Atmospheric Environment 123 (2015) 339–349

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Chemical composition, sources and evolution processes of aerosol at an urban site in Yangtze River Delta, China during wintertime

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HIGHLIGHTS

- ACSM measurement was conducted during wintertime in YRD.
- Secondary inorganics dominate the PM1 pollution in haze periods.
- SOA and aged BBOA significantly contribute to organics in hazy day.
- Secondary low-volatility production was extremely affected by regional transport.

ARTICLE INFO

Article history:
Received 2 December 2014
Received in revised form
25 June 2015
Accepted 5 August 2015
Available online 4 September 2015

Keywords:
Haze
PM_1
Components
Sources
Evolution processes
YRD

ABSTRACT

To investigate the composition, sources and evolution processes of submicron aerosol during wintertime, a field experiment was conducted during December 1–31, 2013 in urban Nanjing, a megacity in Yangtze River Delta of China. Non-refractory submicron aerosol (NR-PM_1) species were measured with an Aerodyne Aerosol Chemical Speciation Monitor. NR-PM_1 is dominated by secondary inorganic aerosol (55%) and organic aerosol (OA, 42%) during haze periods. Six OA components were identified by positive matrix factorization of the OA mass spectra. The hydrocarbon-like OA and cooking-related OA represent the local traffic and cooking sources, respectively. A highly oxidized factor related to biomass burning OA accounted for 15% of the total OA mass during haze periods. Three types of oxygenated OA (OOA), i.e., a less-oxidized OOA (LO-OOA), a more-oxidized OOA (MO-OOA), and a low-volatility OOA (LV-OOA), were identified. LO-OOA is likely associated with fresh urban secondary OA. MO-OOA likely represents photochemical products showing a similar diurnal cycle to nitrate with a pronounced noon peak. LV-OOA appears to be a more oxidized factor with a pronounced noon peak. The OA composition is dominated by secondary species, especially during haze events. LO-OOA, MO-OOA and LV-OOA on average account for 11%, (18%), 24% (21%) and 23% (18%) of the total OA mass for the haze (clean) periods respectively. Analysis of meteorological influence suggested that regional transport from the northern and south-eastern areas of the city is responsible for large secondary and low-volatility aerosol formation.

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http://dx.doi.org/10.1016/j.atmosenv.2015.08.017
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1. Introduction

Atmospheric particulate matter (PM) seriously affects air quality (Sun et al., 2010), climates (IPCC, 2007) and human health (Pope et al., 2002). Recently, the PM pollution is a serious environmental problem in the economic developed areas of China, including the North China Plain (NCP) (Guo et al., 2014; Sun et al., 2014; Zhang et al., 2014a), Yangtze River Delta (YRD) (Ding et al., 2013; Huang et al., 2013) and Pearl River Delta (PRD) (Huang et al., 2014). Improving megacity air quality is still a great challenge in China, due to complex factors including various sources, evolution processes of atmospheric PM and meteorological conditions (Cao et al., 2012; Huang et al., 2013; Guo et al., 2014; Sun et al., 2014).

The PM emissions from anthropogenic activities could have an impact on environmental air quality in industry regions (Zhang et al., 2013), transport regions (Zhang et al., 2012, 2013), urban areas (Zhan et al., 2012; Sun et al., 2014), and even on a global scale (Ma et al., 2011; Zhang et al., 2014b). Zhang et al. (2012, 2013) found that industry emissions and transport sectors contributes a lot of PM on a regional scale. Atmospheric PM is often dominated by high loadings of submicron aerosol (PM_1) during the haze-polluted days (Huang et al., 2013; Sun et al., 2013, 2014; Zhang et al., 2014a). Particularly, atmospheric organic aerosol (OA) can account for more than 50% of PM_1 during haze periods (Sun et al., 2013, 2014; Zhang et al., 2014a). Atmospheric OA can be as the primary OA (POA) and secondary OA (SOA) (Robinson et al., 2007; Jimenez et al., 2009). POA is directly emitted from various sources, such as traffic (Zhang et al., 2007; Liu et al., 2011), cooking (Allan et al., 2010), biomass burning (Alfarra et al., 2007), and coal combustion (Sun et al., 2013) emissions. SOA, composed of thousands of compounds which are poorly known, is formed from the oxidation of gas-phase precursors (Volkamer et al., 2006; Robinson et al., 2007). Therefore, it is crucial to investigate both PM_1 components and OA sources, in order to better understand behavior of aerosol particles in haze days in China.

Understanding of the atmospheric PM_1, including its chemical components, sources and evolution, greatly depends on measurement techniques and instrumentation (Canagaratna et al., 2007). Recently, an Aerodyne Aerosol Chemical Speciation Monitor (ACSM), designed from Aerosol Mass Spectrometers (AMS), was developed for routine and long-term measurements of the ambient non-refractory PM_1 species (NR-PM_1), including organic aerosol (OA), nitrate, sulfate, ammonium, and chloride (Ng et al., 2011). Combing OA mass spectra data with atmospheric receptor models, e.g., positive matrix factorization (PMF, Paatero and Tapper, 1994) and multilinear engine (ME-2, Canonaco et al., 2013), has been successfully utilized to resolve atmospheric OA source apportionment (Sun et al., 2012a, 2013). This is helpful for investigating contribution of various source emissions to OA and quantifying OA components (e.g. Zhang et al., 2005a; Lanz et al., 2007; Ulbrich et al., 2009). In fact, chemical components of PM and sources of OA show obviously regional variability during the winter haze periods in China (Huang et al., 2014; Sun et al., 2014). Many studies of PM_1 have been done in details with the ACSM or AMS in NCP like Beijing (Sun et al., 2010, 2012, 2013, 2014) and PRD like Guangzhou (Huang et al., 2011; Xiao et al., 2011) and Hong Kong (Li et al., 2015). However, the chemical components of PM_1, source and evolution processes of OA remains poorly understood in winter in the YRD region.

In this study, we deployed an Aerodyne ACSM for measuring the ambient non-refractory PM_1 (NR-PM_1) species in urban Nanjing during December 1–31, 2013. Insights into the composition and evolution processes of winter aerosols in YRD region are presented here. The source categories and evolution processes of the dynamic variations of OA are investigated by source apportionment analysis using PMF model.

2. Materials and methods

2.1. Sampling site

The sampling site is located in the downtown area of Nanjing (32°03′ N, 118°46′ E), which is one of the largest cities in western YRD region. Measurements took place during the winter from December 1 to 31, 2013. As shown in Fig. 1b, some open fire locations distributed around the urban Nanjing, especially in the southwestern area of Nanjing. This means that the air quality of the site can be subjected to the influence of biomass burning (BB) plumes. In fact, the fire counts might be underestimated due to the potential influence of haze-polluted weather and cloud. In this sampling site, a series of on-line measurements were performed from rooftop of the six floors building, which housed the Jiangsu Environment Monitoring Center. The aerosols were sub-sampled from the flow through a standard $PM_{2.5}$ inlet, located on the northeastern side of the building at a height of approximately 18 m. More descriptions on this measurement site can be found in Zhang et al. (2015).

2.2. Aerosol monitoring

The atmospheric NR-PM_1 species (organics, sulfate, nitrate, ammonium, and chloride) were measured during this experiment with the ACSM, which operated at a time resolution of ~15 min with a scan from m/z 10–150 amu at 500 ms amu^{-1} rate. The detailed descriptions of ACSM can be found in Ng et al. (2011). The aerosol sampling set-up and the ACSM operations in this study were the same as those in Zhang et al. (2015). An online analyzer, Monitoring of Aerosols and Gases (MARGA, model ADI 2080 Applikon Analytical B. V. Corp., the Netherlands), was deployed to measure the mass concentrations of a major water-soluble inorganic ion (potassium ion, K^+) from $PM_{2.5}$. In addition to the aerosol component deployments, the mass concentration of $PM_{2.5}$ was simultaneously measured with the MET ONE BAM-1020 (Met One Instruments, USA). And collocated gaseous species, NO–NO_2-NO_x was measured by a gas analyzer (Thermo Scientific). Ambient meteorological parameters, including ambient temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD), pressure, precipitation, and visibility, were obtained from a ground meteorology station located on the same six-story building as the sampling site.

2.3. Data analysis

The ACSM spectra were performed using the toolkit provided by Aerodyne, named ACSM standard data analysis software (version 1.5.2.0.0) written in Wavemetrics Igor™. In particular, the mass concentrations of ambient aerosol need to be corrected for particle collection efficiency (CE) (Middlebrook et al., 2011). In this study, we selected the CE value for OA, nitrate, sulfate, ammonium, and chloride, according to the equation $CE = \max(0.45, 0.0833 + 0.9167 \times ANMF)$ (Middlebrook et al., 2011), in which ANMF is the mass fraction of NH_4NO_3measured by the ACSM. In addition, the RIE values of 1.4 (organics), 1.1 (nitrate), 1.2 (sulfate), and 1.4 (chloride) were used for the data calibration.

The PMF with PMF2 executable version 4.2 (Paatero and Tapper, 1994) was performed on ACSM OA mass spectra, running in robust mode. The OA error dataset was resolved via the standard ACSM toolkit software. An IGOR-based PMF Evaluation Toolkit (PET) developed at the University of Colorado and introduced by Ulbrich et al. (2009) was employed. This enabled the facility to run analysis

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