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Seasonal variation of urban carbonaceous aerosols in a typical city Nanjing in Yangtze River Delta, China

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HIGHLIGHTS

- The seasonal character of carboaceous aerosols was provided.
- Emission sources were discussed as reasons for OC/EC seasonal variation.
- Biomass burning is one of the major contributors to both OC and EC.
- Meteorological condition affected SOC level and its contribution in OC.

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ABSTRACT

The Yangtze River Delta (YRD) is one of the regions with the most dynamic economy and severe atmospheric pollution in China. In order to characterize the particle features, especially the carbonaceous component in the YRD, particulate matter smaller than 2.5 μm ($PM_{2.5}$) and 10 μm (PM_{10}) samples in each season were collected in urban Nanjing, a typical city that locates in the west part of the YRD. The organic carbon (OC) and elemental carbon (EC) was differentiated using the thermal optical reflectance method. The average concentrations of $PM_{2.5}$, OC and EC during the study periods were observed to be 117.6, 13.8, and 5.3 μg/m^3 respectively, with all the highest levels in winter. The mass fraction of the Total carbonaceous aerosol (TCA) in $PM_{2.5}$ was estimated at 23% on average, lower than those reported for other cities in the YRD. The OC and EC correlated well in all the seasons, especially in spring and winter, implying that OC and EC were attributed to common emission sources. Good correlation was observed between OC and estimated K^+ from biomass burning in the harvest season in autumn and summer, indicating biomass burning a significant source of carbonaceous aerosols. This could also be confirmed by the lower fraction of OC3 + OC4 in OC during autumn and summer. The secondary organic carbon (SOC) estimated by EC-tracer method was the highest in winter (7.3 μg/m^3) followed by autumn (6.7 μg/m^3), summer (3.7 μg/m^3) and spring (2.0 μg/m^3). However, the SOC/OC in winter was not as high as that in summer and autumn, implying the high concentration of OC in winter was probably due to the stable weather but not mainly caused by SOC formation. The high SOC/OC ratio in summer was attributed to stronger oxidation, which could be suggested by higher sulfur oxidation ratio (SOR).

1. Introduction

Carbonaceous aerosols, including organic carbon (OC) and elemental carbon (EC), come mainly from incomplete fuel combustion of industrial boilers/kilns and residential stoves, iron & steel production, and vehicles (Bond et al., 2007). Additionally secondary organic particle formation is also an important source of OC (Pandis et al., 1992; Turpin and Huntzicker, 1995). The adverse

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effect of carbonaceous aerosols on human health has been confirmed by epidemiological studies, by the increased morbidity and mortality of respiratory and cardiovascular diseases with the enhanced exposure to OC and EC (Valavanidis et al., 2008; Cao et al., 2012). Additionally EC has ineligible effect on regional and even global climate by influencing the precipitation and enhancing the green house effect (Menon et al., 2002; Jacobson, 2001).

The carbonaceous aerosols are one of the major pollutants in China (Wang and Hao, 2012; Zhang et al., 2012). In particular, Yangtze River Delta (YRD) in eastern China was one of the three regions (YRD, Beijing–Tianjin–Hebei and Pearl River Delta) suffering the highest particle concentrations and the longest pollution episodes (Wang et al., 2014). The total amount of OC and EC has been reported at 17.5–20.5 μg/m^3 and the total carbonaceous aerosol (TCA), i.e., sum of EC and organic matter (OM, calculated as 1.6 × OC) contributed about 30% of particulate matter smaller than 2.5 μm ($PM_{2.5}$) in Shanghai, the biggest city in the YRD (Feng et al., 2009). In Hangzhou, another big city in the YRD, the total amount of OC and EC was observed at 25.5 μg/m^3, about 28.5% of particulate matter smaller than 10 μm (PM_{10}) (Cao et al., 2009).

Located in the west part of the YRD, Nanjing (31°14′–32°37′ N, 118°22′–119°14′ E) is the capital city of Jiangsu province with the population of 8.2 million. Due to the high pollutant emissions (Qiu et al., in preparation) and the unfavorable terrain for pollutant diffusion, the city had haze days increased since 1980s and remained at a high level since 2001 (Cheng et al., 2013a). It is reported that Nanjing had 226 haze days in 2012 (data provided by Nanjing Meteorological Bureau), using the definition of haze day by Wu et al. (2012) (the meteorological optical range is less than 10 km while the daily average relative humidity is less than 90%, with precipitation and other occasions that may lead to low visibility excluded).

The purpose of this study is to assess the seasonal variation of the carbonaceous aerosols in urban Nanjing, representative for the pollution caused by industrial, transport, straw burning etc. in the YRD. There are some studies focusing on the particle mass and organic compounds in the city: Wang et al. (2003) and Wang and Kawamura (2005) have reported the levels of N-alkanes, PAHs, and water soluble carbon in PM_{10} and $PM_{2.5}$ in Nanjing. Several researchers studied the particle mass and organic compounds during specific periods, e.g., straw burning seasons and haze episodes (Kang et al., 2013; Wang et al., 2009). Those studies, however, covered only specific periods, and the seasonal differences in the carbonaceous aerosols levels, carbon fractions and sources has not been fully evaluated. In particular, some other chemical indicators, e.g., K^+ and SO_4^{2-}, were seldom applied to help understanding the anthropogenic sources of carbonaceous aerosols in a complex pollution system. Moreover, the secondary organic aerosol (SOA) is the most uncertain and complicated part of particulate matter (PM), while its concentration levels and the reasons affecting the SOA formation in Nanjing have not been well investigated. It is thus important to analyze the seasonal characteristics of OC, EC, OC/EC ratios and secondary organic carbon (SOC) levels, for better understanding the sources, formation mechanisms, and control strategy of carbonaceous aerosols in the typical polluted city.

2. Methods

2.1. Sampling

As shown in Fig. 1, the sampling site is located in the western downtown of Nanjing, on top of the building of Jiangsu Provincial Academy of Environmental Science, 21 m above the ground. It is surrounded by residences, schools, offices and shops, with light traffic. The site is considered representing the pollution condition in the urban area, since it is just downwind of the dominant wind direction (east wind) and there is no obvious emission source nearby.

The 24-h $PM_{2.5}$ and PM_{10} samples were collected using a four-channel sampler at a rate of 16.7 l/min (TH-16A, Tianhong Company, Wuhan, China), using Φ47 mm Quartz and Teflon filters. The samples were collected for about 20 days in each season, i.e., autumn (Nov 10–28, 2011), spring (Mar 12–31, 2012), summer (Jun 11–28, 2012) and winter (Dec 5–19, 2013, Dec 24, 2013 and Jan 23–Feb 1, 2014, only $PM_{2.5}$ samples were collected). The meteorological data during sampling periods can be obtained from the National Climatic

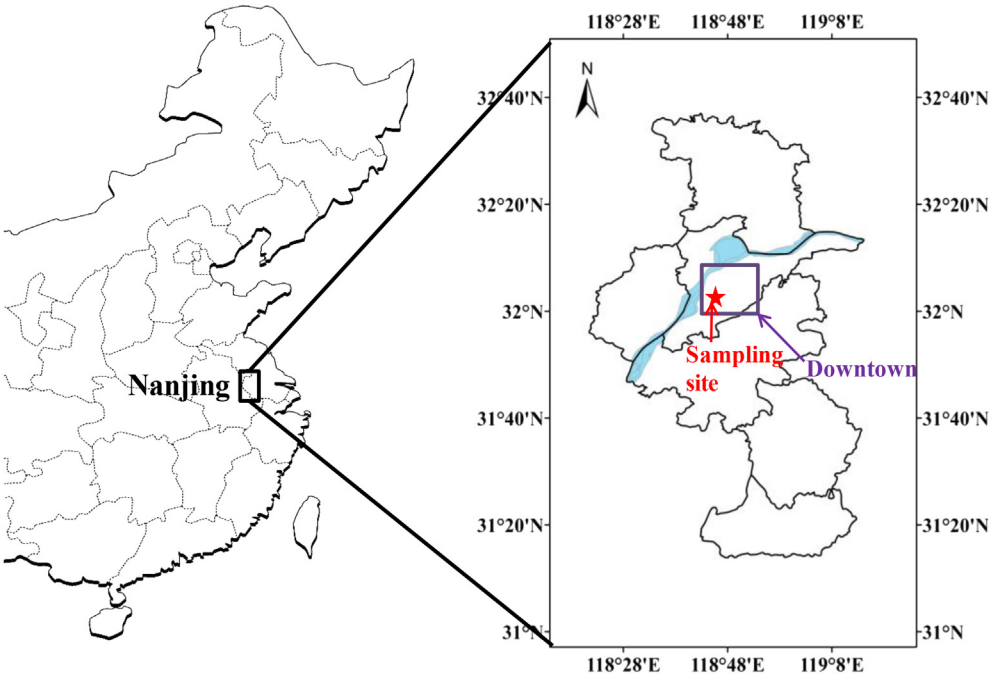

Fig. 1. Location of sampling site in Nanjing.