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# Characteristics and sources of aerosol pollution at a polluted rural site southwest in Beijing, China

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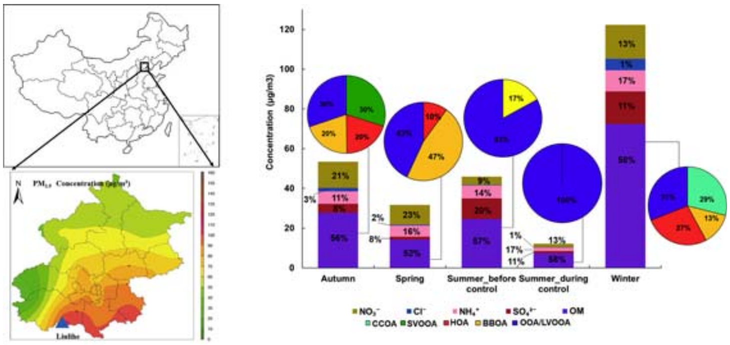
## HIGHLIGHTS

- Four season organic aerosol mass spectra is obtained and source apportionment is conducted in the rural area in Beijing.
- Residential solid fuel burning is the most important source of aerosol pollution in the rural area of Beijing.
- Results focusing on urban Beijing might have underestimate the contribution from residential emissions.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Annual average $PM_{2.5}$ concentration in south Beijing was 30% and 40% higher than the whole Beijing city in 2015 and 2016, respectively. Few studies have been conducted to investigate what leads to the characteristics and sources of heavy pollution in the south rural area of Beijing. This study conducted an observation with Aerosol Chemical Speciation Monitor (ACSM) at a southwest rural site (Liulihe) in Beijing during 2014–2016, to investigate the seasonal aerosol characteristics and their sources. Positive matrix factorization (PMF) algorithm was used to distinguish different components of organic aerosol measured by ACSM. Biomass burning is an important pollution source, mainly due to the open burning after harvest season in autumn, regional transport in spring, and local residential biofuel use in winter. Coal consumption is the largest primary organic aerosol source in winter. Heavy duty diesel trucks contributed significantly to organic aerosol at night-time in the rural area. Results of this study show residential solid fuel burning is the most important source of aerosol pollution in the rural area of Beijing and the results focusing on urban Beijing might have underestimate the contribution from residential emissions in the Beijing-Tianjin-Hebei region.

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## 1. Introduction

As the capital city of China, Beijing burdens 21.5 million residents, 5.3 million vehicles (Beijing Municipal Bureau of Statistics, 2015) and the corresponding high anthropogenic emissions of air pollutants. As a result, Beijing has experienced serious air pollution and associated health impact in recent years (Zheng et al., 2016; Zheng et al., 2015). In 2013, Chinese government released the Air Pollution Prevention and Control Action Plan (2013–2017) to improve the air quality, especially in key regions including Beijing-Tianjin-Hebei area. Annual average $PM_{2.5}$ concentration in Beijing is targeted from 88 $\mu g/m^3$ in 2013 to 60 $\mu g/m^3$ in 2017 (H. Zhang et al., 2016; J.K. Zhang et al., 2016). However, the annual average $PM_{2.5}$ concentration in Beijing was 81 $\mu g/m^3$ with 179 polluted days in 2015. The average concentration decreased to 73 $\mu g/m^3$ with 169 polluted days in 2016, still facing challenges to reach the goal (Beijing Municipal Environmental Protection Bureau, 2016; Beijing Municipal Environmental Protection Bureau, 2017). Air pollution in Beijing exhibits a remarkable spatial distribution characteristic of much higher concentration in south and lower concentration in north. Annual average $PM_{2.5}$ concentration in south Beijing was 30% higher than the whole city in 2015 (Beijing Municipal Environmental Protection Bureau, 2016). This spatial distribution continued in 2016, with annual average $PM_{2.5}$ concentration of the southwest site 40% higher than the whole city (Beijing Municipal Environmental Protection Bureau, 2017). To reduce the pollution concentration of the whole city, much more efforts need to be made on investigation of pollution characteristics and sources of the pollution in the south area of Beijing. Considering that eight out of the most polluted ten cities in China in 2015 located in south of Beijing (Ministry of Environment Protection, 2016), regional transport contributed significantly to pollution in Beijing (Beijing Municipal Environmental Protection Bureau, 2014; Ji et al., 2014; Sun et al., 2015a, 2015b; Wang et al., 2015). As a result, south area of Beijing might be impacted by the regional transport significantly. In addition to the regional transport impact, local emission also needs more investigation. Since the most polluted area in south of Beijing are rural area, with different anthropogenic activities from urban area, the local emission sources might be different. For example, household solid fuel use which was underestimated is proved to be a major ambient pollution source recently (Liu et al., 2016).

Field observations have been carried out at urban sites in Beijing to explore the pollution characteristics and sources. The design of Aerosol Chemical Speciation Monitor (ACSM) enables it easier to conduct long-term continues monitoring of non-refractory particulate matter with aerodynamic diameters smaller than 1 μm ($NR\text{-}PM_1$), providing an advanced technique to look into pollution sources and process (Ng et al., 2011). For example, pollution was characterized by high contribution of secondary species and oxygenated organic aerosol (OOA) from regional scale in summer in Beijing (Sun et al., 2012). In winter, coal combustion organic aerosol (CCOA) was resolved in several studies at urban sites (Sun et al., 2013; H. Zhang et al., 2016; J.K. Zhang et al., 2016; Elser et al., 2016). With the implementation of Air Pollution Prevention and Control Action Plan, coal has been replaced by gas energy for heating season in downtown of Beijing. The contribution from CCOA to $NR\text{-}PM_1$ reduced from 17% (Sun et al., 2013) in 2011–2012 to 12% in 2014 (H. Zhang et al., 2016; J.K. Zhang et al., 2016).

However, most of the previous studies were carried out at urban sites. These results are disable to explain the reason why pollution is much more severe in the rural area than the urban area in Beijing. Few investigations have been conducted in the rural area. To have an insight of the pollution characteristics and sources in the rural area, we conducted continuous sampling in four seasons at a rural site southwest in Beijing, which is the most polluted site in 2016 among all the sites in Beijing (Beijing Municipal Environmental Protection Bureau, 2017). The organic aerosol mass spectra were obtained by ACSM, providing results of pollution characteristics and sources in the most polluted area in Beijing.

## 2. Field observation and analysis methods

### 2.1. Field observation site and sampling methods

The observation was conducted in four seasons (October 22nd to November 11th, 2014; March 30th to April 30th, 2015; August 11th to September 7th, 2015; December 5th, 2015 to January 7th, 2016) from 2014 to 2016.

The sampling site was located at a rural site (Liulihe site, 116°2′E, 39°36′N, Fig. S1) in the southwest in Beijing. The site was located on the border of Beijing and Hebei province. This site is in Fangshan District, which is a heavy polluted region in Beijing. The sampling height was 3 m, on the roof of a one-storey sampling station which was 500 m away from the traffic road. All the time discussed in this article is local time.

The sampling instruments (Table S1) included weather station (WXT520, VAISALA, Finland), gaseous pollutants monitors (API100/200/400E, Teledyne, USA) and $PM_{2.5}/PM_{10}$ monitors (TEOM1405/1400a, Thermo Scientific, USA). Time resolution of these instruments is 5 min and averaged into 1 h.

ACSM was used to measure species including organic matter (OM), nitrate, sulfate, ammonium and chloride of $NR\text{-}PM_1$ (Ng et al., 2011). The ACSM was calibrated with Ionization Efficiency (IE), which was determined with SMPS. DMA (Differential Mobility Analyzer) was generated to select $NH_4NO_3$ particles with a size of 300 nm mobility diameter and counted by CPC (Condensation Particle Counter). The results were compared with ACSM data.

### 2.2. Back trajectory analysis and satellite data

HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model was used to analyze the regional transport. Trajstat, a Geographical Information System (GIS)-based software into which the HYSPLIT model was loaded (Wang et al., 2009) and used to calculate the back trajectory. The model was run every 24 h and four staring height were set to be 300 m, 500 m, 1000 m, and 1500 m above sea level. To investigate the air mass transport impact, the back trajectories were clustered. Euclidean distance mode was selected. The method is described in Wang et al. (2009). The transport differences at different heights are provided in supplement to explain the reason for height selection. Fire point maps were obtained from https://firms.modaps.eosdis.nasa.gov/firemap/. Planetary boundary layer (PBL) height data were obtained from the Global Data Assimilation System (GDAS) model (http://www.ready.noaa.gov/READYamet.php).

### 2.3. Data calibration and PMF analysis

Collection efficiency (CE) was used to calibrate the ACSM data to compensate the particle loss. Based on the monitoring site condition, the following formula was used for calibration (Middlebrook et al., 2012).

$$CE = \max(0.45, 0.0833 + 0.9167 \times ANMF)$$

ANMF is characterized by the ammonium nitrated mass fraction (ANMF). Ionization Efficiency (IE) was determined with SMPS. DMA (Differential Mobility Analyzer) was generated to select $NH_4NO_3$ particles with a size of 300 nm mobility diameter and counted by CPC (Condensation Particle Counter). The results were compared with ACSM data.

The $NR\text{-}PM_1$ concentration $(OM + SO_4^{2-} + NO_3^- + NH_4^+ + Cl^-)$ measured by ACSM tracks well with the $PM_{2.5}$ concentration measured by TEOM during the four seasons (Fig. S2). All the correlation coefficients ($R^2$) are above 0.60. It is noticed $R^2$ are lowest in spring and the slope is 0.32, relatively much lower than other seasons and other studies (Sun et al., 2012; Aurela et al., 2015). It is caused by the dust events in