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Chemical characterization of submicron aerosol particles during wintertime in a northwest city of China using an Aerodyne aerosol mass spectrometry☆

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ABSTRACT

An Aerodyne quadrupole aerosol mass spectrometry (Q-AMS) was utilized to measure the size-resolved chemical composition of non-refractory submicron particles (NR-PM_1) from October 27 to December 3, 2014 at an urban site in Lanzhou, northwest China. The average NR-PM_1 mass concentration was 37.3 μg m^{-3} (ranging from 2.9 to 128.2 μg m^{-3}) under an AMS collection efficiency of unity and was composed of organics (48.4%), sulfate (17.8%), nitrate (14.6%), ammonium (13.7%), and chloride (5.7%). Positive matrix factorization (PMF) with the multi-linear engine (ME-2) solver identified six organic aerosol (OA) factors, including hydrocarbon-like OA (HOA), coal combustion OA (CCOA), cooking-related OA (COA), biomass burning OA (BBOA) and two oxygenated OA (OOA1 and OOA2), which accounted for 8.5%, 20.2%, 18.6%, 12.4%, 17.8% and 22.5% of the total organics mass on average, respectively. Primary emissions were the major sources of fine particulate matter (PM) and played an important role in causing high chemically resolved PM pollution during wintertime in Lanzhou. Back trajectory analysis indicated that the long-range regional transport air mass from the westerly was the key factor that led to severe submicron aerosol pollution during wintertime in Lanzhou.

1. Introduction

Atmospheric aerosols, which are emitted and formed from various natural and anthropogenic sources, are important components of the Earth's atmosphere and currently elicit a significant amount of scientific interests and political attention (Kulmala et al., 2004; Lanz et al., 2010; Sun et al., 2012). Aerosols exert significant effects on climate change through the direct scattering and absorption of solar visible radiation (IPCC, 2007; Myhre, 2009; Zhou et al., 2013). They also have evident effects on atmospheric visibility reduction (Watson, 2002) and can indirectly affect cloud properties and act as cloud condensation nuclei (Merikanto et al., 2009; Zhou et al., 2014). Furthermore, high concentrations of aerosols endanger public health by increasing the risk of morbidity and mortality and reducing life expectancy (Pope et al., 2009).

Rapid economic growth, industrial development, and urbanization in the past few decades in China have resulted in the severe and rapid deterioration of air quality. Air pollution has become a major public environmental concern (Hao and Wang, 2005; Chan and Yao, 2008; Huang et al., 2011; Miao et al., 2014). Particulate matter (PM), particularly fine particles ($PM_{2.5}$, particulates with diameters <2.5 μm) have become the primary pollutants in many urban cities wherein serious PM pollution events are present, such

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as regional haze (Huang et al., 2014; Yang et al., 2015; Zhang et al., 2015a). Lanzhou, which is an important industrial city in northwest China, suffered from serious air pollution caused by large emissions from local pollution sources, special landform, poor atmospheric diffusion conditions, and dust intrusions from upstream regions (Wang et al., 2009). The air pollution in the urban areas of Lanzhou is mainly characterized by the combined results of dust, soot, and motor vehicle exhaust. Although various mitigation strategies have been implemented in Lanzhou in the past few years, such as regular watering on arterial roads to prevent road dust resuspension and the prohibition of large trucks and farm vehicles at the downtown of urban areas, ambient air pollution is still serious at present. The annual average mass concentration of inhalable particles (PM_{10}, particulates with diameters <10 μm) in 2014 was reported to be 126 μg m^{-3} (http://hbj.lanzhou.gov.cn/). This value is approximately twice the secondary concentration limits of China's National Ambient Air Quality Standards (70 μg m^{-3}). In addition, submicron particles can scatter more sun light and influence atmospheric visibility because their sizes are considerably closer to the wavelengths of solar shortwave radiation (Cheng and Tsai, 2000). Thus, investigating the chemical compositions and size distributions of submicron particles in Lanzhou and providing further insight into the possible sources of air pollutants are necessary.

Although many studies on atmospheric aerosols have been conducted in Lanzhou in the past few years, most of them are focused on the mass loadings or optical properties of PM_{10} and total suspended particulate (Wang et al., 2005; Huang et al., 2009; Zhang et al., 2010; Liu et al., 2011) because of its unique semi-arid regions and frequent dust storm events. Few studies concentrated on the chemical characteristic of PM, particularly submicron particles. Previous studies revealed that, the finer the particles are, the more detrimental it will be to human health because of their high toxicity, easy penetration, and deep deposition in the human body (Oberdörster et al., 2005; Nel et al., 2006). Understanding the characteristics of the compositions, sources, and evolution processes of submicron particles are particularly important for scientific understanding and future policy making.

Most previous studies on the physicochemical properties of aerosols were based on off-line filter samplings followed by laboratory analyses, which were mainly in consideration of their easy operation, low cost, unadvanced instrument technology, and more important, wide application of various instruments and techniques together to provide a full characterization of aerosol compositions. However, the off-line filter measurements also provided poor time resolution data and limited size information, and even might have significant losses of semi-volatile species because of evaporative loss during sampling (Sun et al., 2013). In recent years, various online instruments have been widely deployed to real-time measure the aerosol physicochemical properties, such as an Aerodyne aerosol mass spectrometry (AMS), which has been extensively applied in field studies worldwide for its real-time measurements of the non-refractory submicron aerosol (NR-PM_1) species with high time resolution (Jayne et al., 2000; Jimenez et al., 2003; Drewnick et al., 2005; DeCarlo et al., 2006; Canagaratna et al., 2007). Despite this, AMS also has many drawbacks compared with the off-line analysis methods: its size range is limited to 1 μm, many refractory components (dust, black carbon, metals, etc.) are not detected by AMS, as well as AMS also need to be maintained routinely with a series of strict calibrations and rigorous operation environments in order to obtain high-quality data. Several studies have been implemented to survey the characterization of submicron aerosols by AMS in China since 2006. However, almost all studies were conducted in eastern China, such as Beijing (Sun et al., 2010; Zhang et al., 2011b, 2014a; Huang et al., 2010), Yangtze River Delta (YRD) (Huang et al., 2012; Zhang et al., 2015b, 2015c; Wang et al., 2016; Tang et al., 2016), and Pearl River Delta (PRD) (Huang et al., 2011; He et al., 2011; Xiao et al., 2009). These regions have experienced rapid economic development or hosted major events, such as the 2008 Beijing Olympic Games and the 2010 Shanghai World Expo. Few studies have focused on the underdeveloped area in northwest China. Thus far, only two studies regarding the characterization of submicron aerosols by AMS were reported in Lanzhou (Xu et al., 2014, 2016).

Surveying the chemical compositions, sources, and processes is significant to better understand the air pollution and emission sources in Lanzhou. An Aerodyne quadrupole AMS (Q-AMS) was utilized to conduct real-time NR-PM_1 measurements at an urban site in Lanzhou from October 27 to December 3, 2014. The present paper reported (1) the mass concentrations, size distributions, size-resolved chemical compositions, and temporal and diurnal variations of NR-PM_1 species; (2) the characteristics and dynamic variations of six organic aerosol (OA) components, including a hydrocarbon-like OA (HOA), a coal combustion OA (CCOA), a cooking-related OA (COA), a biomass burning OA (BBOA) and two oxygenated OA (OOA1 and OOA2); and (3) a back trajectory clustering analysis to examine the influence of regional transport on PM loading and compositions.

2. Materials and methods

2.1. Sampling site and instrumentation

Lanzhou is located in the northwest part of China and is characterized as a long but narrow northwest–southeast-oriented valley basin, with Baita Mountain in the north and Gaolan Mountain in the south (Chu et al., 2008; Wang et al., 2009), as shown in Fig. 1. The sampling site (36°2′36″N, 103°52′45″E; 1525 m a.s.l.) is in the yard of the Gansu Meteorological Administration, with all instruments installed in a room on the third floor (~12 m above ground level). The site is mainly surrounded by residential and business areas, with an arterial traffic road (east Donggang road) in the south approximately 120 m away.

A Q-AMS (Aerodyne Research Inc., Billerica, MA, USA) was utilized in situ to measure 5 min average size-resolved chemical compositions of NR-PM_1 continuously from October 27 to December 3, 2014. A scanning mobility particle sizer (SMPS; TSI model 3936, TSI Inc., St. Paul, MN, USA), which consisted of an electrostatic classifier (TSI model 3080) equipped with a long differential mobility analyzer (TSI model 3081) and a condensation particle counter (TSI model 3772), was employed simultaneously to quantify the particle number concentrations and size distributions in the size range of 14.6 nm–661.2 nm in the mobility diameter.

A URG cyclone (URG-2000-30ED, URG Inc., Carolina, USA) was used in front of the sampling inlet to remove coarse particles with size cutoffs of 2.5 μm in the aerodynamic diameter. Atmospheric aerosol was introduced to the instruments through a 1/2 inch stainless steel tube at a flow rate of 3 L min^{-1}, in which 0.15 L min^{-1} is for AMS and 0.3 L min^{-1} is for SMPS, and the remaining flow was exhausted by an external pump. A silica gel dryer was used to dry the atmospheric air before entering the instruments during sampling, and the room temperature was maintained at ~25 °C by using an air conditioner.

The meteorological data (temperature, RH, air pressure, wind speed, wind direction, and precipitation) for this study were obtained from the Lanzhou Meteorological Observing Station, China (No. 52889), which is approximately 150 m away from the sampling site. Hourly $PM_{2.5}$ data was provided by one of the China National Environmental Monitoring Center (CNEMC) air quality station, which was approximately 3.7 km away to the east from the sampling site, as shown in Fig. 1. All meteorological parameters are