Science of the Total Environment 618 (2018) 531–541

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Source apportionment of black carbon during winter in Beijing

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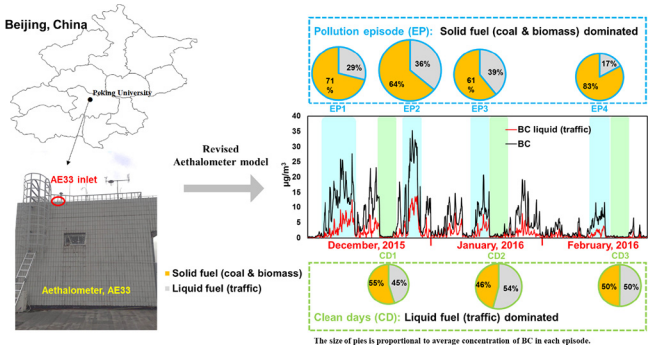
HIGHLIGHTS

- An aethalometer model is combined with tracers to identify BC sources.
- Traffic is one major BC source in Beijing especially during clean days.
- Solid fuel including coal and biomass burning dominates BC during haze episodes.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 September 2017
Received in revised form 2 November 2017
Accepted 4 November 2017
Available online xxxx

Editor: Xuexi Tie

Keywords:
Black carbon
Source apportionment
Haze
Aethalometer model
Beijing

ABSTRACT

Black carbon (BC) in $PM_{2.5}$ was measured at an urban site in Beijing during winter 2015 using an aethalometer. The characteristics and sources of BC during pollution episodes and clean days were analyzed. The average hourly mass concentration of BC during the study period was 5.31 ± 6.27 μg/m^3. BC was highly correlated with $PM_{2.5}$ (R^2 = 0.80), with its concentration ranging from 0.17 μg/m^3 in clean days to 35.33 μg/m^3 in haze days. Source apportionment results showed that the average contribution of liquid fuel source (e.g., vehicle emission) to BC was around 50% in clean days. While during the pollution episodes, solid fuel sources including coal combustion and biomass burning were the predominant sources, accounting for 61–83% of BC. Specific source tracers suggested that coal combustion and biomass burning dominated in different pollution episodes. Ratios of BC/$PM_{2.5}$ and BC/CO as well as source tracers provided further supportive evidences for the source apportionment results. Our findings suggest that it is more important to control solid fuel sources such as coal combustion for BC abatement in Beijing during haze days, while liquid fuel source (e.g., vehicle emission) plays a relatively more important role in clean days compared to pollution episodes.

1. Introduction

Black carbon (BC) aerosol, an atmospheric pollutant mainly consisting of carbon, has attracted much attention over the last several decades. BC in the atmosphere has relatively stable chemical property with large surface area and strong adsorbability (Croft et al., 2005; Moffet and Prather, 2009; US EPA, 2010). Due to its unique property, BC can adsorb primary and secondary pollutants, and serve as substrate or catalyst for various chemical processes (R. Wang et al., 2016). Other effects of BC include reducing atmospheric visibility and stunting plant growth by adhering to plant surface (Auffhammer et al., 2006; Chameides and Giorgi, 1999). Due to small particle size and large specific surface area of BC, it can adsorb carcinogenic pollutants such as volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs) and deposit in weasands and lungs, causing severe health effects (Cao et al., 2012; Dachs and Eisenreich, 2000). Furthermore, BC has

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https://doi.org/10.1016/j.scitotenv.2017.11.053
0048-9697/

received much attention because of its climate effect. BC in the atmosphere can strongly absorb solar radiation, thus it has positive radiative forcing (Chung and Ramanathan, 2010; Jacobson, 2001; R. Wang et al., 2016). Bond et al. (2011) estimated the total climate forcing of BC as $+1.1\ Wm^{-2}$, which was considered as the second largest contributor to anthropogenic radiative forcing after CO_2. The indirect forcing of BC mainly includes formation of cloud condensation nuclei (CCN) (Chýlek et al., 2010; Twohy and Poellot, 2005) and the melting of snow and ice surface (Menon et al., 2002).

Due to significant effects of BC on air quality, climate, and human health, it is necessary to investigate sources of BC to better control and reduce its adverse effects. BC are mainly emitted from incomplete combustion of biomass and fossil fuel, including open biomass burning, mobile/transportation sources, electricity generating units and other power production sources, and residential heating and cooking sources (Bond et al., 2011; Briggs and Long, 2016; Cooke et al., 1999; US EPA, 2010). The major methods of BC source apportionment include the aethalometer model, Chemical Mass Balance (CMB) model (Favez et al., 2010), the macro-tracer method (Larsen et al., 2012), Positive Matrix Factorization (PMF) model (Belis et al., 2014), the radiocarbon method and other specialized models (Belis et al., 2013; Briggs and Long, 2016). Among them, the CMB and PMF method can identify multiple source categories but they require large data set with many other chemical species information (Belis et al., 2013; Favez et al., 2010). The aethalometer model can identify less source categories (e.g. fossil fuel and biomass burning), however, it only needs multi-wavelength BC data to obtain source information (Chen et al., 2013; Favez et al., 2010; Larsen et al., 2012; Sandradewi et al., 2008a,b).

China has been considered as the largest emitter of BC in Southeast Asia, with estimated annual emissions of 1.8 Tg of BC in 2006 (Zhang et al., 2009), of which about 55%, 32%, 11% and 2% emitted from the residential, industrial, transportation, and power sectors, respectively. With the increased use of coal and vehicle population in China (China Statistical Yearbook, 2015), BC pollution has become more severe in multiple cities (Cao et al., 2012; Q. Wang et al., 2016; Zhao et al., 2013) such as Beijing, one of the global financial centers. Due to the rapid increase of vehicle number and recent changes of control policies, the sources of BC in Beijing might vary from year to year. It is essential to study current sources of BC in Beijing to reduce pollution level. By now, several studies have been conducted on BC aerosol in Beijing, focusing on its temporal variation, size distribution, optical properties, and emission inventory (Gao et al., 2007; Liu and Shao, 2007; Wu, 2015; Xu et al., 2015; Zhao et al., 2013; Zhou et al., 2009). Chen et al. (2013) investigated the sources of BC based on the ^{14}C method and identified that fossil fuel combustion was predominant in Beijing. Some studies applied models for BC source apportionment in Beijing (Q. Wang et al., 2016; Zhang et al., 2016). However, considering the limitation and uncertainty of each method (e.g., emission inventory in model and the complex operation in isotope methods), more methods are needed to confirm the sources of BC. Therefore, the primary goal of this study is to apply the aethalometer model with continuous BC data combined with tracer analysis in order to better understand the main sources and relative contributions of BC during pollution episodes and clean days in Beijing in winter.

2. Materials and methods

2.1. Sampling

On-line measurement of BC was conducted during the heating period from December 2015 to February 2016 at the Peking University monitoring station on campus of Peking University (PKU, 39°59′21″N, 116°18′25″E), in the northwestern area of Beijing city. The sampler was located on the roof top of a 6-story building on campus and the inlet of the instrument was about 20 m above the ground. There were no obvious emission sources around (see Fig. 1), except two roads (150 m to the east and 200 m to the south). Situated in a mixed district of teaching, residential, and commercial areas, the sampling site is representative of the Beijing urban area.

BC mass concentrations were continually measured by a multi-wavelength Aethalometer (AE33, Magee Scientific, USA), following the same basic measurement principle as the older models (Drinovec et al., 2015). Aerosol particles are continually collected on the filter and the optical attenuation (ATN) is measured with a time resolution of 1 min. Attenuation is measured on two spots simultaneously with different sample flows and there is no flow on the reference spot. The BC mass concentration is calculated based on the change of optical attenuation at 880 nm in the selected time interval using the mass absorption cross section (MAC) 7.77 $m^2\ g^{-1}$ (Drinovec et al., 2015). According to previous studies (Fialho et al., 2005; Sandradewi et al., 2008a,b; Yang et al., 2009), the absorption of other aerosol particles (e.g., carbonaceous aerosol or mineral) is not significant at 880 nm, and absorption at this wavelength is primarily due to BC. The measurements of AE33 at different wavelengths (i.e., 370, 470, 520, 590, 660, 880 and 950 nm) can be used for studying brown carbon, which is useful for assessing climate forcing of carbonaceous particles (Bond et al., 2011; IPCC, 2013). However, in this study, our goal is to identify BC sources using the continuous ambient BC data with the aethalometer model. Other pollutants including elements (e.g., K, As) and gas precursors (e.g., NO_x, SO_2) were measured simultaneously at the same sampling site by the Xact™ 625 (Cooper Environmental Services LLC, USA) and NO-NO_2-NO_x Analyzer (Model 42i, HL, Thermo Fisher Scientific Inc.). The detailed introductions of Xact could be found in Green et al. (2015).

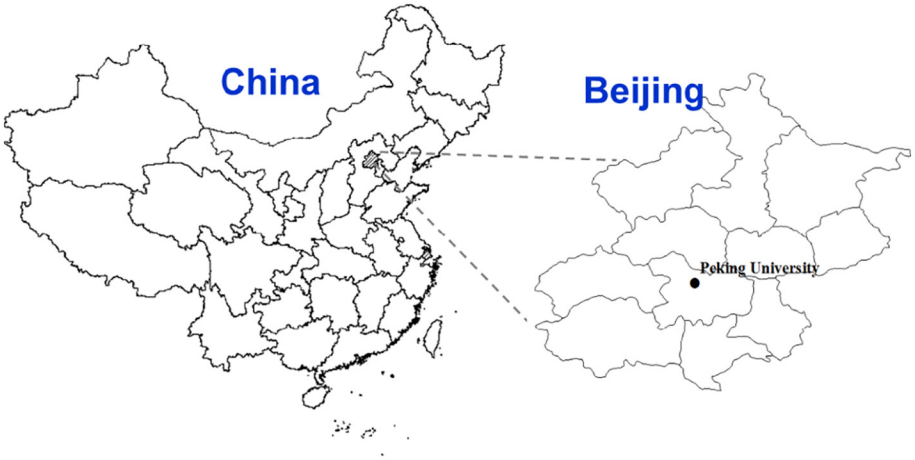

Fig. 1. Location of the sampling site in this study.