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Six sources mainly contributing to the haze episodes and health risk assessment of $PM_{2.5}$ at Beijing suburb in winter 2016



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ABSTRACT

Aiming to a better understanding sources contributions and regional sources of fine particles, a total of 273 filter samples (159 of PM2.5 and 114 of PM1.0) were collected per 8 h during the winter 2016 at a southwest suburb of Beijing. Chemical compositions, including water soluble ions, organic carbon (OC), and elemental carbon (EC), as well as secondary organic carbon (SOC), were systematically analyzed and estimated. The total ions concentrations (TIC), OC, and SOC of $PM_{2.5}$ were with the following order: 16:00-24:00 > 08:00-16:00 > 00:00-08:00. Since primary OC and EC were mainly attributed to the residential combustion in the night time, their valley values were observed in the daytime (08:00-16:00). However, the highest ratio value of SOC/OC was observed in the daytime. It is because that SOC is easily formed under sunshine and relatively high temperature in the daytime. Positive matrix factorization (PMF), clustering, and potential source contribution function (PSCF) were employed for apportioning sources contributions and speculating potential sources spatial distributions. The average concentrations of each species and the source contributions to each species were calculated based on the data of species concentrations with an 8 h period simulated by PMF model. Six likely sources, including secondary inorganic aerosols, coal combustion, industrial and traffic emissions, road dust, soil and construction dust, and biomass burning, were contributed to PM25 accounting for 29%, 21%, 17%, 16%, 9%, 8%, respectively. The results of cluster analysis indicated that most of air masses were transported from West and Northwest directions to the sampling location during the observation campaign. Several seriously polluted areas that might affect the air quality of Beijing by long-range transport were identified. Most of air masses were transported from Western and Northwestern China. According to the results of PSCF analysis, Western Shandong, Southern Hebei, Northern Henan, Western Inner Mongolia, Northern Shaanxi, and the whole Shanxi provinces should be the key areas of air pollution control in China. The exposure-response function was used to estimate the health impact associated with $PM_{2.5}$ pollution. The population affected by $PM_{2.5}$ during haze episodes reached 0.31 million, the premature death cases associated with PM2.5 reached 2032. These results provided important implication for making environmental policies to improve air quality in China.

1. Introduction

In the past decades, the quantity of motor vehicles and energy consumption have an escalating increase by reasons of the rapid development of economy and industrialism and urbanization in China (Zhang et al., 2013). Meanwhile, air pollution is becoming increasingly severe with the progress and development of the human society. Pollution of fine particulate matter ($PM_{2.5}$) is one of the serious air pollution problems in northern China, especially in winter. $PM_{2.5}$ has been recognized as the key air pollutant of reducing visibility and harming human health (Tao et al., 2014; Chen et al., 2015; Fu and Chen, 2017). Several recent studies have indicated that many adverse health outcomes such as respiratory and cardiovascular morbidity and mortality are related with long-term exposure to highly PM (Hoek et al.,

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2013; Weichenthal et al., 2014). The outdoor PM has been classified as one of carcinogens by the International Agency for Research on Cancer (IARC) which is a specialized cancer agency of the World Health Organization (WHO). PM_{2.5} usually have a long atmospheric lifetime of days, which is beneficial to long-range transport in atmosphere and to deposition toward remote areas. During the long-range transport, PM_{2.5} carries abundant anthropogenic pollutants and affects the global ecosystems (Mahowald, 2011). Those adverse health and environment problems are attributed to the harmful components in PM_{2.5}, including water soluble ions, OC, EC, and trace elements etc. (Zhang et al., 2013; Shi et al., 2014; Li et al., 2015).

To identify the source contributions to PM_{2.5}, the technique of source apportionment has been widely used around the world and also increasingly applied for the past decade in China (Hueglin et al., 2005; Chen et al., 2010; Tao et al., 2012; Zhang et al., 2013; Cao et al., 2011a, 2011b). Through statistical interpretation of ambient measurement, the contribution levels of different sources are quantitatively estimated by using receptor models. Generally, scholars identified the possible sources of PM2.5 to be traffic and industrial emissions, dust storms, coal combustion, secondary inorganic aerosols, and biomass burning (He et al., 2001; Song et al., 2006; Tao et al., 2014; Chen et al., 2017). Secondary inorganic aerosols were divided into secondary sulfate, secondary nitrate, and secondary ammonium by using the chemical mass balance receptor model (CMB) (Zheng et al., 2005). Unfortunately, the source profiles must be provided when the source apportionment is concluded with CMB model. The measurements of source profiles are time-consuming and difficult work. A convenient and efficient analysis method is provided by positive matrix factorization (PMF) model. The PMF model developed by the Environmental Protection Agency (EPA) of USA is an easy and powerful tool in using factor analysis to identify the possible source contributions without the source profiles. Many cities such as Pittsburg (Zhou et al., 2004), Hong Kong (Lee et al., 1999), Chengdu (Tao et al., 2014), and Beijing (Wang et al., 2008; Song et al., 2006) have successfully applied the PMF model to identify the major sources and apportion source contributions. For example, Tao et al. (2014) identified six major sources including secondary inorganic aerosols, coal combustion, biomass burning, iron and steel manufacturing, Mo-related industries, and soil dust, and accounting for 37 \pm 18, 20 \pm 12, 11 \pm 10, 11 \pm 9, 11 \pm 9, and $10 \pm 12\%$, respectively, to PM_{2.5} in Chengdu (an inland city in southwest China). This model provides a feasible alternative for the place lacked the local source profiles.

Several recent studies have attempted to identify the major sources of $PM_{2.5}$ based on the daily chemical compositions of $PM_{2.5}$ in different seasons in Beijing (Song et al., 2006; Yu et al., 2013; Zhang et al., 2013). Some sources and their contribution rates to $PM_{2.5}$ were simulated by PMF model. However, most of their studies focused on the chemical characterization and sources of $PM_{2.5}$ from seasonal perspective. In fact, the effect of human activities on the chemical compositions of $PM_{2.5}$ at different time periods of the day exhibits large diurnal variability. Better understanding of chemical characterization and source apportionment of $PM_{2.5}$ in haze episodes based on the diurnal variation will be high importance for air pollution control and human health protection.

In addition to source apportionment, the regional distribution of emission sources was also concerned by scholars. The potential source contribution function (PSCF) analysis has been successfully applied to identify potential regional sources (Polissar et al., 2001; Zhang et al., 2013; Li et al., 2015). The PSCF was computed by considering the backward trajectories and measured atmospheric pollutant concentrations using a geographical information system-based software, TrajStat (Wang et al., 2009). It is a conditional probability that air masses are probably responsible for pollutant concentrations higher than the criterion level when air masses arrive at the receptor site (Li et al., 2015). The operation of PSCF analysis was expound in Section 2.4.3.

Moreover, the health risk assessment associated with PM2.5 is also one of the hot topics in air pollution studies. Generally, PM_{2.5} has a higher adverse effect on human body than coarse particles (PM₁₀), which is because PM2.5 can pass to the breathing system into human tissue and has systemic effects (Yin et al., 2017; Li et al., 2017; Miri et al., 2016, 2017). Epidemiological studies show PM_{2.5} exposure can increase the risks of mortality and morbidity related to respiratory and cardiopulmonary diseases (Hammitt and Zhou, 2006; Xie et al., 2009; Liu et al., 2010; Huang and Zhang, 2013). Toxicological studies also suggest that PM2.5 exposure can cause platelet activation and inflammation in lungs regions, which is closely related to increased risks of cardiovascular diseases and lung cancer (Frampton et al., 2012; Lippmann, 2014; Van Winkle et al., 2015; Miri et al., 2018), Heavy elements and polycyclic aromatic hydrocarbons (PAHs) in particles can also cause health risks to human body (Wang et al., 2018; Xu et al., 2016). Therefore, scholars hoped to estimate the health risk and help governments make environmental policies (Kan and Chen, 2004; Zhang et al., 2007; Li et al., 2013; Yin et al., 2015; Du and Li, 2016). Beijing as one of the polluted mega-cities in China, it is of great importance to evaluate and analyze the risk and cost of the health effects associated with PM_{2.5} pollution in Beijing.

In this study, we continuously collected daily $PM_{2.5}$ and $PM_{1.0}$ samples with an eight-hour cycle during the winter in 2016 at a suburb site in Beijing. A suite of chemical species in particles including the major water soluble ions, OC, and EC were measured and analyzed. Furthermore, the major sources and source contributions of $PM_{2.5}$ were identified by using the PMF model. The potential source regions of chemical species in $PM_{2.5}$ were identified by employing the potential source contribution function analysis. Furthermore, the health exposure-response function was used to estimate the health risk due to $PM_{2.5}$ pollution. This study will provide significant information in making environmental policies and air management framework to reduce the current pollution level of $PM_{2.5}$ and improve the air quality in China.

2. Methodology

2.1. Site description

Beijing is the capital of China and located on the northern edge of the North China Plain. The total population of Beijing was 21.73 million in 2016 (Beijing Statistics Yearbook). The total motor vehicles increased to 5.72 million in 2016 compared to 5.20 million in 2012, and energy consumption was equivalent to 69.62 million tons of standard coal (Beijing Statistics Yearbook). The annual average concentration of $PM_{2.5}$ was 73 $\mu g\,m^{-3}$ in Beijing in 2016 and still fell short the national standard (35 $\mu g\,m^{-3}$), although all levels of government had made great efforts to reduce the levels of PM2.5. The sampling station was located at southwest of Beijing (116°07' E, 39°43' N). The PM25 transported from the southern of Hebei province where is regarded as one of the most polluted regions in China, would be collected when air masses pass through this sampling location. The samples were collected on the roof (21 m above ground) of an office building. There are few high-rise buildings within 100 m around the sampling station and no main pollution sources exist nearby. Thus, this observation could be considered as typical of the long-range transport pollution in Beijing.

2.2. Sample collection

Daily $PM_{2.5}$ and $PM_{1.0}$ samples were collected using two high volume air samplers (TH-1000F; Wuhan Tianhong Instruments Co. Ltd., China). The flow rate of samplers was calibrated before the start of the sampling campaign. The fine particles were collected on quartz fibre filters (Pallflex 2500 QAT-UP 8 × 10 in.) at a flow rate of $1.05 \text{ m}^3 \text{ min}^{-1}$. The quartz filters were baked at 500 °C for 3 h prior to sampling to remove carbonaceous impurities. The collected samples were stored

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