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Characterization of fine particulate black carbon in Guangzhou, a megacity of South China

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ABSTRACT

Continuous measurement of fine particulate black carbon (BC) was conducted at an urban site of Guangzhou in South China from December 2007 to December 2008. The daily average BC concentrations ranged from 0.6 to 20.5 μg m^{-3}, with an average value of 4.7 μg m^{-3}, which was substantially higher than those observed in the urban areas of other developed countries. Diurnal fluctuations of BC were marked with two peaks, one in the morning rush hour (08:00 LT) and the other in the late evening hour (21:00–22:00 LT), while the lowest BC concentrations were observed in the afternoon. Ambient BC concentrations displayed significant seasonal and diurnal variations with higher values in winter and spring, followed by lower concentrations during autumn and summer. Wind speed, wind direction and temperature were important meteorological factors that affected BC concentrations. A clearly negative correlation (r=–0.50, p<0.01) between BC concentrations and wind speed was found during the study period. A specific investigation was conducted to determine the relationship between optical BC and thermal–optical–reflectance elemental carbon (TOR EC) in distinct seasons. Although significant correlations between BC and EC were obtained (r>0.92, p<0.01), the regression slopes (ΔBC/ΔEC) slightly deviated from each other with values of 0.79, 1.18, and 0.81 in winter, spring and summer, respectively, possibly due to the distinct mixing states and source variations in different seasons. The calculated experimental attenuation coefficient showed a higher value (19.3 m^2 g^{-1}) in Guangzhou than the one recommended for typical Aethalometer measurements.

Keywords: *BC, temporal variations, meteorological parameters, elemental carbon*

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1. Introduction

Black carbon (BC) aerosols, primarily originate from anthropogenic activities, such as open biomass burning, combustion of fossil fuels and biofuels (Streets et al., 2001), and have significant impacts on local air quality, regional and global climate (Menon et al., 2002; Jacobson, 2004; Ramanathan and Carmichael, 2008). Characterization of BC has recently drawn considerable concerns because of its environmental significance (Ramachandran and Rajesh, 2007; Huang et al., 2012), as well as its adverse health effects (Ramanathan and Carmichael, 2008; Kopp and Mauzerall, 2010; Janssen et al., 2012). BC emissions reduction would have large benefits to near–term climate change, human health and food security (Shindell et al., 2012). BC has a typical atmospheric lifetime of about one week in the absence of precipitation (Ramanathan and Carmichael, 2008). Therefore it is subject to mixing with other aerosol components along the transport pathway (Wang et al., 2011a; Cape et al., 2012). It is well recognized that BC is the dominant light absorbing aerosol component, warming the atmosphere, and thus affecting the earth's radiation balance on a global scale (Ramanathan and Carmichael, 2008; Shindell et al., 2012). Consequently, recent estimates suggest that BC is the second most critical factor, following carbon dioxide, contributing to global warming in form of direct forcing (Jacobson, 2001; Jacobson, 2004). BC is mainly present in fine particulates in urban environments (Sahu et al., 2011; Wang et al., 2011b), and thus it is small enough to be readily inhaled into the human body and affects the human respiratory system. Epidemiological studies have associated BC containing particles, in particular, with allergies, respiratory and cardiovascular diseases (Smith et al., 2009; Herich et al., 2011). Therefore, a large number of BC measurements have been conducted globally in urban locations (Ramachandran and Rajesh, 2007; Cao et al., 2009; Tiwari et al., 2009; Wang et al., 2011b; Rattigan et al., 2013), rural sites (Cheng et al., 2006; Wang et al., 2011a), and background areas (Zhao et al., 2012).

BC and EC represent a similar fraction of the carbonaceous aerosol, although they are different depending on their physical and chemical behavior (Streets et al., 2001; Dutkiewicz et al., 2009; Petzold et al., 2013). BC is typically defined as the amount of strongly light–absorbing carbon, that is generally measured by optical instruments (Andreae and Gelencser, 2006). EC, on the other hand, refers to the fraction of carbon that is thermally stable in an inert atmosphere to high temperatures of about 4 000 K and can only be gasified by oxidation starting at temperatures above

340 °C (Andreae and Gelencser, 2006; Petzold et al., 2013). The Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance (TOR) protocol (Chow et al., 1993; Cao et al., 2004; Chow et al., 2004) and National Institute of Occupational Safety and Health (NOISH) thermal–optical transmittance (TOT) method (Birch and Cary, 1996; Chow et al., 2001) have commonly been used for OC/EC determination. EUSAAR_1 and EUSAAR_2 protocols have been applied for OC and EC analysis for the European Supersites for Atmospheric Aerosol Research (EUSAAR) project (Cavalli et al., 2010; Reche et al., 2011). The Aethalometer uses an attenuation measurement method to obtain semi–continuous real–time BC data (Hansen et al., 1984; Cheng et al., 2006; Cao et al., 2009; Cheng et al., 2010). Other instruments such as the Particle Soot Absorption Photometer (PSAP) (Sharma et al., 2002), Multi–Angle Absorption Photometer (MAAP) (Hitzenberger et al., 2006), EEL type Smoke Stain (EELS) Reflectometer (Salako et al., 2012), Continuous Soot Monitoring System (COSMOS) (Sahu et al., 2011), and Single–Particle Soot Photometer (SP2) (Huang et al., 2011) have also been used for BC measurements. Optical measurements obtained by Mm^{-1} but calibrated with respect to TOR EC or TOT EC protocols are named as Equivalent Black Carbon (EBC) (Reche et al., 2011; Petzold et al., 2013). In this study, BC was measured by an Aethalometer and reported as BC, instead of EBC, while EC was determined following the TOR protocol.

As BC emissions are particularly large in China (Streets et al., 2001; Ohara et al., 2007), it is reasonable to anticipate that aerosols containing BC contribute substantially to regional climate change and severe health issues in China. Guangzhou is one of the most urbanized cities in the Pearl River Delta region. The rapid industrial and economic development in this region has been accompanied by serious fine particle pollution that has gained high attention in recent years (Cao et al., 2004; Andreae et al., 2008; Zheng et al., 2009; Huang et al., 2012; Zhang et al., 2013). Thus, due to such importance in the atmosphere, determination of BC concentrations and its atmospheric behavior is urgently required in this region. Previous studies in China have focused on thermal carbonaceous measurements (Cao et al., 2004; Cao et al., 2007; Zhang et al., 2013), and most other research efforts addressed the radiative properties of optical BC (Wu et al., 2009). Although many uncertainties remain with BC analysis techniques, thermal elemental carbon (EC) has mostly been used as surrogate for BC (Wu et al., 2009; Verma et al., 2010; Huang et al., 2011). Short term inter–comparison measurements of BC and EC have been conducted at other locations in different countries (Jeong et al., 2004; Ahmed et al., 2009; Salako et al., 2012; Rattigan et al., 2013), and considerable differences must be expected when inferring site–specific absorption coefficients from filter samples. To date, few studies have assessed the long–term characteristics of optical BC in fine particles in South China. The purpose of this study is, therefore, to describe and examine the seasonal and diurnal variability of BC concentrations, as well as illustrate its variation with emission sources and meteorological conditions. This paper also discusses the variability between the higher time–resolution optical BC and filter based TOR EC mass concentrations in fine particles. In order to further understand the potential influence factors, the variations of BC/EC ratios during different seasons in fine particles were investigated as well.

2. Experimental Procedures

2.1. Study site

Guangzhou covers an area of 7 434 km^2 and had a registered population exceeding 12.7 million in 2010 (Guangzhou International, 2010). It is the largest city in southern China, and generally represents a typical urban area in the PRD region. The city often faces serious traffic congestion problems because of the increased vehicle numbers; the total count of vehicles was 1.82 million by the end of 2007 (GTPRI, 2010). Black carbon mass concentration measurements were conducted at the rooftop of a fourteen–story building (~53 m above the ground level) at the South China Institute of Environmental Sciences (23°07′N, 113°21′E, Figure 1) in Guangzhou. The surrounding area of the study site is characterized by commercial and residential land use. At about 400 m to the north and 1 km to the west of the measurement site are the South China Expressway and Huangpu Highway, respectively. More detailed information about the site can be found in the literature (Tao et al., 2009).

2.2. Data collection

BC mass concentrations were obtained by an Aethalometer (AE–31, Magee Scientific, USA), equipped with a 2.5 µm cut cyclone (Model SCC 1 829, BGI Inc., Waltham, MA, USA). It operated at a flow rate of 5 L min^{-1} with a time resolution of 5 min for twenty–four hours (24 hrs). The AE–31 measured BC attenuation at seven different wavelengths (370, 450, 520, 590, 660, 880 and 950 nm). The Aethalometer measurement is based on the optical method, i.e., by determining light beam attenuation, ATN (ATN=–ln(I/I_o)), where I and I_o were the respective transmission optical densities through the deposit and blank filters, respectively. The ATN through the filter is proportional to the amount of BC mass loading of the particle deposition; the ATN is then converted into BC mass concentration (BC=ATN/σ_{ATN}) (Hansen and Schnell, 2005). The attenuation coefficient (σ_{ATN}) is typically related to lifetime, emission sources, and chemical composition of the BC containing aerosol (Jeong et al., 2004; Salako et al., 2012), which have been shown to vary from 6.4 to 20.1 $m^2\ g^{-1}$ for the measurements conducted at urban sites and with a range of 5.2–19.3 $m^2\ g^{-1}$ for various types of biomass fires (Ramachandran and Rajesh, 2007). In this study, the ATN was converted to BC concentration using σ_{ATN}=16.6 $m^2\ g^{-1}$ at the wavelength of 880 nm. The detection limit for BC was 5 ng m^{-3} and the missing data due to instrument failure were excluded from further analysis.

Filter–based fine particle ($PM_{2.5}$) samples for thermal carbonaceous measurement were collected simultaneously with BC monitoring, using a Partisol air sampler (Model 2000H, Thermo Electron Corporation, USA), fitted with a $PM_{2.5}$ cyclone (Model PQ200, BGI Inc., Waltham, MA, USA), loaded with 47 mm quartz fiber filters (Whatman, UK), and operated at a flow–rate of 16.7 L min^{-1} for 24 hrs a day (starting at 10:00 LT each day and ending at 10:00 LT the next day). A total of 91 $PM_{2.5}$ samples, including 10 blank samples, were collected from January to July 2008. The sampling dates were from the first day to the last day of the months during January (representative of winter), April (spring), and July (summer). All the filters were pre–baked at 800 °C for 4 hrs before sample collection in order to remove potential organic contaminants.

The basic meteorological parameters, such as wind speed (WS), wind direction (WD), temperature (T), relative humidity (RH), and precipitation (PR), were obtained every 30 min during the study period. Wind speed and wind direction were recorded by a wind monitor (Model QMW110A, Vaisala, Helsinki, Finland). Ambient temperature and relative humidity were measured by a temperature probe (Model QMH102, Vaisala, Helsinki, Finland). Precipitation was measured by a rain gauge (Model RG13, Vaisala, Helsinki, Finland). The boundary layer depths were calculated by the U.S. National Oceanic and Atmospheric Administration (NOAA) READY archived meteorological data (Cao et al., 2009). The program used the archived data set GDAS (1 deg, 3–hourly, Global) based on Coordinated Universal Time (UTC), and the time–series of calculated daily boundary layer depths were obtained. All UTC values were converted to local time.