temperature; RH: Relative humidity; WS: Wind speed.

### 3.2. Concentrations and molecular compositions of organic aerosols

#### 3.2.1. *n*-Alkanes

Together with the reduction in PM concentrations, concentrations of airborne particulate organic matters also reduced significantly. As seen in Fig. 1a and Table S3, PM<sub>9</sub>-equivalent concentration of *n*-alkanes varied from 172 to 830 ng m<sup>-3</sup> with an average of 489 ± 262 ng m<sup>-3</sup> during heating season I, and decreased by 74% during heating season II (126 ± 44 ng m<sup>-3</sup>). The total *n*-alkanes are 296 ± 158, 149 ± 103, 44 ± 10 ng m<sup>-3</sup> for PM<sub>1.1</sub>, PM<sub>1.1–3.3</sub> and PM<sub>3.3–9</sub> during heating season I (Table S3), and decreased by 76%, 71% and 68% during heating season II, respectively (Fig. 1b–d). In addition, the highest concentrations of *n*-alkanes in PM<sub>1.1</sub> indicate they tend to stay in fine particles.

*n*-Alkanes derived from fossil fuel combustion are dominated by low molecular weight (LMW) ones (<C<sub>25</sub>), in contrast to *n*-alkanes derived from plant wax, which are dominated by high molecular weight (HMW) ones (>C<sub>25</sub>) (Rogge et al., 1993; Simoneit et al., 2004). Moreover, the carbon preference index (CPI, a mass ratio of *n*-alkanes with odd carbon numbers to those with even carbon numbers) of the anthropogenic *n*-alkanes is close to unity, whereas CPI of the natural *n*-alkanes is far beyond 1.0 (Rogge et al., 1993; Simoneit et al., 2004). In this study, concentrations of LMW *n*-alkanes (C<sub>18</sub>–C<sub>25</sub>) for PM<sub>9</sub>-equivalent decreased by 80% during

heating season II with CPI values ranging from 1.0 to 1.1 and accounted for 82–93% of the total *n*-alkanes during both heating seasons (Table S3). Such molecular distribution pattern in heating seasons suggests that emission from fossil fuel combustion is the dominant source of *n*-alkanes in Urumqi. Fig. 2a shows that relative abundance of PM<sub>9</sub>-equivalent LMW *n*-alkanes decreased by 12–27%, mainly due to the replacement of coal by natural gas for house heating. In contrast, those of the HMW *n*-alkanes increased by 2.0–49% with no significant odd/even predominance. LMW *n*-alkanes are 218 ± 123, 106 ± 78, 30 ± 6 ng m<sup>-3</sup> for PM<sub>1.1</sub>, PM<sub>1.1–3.3</sub> and PM<sub>3.3–9</sub> during heating season I (Table S3), and decreased by 51%, 57% and 33% during heating season II, respectively. Similar to the PM<sub>9</sub>-equivalent, relative abundances of LMW *n*-alkanes decrease and those of HMW *n*-alkanes increase by varying degrees for PM<sub>1.1</sub>, PM<sub>1.1–3.3</sub> and PM<sub>3.3–9</sub> during heating season II (Fig. 2b–d).

#### 3.2.2. PAHs

PM<sub>9</sub>-equivalent concentration of the total 14 PAHs is 216 ± 112 ng m<sup>-3</sup> during heating season I, and decreases by around 74% during heating season II with concentration of 56 ± 21 ng m<sup>-3</sup>, which is comparable to that in Xi'an (Table S3) (Ren et al., 2017). Since Flu, Pyr, Chr, BbF, BkF, BaA, BeP, BaP, IP and BghiP are taken as combustion derived PAHs (COMPAHs) (Kong et al., 2011), and the high ratio (0.9 ± 0.01) of COMPAHs to total PAHs for PM<sub>9</sub>-equivalent

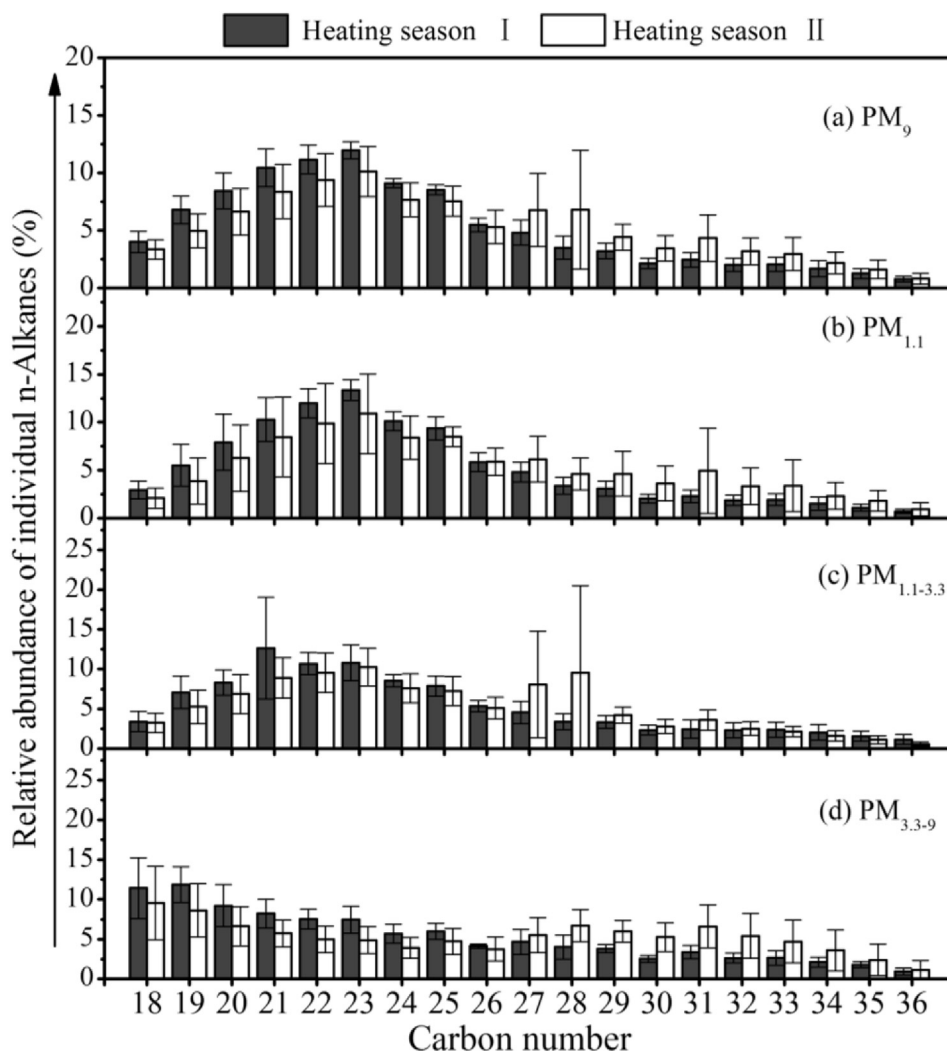


Fig. 2. Relative abundances of *n*-alkanes in different size range of particles during the heating seasons in Urumqi, China (PM<sub>9</sub>: the sum of the three grouped size ranges).

suggests that combustion emission is the dominant source for PAHs in Urumqi city. As for carcinogenic PAHs (C-PAHs), including BaA, Chr, BbF, BkF, BaP, IP and DBA (Kong et al., 2012), their contents are in the ranges of 20–177 and 16–39 ng m<sup>-3</sup> during the two heating seasons, accounting for 43–53% and 40–51% of the total, respectively. The total PAHs are 123 ± 72, 78 ± 58, 14 ± 7 ng m<sup>-3</sup> for PM<sub>1,1</sub>, PM<sub>1,1-3,3</sub> and PM<sub>3,3-9</sub> during heating season I, and decreases by 72%, 76% and 77% during heating season II, respectively ( $p < 0.05$ ) (Fig. 1). The higher concentration of PAHs in PM<sub>1,1</sub> indicates that they tend to stay in fine particle (Zhou et al., 2005), being similar to LMW *n*-alkanes.

Fig. 3 shows the molecular composition of PAHs in each size range. BbF, Flu, Pyr and Chr are the major compounds for PM<sub>9</sub>-equivalent (Fig. 3a), consistent with those reported from other Chinese cities such as Nanjing (Wang et al., 2007) and Baoji (Xie et al., 2009). BbF is the most abundant congener for PM<sub>9</sub>-equivalent during heating season I, accounting for 18 ± 3.0% of the total, but became the second most abundant PAH during heating season II, accounting for 14 ± 2.0% of the total. In contrast, relative abundances of IP and BghiP to the total PAHs increases by about 30%, from 6.4 ± 1.8% and 5.7 ± 2.2% in heating season I to 7.8 ± 1.2% and 6.2 ± 1.1% in heating season II, respectively (Fig. 3a). However, relative abundances of Flu and Pyr to total PAHs between the two heating seasons are almost constant (18 ± 3.5% and 13 ± 3.5% during heating season I versus 19 ± 3.4% and 14 ± 3.0% during

heating season II, respectively) (Fig. 3a). Particulate PAHs emitted from domestic coal burning is enriched in BbF (Wang et al., 2006), while PAHs derived from traffic emissions are dominated by IP and BghiP (Kong et al., 2010; Mostafa et al., 2009) and those from biomass burning smoke are dominated by Flu and Pyr (Zhang et al., 2008a). Therefore, the above changes in relative abundances of BbF, IP, BghiP, Flu and Pyr in the Urumqi samples are mainly attributed to the replacement of coal with natural gas for house heating, resulting in a decrease in coal combustion emission and a relative increase in vehicle exhaust. Similar to PM<sub>9</sub>-equivalent, relative abundance of BbF decreased and that of IP/BghiP increased during heating season II for PM<sub>1,1</sub>, PM<sub>1,1-3,3</sub> and PM<sub>3,3-9</sub>, respectively. However, molecular distributions of PAHs are different for the three size ranges. HMW PAHs (i.e., 5/6 ring PAHs) are enriched in fine particles (e.g. PM<sub>1,1</sub> and PM<sub>1,1-3,3</sub>) (Fig. 3b–c) while LMW PAHs (i.e., 3/4-ring PAHs) are enriched coarse particles (PM<sub>3,3-9</sub>) (Fig. 3d). LMW PAHs such as 3/4-ring ones are much more volatile in comparison with HMW PAHs. After being emitted into the ambient air as fine particles, they can evaporate into the gas phase and re-condense and/or adsorb onto coarse particles (e.g. PM<sub>3,3-9</sub>) (Wang et al., 2009b).

### 3.2.3. OPAHs

Seven OPAHs were detected during both heating seasons in Urumqi (Table S3). PM<sub>9</sub>-equivalent concentrations of the total

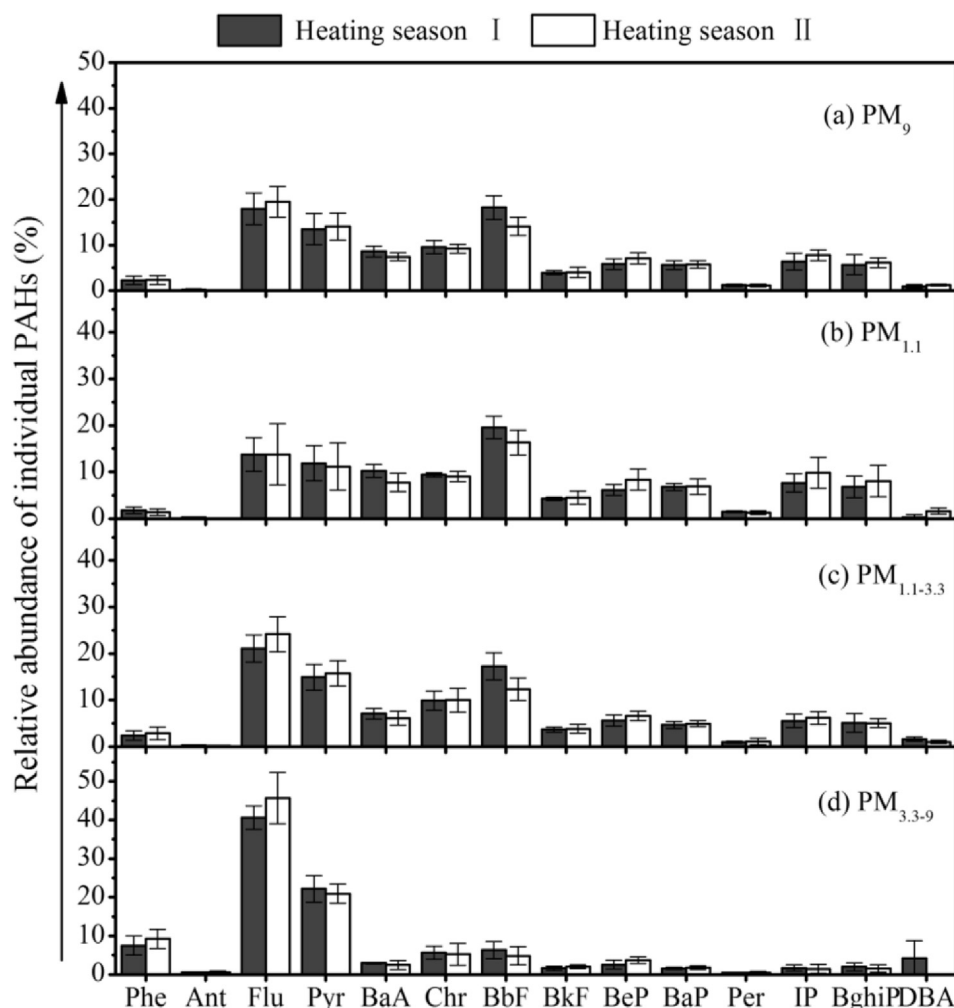


Fig. 3. Relative abundances of PAHs in different size range of particles during the heating seasons in Urumqi, China (PM<sub>9</sub>: the sum of the three grouped size ranges).



OPAHs are  $212 \pm 214 \text{ ng m}^{-3}$  during heating season I, and decreased by around 82% during heating season II ( $39 \pm 10 \text{ ng m}^{-3}$ ) (Fig. 1). OPAHs during heating season II in Urumqi are lower than those ( $3.5\text{--}904 \text{ ng m}^{-3}$ ) in Beijing (Lin et al., 2015) but are still about two times higher than those in other cities in developed countries, such as Umea, Sweden ( $0.9\text{--}7.9 \text{ ng m}^{-3}$ ) (Wingfors et al., 2011) and Augsburg, Germany ( $11 \pm 10 \text{ ng m}^{-3}$  in winter) (Pietrogrande et al., 2011). The total OPAHs are  $105 \pm 72$ ,  $89 \pm 86$ ,  $19 \pm 8 \text{ ng m}^{-3}$  for  $\text{PM}_{1.1}$ ,  $\text{PM}_{1.1-3.3}$  and  $\text{PM}_{3.3-9}$  during heating season I, and decreased by 77%, 89% and 72% during heating season II, respectively (Fig. 1).

There is no significant change in molecular compositions of OPAHs between the two heating seasons (Fig. 4). ATQ is the most abundant OPAH for  $\text{PM}_9$ -equivalent in the heating seasons and accounts for nearly 40% of the total OPAHs, followed by BPyRone and 9-FO (Fig. 4a). HMW molecules such as BPyRone (5-ring OPAH) and BZA (4-ring OPAH) in Urumqi are enriched in the fine mode particles ( $<3.3 \mu\text{m}$ ) (Fig. 4b and c). LMW molecules such as 9-FO (3-ring OPAHs) accounted for  $<9\%$  of the total in  $\text{PM}_{1.1}$  but increased to  $>33\%$  of the total in coarse particles ( $>3.3 \mu\text{m}$ ) (Fig. 4b–d). Vapor pressure of 9-FO is  $1.6 \times 10^{-2} \text{ Pa}$  at 298 K (Hansen and Eckert, 1986), which is three orders of magnitude higher than that ( $2 \times 10^{-5} \text{ Pa}$  at 298 K) of BZA (Inokuchi et al., 1952). Thus, 9-FO may shift to coarse mode aerosols by evaporation from fine particles and subsequent re-adsorbed onto large particles, leading to a large fraction of 9-FO in the coarse mode (Fig. 4d), whereas BZA almost entirely stays in

the fine mode ( $<3.3 \mu\text{m}$ ) due to its low volatility (Fig. 4b and c).

### 3.3. Positive matrix factorization (PMF) analysis for source apportionment

In the current work we performed source apportionment for *n*-alkanes, PAHs and OPAHs by using the EPA PMF 5.0 version. PMF developed by Paatero and Tapper (Paatero and Tapper, 1994) is a multivariate factor analysis model, which decomposes the data matrix into two matrices: factor contributions and factor profiles (Paatero, 1997; Paatero and Tapper, 1994), and has widely been used for the source apportionment of air pollutants (Hu et al., 2010; Lin et al., 2013). The values of *Q* and  $r^2$  are indicative of the agreement of the model fit and the correlation between measured and estimated concentrations, respectively (Comero et al., 2009; Wang et al., 2009a). These values are useful for choosing the proper factor number for modeling. In this work, the results of *Q* value, *Q* theoretical value and average  $r^2$  for each heating season are given in Table S4. From Table S4, it can be seen that the *Q* and *Q* theoretical values are similar with a robust correlation ( $r^2 > 0.98$ ), although the data sets used is limited, indicating that the number of factors chosen is appropriate (Teixeira et al., 2013). We choose four factors for these two heating seasons. But during the process of analyzing the generated results, two profiles are identified as the same sources, which have similar composition and present same

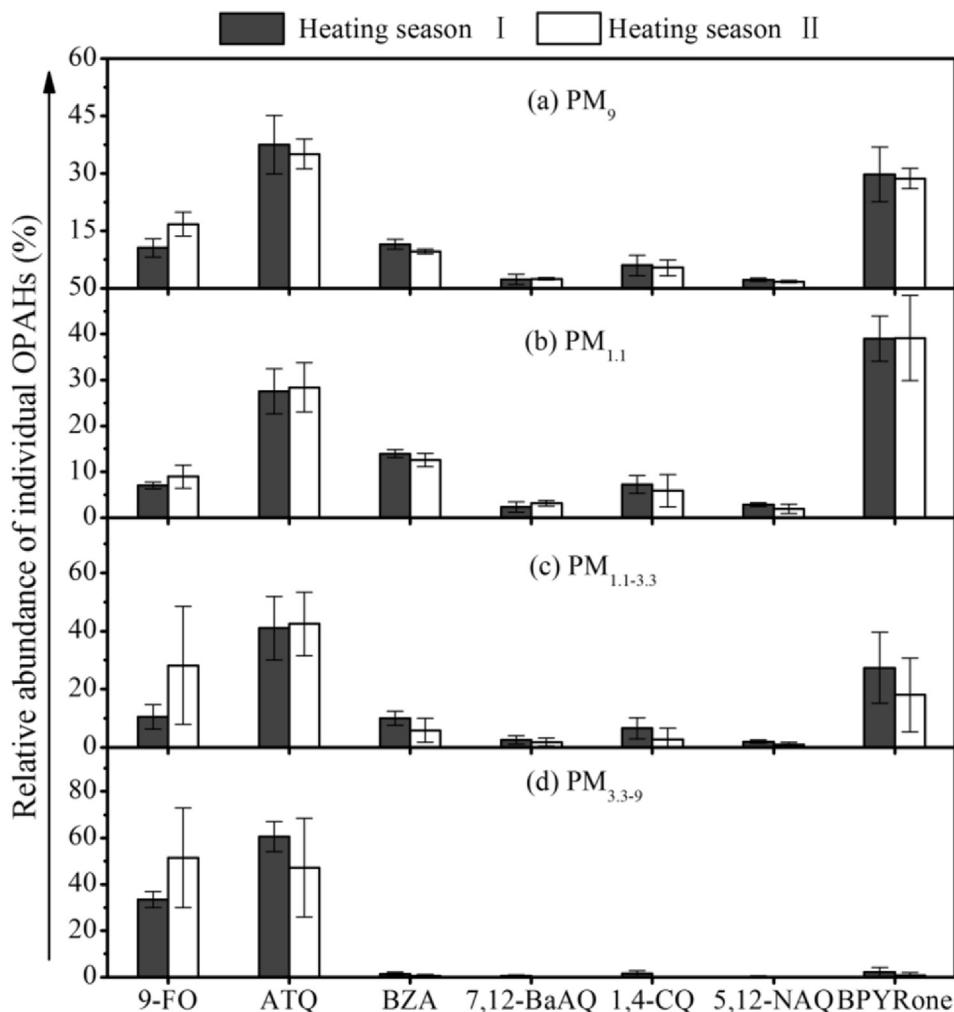


Fig. 4. Relative abundances of OPAHs in different size range of particles during the heating seasons in Urumqi, China ( $\text{PM}_9$ : the sum of the three grouped size ranges).

tracer species. Thus the results pointed out three sources for the organic aerosols in both heating seasons, which are identified as coal combustion, traffic emission, and biomass burning, respectively (Fig. S4).

As shown in Fig. S4a and b, the profile of source #1 in both heating seasons represent the following PAHs: BbF, BeP, Chr and Flu, which are dominant species in coal combustion smokes (Lin et al., 2015; Zhang et al., 2008b). Thus source #1 is identified as coal combustion. The profile of source #2 shows high loadings with IP and BghiP (Figure S4c, d). Since atmospheric IP and BghiP in urban area are largely derived from vehicle exhausts (Kong et al., 2010; Mostafa et al., 2009), the source # 2 is identified as traffic emissions. The profile of source #3 shows high loading with Flu and Pyr (Figs. S4e and f) and is similar to the composition pattern of PAHs emitted from biomass burning (Lin et al., 2015; Zhang et al., 2008a). Thus source # 3 is identified as biomass burning. In heating season I contributions from coal combustion are 21%, 41% and 75% for *n*-alkanes, PAHs and OPAHs, respectively (Fig. 5a, c and e). In contrast, organic aerosols from coal combustion source sharply decreased in heating season II, contributing to 4%, 4% and 21% of the total *n*-alkanes, PAHs and OPAHs, respectively (Fig. 5b, d and f). Traffic emission became the major source for the organic aerosols in heating season II, contributing to 53% of *n*-alkanes, 59% of PAHs and 53% of OPAHs, respectively. Biomass burning is the second

most important source in heating season II, contributing to 43% of *n*-alkanes, 37% of PAHs and 26% of OPAHs, respectively.

### 3.4. Toxicity and health implications

BaP-based toxic equivalent factor (BaPeq) has been frequently utilized to indicate the potential health risk of PAHs to ecosystems and human beings (Agudelo-Castaneda and Teixeira, 2014; Chen et al., 2004; Kong et al., 2011; Liu et al., 2009). As seen in Table 2 the PM<sub>10</sub>-equivalent BaPeq contents are  $21 \pm 11 \text{ ng m}^{-3}$  in heating season I and three times higher than that ( $5.8 \pm 2.0 \text{ ng m}^{-3}$ ) in heating season II. The relative contribution of each PAH to the BaPeq levels is also shown in Table 2. DBA dominates the BaPeq levels in this study, followed by Flu, BbF and IP. The strongest contribution of DBA (even at much lower concentrations) to the carcinogenic potential is due to its high toxicity equivalent factor (TEF) (Kong et al., 2012). Size distribution of BaPeq shows a decreasing trend as  $\text{PM}_{1.1} > \text{PM}_{1.1-3.3} > \text{PM}_{3.3-9}$  (Table 2), suggesting that fine particles are more toxic (Zhang et al., 2015). Moreover, the linear correlation coefficient of PAHs and BaPeq is 0.88–1.0 for the three size ranges of particles, indicating that concentrations of PAHs could be used as an indicator of the toxicity of PAHs (Kong et al., 2012).

Cancer is the leading cause of death in China and is a major

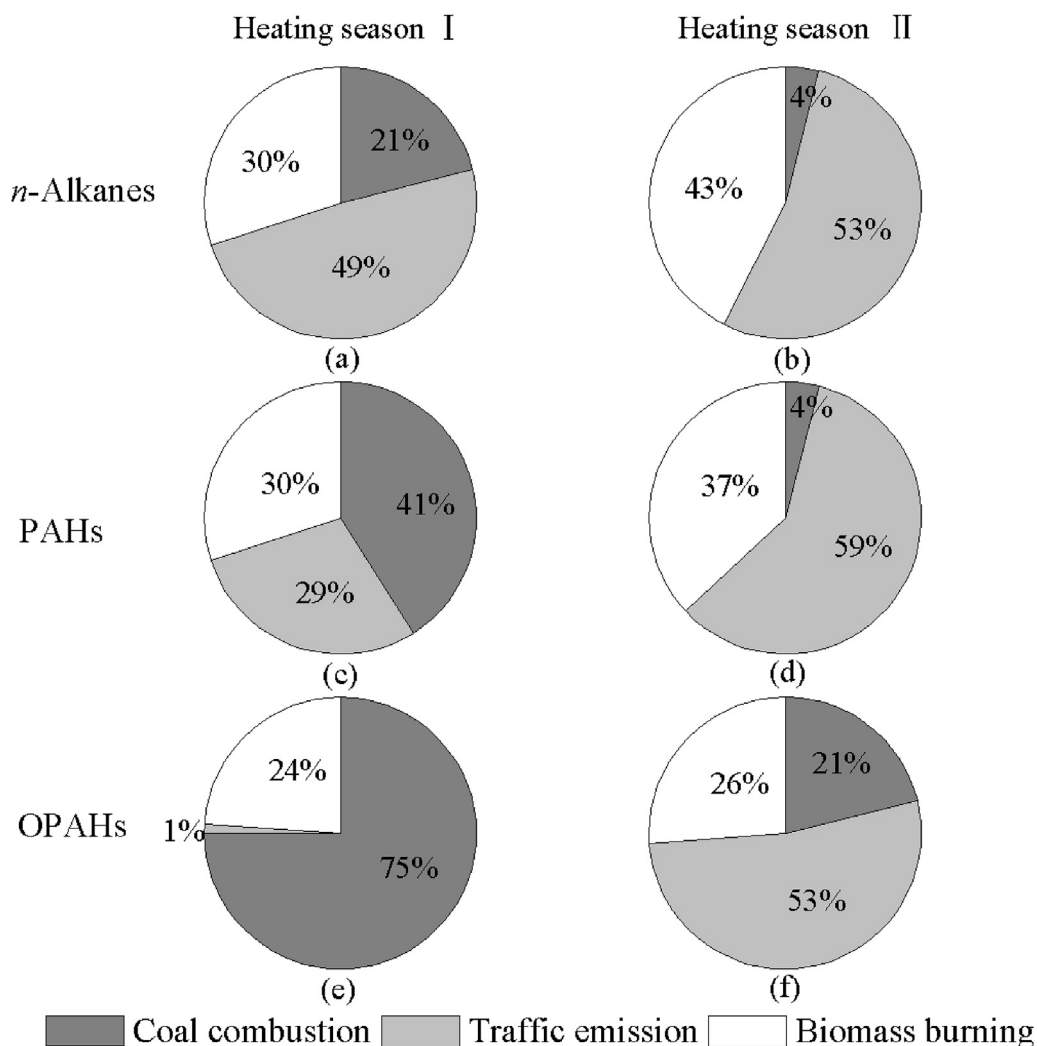


Fig. 5. Source apportionment (%) for *n*-alkanes, PAHs and OPAHs in the heating season in Urumqi, China.

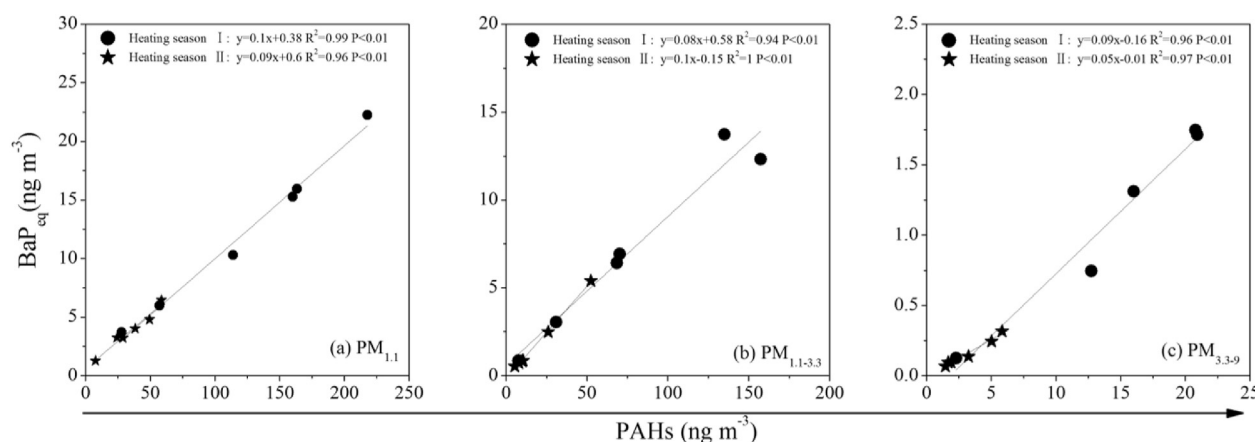
**Table 2**

Relative toxicity of each PAH to BaP (BaP-based toxic equivalent factor, BaP<sub>eq</sub>, %<sup>a</sup>) and the total toxicity concentration (BaP<sub>eq</sub>, ng m<sup>-3</sup>) in the size-segregated samples (For compound abbreviations, see Table S3).

	TEF <sup>b</sup>	Heating season I (January–March 2012)				Heating season II (January–March 2014)			
		PM <sub>1,1</sub>	PM <sub>1,1-3.3</sub>	PM <sub>3,3-9</sub>	PM <sub>9</sub>	PM <sub>1,1</sub>	PM <sub>1,1-3.3</sub>	PM <sub>3,3-9</sub>	PM <sub>9</sub>
Phe	0.0005	0.01 ± 0.006 (0.004–0.02)	0.03 ± 0.01 (0.01–0.04)	0.3 ± 0.1 (0.1–0.4)	0.02 ± 0.01 (0.01–0.03)	0.01 ± 0.008 (0.004–0.03)	0.03 ± 0.02 (0.01–0.07)	0.3 ± 0.1 (0.1–0.5)	0.02 ± 0.01 (0.007–0.04)
Ant	0.0005	0.002 ± 0.001 (0.0003–0.003)	0.003 ± 0.001 (0.001–0.004)	0.02 ± 0.01 (0.01–0.03)	0.002 ± 0.001 (0.0007–0.004)	0.0008 ± 0.0006 (0.0002–0.002)	0.002 ± 0.0007 (0.0009–0.003)	0.02 ± 0.01 (0.008–0.04)	0.002 ± 0.0007 (0.0005–0.003)
Flu	0.05	10 ± 3.6 (4.8–15)	23 ± 6.2 (16–32)	134 ± 34 (101–188)	17 ± 5.3 (8–23)	11 ± 6.7 (1.4–20)	25 ± 7.5 (18–39)	137 ± 51 (87–199)	17 ± 4.9 (9.5–25)
Pyr	0.001	0.2 ± 0.07 (0.07–0.27)	0.33 ± 0.07 (0.19–0.4)	1.5 ± 0.5 (0.9–2.3)	0.25 ± 0.08 (0.1–0.32)	0.2 ± 0.1 (0.03–0.3)	0.3 ± 0.1 (0.2–0.5)	1.3 ± 0.4 (0.7–1.9)	0.3 ± 0.1 (0.1–0.3)
BaA	0.005	0.8 ± 0.2 (0.5–0.9)	0.78 ± 0.19 (0.45–1)	1 ± 0.2 (0.8–1.2)	0.79 ± 0.17 (0.46–0.92)	0.6 ± 0.2 (0.2–0.9)	0.6 ± 0.1 (0.4–0.7)	0.7 ± 0.3 (0.1–1.1)	0.7 ± 0.1 (0.5–0.8)
Chr	0.03	4.2 ± 0.5 (3.2–4.8)	6.7 ± 2.9 (5.2–13)	11 ± 1.4 (9.1–13)	5.3 ± 1.8 (3.7–8.9)	4.2 ± 1.2 (2.1–5.6)	6.2 ± 2 (4.2–9.9)	8.7 ± 4.3 (1.2–13)	4.9 ± 0.7 (3.9–5.8)
BbF	0.05	14 ± 0.7 (13–15)	19 ± 6.7 (16–33)	20 ± 4.8 (10–24)	17 ± 4 (14–25)	12 ± 1.3 (10–14)	13 ± 2 (11–16)	13 ± 5.8 (2.3–19)	12 ± 0.9 (11–14)
BkF	0.1	6.3 ± 0.2 (6.1–6.6)	8.2 ± 2.8 (6.8–14)	10 ± 2.1 (8.2–13)	7.2 ± 1.6 (6.4–11)	6.6 ± 1.3 (5.5–8.3)	8 ± 2.5 (6.2–13)	12 ± 3.5 (7.4–18)	6.9 ± 1.3 (5.8–9.1)
BaP	1	100	100	100	100	100	100	100	100
IP	0.1	11 ± 1.7 (8.9–14)	12 ± 1.7 (9–14)	11 ± 3.7 (4.2–15)	11 ± 1.5 (8.9–14)	14 ± 1.4 (13–17)	13 ± 1.7 (10–15)	7.7 ± 5.1 (0–14)	14 ± 0.8 (12–14)
BghiP	0.02	2 ± 0.4 (1.5–2.7)	2.1 ± 0.6 (1.6–3)	2.4 ± 0.9 (1.3–3.9)	2 ± 0.4 (1.5–2.8)	2.3 ± 0.4 (2–3)	2.1 ± 0.3 (1.5–2.5)	1.6 ± 0.9 (0–2.3)	2.1 ± 0.2 (1.9–2.4)
DBA	1.1	3.7 ± 8.5 (0.04–21)	38 ± 7.8 (30–52)	309 ± 318 (14–908)	18 ± 8.5 (11–33)	26 ± 3.6 (22–32)	23 ± 8.3 (9.7–34)	5.2 ± 8.1 (0–20)	24 ± 2.9 (20–28)
BaP <sub>eq</sub> (ng m <sup>-3</sup> )		12 ± 6.9 (3.7–22)	7.2 ± 5 (0.9–14)	1.3 ± 0.7 (0.1–1.9)	21 ± 11 (4.7–38)	3.8 ± 1.7 (1.3–6.5)	1.8 ± 1.9 (0.5–5.4)	0.2 ± 0.1 (0.1–0.3)	5.8 ± 2 (3.9–9.2)

<sup>a</sup> BaP<sub>eq</sub>(%) = (X<sub>TEF</sub> × X<sub>Con.</sub>) / (BaP<sub>TEF</sub> × BaP<sub>Con.</sub>) × 100, where X<sub>TEF</sub> and X<sub>Con.</sub> are the toxicity and the mass concentration of a specific PAH in the size-segregated samples.

<sup>b</sup> BaP-based toxicity equivalent factors (TEF) are referred to Larsen and Larsen (1998) and Chen et al. (2004).



**Fig. 6.** Linear fit regression for BaP<sub>eq</sub> and PAHs in the three size ranges of particles during the heating seasons in Urumqi, China.

**Table 3**

The population attributable fraction (PAF) and lung cancer incidence rate induced by PAHs exposure (LCR<sub>PAHs</sub>) based on the average BaP<sub>eq</sub> in this work.

Site	Time	BaP <sub>eq</sub> (ng m <sup>-3</sup> ) <sup>a</sup>	LCR <sup>b</sup>	PAF <sup>c</sup>	LCR <sub>PAHs</sub> <sup>d</sup>	The number of lung cancer <sup>e</sup>	Reference
The whole of China	2003	7.64	40.7 × 10 <sup>-5</sup>	1.60%	0.65 × 10 <sup>-5</sup>		Zhang et al., 2009
Urumqi	January–March 2012 (Heating season I)	21		4.40%	1.8 × 10 <sup>-5</sup>	63	This work
Urumqi	January–March 2014 (Heating season II)	5.8		1.20%	0.49 × 10 <sup>-5</sup>	17	This work

<sup>a</sup> See Table 2.

<sup>b</sup> Lung cancer incidence rate; Data from Zhang et al., 2009.

<sup>c</sup> Population attributable fraction; Values of PAF in our study were calculated by the PAF from Zhang et al., 2009 and based on BaP<sub>eq</sub> in our study.

<sup>d</sup> Lung cancer incidence rate induced by PAHs exposure; LCR<sub>PAHs</sub> = LCR × PAF.

<sup>e</sup> Based on the hypothesis of the population of 3.5 million constant for Urumqi in our sampling periods.



public health problem, with lung cancer being the most common incident and the leading cause of cancer death (Chen et al., 2016). Cause of human lung cancer has been found to be associated with the inhalation exposure to PAHs (Bai et al., 2009; Chen and Liao, 2006; Xia et al., 2013; Zhang et al., 2009). In this study, we evaluated lung cancer risk for the Urumqi population caused by inhalation exposure to PAHs based on the BaP<sub>eq</sub> (Table 3). We used values of lung cancer incidence rate (LCR), population attributable fraction (PAF), and lung cancer incidence rate induced by PAHs exposure (LCR<sub>PAHs</sub>) from Zhang et al. (Zhang et al., 2009). As seen in Table 3, we predict that the PAF and LCR<sub>PAHs</sub> would decrease from 4.4% to  $1.8 \times 10^{-5}$  in heating season I to 1.2% and  $0.49 \times 10^{-5}$  in heating season II, respectively. If the population of Urumqi remains the same as 3.5 million in our sampling periods, the number of lung cancer related to PAHs exposure decreased by 73% with from 63 persons in heating season I to 17 persons in heating season II. Although these studies are based on a limited number of samples, the results clearly indicate that we will benefit greatly by using clean energy, such as “shifting coal to natural gas” in Urumqi.

#### 4. Conclusion

Size-resolved airborne particulate *n*-alkanes, PAHs and OPAHs in Urumqi atmosphere during the two heating seasons were determined in order to examine the relationship between substantial changes in the energy sources and changes in the levels and composition of organic aerosols. Our results showed that *n*-alkanes, PAHs and OPAHs are  $489 \pm 262$ ,  $216 \pm 112$  and  $212 \pm 124$  ng m<sup>-3</sup> for PM<sub>2.5</sub>-equivalent during heating season I, and decreased by 74%, 74% and 82% during heating season II, respectively. These compounds are enriched in PM<sub>1.1</sub>. Molecular composition analysis showed that relative abundances of LMW *n*-alkanes and BbF decreased and those of HMW *n*-alkanes, IP and BghiP increased during heating season II compared with those in heating season I due to the replacement of coal with natural gas for house heating. Source apportionment showed that *n*-alkanes, PAHs and OPAHs contributed by coal combustion decreased from 21%, 41% and 75% in heating season I to 4%, 4% and 21% in heating season II, respectively. The health risk assessment further suggested that due to the implementation of the project of “shifting coal to natural gas”, the number of lung cancer related to PAHs exposure in Urumqi decreased by 73% from 63 persons in heating season I to 17 persons in heating season II, demonstrating a significant improvement of human health by using natural gas for house heating in the city.

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#### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2016.10.053>.

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