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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

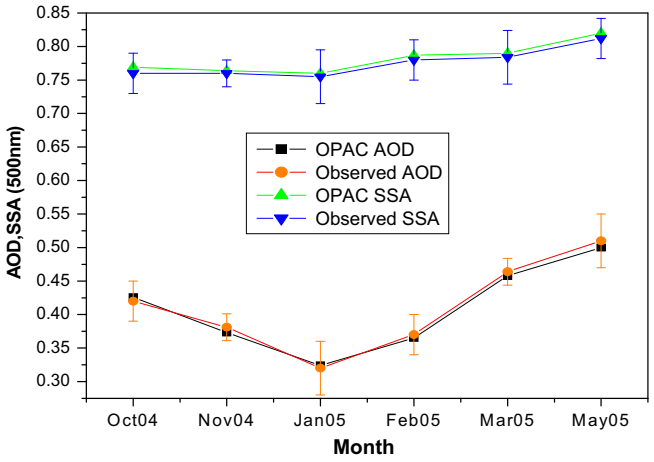

Fig. 1. Comparison between OPAC-modeled and observed AOD, SSA at 0.5 μm for different months.

aerosol optical properties from the chemical composition data are reasonable to utilize in modeling radiative fluxes.

It can be seen that AOD values were highest in pre-monsoon (March and May), especially in the month of May. This may be due to strong convection and surface winds which raise more dust particles. Also, influx of transported particles from arid regions through northwesterly winds is the other source for enhanced aerosol load during pre-monsoon. The SSA values were observed to be lowest in January, obviously due to the higher BC mass concentration. The seasonal mean AOD values at 0.5 μm were found to be 0.41, 0.35 and 0.48 during post-monsoon (October and November), winter (January and February) and pre-monsoon respectively. The corresponding SSA values were found to be 0.77, 0.76 and 0.80 respectively for the above seasons.

4.2. Radiative forcing estimates

Radiative forcing for both composite and BC only aerosols were estimated as explained in section 3. The monthly averaged surface radiative forcing values for composite and BC aerosols from October 2004 to May 2005 are shown in Fig. 2. Composite aerosol radiative forcing at the surface was found to be high during pre-monsoon season, which may be due to the observed higher AOD values (Fig. 1). The forcing value during pre-monsoon season was $-47.2\ Wm^{-2}$ at the surface and $+0.4\ Wm^{-2}$ at TOA. The negative forcing values observed at the surface imply a net cooling effect and the positive sign for the TOA forcing arises due to highly reflecting continental surface albedo and strong aerosol absorption, reducing the back scattered radiation reaching TOA. The forcing values were -33.5 and $-34.9\ Wm^{-2}$ at the surface respectively during winter and post-monsoon and the corresponding TOA forcing values were -0.55 and $+0.56\ Wm^{-2}$. The difference between the TOA and the surface gives the atmospheric forcing, indicating the net atmospheric absorption, which is $+47.6$, $+32.9$ and $+35.5\ Wm^{-2}$ during pre-monsoon, winter and post-monsoon, respectively.

On the contrary to composite aerosol forcing, higher BC forcing was observed during winter than in pre-monsoon season. The forcing solely due to BC during winter was found to be $-19.5\ Wm^{-2}$ at the surface and $+3.9\ Wm^{-2}$ at TOA, indicating strong radiative absorption by BC. The highest BC forcing in winter is associated with the highest BC mass fraction observed during winter compared to other two seasons (Table 1). The respective forcing values during post-monsoon and pre-monsoon were found to be

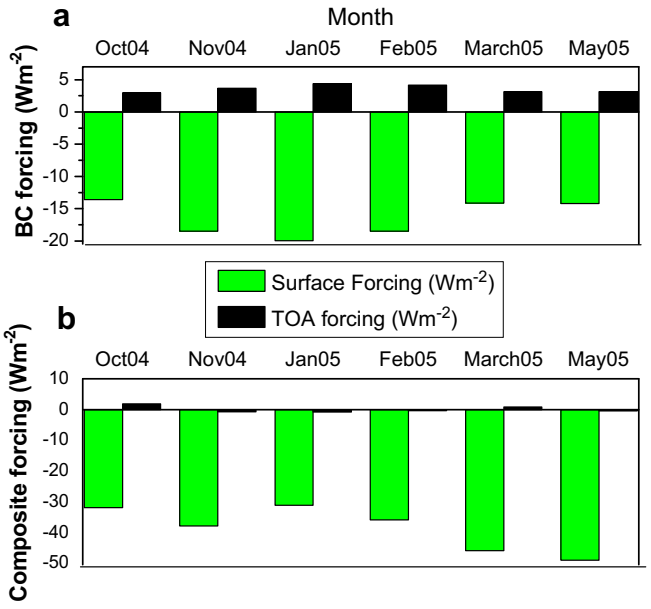

Fig. 2. Shortwave aerosol radiative forcing estimated at the surface and at the TOA for (a) BC aerosols and (b) Composite aerosols.

-15.5 and $-14.1\ Wm^{-2}$ at the surface and $+3.4$ and $+3.1\ Wm^{-2}$ at TOA, inducing an atmospheric forcing of $+23.4$, $+18.9$, $+17.2\ Wm^{-2}$ respectively during winter, post-monsoon and pre-monsoon. The composite and BC aerosol atmospheric radiative forcing is shown in Fig. 3. It can be seen that on an average, the BC atmospheric forcing contributes around 55% of total aerosol atmospheric radiative absorption integrated over all the three seasons. The main reason for remaining 45% of the forcing may be due to other absorbing aerosol species such as mineral dusts, originated due to convectively activated mineral soil present over and around the station and transported desert dust from Arabian/African regions.

It could be seen that on the contrary to the Indian Ocean Experiment (INDOEX) results, which reports negative radiative forcing at TOA (Rajeev and Ramanathan, 2001; Satheesh et al., 2006), positive TOA forcing estimates were observed in this study. Differences in sign of TOA forcing are due to highly reflecting land surface in contrast to dark ocean surface albedo and higher fraction of pollutants over the experimental station. A typical study

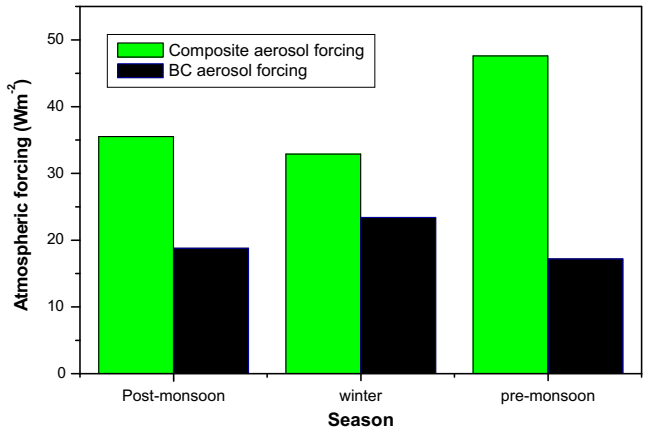

Fig. 3. Seasonal averages of shortwave atmospheric forcing for composite and solely for BC aerosols.

by Gonzi et al. (2007) has reported aerosol radiative forcing values over different AERONET stations in Europe. They have performed the experiments for different albedo conditions and reported that albedo is a strong factor in modulating the direct aerosol radiative forcing. The surface and TOA forcing values obtained found to be less compared to the present study. This may be due to experimental locations over Europe considered in Gonzi et al. (2007) was relatively of less polluted regions. Marmer et al. (2007) reported positive TOA forcing value over eastern Europe in spring and winter because of the contribution of black carbon. Higher surface albedo and strongly absorbing aerosols over continental urban environment may be the main reason for positive forcing obtained at TOA (e.g., Sreekanth et al., 2007; Pandithurai et al., 2008; Badrinath and Latha, 2006).

The mass fraction of BC to TSP is reported to be less (Ramanathan and Crutzen, 2003). In spite of this small mass fraction, BC contribution to AOD and consequently to radiative forcing is high (Satheesh, 2002; Podgorny et al., 2000). The percentage contribution of BC to TSP is found to be 2.2% during pre-monsoon, 3.5% during post-monsoon and 5.8% during winter over Pune. Our study suggests that, this percentage of BC is able to contribute to total AOD by around 7% during pre-monsoon, 10% during post-monsoon and 15% during winter and over all contributing to around 55% of total atmospheric aerosol radiative absorption averaged over all the three seasons. This strong atmospheric heating along with surface reduction in solar flux can intensify low level inversion, which slowdown convection and in turn can inhibit cloud formation (Chou et al., 2002).

5. Summary and conclusions

OPAC derived aerosol optical properties for composite aerosols and for BC fraction alone has been incorporated in SBDART to derive composite and BC only aerosol forcing respectively, over Pune for different seasons. The atmospheric forcing derived for composite aerosols were found to be +35.5, +32.9, +47.6 Wm^{-2} and for BC fraction alone these were found to be +18.8, +23.4 and +17.2 Wm^{-2} during post-monsoon, winter and pre-monsoon, respectively. The study suggests that on an average, BC atmospheric shortwave radiative forcing over the station was around 55% of the total atmospheric aerosol forcing, there by enhancing greenhouse warming.

Black carbon aerosols earlier have been considered as a potential source of air pollution alone. But now it is found that BC is the most important constituent in inducing direct aerosol radiative forcing (Khatri et al., 2009). It has also been reported that, BC has been a strong component in altering cloud microphysics and hence inducing aerosol indirect forcing (Conant et al., 2002). Apart from inducing local radiative effects, BC is found to influence even large-scale phenomenon such as monsoon due to its strong radiative absorption (Lau and Kim, 2006). Our study suggests that, even though BC contribution to total aerosol loading is less, it contributes to around 55% of atmospheric forcing due to its strong radiative absorption nature, which can alter tropospheric temperature profile and cloud formation processes.

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