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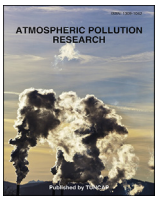


# Characteristics and origins of air pollutants and carbonaceous aerosols during wintertime haze episodes at a rural site in the Yangtze River Delta, China

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ABSTRACT

China has frequently suffered regional-scale haze pollution in recent years. In this study, real-time observation data such as $PM_{2.5}$, $PM_{10}$, $SO_2$, $NO_2$, CO and $O_3$ were used to analyze wintertime haze events at a rural site (Dongshan) in the Yangtze River Delta (YRD). 3-hour resolution organic carbon (OC) and elemental carbon (EC) were also measured to further investigate the sources of $PM_{2.5}$. The hybrid receptor models were used to identify source regions of $PM_{2.5}$. The results showed that both regional transport and local emissions significantly contribute to air pollution at Dongshan during haze periods. The source areas affecting high $PM_{2.5}$ concentrations were mainly located in nearby urbanized provinces (i.e., Jiangsu, Anhui and Zhejiang) and industrial provinces (i.e., Shandong and Hebei) in eastern China. Furthermore, open biomass-burning emissions in south China (i.e., Jiangxi, Hunan, Guangdong and Fujian) decreased regional air quality, which was supported by MODIS fire spots and receptor models. During clean periods, air masses were originated from remote regions such as Mongolia and oceanic areas (i.e., the Yellow Sea and the East Sea). Enhanced secondary organic carbon (SOC) formation was found under long-range transport when OC aging was favorable. Contrarily, relatively low SOC formation was found when the site was dominated by local emissions. In addition to local emissions, high $PM_{2.5}$ concentrations at Dongshan were apparently affected by either regional or long-range transport, which were characterized by relatively low and high wind speeds, respectively. It is necessary to implement the emission control strategies for the industrial and urbanized areas.


## 1. Introduction

Due to the rapid economic growth and urbanization over the last few decades in China, air pollution and heavy haze have become a serious environmental issue and led to a global concern (Yu, 2014; Chan and Yao, 2008). Haze is defined as a weather phenomenon with low atmospheric visibility (less than 10 km) under the conditions of 80% relative humidity (Yu et al., 2014). Previous studies have shown that anthropogenic emissions and meteorological conditions are the two most important factors causing extreme haze pollution (Xu et al., 2015; Zhang et al., 2015a). Fine aerosols (particulate matter with aerodynamic diameters equivalent to or less than 2.5 μm; $PM_{2.5}$) have been reported to be an important determinant for the formation of regional haze events (Wang et al., 2014a; Yang et al., 2011; Cao et al., 2012). Gaseous species such as $NO_2$, $SO_2$ as well as volatile organic compounds can

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react in the atmosphere and produce secondary PM pollution (Chang et al., 2011; Huang et al., 2014; Yu et al., 2007). High occurrence of haze has important impacts on visibility, air quality, climate change and human health (Yu et al., 2013, 2001, 2010; Chen et al., 2013). Coal combustion, biomass burning, traffic and industrial emissions are the main contributors of extreme haze pollution in China (Adame et al., 2012; Zhang et al., 2015c, 2015d; Cao et al., 2015).

Carbonaceous species, as significant chemical components of $PM_{2.5}$, have also been found to be widely associated with global radiative transfer, health problems and visibility deterioration (Ramanathan et al., 2007; Mcconnell et al., 2007; Cao et al., 2005). Carbonaceous aerosol is usually divided into organic carbon (OC) and elemental carbon (EC) fractions. EC is mainly emitted from fossil fuel and biomass combustion, and is of special interest because it could cause positive radiative forcing and was found to be the second most important factor of global warming behind $CO_2$ (Jacobson, 2001). OC originates both from direct emissions as primary organic carbon (POC) and gas-to-particle conversion as secondary organic carbon (SOC) and can cause negative forcing due to its scattering of sunlight (Cao et al., 2007; Mcconnell et al., 2007; Kaiser and Yun, 2002). The knowledge on carbonaceous aerosols is important for understanding the impacts of emissions on regional air quality and climate change.

There have been a large number of studies focusing on the physical and chemical characteristics (Yu and Zhang, 2011), formation mechanisms (Yu et al., 2008; Wang et al., 2014c; Guo et al., 2014; Yang et al., 2005), climate effect (Yu et al., 2013; Ramanathan et al., 2007) and source apportionments (Zhang et al., 2015c) of aerosol particles in China, especially in the high-populated city clusters such as the Beijing-Tianjin-Hebei (BTH) regions, the Yangtze River Delta (YRD) and the Pearl River Delta (PRD) (Wang, 2015; Cao et al., 2013; Tan et al., 2009). For example, Zhang and Cao (2015) investigated the spatial and seasonal distribution of $PM_{2.5}$ in 190 cities of China and found that high $PM_{2.5}$ level appears in the spring of the Northwest and West Central China, and autumn of the East China. The $PM_{2.5}$ concentrations were also found closely associated with variations of the boundary layer depth and human activities. Zhang et al. (2007a) investigated the characteristics of carbonaceous species in $PM_{10}$ and trace gases in winter in Beijing and found strong correlation between OC, EC with $PM_{10}$, $SO_2$ and CO, indicating the similar source of them. Vehicle emission with low OC/EC ratio and coal combustion with high OC/EC ratio were the main sources for carbonaceous aerosols in winter in Beijing. Wang et al. (2015) discussed possible causes of a severe haze episode in the Yangtze River Delta. It was found that meteorological conditions played a very important role in the formation of haze. Strong relationship between $PM_{10}$, $PM_{2.5}$, $SO_2$, $NO_2$ and CO was found, indicating the great contributors of vehicular emissions and biomass burning for the haze episode. Most of these studies reported air pollution on the regional or national scale; however, only a few studies have been conducted at rural sites in China, especially on the transport characteristics of air pollutants between rural sites and surrounded urban areas.

Air pollution is not only a local but also a regional or even a global-scale problem (Wang et al., 2010). Some air pollutants can be transported over hundreds of or even thousands of kilometers. (Bergin et al., 2005). Back trajectory and cluster analysis as well as hybrid receptor models such as the potential source contribution function (PSCF) and the concentration weighted trajectory (CWT) methods have been widely used to assess the transport pathways and the source regions of air pollutants (Yu et al., 2014; Zhang et al., 2014, 2015b; Yan et al., 2015; Sadyś et al., 2014). For example, Yu et al. (2014) reported that the major contributions to air pollutants in Hangzhou were from the southeastern sources based on the back trajectories and receptor model analysis. Karaca et al. (2009) evaluated long-range source contributions to the $PM_{10}$ profile of Istanbul, Turkey in 2008. Their results showed that air masses arriving at Istanbul were seasonally dependent and the potential source region of $PM_{10}$ in Istanbul included Europe, Asia and the Mediterranean region. Kim et al. (2009) investigated transport patterns of air pollution in Korea and found that long-range or regional transport between heavily polluted urban and rural area can significantly affect the air pollution level in rural sites.

In this study we analyzed 6 major air pollutants (i.e., $PM_{2.5}$, $PM_{10}$, CO, $SO_2$, $NO_2$ and $O_3$) and carbonaceous species (i.e., OC and EC) in $PM_{2.5}$ from January 15 to 28, 2015, at Dongshan. The objectives of this study are 1) to investigate the temporal variations and characteristics of major air pollutants and the carbonaceous components in $PM_{2.5}$ during wintertime haze events; 2) to evaluate the impact of local and regional transport sources on the formation of haze episodes in Dongshan using the air mass back trajectory and hybrid receptor models (PSCF and CWT).

## 2. Measurement and methods

### 2.1. Sampling site and observational data

Our measurement site Dongshan (31.04°N, 120.26°E) is located on the southeast bank of the Taihu Lake in eastern China, a rural area of Suzhou, which is the fifth largest city in the Yangtze River Delta (YRD) region (see Fig. 1). Dongshan has an eastern Asian monsoon climate accompanying seasonally changing prevailing winds which is warm and humid with four clear seasons. Due to its unique topography and humid climate, Dongshan is very sensitive to regional transport of air masses from its surrounding industrial areas and population centers.

Hourly mass concentrations of $PM_{2.5}$ and $PM_{10}$ were determined by the β-Ray method (BAM-1020, MetOne, America) at the Dongshan Automatic Meteorological Station (DAMS). Meanwhile, real-time hourly concentrations of gaseous species including $SO_2$, $NO_2$, CO and $O_3$ were measured using the ultraviolet fluorescence method, the chemiluminescence method, the gas filter correlation analysis method and the ultraviolet photometric method (EC9800 series, Ecotech, Australia), respectively. The instrumental operation maintenance, data assurance and quality control were performed according to the Chinese Ministry of Environmental Protection Standards for $PM_{10}$, $PM_{2.5}$ (MEP, 2013a) and for $SO_2$, $NO_2$, $O_3$ and CO (MEP, 2013b) (Zhang and Cao, 2015). The wind speed data were obtained from the DAMS.

### 2.2. Filter-based $PM_{2.5}$ sampling

The 3-h $PM_{2.5}$ samples were collected on prebaked quartz fiber filters (QFF, PALL, America) with 8 × 10 inches by a high volume air sampler (KC-1000, Laoshan, China) at a flow rate of 1.05 $m^3$ $min^{-1}$ in the DAMS (31.04°N, 120.26°E) from January 15 to 28, 2015. A total of 99 samples were collected including two filed bank filters collected following 10 min exposures to ambient air without active sampling.

All QFFs were pre-baked at 450 °C for 6 h before sampling to remove carbon contaminations. Before and after sampling, all QFFs were weighed by electronic balance (Sartorius, 0.1 mg, Germany). To avoid the errors introduces by variations of room temperature and relative humidity, the filters were equilibrated at 25 ± 0.5 °C and 30 ± 5% relative humidity for 72 h before being weighed. After weighting, the filters were wrapped in aluminum foils, packed in sealable bags and stored at −20 °C for further analysis. All procedures during sampling and analysis were strictly quality

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