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Long-range and regional transported size-resolved atmospheric aerosols during summertime in urban Shanghai

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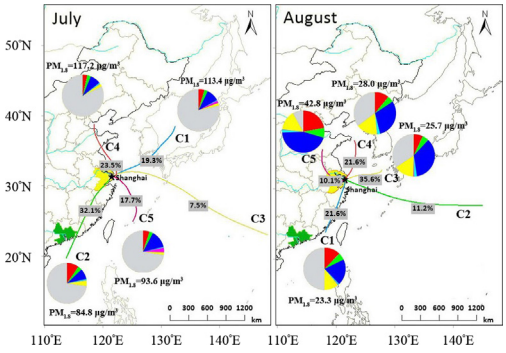
HIGHLIGHTS

- Spatial source distributions of size-resolved particles were studied.
- Potential pollution sources of inorganic and carbonaceous aerosols in $PM_{1.8}$-associated particles were studied.
- Important role of ship emission in the air quality of Shanghai in summer.
- Significant contribution of particulate pollutants from long-range transported air masses to PM levels of Shanghai in summer.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the concentrations of water soluble ions (WSI), organic carbon (OC), and elemental carbon (EC) of size-resolved (0.056–18 μm) atmospheric aerosols were measured in July and August 2015 in Shanghai, China. Backward trajectory model and potential source contribution function (PSCF) model were used to identify the potential source distributions of size-resolved particles and $PM_{1.8}$-associated atmospheric inorganic and carbonaceous aerosols. The results showed that the average mass concentrations of $PM_{0.1}$, PM_1, and $PM_{1.8}$ were 21.21, 82.90, and 100.1 μg m^{-3} in July and 7.00, 29.21, and 35.10 μg m^{-3} in August, respectively, indicating that the particulate matter pollution was more serious in July than in August in this study due to the strong dependence of the aerosol species on the air mass origins. The trajectory cluster analysis revealed that the air masses originated from heavily industrialized areas including the Pearl River Delta (PRD) region, the Yangtze River Delta (YRD) region and the Beijing-Tianjin region were characterised with high OC and SO_4^{2-} loadings. The results of PSCF showed that the pollution in July was mainly influenced by long-range transport while it was mainly associated to local and intra-regional transport in August. Besides the contributions of anthropogenic sources from YRD and PRD region, ship emissions from the East China Sea also made a great contribution to the high loadings of $PM_{1.8}$ and $PM_{1.8}$-associated NO_3^-, NH_4^+, and EC in July. SO_4^{2-} in Shanghai was dominantly ascribed to anthropogenic sources and the high PSCF values for $PM_{1.8}$-associated SO_4^{2-} observed in August was mainly due to the ship emissions of Shanghai port, such as Wusong port and Yangshan deep-water port. These results indicated that the particulate pollutants from long-range transported air masses and shipping made a significant contribution to Shanghai's air pollution.

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1. Introduction

Located on the eastern tip of the Yangtze River Delta (YRD), e.g. the triangle-shaped territory of Shanghai, southern Jiangsu province, and northern Zhejiang province of China, Shanghai develops with China's largest petrochemical complex, the largest steel plant and many other industries. And it owns the world's busiest container port, whose container throughput totalled 36.5 million TEU (twenty-foot-equivalent units) in 2015. Despite local pollution and intra-regional transport in YRD (Ying et al., 2014), the dust storm from the northwest of China (Wang et al., 2013a) and the polluted air masses from the PRD region, e.g. the dense network of cities that covers nine prefectures of the province of Guangdong and the special administrative regions of Hong Kong and Macau further deteriorates the atmospheric environmental quality of Shanghai. In addition, Shanghai may also under the impact of the vast anthropogenic emissions from East Asian region via long- and medium-range transport due to the monsoon system, which could be associated with the high observed aerosol loadings in Shanghai. Long range transport should contribute a lot to the increase of particulate matter (PM) concentrations in Shanghai.

The PM pollution in Shanghai has drawn increasingly attention in recent years due to its impacts on visibility degradation (Song et al., 2003), climate change (Cifuentes et al., 2001; Forster and Taylor, 2006) and human health (Bell et al., 2006; Iii et al., 2002; Sheppard et al., 1999). The visibility degradation due to PM is generally related to scattering by sulfate, nitrate, ammonium, and certain organic carbon (OC) as well as absorption by elemental carbon (EC) and some other OC (Andreae et al., 2008; Laskin et al., 2010; Sun et al., 2005).

Studies dealt with local pollution sources and long-range transport of the above mentioned aerosol species has been reported during the recent years (Wang et al., 2013a; Zhao et al., 2015). And there are a few studies using a variety of receptor model to determine the sources of aerosols and estimate their contributions to aerosol concentration at receptor site and downwind areas in East Asia (Han et al., 2005; Huang et al., 2007; Jeong et al., 2011; Mochida et al., 2003). However, few study on the characteristics of the long-range transported size-resolved atmospheric inorganic and carbonaceous aerosols in urban Shanghai have been reported. Besides, many studies just take one month of summer as a representative to demonstrate the characteristics of the atmospheric pollution of summer, which has some limitations because great changes could take place due to the differences of the dominant air mass origins among the months even though they all belong to summer.

Therefore, in this study, the concentration of PM in the size range of 0.056–18 μm and its compositions including water soluble ions (F^-, Cl^-, SO_4^{2-}, NO_3^-, Li^+, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, formate, acetate, methanesulfonic acid, oxalate) as well as carbonaceous species (OC, EC, SOC) were measured in Shanghai in two typical months (July and August) in summer and the spatial distributions of apportioned anthropogenic sources were analysed by PSCF model. The purposes of the study are 1) to provide the levels of size-resolved PM and its compositions in the urban areas of Shanghai; 2) to learn the contributing sources of size-resolved atmospheric inorganic and carbonaceous aerosols; 3) to get a better understanding of the role of the air mass parcels in facilitating long range transport to the receptor site.

2. Methods

2.1. Particle sampling

Size-segregated aerosol particle samples were collected on the roof (~15 m) of 4th Teaching Building in Fudan University (31.30°N, 121.50°E), Shanghai, China (Fig. 1). The sampling site was under the impact of residential, traffic, industrial and construction emissions, and it was located approximately 90 km to Yangshan Port, one of the largest container ship ports in the world.

Sampling was conducted in July and August, two representative months in the summer of 2015. Sampling of size-segregated aerosol particles was performed on 47 mm quartz filter membranes (PALLFLEX, USA) using the Micro-Orifice Uniform Deposit Impactor (MOUDI) Sampler (MODEL 110-R, MSP CORP., USA), a ten-stage cascade impactor with 50% cut-offs ranging from 0.056 to 18 μm at a flow rate of 30 L min^{-1}. The nominal cut-offs being 18, 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.10, 0.056 μm in aerodynamic diameters respectively. Here the inclusion of inlet and backup stages allowed collection of particles in 12 size fractions between <0.056 and >18 μm. One additional backup filter was applied. Prior to use, the quartz membranes were pre-heated at 500 °C for 4 h in a muffle furnace to remove original organic traces. Samples were put in membrane filters boxes right after sampling and reserved in a refrigerator at −18 °C until analysis. The sampling period was 24 h and the sampling work was taken only when the meteorological conditions was favorable (i.e. no wet precipitation and a constant air mass origin throughout the sampling day). 168 and 264 size-segregated aerosol samples were collected in July and August, respectively. All the procedures were strictly quality controlled to avoid any possible contamination of the samples. It should be noted that the experimental study was interrupted on 4–11, 22–26 July due to typhoon "Chanhom" and "Linfa" while 6–11, 20–25 August was under the impact of typhoon "Soudelor" and "Goni", respectively.

The determination of the collected particle mass was done gravimetrically using an intelligent weighing system (Hangzhou Wmade Intelligent Technology co., LTD, reading precision 10 μg) after at least 24 h of equilibration at (20 ± 1) °C with a relative humidity of (40 ± 1) %. After weighing, the quartz filter membranes were cut with ceramic scissors and chemically analysed.

2.2. Chemical analysis

OC and EC in particles were determined by Thermal/Optical Carbon Analyzer (Desert Research Institute (DRI) Model 2001, Atmoslytic Inc., Calabasas, CA, USA). A 0.506 cm^2 punch of each sample was analysed for eight carbon fractions following the IMPROVE TOR protocol. Four OC fractions (OC1, OC2, OC3, and OC4 at 120 °C, 250 °C, 450 °C, and 550 °C, respectively, in a helium atmosphere), and three EC fractions (EC1, EC2, and EC3 at 550 °C, 700 °C, and 800 °C, respectively, in a 2% oxygen/98% helium atmosphere) were produced. During volatilization of organic carbon, part of organic carbon was converted pyrolytically to EC, this fraction of pyrolized organic carbon (PC) is determined by transmittance. A He–Ne laser was employed to monitor the sample reflectance. As pyrolysis occurs, the reflectance decreases. Then, as EC is removed from the filter, the reflectance will increase until all carbon is removed from the filter. IMPROVE OC is operationally defined as OC1 + OC2 + OC3 + OC4 + PC and EC is defined as EC1 + EC2 + EC3 − PC. The concentrations of OC and EC reported here are all corrected for the field blanks. SOC was estimated by EC-based method (Turpin and Huntzicker, 1995) in this study, the primary OC/EC ratio used in the equation was replaced by observed minimum ratio of OC/EC which has been discussed in detail by Castro et al. (1999) and the estimation equation could be written as

$$OC_{sec} = OC_{tot} - EC \times (OC/EC)_{min} \quad (1)$$

For ion analysis, one fourth of each aerosol sample and the blank filter were extracted ultrasonically by 5 mL ultra-pure deionized distilled water (specific resistance ≥18.1 MΩ, Millipore) for 40 min. Water soluble ions F^-, Cl^-, Br^-, SO_4^{2-}, NO_3^-, PO_4^{3-}, Li^+, Na^+, NH_4^+, K^+, Ca^{2+}, and Mg^{2+} as well as formate, acetate, methanesulfonic acid, and oxalate were analysed by an ion chromatography (940 Professional IC, Metrohm, Switzerland.), with an separation columns of Metrosep A supp 16–250, a Metrosep A supp 16 guard column for anion and a Metrosep C6 analytical column, a Metrosep C4 guard column for cation.

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