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Black carbon aerosol characterization in a coastal city in South China using a single particle soot photometer

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ABSTRACT

Black carbon (BC) is the dominant light-absorbing aerosol component in the atmosphere and plays an important role in atmospheric pollution and climate change. The light-absorbing properties of BC rely on particle size, shape, composition, as well as the BC mixing state with other aerosol components, thus more thorough exploration of BC aerosol characteristics is critical in understanding its atmospheric sources and effects. In this study, a newly-developed Single Particle Soot Photometer (SP2) was deployed in Shenzhen, China, for continuous BC measurements to obtain the important information about size distribution and mixing state of BC under severe air pollution conditions of China. The mean BC mass concentrations were found to be 6.0 and 4.1 μg m^{-3} at an urban site (UT) in the fall and winter, respectively, while it is much lower (2.6 μg m^{-3}) at a rural site (BG) in the fall. The mass size distributions of BC in volume equivalent diameter (VED) at the three sites showed a similar lognormal pattern, with the peak diameter at BG (222 nm) slightly larger than at the UT (210 nm) site. As to mixing state, the average percentage of internally mixed BC at the UT site was detected to be 40% and 46% in the fall and winter, respectively, while that at the BG site in the fall was only a slightly higher (47%), which implies that fresh local fossil fuel combustions were still significant at this rural site. The analysis of extremely high BC concentrations (>20 μg m^{-3}) at UT indicates that they were a complex of comparable contributions from both local fresh emissions and regional transport under unfavorable meteorology. Other characteristics of BC aerosol and their influencing factors in Shenzhen were also discussed.

1. Introduction

Black carbon (BC), usually defined as highly condensed carbonaceous residues in incomplete combustion, is the dominant light-absorbing aerosol component in the atmosphere (Jacobson, 2001). It plays an important role in the atmosphere and climate change through radiative absorption and changing surface albedo (Hansen et al., 1997; Haywood et al., 1997). Observational and simulation studies over East Asia indicate that BC may be responsible for the reduction of monsoon rainfall in this region as well as the increase of droughts in northeast China and the flooding in southeast China (Lau and Kim, 2006; Menon et al., 2002).

The light-absorbing properties of BC rely on particle size, shape, composition, as well as the BC mixing state with other aerosol components (Fuller et al., 1999; Jacobson, 2001; Chuang et al., 2003). Coatings on BC particles could enhance their light absorption efficiency. Bond et al. (2006) found that internally mixed BC coated by ammonium sulfate or organic compounds can absorb up to 50% more solar radiation than externally mixed BC. Global model simulations showed that the absorption cross-section of internally mixed BC particles was larger and the positive radiative forcing of these particles was about three times higher than that of the externally mixed BC (Jacobson, 2000). Generally, BC particles freshly emitted from vehicles are externally mixed with other components in the air, being hydrophobic and bare of non-refractory materials (Weingartner et al., 1997). After emissions, various processes in the ambient atmosphere like condensation of secondary materials would combine BC with sulfate, nitrate and/or organic matter, and thus transform it to internal mixing state. In biomass burning plumes, however, the internally mixed BC particles were reported to account for a fraction of ~70% (Schwarz et al., 2008a). The knowledge of the mixing state of BC is of great importance to understand the atmospheric behaviors and radiative forcing of BC aerosol.

Previous BC measurements in China have been mostly limited by filter and/or optical methods, which both measure bulk BC mass

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and fail to provide the size distribution and mixing state information of BC particles. In order to improve the understanding of BC mass concentrations, size distributions, and mixing state, an advanced on-line BC analyzer named Single Particle Soot Photometer (SP2) was deployed by the company of Droplet Measurement Technologies (DMT) in Boulder, Colorado. With high sensitivity and resolution, SP2 can provide direct single particle measurement, including BC concentration, size distribution, and mixing state (Schwarz et al., 2006; Gao et al., 2007). In this study, we deployed an SP2 in Shenzhen, China, for continuous BC measurements to obtain the important information about size distribution and mixing state of BC under severe air pollution conditions of China. Shenzhen is a mega-city located in the southeast of Pearl River Delta (PRD) along the southeast coast of China, neighboring Hong Kong to the south. The PRD region has been considered as one of the world's largest sources of anthropogenic soot emissions (Streets et al., 2001; Bond et al., 2004; Koch and Hansen, 2005). The annual mean BC concentration in Shenzhen was reported to be as high as $\sim$4 μg m^{-3} (Hagler et al., 2006). The SP2 measurement results in Shenzhen will help improve understanding of the formation processes of high BC loading in PRD and the parameterization in modeling its radiative effects.

2. Experimental methods

2.1. Sampling sites and periods

The SP2 measurement was performed in Shenzhen during the fall and winter of 2009–2010, which is the dry season of this region with prevailing wind of northerly to northeasterly. Two sampling sites were selected to observe BC aerosol characteristics in this area, including an urban site in Shenzhen University Town (UT) and a rural site in Ba Guang village (BG), as shown in Fig. 1. The UT site was on the campus of Peking University Shenzhen Graduate School (22.60 °N, 113.97 °E) located in the west of Shenzhen. The sampling periods at this site was from 25 October to 23 November 2009 in the fall and from 14 January to 27 February 2010 in the winter. The BG site was located at a holiday resort (22.65 °N, 114.53 °E) at the eastern coast of Shenzhen, with much less traffic and other pollution sources compared to the urban areas. The distance between the two sites was about 60 km. The sampling at this site was only conducted in a short period of 24 November to 2 December 2009 in the fall.

2.2. SP2 operation and data processing

In this study, the SP2 instrument sampled air through a $PM_{2.5}$ cyclone, limiting the sizes of ambient particles entering the instrument to below 2.5 μm in aerodynamic diameter. The sampling flow rate was set to 30 ml min^{-1}. The technical details of SP2 have been described elsewhere (Schwarz et al., 2006, 2008b). In brief, the instrument detects incandescence and scattering signals of BC-containing particles induced by a 1064 nm Nd:YAG intracavity laser. The mass of BC is proportional to the peak of the incandescence signal, irrespective of the presence of non-BC material or the morphology of the particle. The volume equivalent diameter (VED) of BC can be derived from the measured BC mass by assuming a density of 2 g cm^{-3}. The calibration of the SP2 was conducted with fullerene soot (Alpha Aesar, Inc., Ward Hill, MA), which were size-selected by introducing a differential mobility analyzer (DMA) upstream of the SP2, and the calibration curve was obtained by recording the peak intensity of the incandescence signal for fullerene soot particles of known diameters. The BC particle detection limit in this study was about 0.07 μm in VED. More details about the SP2 operation can be found in our previous publication (Huang et al., 2011).

The SP2 also has the capability to detect the mixing state of BC. The mixing state is identified by the separation of the scattering

Fig. 1. The map of the sampling sites in Shenzhen (UT and BG). (map source: www.baidu.com).