



Aerosol characteristics and sources in Yangzhou, China resolved by offline aerosol mass spectrometry and other techniques[☆]



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ABSTRACT

Detailed chemical characterization of fine aerosols (PM_{2.5}) is important for reducing air pollution in densely populated areas, such as the Yangtze River Delta region in China. This study systematically analyzed PM_{2.5} samples collected during November 2015 to April 2016 in urban Yangzhou using a suite of techniques, in particular, an Aerodyne soot particle aerosol mass spectrometry (SP-AMS). The techniques used here reconstructed the majority of total PM_{2.5} measured where extracted species comprised on average 91.2%. Source analyses of inorganic components showed that secondary nitrate, sulfate and chloride were the major species, while primary sources including biomass burning, coal combustion, traffic, industry and re-suspended dust due to nearby demolition activities, could contribute to other species. EC-tracer method estimated that the organic matter (OM) was composed of 65.4% secondary OM (SOM) and 34.6% primary OM (POM), while the SP-AMS analyses showed that the OM was comprised of 60.3% water-soluble OM (WSOM) and 39.7% water-insoluble OM (WIOM). Correlation analyses suggested that WSOM might be rich in secondary organic species, while WIOM was likely mainly comprised of primary organic species. We further conducted positive matrix factorization (PMF) analyses on the WSOM, and identified three primary factors including traffic, cooking and biomass burning, and two secondary factors. We found the secondary factors dominated WSOM mass (68.1%), and their mass contributions increased with the increase of WSOM concentrations. Relatively small contribution of primary sources to WSOM was probably due to their low water solubility, which should be investigated further in future. Overall, our findings improve understanding of the complex aerosol sources and chemistry in this region.

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1. Introduction

Yangtze River Delta (YRD) region is one of the major economically developed and populated areas in China. Yangzhou is located in the north of YRD with a population of 4.6 million and an area of 6634 km². Due to dramatic increase of energy consumption and

emissions, air pollution in Yangzhou is emerging as a serious issue recently. In order to implement proper and effective measures to improve the air quality, knowledge of the physical, chemical characteristics and long-term temporal variations of fine particulate matter (PM_{2.5}) is essential. Traditionally, aerosol particles have been collected on filters, and then characterized by a variety of analytical techniques, such as total organic carbon (TOC) analyzer, ion chromatography (IC), gas/liquid chromatography mass spectrometry, etc. The organic fraction of aerosols, namely organic matter (OM), is an important and often dominant component of fine aerosols (Zhang et al., 2007). Nevertheless, due to the

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complexity of OM functionalities and properties, current methods are limited to determine the bulk properties of OM, such as total mass, elemental ratios, oxidation degrees etc., although some advanced mass spectrometric techniques can identify or quantify a subset of the organic components on a molecular level (Pratt and Prather, 2012).

Since the year 2000, the Aerodyne Aerosol Mass Spectrometer (AMS) has been widely used in characterizing fine aerosol particles (Canagaratna et al., 2007; Jayne et al., 2000). By using a thermal vaporizer, 70eV electron impact ionization, and a mass spectrometer, aerosol species can be quantified. Particularly, the AMS technique is powerful in determining the total OM mass. The advanced versions of AMS, such as high-resolution time-of-flight AMS (HR-ToF-AMS) (DeCarlo et al., 2006), or soot particle - AMS (SP-AMS) (Onasch et al., 2012; Wang et al., 2016c) can further gain detailed chemical information of OM, owing to its enhanced mass resolution (up to 4300) (DeCarlo et al., 2006) and wide detection range of mass-to-charge (m/z) ratios (up to a few thousands amu). It should be noted that Aerodyne AMS is designed specifically for online and intense field campaigns with very fine time resolution (typically a few minutes). Beside applications of the simplified versions of AMS – the aerosol chemical speciation monitor (ACSM) (Ng et al., 2011) and time-of-flight ACSM (ToF-ACSM) (Fröhlich et al., 2013), long-term (such as more than half a year) continuous deployment of AMS is often unrealistic (Daellenbach et al., 2016), due to its high operating cost, complex maintenance, requirement of trained professionals etc. However, through the re-aerosolization of liquid extracts, analysis of filter-collected PM samples via AMS is achievable. Data obtained by such offline AMS technique is in a low time resolution as each filter is typically collected every a few hours or days. Nevertheless, in this case, the AMS can be used to probe aerosol characteristics across a relatively long period. Offline AMS technique also extends the size range of measured particles as online AMS typically only measures submicron aerosols (Canagaratna et al., 2007). Moreover, filter sampling is very simple and easy to conduct, which can greatly save the costs of manpower and instrument maintenance. In these regards, development and application of offline AMS technique are necessary and valuable.

Recently, a number of studies applied the offline AMS technique for ambient PM characterization and source apportionment. For example, Sun et al. (2011b) analyzed 24 $PM_{2.5}$ samples collected in four US sites; Mihara and Mochida (2011) analyzed urban OM in Japan; Huang et al. (2014) characterized $PM_{2.5}$ samples from Beijing, Shanghai, Guangzhou and Xi'an collected during January 2013; Xu et al. (2013, 2015) used the HR-AMS to investigate the chemical characteristics of snow samples and $PM_{2.5}$ samples from Tibet plateau, respectively; Ye et al. (2017) employed the SP-AMS to investigate $PM_{2.5}$ in Changzhou; Daellenbach et al. (2016) analyzed 256 samples (including PM_1 , $PM_{2.5}$, PM_{10}) from 16 European urban and rural sites during summer and winter, and demonstrated that the results obtained by the offline AMS compared well with the online measurement results obtained by the HR-AMS or the ACSM.

In this study, the SP-AMS in combination with other analytical instruments were used to probe the characteristics and sources of $PM_{2.5}$ filter samples collected in urban Yangzhou during 2015–2016. As aerosol chemistry studies were very scarce in Yangzhou, our results are valuable in providing scientific basis for policymakers to abate the air pollution in this city and its surrounding areas.

2. Experimental methods

2.1. Sample collection

The $PM_{2.5}$ samples were collected at an urban site in Yangzhou

(119.40°N, 32.38°E), which was surrounded by residential buildings (see Supplemental Fig. S1). The site is located south of an arterial road - Yangtze Road (~300 m), and east of the Slender West lake (~1 km). The sampling was conducted from 13 November 2015 till 5 April 2016. $PM_{2.5}$ samples were collected using a high volume (flow rate of $1.05\text{ m}^3\text{ min}^{-1}$) sampler (Jinshida Ltd. Qingdao, model KB-1000), from 9:00 a.m. to 7:00 a.m. of the next day with a duration time of 22 h. All particles were deposited on 8×10 inch quartz fiber filters (Pallflex, USA), which were pre-baked inside a muffle furnace at $450\text{ }^\circ\text{C}$ for 4 h prior to use. The sampler was placed on the rooftop of an office building (~10 m above the ground), and a total of 103 samples were obtained.

The $PM_{2.5}$ mass was determined gravimetrically using a digital balance (OHAUS DV215CD, precision 0.01 mg) at 45% RH (relative humidity) and room temperature, immediately after the filter collection, and then was wrapped in aluminum foil and stored in a refrigerator at $-18\text{ }^\circ\text{C}$. The filters were extracted with 100 mL ultrapure water, sonicated for 30 min at $0\text{ }^\circ\text{C}$ in an ice-water bath, filtrated through $0.45\text{ }\mu\text{m}$ syringe filters (PES, ANPEL) and then were stored in sealed bottles maintained at $-18\text{ }^\circ\text{C}$ until analyses. Filter blanks were treated in the same manner as samples.

2.2. Chemical analyses

The aqueous extracts were analyzed by the SP-AMS, ion chromatography (IC), total water-soluble organic carbon (TOC) analyzer, and inductively coupled plasma mass spectrometer (ICP-MS). In addition, a portion of each filter was directly cut and measured by an organic carbon/element carbon (OC/EC) analyzer for the OC and EC contents. Meteorological parameters and concentrations of gaseous pollutants during the sampling period were acquired from the official website of Yangzhou Environmental Protection Agency.

2.2.1. Offline SP-AMS analyses

The operational procedures were similar to those reported previously (Ge et al., 2014; Sun et al., 2011b). Briefly, aliquots of aerosol extracts were nebulized using a constant output atomizer (TSI Model 3076), dehumidified by a diffusion dryer filled with silica-gel, and the dried particles were sent to the SP-AMS. Between every two samples, purified water was aerosolized to clean the system, and extracts of blank filters were treated in the same way as a system blank. The SP-AMS was operated with only the thermal vaporizer (at $600\text{ }^\circ\text{C}$) thus it measured non-refractory species as the HR-AMS does. The instrument was switched between V-mode (sensitive for mass quantification) and W-mode (with high mass resolution of ~4000) every 5 min. The m/z ranges were up to ~1000 for V-mode and ~450 for W-mode. As we focused on the aerosol chemical compositions, W-mode data was used in this work. Note similar to Daellenbach et al. (2016), we only analyzed the water-soluble fraction of $PM_{2.5}$. Extraction of $PM_{2.5}$ with other solvents such as methanol and ethyl acetate is also possible (Mihara and Mochida, 2011), but requires additional materials such as activated carbon and specific setup to efficiently remove and eliminate influences from the organic solvents. In addition, size distribution of the re-atomized aerosols, and its influences on the measurement results should also be investigated. Such studies are subject of our future work.

The SP-AMS data were post-processed by using the Igor-based ToF-AMS Analysis Toolkit (SQUIRREL version 1.56D and PIKA version 1.15D, available at: <http://cires1.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/index.html>). Due to possible loss of nitrate/chloride during nebulization, and incapability of the AMS in quantifying metals, concentrations of inorganic species were determined by IC and ICP-MS in this study, and the SP-AMS was used only to measure the water-soluble organic matter

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