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Contribution of regional transport to the black carbon aerosol during winter haze period in Beijing

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HIGHLIGHTS

- High BC episode with strong variability in Beijing was analyzed.
- WRF-BC model combined with a single particle soot photometer was used to analyze the regional transport.
- 47.9–56.8% of the BC in Beijing was attributed to sources in the central North China Plain.

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ABSTRACT

The mass concentrations of atmospheric refractory black carbon (rBC), an important absorber of solar radiation, were continuously measured with a single particle soot photometer (SP2) during wintertime haze period to investigate the transport of pollution to Beijing. The average mass concentration of rBC was 6.1 ± 3.9 μg m^{-3} during hazy periods, which was 4.7 times higher than it during non-hazy periods. Cluster analysis showed that the air parcels arriving at Beijing mainly originated from the northwest, passed through the south and brought the most polluted air to Beijing. Concentration-weighted trajectory analyses indicated that the central North China Plain were the most likely source region for the rBC that impacted Beijing. Furthermore, the Weather Research and Forecasting-Black Carbon model showed that 71.4–82.0% of the rBC at Beijing was from regional transport during the high rBC episodes and that 47.9–56.8% of the rBC can be attributed to sources in the central North China Plain. These results suggest that regional transport from the central North China Plain, rather than local emissions, was a more important source for rBC pollution in Beijing.

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1. Introduction

The black carbon (BC) aerosol is a strong absorber of solar radiation and largely because of this, it plays a unique and important role in Earth's climate system (IPCC, 2013). BC aerosols are emitted directly into the atmosphere as a byproduct of the incomplete combustion of fossil fuels and biomass. Globally, the largest BC sources are the open burning of forests and savannas (Bond et al.,

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2013). Over smaller scales, the impacts of other types of combustion on BC vary strongly with location. For example, on-road and off-road diesel engines account for ~70% of BC emissions in Europe, North America, and Latin America whereas the burning of residential solid fuels, especially coal and biomass, contribute ~60–80% of the Asian and African BC emissions (Bond et al., 2013).

In the boundary layer, BC particles absorb solar radiation, which leads to surface dimming, but at the top of atmosphere, the absorption of radiant energy by the BC aerosol enhances climate warming (Chung et al., 2010; Ramanathan and Carmichael, 2008). The total climate forcing of BC aerosol has been estimated to be $+1.1$ W m^{-2}, which is ranked as the second largest contributor to anthropogenic radiative forcing after CO_2 (Bond et al., 2013). Coated BC particles can act as cloud condensation nuclei (CCN), and the production of CCN leads to indirect climate forcing (Twohy and Poellot, 2005). The BC aerosols also can serve as substrates for various chemical transformations due to their porous and adsorptive nature. As a result, BC plays an important role in atmospheric chemistry (Fenidel et al., 1995). Furthermore, the absorption of sunlight by the BC aerosol can contribute to the degradation of visibility in polluted regions (Wang et al., 2013a), decrease crop yields (Chameides et al., 1999), and cause negative impacts on terrestrial and aquatic ecosystems (Forbes et al., 2006). Beyond the effects on the environment, epidemiological research suggests that BC particles may cause adverse health effects, including respiratory, cardiovascular and cardiopulmonary disease (Cao et al., 2012a; Mordukhovich et al., 2009; Smith et al., 2010).

China is the largest emitter of BC in eastern-southern Asia, with estimated annual emissions of 1.8 Tg of BC in 2006 (Zhang et al., 2009), which about 55.3%, 31.8%, 10.9% and 2.0% comes from the residential, industrial, transportation, and power sectors, respectively. Once emitted into the atmosphere, the BC aerosol can be transported over regional to intercontinental scales, and the particles are ultimately removed from the atmosphere through wet (e.g. in rain and snowfall) and dry deposition to the Earth's surface. These removal processes limit the atmospheric lifetime of BC particles to about one ($\pm$1) week (Bond et al., 2013). Therefore, reductions in BC aerosol emissions are an attractive option not only for mitigating global warming but also for improving environmental conditions in other ways (Shindell et al., 2012).

Numerous Chinese cities have suffered from serious air pollution in recent years (e.g. Cao et al., 2012b; Huang et al., 2014; Wang et al., 2013b), which has become one of the nation's top environmental concerns. However, the studies that have investigated the impacts of regional transport on BC aerosol pollution urban areas of China are limited, and thus it is important to identify the sources affecting the BC particles. Our study focused on Beijing, which is located in an area bordering the North China Plain, surrounded by the Yanshan Mountains to the north, and the Taihang Mountains to the west. Beijing is one of the world's largest cities, which has a population of >21 million. The number of motor vehicles reached 5.2 million in 2012, and the energy consumption was equivalent to 71.777 million tons of Standard Coal Equivalents (Beijing Statistics Yearbook, 2013). As a result of the rapid economic development, population expansion, and urbanization over the past several decades, Beijing has been experiencing severe $PM_{2.5}$ (particulate matter with aerodynamic diameter $\leq$ 2.5 μm) pollution (Tie et al., 2015).

Previous work at Beijing has shown that the mean concentration of $PM_{2.5}$ from April 2009 to January 2010 was 135 ± 63 μg m^{-3}, and 26% of this was secondary inorganic aerosol, 25% was from industrial pollution, 18% from coal combustion, 15% from soil dust, 12% from biomass burning, and 4% from traffic and waste incineration emissions (Zhang et al., 2013). Air pollution in Beijing is not only a local issue but also a regional problem due to various emission sources that mix together from local and surrounding areas (e.g, Garland et al., 2009; Matsui et al., 2009; Zhang et al., 2013). BC as the main absorbing material in $PM_{2.5}$ plays an important role in the air pollution and regional climate change. In this study, a single particle soot photometer (SP2) was deployed in Beijing to investigate the characteristics of refractory black carbon (rBC) particles in winter. The primary objectives were to investigate the temporal variations of rBC in a polluted atmosphere and to use several different methods to identify the sources for the rBC pollution.

2. Experimental

2.1. Research site

The rBC measurements were made from the rooftop of the Institute of Remote Sensing Applications (IRSA, 116.39°E, 40.01°N, ~20 m above ground level), Chinese Academy of Sciences (see Fig. S1), from 11 to 30 January 2014. Several field experimental campaigns have been previously conducted at this location (e.g., Li et al., 2013). The sampling site is surrounded by educational, commercial and residential districts, and no major industrial sources are located nearby. Thus, the observations are generally representative of urban conditions in Beijing.

2.2. rBC mass measurements

The rBC mass concentrations were measured with the use of a commercially available SP2 instrument (Droplet Measurement Technology, Boulder, CO, USA). This instrument has been carefully assessed for its ability to quantify rBC mass (e.g., Gao et al., 2007; Moteki and Kondo, 2007), and the operating principles of the SP2 have been previously described in detail (e.g., Stephens et al., 2003; Schwarz et al., 2006). In short, the SP2 measures the thermal radiation emitted by rBC-containing particles at their vaporization point. It relies on a continuous intra-cavity Nd:YAG laser beam which operates at a wavelength of $\lambda = 1064$ nm. The peak thermal radiation is linearly related to the rBC mass concentration regardless of the particle's morphology or mixing state (Slowik et al., 2007). The thermal radiation signal is then used to obtain the single particle's rBC mass after calibration using a fullerene soot standard reference sample (Lot F12S011, AlphaAesar, Inc., Ward Hill, Massachusetts).

For calibration, the fullerene soot was separated into different sizes by installing a differential mobility analyzer (EPS-20 Electrical Particle Size, HCT Co. Ltd., Korea) upstream of the SP2. Calibration curves were obtained from the peak intensities of the thermal radiation signals for the fullerene soot particles over a range corresponding to rBC masses of ~0.35–30 fg. This conversion was based on the mass-mobility relationships for the test material as described in Moteki and Kondo (2010). The rBC mass range corresponds to volume equivalent diameters of ~80–450 nm assuming a void-free density of 2.0 g cm^{-3} (Schwarz et al., 2008). The calibration for this size interval was close to purely linear, and various determinations of the mass-to-mobility relationship for this material have been found to be in good agreement (Gysel et al., 2011; Moteki and Kondo, 2010). The laser intensity was monitored using 269 nm monodisperse polystyrene latex spheres throughout the campaign period to check the stability of the SP2 laser power (Schwarz et al., 2010). More details concerning the SP2 operation can be found in our previous publication (Wang et al., 2014a).

2.3. Local meteorological measurements

Visual range (VR) and relative humidity (RH) were measured every minute with the use of an Automatic Weather Station

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