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On the contribution of black carbon to the composite aerosol radiative forcing over an urban environment

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ABSTRACT

This paper discusses the extent of Black Carbon (BC) radiative forcing in the total aerosol atmospheric radiative forcing over Pune, an urban site in India. Collocated measurements of aerosol optical properties, chemical composition and BC were carried out for a period of six months (during October 2004 to May 2005) over the site. Observed aerosol chemical composition in terms of water soluble, insoluble and BC components were used in Optical Properties of Aerosols and Clouds (OPAC) to derive aerosol optical properties of composite aerosols. The BC fraction alone was used in OPAC to derive optical properties of BC aerosols. The aerosol optical properties for composite and BC aerosols were separately used in SBDART model to derive direct aerosol radiative forcing due to composite and BC aerosols. The atmospheric radiative forcing for composite aerosols were found to be +35.5, +32.9 and +47.6 Wm^{-2} during post-monsoon, winter and pre-monsoon seasons, respectively. The average BC mass fraction found to be 4.83, 6.33 and 4 $\mu g\, m^{-3}$ during the above seasons contributing around 2.2 to 5.8% to the total aerosol load. The atmospheric radiative forcing estimated due to BC aerosols was +18.8, +23.4 and +17.2 Wm^{-2}, respectively during the above seasons. The study suggests that even though BC contributes only 2.2–6% to the total aerosol load; it is contributing an average of around 55% to the total lower atmospheric aerosol forcing due to strong radiative absorption, and thus enhancing greenhouse warming.

1. Introduction

Atmospheric aerosols affect the climate directly by scattering and absorbing the incoming solar radiation, known as direct aerosol radiative forcing (Ramanathan and Crutzen, 2003; Takamura et al., 2007). Out of many species of aerosols which induce direct radiative forcing, Black Carbon (BC) play a major role in the forcing by partly shielding the surface from the intense tropical solar radiation (Ramanathan and Crutzen, 2003). BC aerosols have been found to be contributing significantly to the atmospheric warming both globally and regionally (Ramanathan and Carmichael, 2008). The total BC emissions over India from all sources such as fossil fuel, biomass burning and biofuel combustion are a large fraction of the total global emissions (Reddy and Venketaraman, 1999). Therefore, monitoring of BC aerosol across various parts of the country is important to assess the radiative effects on regional as well as global scale (Sreekanth et al., 2007). However, studies focusing on the radiative impacts of BC aerosols are sparse over the Indian region. Some limited studies (Babu et al., 2002; Tripathi et al., 2005; Latha and Badrinath, 2003) have reported characteristics of BC aerosols over the country, especially over southern and northern regions of Indian subcontinent. The characteristic variation of BC over Pune has been already reported in Safai et al. (2007). But the extent of radiative forcing due to BC and contribution of BC fraction to total aerosol radiative forcing has not been examined so far over the site. Hence an attempt has been made to estimate the radiative forcing due to composite aerosols (total aerosol fraction) and the percentage contribution of forcing solely due to BC aerosols in total forcing, over the site.

2. Instrumentation

Measurements of aerosol chemical and optical properties were carried out over Pune, an urban site located in the western part of India [18° 32′ N, 73° 51′ E and 559 m AMSL]. Measurements of Total Suspended Particulates (TSP) were carried out using a high volume air sampler [Model 401, Envirotech, India]. Whatman-41 filter papers were used for sampling at a flow rate of about 1 $m^3 min^{-1}$. All the samples were extracted for water-soluble and water-insoluble

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components and were further analyzed for different chemical components using atomic absorption spectrophotometric and ion chromatographic techniques (Rengarajan et al., 2007; George et al., 2008; Safai et al., 2005). The detection limit for ion chromatographic analysis was about 0.02 ppm whereas, that for the atomic absorption spectrophotometric analysis varied from 0.0002 to 0.02 ppm.

Soot (BC) data was obtained from an Aethalometer (Magee Scientific, AE - 42). In this method, atmospheric air is pumped through an inlet at the desired flow rate (3 $Lmin^{-1}$ in present study), which impinges on a quartz micro fiber strip. A light beam from a high-intensity LED lamp is transmitted through the sample deposit on the filter strip, at 880 nm. The measurement of the attenuation of light beam is linearly proportional to the amount of BC deposited on filter strip. The specific absorption of BC is 16.6 m^2g^{-1} used in calculation of BC mass concentration and also the aethalometer measurements were dry. Observations were recorded at the time base of 5-minute interval (Safai et al., 2007). Uncertainty in the observations was reported up to ±10% by the manufacturer after the calibration, which was carried out in May 2004. The filter based absorption technique used in Aethalometer is widely used and reported to have shown good comparison with the other methods used for monitoring of BC particles viz., coefficient of haze tape sampler, particle soot absorption photometer, thermal oxidation/reflectance technique, etc. (Allen et al., 1999; Babich et al., 2000). Also, as the instrument was calibrated for flow rate and optical attenuation test just five months before the present set of observations, hence the instrumental error bias can be minimal. However, aerosol parameters, such as scattering interference, filter loading, composition, size, etc., systematically bias this filter absorption measurement method. Scattering and filter loading are the major primary corrections used (Corrigan et al., 2006). Also, there are other methods of corrections suggested by Arnott et al. (2005) and Weingartner et al. (2003). The uncertainties arising from the above mentioned factors could not be characterized for this data as they depend upon information on aerosol type (fresh/aged), size, chemical composition and scattering coefficients, which are presently not available. Details of Aethalometer and its operation have been discussed elsewhere (Hansen et al., 1984).

A Prede Sun/sky radiometer which derives aerosol optical properties such as aerosol optical depth (AOD), single scattering albedo (SSA) and asymmetry parameter (ASP) has been in operation over the site since December 2000. A detailed description of calibration, methodology and data reduction procedures of this instrument are presented in Nakajima et al. (1996). Details of the error analysis on retrievals of aerosol optical properties can be found in our earlier work (Pandithurai et al., 2008). The instrument was last calibrated in December 2003 at the factory (Prede Co. Ltd, Tokyo, Japan) and was used in this study for deriving aerosol scale heights and also for the comparison of modeled aerosol optical properties.

3. Methodology

The mass concentrations of insoluble, water-soluble aerosols derived from the chemical composition of TSP and BC from aethalometer is converted in to number density as prescribed by Hess et al. (1998). Then these number densities of corresponding components were combinely used as inputs in the urban aerosol model of Optical Properties of Aerosols and Clouds (OPAC) (Hess et al., 1998) software package to derive the aerosol optical parameters such as AOD, SSA and ASP for composite aerosols. Also, the aerosol scale heights were adjusted in OPAC, which were derived by taking the ratio between AOD and extinction coefficient (σ) as explained in Hayasaka et al. (2007) for each month. AOD at 500 nm observed from sky radiometer and σ derived using the visibility observations from India Meteorological Department were used to estimate aerosol scale height using the relation as suggested by Ricchiazzi et al. (1998) and Hess et al. (1998) i.e.,

$$\sigma = 3.912/\text{visibility} \quad (1)$$

OPAC-modeled AOD and SSA values were compared with those of the Sun/sky radiometer over the site.

For deriving spectral variation of BC optical properties such as AOD, SSA and ASP, BC number density derived from BC mass fraction was used in OPAC. The mass fractions of different components derived from chemical measurements which are used in OPAC to derive spectral aerosol optical properties during different months are shown in Table 1.

Modeled spectral aerosol optical properties for composite aerosols (Insoluble, soluble and BC), MODIS derived spectral surface albedo, column water vapour, and OMI column ozone over the station were incorporated in Santa Barbara Discrete-ordinate Atmospheric Radiative Transfer model (SBDART) (Ricchiazzi et al., 1998) to derive net fluxes in the spectral range 0.3 to 3 μm (shortwave range) at the surface and at the Top of the Atmosphere (TOA). This model accounts for multiple scattering in a vertically inhomogeneous non-isothermal plane-parallel atmosphere. Tropical model atmospheric profiles of temperature and humidity were used in SBDART for this study. The model simulations were carried out for aerosol free conditions and the differences in net short wave radiative fluxes with and without aerosols were calculated to estimate composite direct aerosol radiative forcing, both at the surface and at the TOA.

Subsequently for deriving direct radiative forcing solely due to BC fraction, modeled BC optical properties (AOD, SSA and ASP), MODIS derived spectral surface albedo, column water vapour, and OMI column ozone were used in SBDART to derive net fluxes in the range 0.3 to 3 μm (shortwave range) at the surface and at the Top of the Atmosphere (TOA). No aerosol fluxes also were derived and direct radiative forcing due to BC aerosols was calculated at surface and TOA.

4. Results and discussions

4.1. Comparison between observed and modeled aerosol optical properties

OPAC can model the aerosol optical properties in the entire solar spectrum at eight different relative humidity conditions for a given aerosol chemical composition. In this study, we modeled the aerosol optical properties (AOD, SSA and ASP) at 50% RH as the average relative humidity condition over the experimental station from October to May used to be around 50%. The comparison of OPAC derived AOD and SSA values for composite aerosols at mid-visible wavelength (0.5 μm) and those obtained from Sun/Sky radiometer are shown in Fig. 1. The OPAC derived AOD and SSA values were found to match closely with Sky radiometer observed values at mid-visible wavelength 0.5 μm (Fig. 1). The mean difference in AOD values was 0.02 where as in SSA it was 0.03, which are well within the retrieval uncertainties. This ensures that the modeled composite

Table 1
Mass fraction of different components used in OPAC for different months.

Months	Mass fraction (μgm^{-3})		
	Insoluble	Water-Soluble	BC
October -04	140	31.3	3.91
November-04	139.8	44.9	5.76
January-05	85.3	56.9	6.79
February-05	114.1	53.8	5.87
March-05	184.2	46.8	4.10
May-05	188.9	55.3	3.90