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Comparative analysis of chemical composition and sources of aerosol particles in urban Beijing during clear, hazy, and dusty days using single particle aerosol mass spectrometry

Li Ma [a, 1], Mei Li [a, 1], Hefeng Zhang [b], Lei Li [a], Zhengxu Huang [a], Wei Gao [a], Duohong Chen [c], Zhong Fu [d], Huiqing Nian [e], Lilin Zou [e], Jian Gao [b], Fahe Chai [b], Zhen Zhou [a, *]

[a] Institute of Atmospheric Environment Safety and Pollution Control, Jinan University, Guangdong 510632, China
[b] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science, Beijing 100012, China
[c] Guangdong Environmental Monitoring Center, State Environmental Protection Key Laboratory of Regional Air Quality Monitoring, Guangzhou 510308, China
[d] School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China
[e] Guangzhou Hexin Analytical Instrument Company Limited, Guangzhou 510530, China

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ABSTRACT

A single particle aerosol mass spectrometer (SPAMS) was employed to characterize the particles in urban Beijing in the spring of 2011. The whole sampling time was divided into clear, hazy, and dusty day periods. Based on SPAMS results, the chemical composition of size-resolved particles in five major classes, including K-rich, carbonaceous, industry metal, dust, and Na-rich particles, and their corresponding sources were compared during clear, hazy, and dusty days. Air mass back trajectories are used to identify likely source regions for each period. Under stagnant meteorological conditions, local and regional emissions are a key factor in haze formation. K-rich and carbonaceous particles dominated under all conditions (25–54%), with higher contributions (23% and 29%) from industrial metal particles during hazy days, which could be attributed to the emission of biomass burning, coal combustion, and vehicle exhaust during all periods, whereas industrial metal processes are also a likely source for K-rich particles during hazy days. The aging of particles was observed over the whole sampling period, as evidenced by the presence of secondary species in all particle classes. Industrial metal particles possibly originate from industrial metal processes, vehicle exhaust, and coal combustion etc, while road and soil dust is also a possible source during dusty days. Dust particles increased greatly (17%) in a dust storm event with a coarse mode distribution, which are mainly associated with the mineral dust from deserted regions, whereas they could originate from local dust and coal combustion during non-dusty days. Last, Na-rich particles are mainly associated with industrial metal particles, coal, and fuel combustion during hazy days, and mineral dust during dusty days, respectively. Sea salt is a possible source of Na-rich particles under all conditions. Our analysis could shed a light on the understanding of the different source apportionment of aerosol particles under different meteorology/pollution conditions.

1. Introduction

Beijing, the capital city in China, is experiencing a rapid increase of population and transportation, with a population of more than 17 million people and five million vehicles in 2011 (http://www.stats.gov.cn). Over 60% of the geographical area of Beijing is a mountainous area, but the activities of production and service are mainly in the plain region. In addition, many new tall buildings can hinder the dispersion of particulate matter (PM) pollutants. The above mentioned factors led to the high intensity of pollutant emissions, the weak air self-purification of ecological system, and the lack of environmental bearing capacity for PM pollutants. PM is a mixture of solid and liquid particles suspended in the air, which not only has a significant impact on air quality and visibility (Kanakidou et al., 2005; Prather, 2009), but also is detrimental to human health (Buonanno et al., 2013; Pope et al., 2006; Pope III et al., 2002; Wu et al., 2013b).

* Corresponding author. Tel.: +86 20 82071910; fax: +86 20 82071902.
E-mail address: zhouzhen@gig.ac.cn (Z. Zhou).
[1] The authors contributed equally to this work.

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Currently air quality standards are based on the size range of PM (PM_{10} or $PM_{2.5}$) (WHO, 2005). In China the yearly limit for $PM_{2.5}$ has been set as 35 μg/m^3 since the beginning of 2013 (http://kjs.mep.gov.cn/). Currently, $PM_{2.5}$, together with PM_{10} and the gases of SO_2, CO, NO_2, and O_3 are routinely monitored in 367 cities (http://www.pm25s.com/). Although tremendous efforts have been made to control air pollution in Beijing, severe air pollution still occurred. In 2014 Beijing Environmental Protection Bureau officially announced that annual median mass concentration of $PM_{2.5}$ in 2013 was 89.5 μg/m^3, which was much higher than the yearly limit of $PM_{2.5}$. The exposure to PM pollution has been linked to increased mortality and morbidity worldwide (Buonanno et al., 2013; Chen et al., 2012; Pope et al., 2006; Vanos et al., 2014). Some studies have investigated the adverse effects of chemical components in $PM_{2.5}$ on human health (Buonanno et al., 2013; Gennart et al., 1992; Mordukhovich et al., 2012), indicating that excess health risks may vary because of specific $PM_{2.5}$ components. More importantly, all levels of exposure to air pollution can cause potential health problems (Wan Mahiyuddin et al., 2013; Zhang et al., 2013).

The strategies to be taken to control PM pollution strongly depends on the knowledge of the chemical composition, sources, and processing of aerosol particles. Single particle aerosol mass spectrometry is a powerful tool to characterize aerosol particles in real time, including the analysis of particle chemical and physical properties, and the identification of particle sources and mixing state (Gard et al., 1997; Hinz et al., 1996; Laskin et al., 2012; Zhang et al., 2014, 2009b). Using an aerosol time-of-flight mass spectrometer (ATOFMS), a variety of studies, such as the chemical composition, sources, and mixing state of the particles, have been performed in Shanghai (Huang et al., 2013; Tao et al., 2011; Wang et al., 2009; Yang et al., 2009; Zhang et al., 2014, 2009b), in Guangzhou (Bi et al., 2011; Zhang et al., 2013), and in Beijing (Li et al., 2014), which would be beneficial for deep understanding of the chemical composition, sources, and processing of aerosol particles.

In this study, a single aerosol particle mass spectrometer (SPAMS) was employed to characterize the chemical composition and size distributions of particles during clear, hazy, and dusty days, to elucidate the impacts of meteorological factors on particle chemical composition and size distributions, and further to identify and evaluate particle sources. This study could help people understand the different source apportionment of aerosol particles under different meteorology/pollution conditions.

2. Methodology

2.1. SPAMS

The SPAMS (Hexin Analytical Instrument Co., Ltd., China) was described in detail previously (Li et al., 2011). A brief description about the SPAMS is given here. The particles in the size range of 0.1–2.0 μm were effectively drawn from ambient atmosphere into the vacuum system through a ~100 μm critical orifice at a flow rate of 80 mL/min, and then gradually were focused onto the axis of aerodynamic lens (Su et al., 2004). The velocities of the particles were subsequently determined by two continuous-wave 532 nm green lasers, and then the particles were desorbed/ionized at ion source region by a pulsed 266 nm Nd:YAG laser to obtain positive and negative ions, which were simultaneously analyzed by SPAMS. The power energy density of desorption/ionization laser was kept at ~1.6*10^8 W/cm^2 per pulse.

2.2. Sampling

Ambient aerosol measurement of single particles was performed from April 22 to May 4, 2011 in Chinese Research Academy of Environmental Sciences (40°02′ N, 116°24′ E) located in the northern part of Beijing outside of the 5th ring road (Streets et al., 2007). A residential area is nearby and the nearest traffic road is ~40 m away. The air was sampled into the SPAMS inlet using a conductive silicone tube with an inner diameter of 6 mm and a length of ~2 m on the roof of a 15 m high three-floor building. A $PM_{2.5}$ cyclone (URG Corp., USA) was installed to exclude coarse particles. An additional sampling pump was employed to shorten the residence time of air in the sampling tube.

2.3. Data processing

All the acquired mass spectra were converted into a list of peaks at each m/z using in-house software with a minimum signal threshold of 30 arbitrary units above the baseline. The resulting peak lists were analyzed using the Matlab® based toolkit YAADA 2.1 (http://www.yaada.org) (Allen, 2005). Based on the similarities of mass-to-charge ratio and peak intensity, mass spectral data were grouped into the clusters of particles using an adaptive resonance theory-based neural network algorithm (ART-2a) (Song et al., 1999) with a vigilance factor of 0.6, a learning rate of 0.05, a maximum of 20 iterations, and a range of the first 250 mass-to-charge (m/z) units of mass spectra.

2.4. Meteorological data

Meteorological data, including temperature (T), relative humidity (RH), wind speed (WS), wind direction, and visibility in Fig. S1, was obtained online from Weather Underground (http://www.wunderground.com/). Based on meteorological conditions, six typical weather periods were chosen, including three clear day periods (CD1: Apr 23 20:00–Apr 24 22:00; CD2: Apr 26 20:00–Apr 27 17:00; CD3: May 2 17:00–May 3 19:00), two hazy day periods (HD1: Apr 25 9:00–Apr 26 8:00; HD2: Apr 29 2:00–Apr 30 07:00), and one dusty day period (DD: Apr 30 17:00 – May 1 11:00) (Table 1).

2.5. Back trajectory analysis

Generally, the source regions affect the sampled aerosol particles, thus backward trajectories were analyzed by HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory version) model (http://www.arl.noaa.gov/HYSPLIT.php), which was developed by Draxler, R.R. and Rolph, G.D. in NOAA Air Resources Laboratory. 48 h air mass backward trajectories of each air mass were calculated every 2 h. The trajectories were terminated at a height of 500 m above ground level and ended at local time 22:00 on April 23, 8:00 on April 25, 17:00 on April 26, 7:00 on April 29, 11:00 on April 30, and 19:00 on May 2, corresponding to CD1, HD1, CD2, HD2, DD, and CD3, respectively (Fig. 1).

3. Results and discussion

3.1. Meteorological observation

Meteorological information during the whole sampling period is shown in Fig. S1 and six typical weather periods were summarized in Table 1. There was no rainfall during the six typical weather periods. During dusty days, a dust storm whirled through Beijing in April 30 to May 1and the highest $PM_{2.5}$ concentration reached 589 μg/m^3. During whole sampling period, the wind had a high speed from northwest direction (20–57) km/h and had a low speed from southeast direction (<10 km/h) (Fig. S1a). Higher speed wind favored pollutant dilution, leading to low aerosol levels. Anti-correlated diurnal variation of temperature and RH was observed