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Sources and radiative effects of wintertime black carbon aerosols in an urban atmosphere in east India

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HIGHLIGHTS

- BC surface mass concentration exhibited diurnal variation associated with local meteorology.
- Daytime mean BC surface mass concentration was 11 μg m^{-3}.
- Mean BC-AOD estimated in OPAC at 0.5 μm was 0.11.
- Mean shortwave BC radiative forcing at the TOA was +0.94 W m^{-2}.
- BC emissions from IGP and biofuel and fossil fuel combustion contributed majorly to BC loading.

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ABSTRACT

We carried out an analysis of black carbon (BC) surface mass concentration, its radiative effects, and sources of origin in an urban atmosphere in east India, during winter season, through ground-based measurements and application of modelling tools. BC surface mass concentration exhibited diurnal variation with their higher values and a larger variability during evening to early morning hours than during daytime (1100–1600 h, Local Time, LT) hours. Daytime mean surface BC mass concentration and BC mass fraction in total aerosol (size range 0.23–20 μm) and in submicronic aerosol (size range 0.23–1 μm) during the study period, corresponding to the well-mixed atmospheric layer were 11 μg m^{-3}, 3–10%, and 9–16% respectively. The mean BC optical depth (BC-AOD) and BC-AOD fraction at 0.5 μm were estimated in an optical model as 0.11 and 13% respectively. Mean shortwave aerosol radiative forcing due to BC at top-of-atmosphere (TOA) during the study period was found to be +0.94 W m^{-2}, which is about 59% the global mean radiative forcing due to carbon-dioxide gases. Estimates from BC simulations in a general circulation model showed BC surface concentration and BC optical depth in east India are primarily attributed to emissions from biofuel and fossil fuel combustion. Most of BC surface concentration (95%) and BC optical depth (60%) are contributed by emissions arising from the Indo-Gangetic plain (IGP) but there is a significant influence to BC columnar loading through elevated transport channels attributed mainly to emissions from open biomass burning from distant regions outside IGP.

1. Introduction

Emissions of black carbon (BC) aerosols arise from the combustion of fuel for residential cooking, industrial and transportation uses, and from open biomass burning. BC aerosols contribute significantly to atmospheric warming, constituting about 55% of the CO_2 forcing on the global scale (Ramanathan and Carmichael, 2008). Amplified warming of the upper troposphere and cooling near the surface due to the presence of BC aerosols in the atmosphere (Lau et al., 2005) could have substantial impact on south Asian monsoon circulation, potentially influencing rainfall distribution patterns (Menon et al., 2002). Emission inventories infer particularly large BC emissions due to low-temperature burning residential biofuel (BF) in south Asia, specifically over the entire Indo-Gangetic plain (IGP) in the Indian subcontinent region (Venkataraman et al., 2005). Recent studies on BC measurements in southeast Asia, at Bangkok showed seasonality in BC surface mass concentration with their highest value during the dry season, including December, mostly attributed to biomass burning activities prevalent over the region in addition to influence of long range transport mainly from China (Sahu et al., 2011a). Measurements of BC mass concentration at other location, Singapore, in southeast

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Asia, were found to be mostly higher by a factor of two on hazy days than on nonhazy days mostly attributable to large-scale forest fires in Sumatra (See et al., 2006).

Measurements of BC carried out at different locations in India have shown the presence of high BC surface mass concentration, specifically in the IGP during winter (Tripathi et al., 2005; Ganguly et al., 2006; Nair, 2007; Ramachandran and Kedia, 2010). However, there are very few data records on evaluation of BC aerosols in winter season in east India, with the available information limited to monthly mean value in a semi-urban atmosphere at an outflow region, Kharagpur (22.3°N, 87.3°E) (Nair, 2007) for December 2004. Studies from satellite-based measurements have inferred the presence of concentrated pollution pool in east India and postulated its tremendous impacts on the local climate (Girolamo, 2004). In this context, it is necessary to carry out a detailed investigation of BC aerosols concentration, including the pattern of its diurnal variation and extent of its contribution to aerosol loading in east India, a potentially strong source region over the IGP. It has also been inferred that emissions from different regions and sources influence the surface and columnar aerosol loading disproportionately (Verma et al., 2011). Hence, it is also necessary to understand sources contributing to BC aerosols measured in east India through integration of measurement and modelling studies.

In the present study, we carry out an analysis of black carbon (BC) aerosols and evaluate their radiative effects in an urban atmosphere in east India, during the winter month of December, when there is likely a strong influence of emissions arising from anthropogenic sources such as fuel combustion to atmospheric BC loading. The specific objectives of the present study are to (a) analyse the measured BC surface mass concentration in an urban atmosphere in east India at Kolkata, during the winter of December 2009 and 2010, (b) examine the aerosol optical depth due to BC (BC-AOD) and its radiative effects, (c) evaluate the mass fraction of BC (contribution of BC mass concentration to total aerosol mass concentration) and BC-AOD fraction (contribution of BC-AOD to total AOD), and (d) identify the sources contributing to BC loading in east India.

2. Method of study

2.1. Measurement of BC aerosols in east India

We carried out ground-based measurements of BC from the campus of Indian Institute of Technology Kharagpur Extension Centre (22.57°N, 88.42°E), which is on the outskirts of Kolkata city, in east India. The city map of Kolkata showing the measurement location is given in Fig. 1a. Study site, Kolkata (KOL) and stations over Indian region with available information on BC mass concentration are shown in Fig. 1b. Seven-days back trajectories (discussed in Section 2.3) showing representative pathway of back trajectories at study site are also presented in Fig. 1b. In the present study, observations were carried out on the rooftop of a building, about 12 m above the ground level. Continuous and near-real-time measurements of BC mass concentration were carried out using an aethalometer (model AE-42, Magee scientific, USA) operated at 3 L min^{-1} (LPM) at an average time of 5 min, for a total of 27 d during the period of December 2009 and 2010. The operating principle of the aethalometer is based on the measurement of the optical attenuation of a beam of light transmitted through a sample collected on a filter. Measurements from aethalometer are reported in terms of atmospheric black carbon aerosol (BC) concentration using the aethalometer specific attenuation coefficient recommended by the manufacturer which convert on-filter 'attenuation' (ATN) into a mass of BC (Hansen et al., 1984). The instrument measures attenuation of light beam at seven different wavelengths, namely, 0.37, 0.47, 0.52, 0.59, 0.66, 0.88 and 0.95 μm. Observation

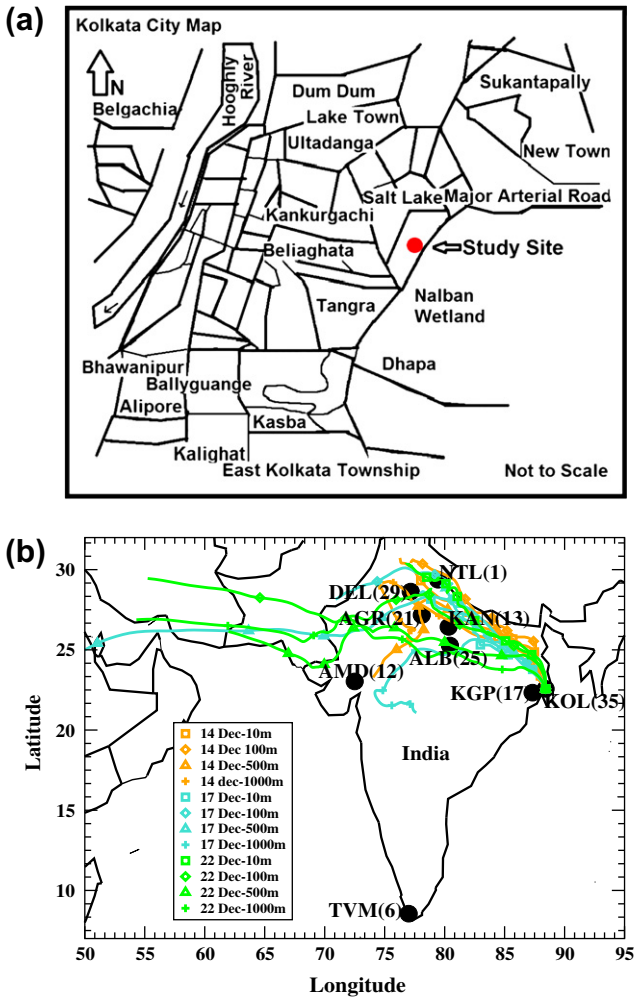

Fig. 1. (a) The city map of Kolkata showing the study site. (b) Study site, Kolkata (KOL) and stations over Indian region, including Kharagpur (KGP), Allahabad (ALB), Kanpur (KAN), Agra (AGR), Delhi (DEL), Nainital (NTL), Ahmedabad (AMD), and Thiruvananthapuram (TVM) with available information on BC mass concentration within parentheses at a respective station. Lines represent the 7-d back trajectories for Kolkata at 10 m, 100 m, 500 m, and 1000 m heights during days of measurement on 14, 17 and 22 December 2009.

at 0.88 μm wavelength is considered standard for BC measurement because BC is the principal absorber of light at this wavelength (other aerosol components have negligible absorption) (Weingartner et al., 2003; Hansen et al., 1984). An attenuation coefficient of 16.6 m^2 g^{-1} at 0.88 μm was used in this study, as recommended by the manufacturer. Recently several investigators have inferred possible errors in aethalometer measurements employing filter absorption method to obtain atmospheric BC concentration (Bodhaine, 1995; Weingartner et al., 2003; Arnott et al., 1999, 2005; Corrigan et al., 2006; Kirchstetter and Novakov, 2007; Virkkula et al., 2007; Coen, 2010). To account for the multiple light scattering effects inside the filter matrix leading to amplified absorption, correction is included in the aethalometer attenuation coefficient recommended by the manufacturer which is in correspondence with Bodhaine (1995). To account for the shadowing effect (underestimation of BC mass concentration with increasing filter loads), the loading correction, R (ATN) was considered as suggested by Weingartner et al. (2003). The shadowing effect has been found to be more prominent for purely soot particles while being almost negligible for aged atmospheric aerosols in ambient data sets (Weingartner et al., 2003) where the single scattering albedo is generally in the range 0.80–0.95 (Kirchstetter and Novakov,