Radiative forcing of black carbon over eastern India

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[1] Measurements of aerosol Black Carbon using a 7 channel Aethalometer at Visakhapatnam, a coastal tropical station on the east coast of India are used to study the temporal variation of surface BC mass concentration. The surface BC mass concentrations show a significant diurnal variation which is seasonally dependant. Analysis using the multi spectral data indicates that the BC mass does not show significant absorption due to non-BC aerosol species which indicates that the surface BC mass is dominated by fossil fuel sources with no indication of any other strong anthropogenic source. The Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model was used in conjunction with Optical Properties of Aerosols and Clouds (OPAC) to estimate the BC radiative forcing. The results show large negative surface forcing during winter (-35.78 W/m^2), moderate during summer (-16.8 W/m^2) and lower forcing during monsoon (-9.9 W/m^2) and post monsoon (-2.81 W/m^2). The forcing at the top of the atmosphere is positive for all the seasons. **Citation:** Sreekanth, V., K. Niranjan, and B. L. Madhavan (2007), Radiative forcing of black carbon over eastern India, *Geophys. Res. Lett.*, *34*, L17818, doi:10.1029/2007GL030377.

1. Introduction

[2] Historical changes in fuel utilization have caused large temporal changes in atmospheric aerosol absorption which suggests that the absorbing aerosol species in atmosphere like the Black Carbon (BC) aerosol may have contributed to global temperature changes in the past century. It was reported that qualitative features of historical changes in BC emissions as quantified in terms of BC emission factors segregated by utilization sectors such as transportation for fossil fuel, industrial, residential/commercial and power generation show rapid increase in the past 50 years implying that BC history needs to be represented realistically in climate change assessments [*Novakov et al.*, 2003], which demand a comprehensive set of measurements on BC mass. Many reports are available in literature quantifying the BC mass fraction of the composite aerosol and its radiative forcing efficiency which show large spatio-temporal variability in the BC mass [*Babu et al.*, 2004; *Tripathi et al.*, 2005]. It was also reported that BC mass also exhibits a well defined seasonal variation at some locations with variability from 10% share to total mass during dry months to 3% during monsoon months [*Babu and Moorthy*, 2002]. The total BC emission from India from all sources such as fossil fuel, biomass burning and bio fuel combustion is a large fraction of the total global emission [*Reddy and Venkataraman*, 1999] and shows large spatio-temporal variability due its varied land use and agricultural patterns. 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. *Krishnan and Ramanathan* [2002] reported that the Indian haze with as much as 10–13% BC by mass is known to reduce the surface solar insolation by about 10% and nearly double the lower atmospheric solar heating. The seasonally asymmetric cooling consistent with the seasonality in the south Asian aerosol forcing indicates that the surface cooling underneath the polluted regions is balanced by warming elsewhere. Secondly the spectral dependence of BC absorption is generally considered to vary weakly [*Bergstrom et al.*, 2002] whereas other aerosol absorbing components show stronger spectral dependence. Hence multispectral aerosol BC measurements can provide information on the regional presence of strong absorbing aerosols sources. In view of the importance of the characterization of aerosol BC mass concentration and its impact on aerosol radiative forcing, we present here the results on the measurements of near surface aerosol BC mass and its radiative forcing at Visakhapatnam (17.7°N, 83.3°E), a tropical Indian station on the east coast of India located in the air mass path ways into Bay of Bengal. These are the only measurements that are available from a location on the east coast of India situated in the outflow region into Bay of Bengal from peninsular India.

2. Instrumentation and Data

[3] A seven channel Aethalometer (Model AE-47) of Magee Scientific Company, USA was set operational at Visakhapatnam in December 2005 for the measurement of BC aerosol mass concentration and the data base with a temporal resolution of 5 minutes, for the period December 2005 to September 2006 is used in the present study. The BC mass was estimated from the attenuation of light transmitted at seven wavelengths viz., 370, 470, 520, 590,660,880 and 950 nm through its filter tape on which aerosol mass is deposited by a uniform air sample flow rate using a small pump. The optical attenuation is converted into mass using a set of wavelength dependant calibration factors for specific absorption efficiency at the respective wavelength (http://www.mageesci.com/Aethalometer_book_2009.pdf). Detailed report on the error analysis is given by *Moorthy and Babu* [2006].

[4] The spectral absorption of BC from fossil fuel sources peaks at 830 nm while other aerosol species have negligible absorption at this wavelength and hence the channel 880 nm close to this peak is considered as the standard channel for BC measurement. BC data from other channels can be considered as equivalent BC mass that will produce the same absorption at this wavelength. If there are sources

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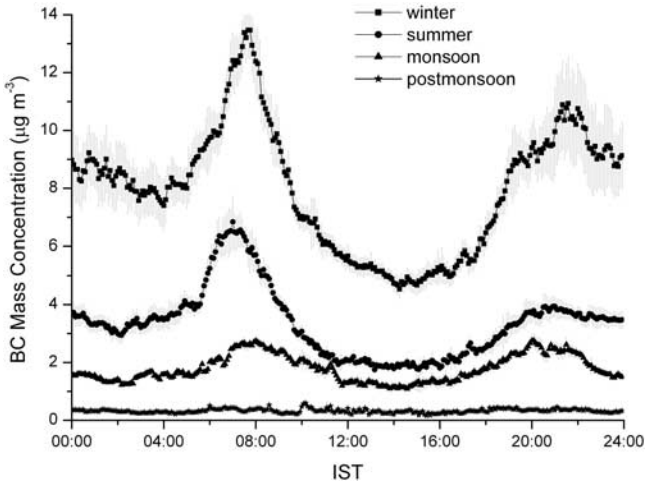

Figure 1. Season wise mean diurnal variation of near surface BC mass concentration.

other than fossil fuel that produce the absorbing aerosol, data from different channels show different BC mass concentrations and the spectral dependence of aerosol BC mass can be used for source identification [*Kirchstetter et al.*, 2004] as reported in section 3.2.

3. Results and Discussion

3.1. Temporal Variation in BC Mass Concentrations

[5] Near surface BC mass concentration data as measured by the aethalometer were sorted into four seasons namely Winter (November, December, January, February), summer (March, April, May), Monsoon (June, July, August) and Post Monsoon (September, October) and the season mean diurnal variation is shown in Figure 1 along with the standard deviations, which indicates a significant diurnal variation during the winter month with a morning peak around 0700 IST with BC mass concentration of 13.47 $\mu g/m^3$, a gradual decrease with minimum BC mass concentration of 4.76 $\mu g/m^3$ during the afternoon hours followed by an increasing trend after the local sunset with a nocturnal peak of 10.39 $\mu g/m^3$. The diurnal mean BC mass concentration was around 8.01 $\mu g/m^3$ in winter, 3.33 $\mu g/m^3$ in summer, 1.67 $\mu g/m^3$ in monsoon and 0.43 $\mu g/m^3$ during the post monsoon season. The prominence of this type of diurnal variation as well the magnitude of BC mass concentration decreases during the summer months with morning peak value of 6.55 $\mu g/m^3$, noon value of 1.79 $\mu g/m^3$ and nocturnal peak value of 3.94 $\mu g/m^3$. The ambient BC mass concentrations was low during monsoon months and during the post monsoon months practically no diurnal variability was observed in the near surface BC mass concentration. Figure 2 shows the colour map of the BC mass concentration during the whole period of observation which brings out the above mentioned features. One of the important features to be noted is the reduction in diurnal variation as we move from winter (December) towards the post monsoon season (September).

[6] The diurnal variations are associated with combined effect of variations in production, surface meteorology and the associated boundary layer dynamics, which is enhanced at coastal stations due to land sea breeze activity. Since the atmospheric boundary layer becomes shallower during night time than during day time and due to low wind speeds during the winter season, there is a rapid reduction in the ventilation of aerosols which results in confinement of aerosols and subsequent increase in the near surface mass concentrations during the night times. Shallow boundary layer acts as a capping inversion leading to the accumulation of aerosol in the near surface regions. As night progresses, due to the reduction in the anthropogenic activity, there is a reduction in the mass concentrations during early hours which subsequently increases with the increase in the anthropogenic activity during the morning hours. However, with the local sunrise and subsequent land surface heating, the confinement of aerosol breaks leading to the dispersion and decrease in the surface concentration takes place due to aerosol ventilation. The heating of the land surface increases the convective activity, vertical mixing and dilution of near surface aerosol concentration. This type of diurnal variation is weak during the summer months since the nocturnal boundary layer during this season does not come down to very low altitudes and hence the diurnal ratio between the early morning high aerosol concentration and the noon time minimum remains low. During the monsoon and post monsoon months, the atmosphere is generally wet with periodic cleansing of the atmosphere and hence the mean BC concentrations are low and the boundary layer dynamics are minimum during this season resulting in insignificant diurnal variation. However, the BC mass concentrations show a significant annual variation with a mean diurnal maximum (winter: 8.0) to minimum (Monsoon: 1.7)) ratio of 4.8. No other station has reported such a large annual variability in the BC mass concentration which is important in terms of regional radiative effects. Further it may be noted that the BC mass fraction was consistently 10% of the total fine mode particle mass as measured from a collocated near surface size segregated aerosol mass size distributions using a Quartz Crystal Microbalance, which indicates that the BC aerosol is well mixed in the ambient fine mode aerosol. During the post monsoon months, the wind is predominantly from eastern oceanic regions before it turns to North east in the winter months. Therefore, the station encounters a relatively pristine air from the Bay of Bengal and hence the BC mass concentrations were at their annual low.

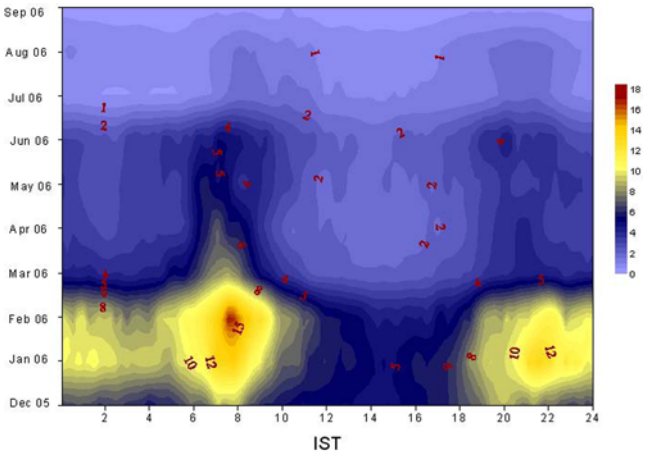

Figure 2. Color map of BC mass concentration against IST (Indian Standard Time) and month.

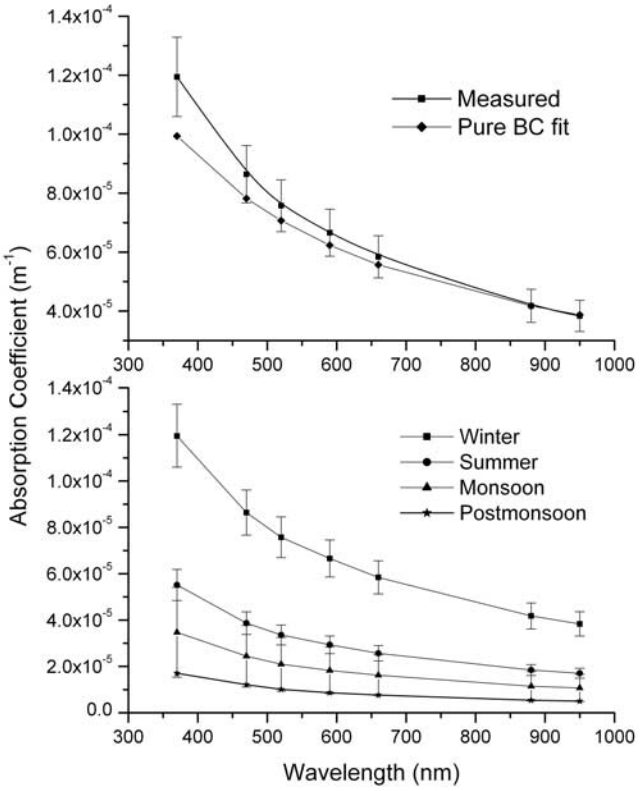

Figure 3. (top) Spectral variation absorption coefficient measured from Aethalometer for winter season and evaluated for BC (fossil fuel alone) from *Kirchstetter et al.*, [2004]. (bottom) Measured absorption coefficient for different seasons.

From earlier land campaigns covering the regions of the three states with 36 stations around the present location and from a systematic observation of aerosol physical properties over Kharagpur in the Indo-Gangetic plains, it was reported that the high pollution region in the northern India has the potential to effect the aerosol physical properties at southern locations in the peninsular India [*Niranjan et al.*, 2005, 2006] and hence the high aerosol BC mass concentrations could partly be contributed by the aerosol transport from north Indian regions via Bay of Bengal.

[7] It is observed that the BC mass observed over Visakhapatnam is due to the fossil fuel emission (please see section 3.2), which is mainly of local origin. During winter season, a fraction of the aerosol mass is expected to be contributed by long range transport from north Indian polluted region. But it is reported that the fraction of the BC mass in these regions during winter is also 10% [*Niranjan et al.*, 2006] and hence the BC mass fraction does not show a seasonal variability at Visakhapatnam. This is in contrast to the reports from other stations in India like Trivandrum and island station Port Blair where the BC aerosol mass fraction is seasonally dependant as the air mass pathways show continental aerosol transport over the observing locations during seasons when high BC mass fraction is observed.

3.2. Angstrom Absorption Coefficient

[8] Using the BC mass concentration measured at different wavelengths, the BC absorption coefficients at the operational wavelengths [$\beta_{abs}(\lambda)$] were calculated using the following equation [*Bodhaine*, 1995, *Weingartner et al.*, 2003]

$$\beta_{abs}(\lambda) = -\frac{1}{C.R}\frac{A.\ln(I_2/I_1)}{Q.\Delta t}$$

where I_1 and I_2 are the ratios of the intensities recorded by the detector for the sensing beam to the reference beam before and after each sampling interval of time Δt. The value of I_2/I_1 is given by $\Delta ATN = -100[\ln (I_2/I_1)]$, where ΔATN is the change in attenuation prior to and after the deposition of particles on the quartz filter tape, which is one of the aethalometer output parameters. Q is the volume of air sampled through the filter during interval Δt and A is the area of the spot where aerosols are collected. We have used the wavelength dependent values C(λ) obtained from the work of *Bodhaine* [1995] and R, an empirical correction factor describing the change in instrumental response with increased particle loading which is assumed to be equal to unity for this instrument due to its large dynamic range.

[9] The dependence of aerosol light absorption on wavelength is parameterized using a power law relationship: $\beta_{abs}(\lambda) = K.\lambda^{-\alpha}$ where $\beta_{abs}(\lambda)$ are the spectral absorption coefficients and K, α are Angstrom absorption coefficients. α is a measure of spectral dependence of aerosol absorption, which was determined by performing a linear regression of $\ln(\beta_{abs})$ and $\ln(\lambda)$. Figure 3 (bottom) shows the season-wise spectra for absorption coefficients. The values of α are found to be around 1.2 for all the seasons exhibiting a relatively weak wavelength dependence, confirming that light absorption by black carbon is generally considered to vary weakly ($\alpha = \sim 1$) with wavelength [*Bergstrom et al.*, 2002]. The value of $\alpha(\sim 1.2)$ here indicates that black carbon was the dominant absorbing aerosol component which is mostly due to motor vehicle exhausts and fossil fuel, typical of an urban environment.

[10] Following *Kirchstetter et al.* [2004], we have estimated the absorption due to non-BC aerosol species, keeping λ^{-1} dependence (pure BC component) as reference. Figure 3 (top) shows the typical spectrum of absorption coefficients and fit for pure BC component for winter 2006. The percentage absorption due to non-BC aerosols was not significant, with around 8% in winter, 9% in summer, 10% in monsoon and 12% in post monsoon, which indicates a constant small amount of absorbing aerosol other than black carbon in all the seasons due to the proximity of industrial area in the city. Previously, *Ganguly et al.* [2005] reported a maximum of 30% excess absorption during a winter month of 2004 by measuring the spectral dependence of absorption and BC mass concentrations at various locations over central India and inferred that major source of BC contributing to this excess absorption was not fossil fuel. From this study we may say that the variation in the excess absorption was much more location specific (spatial) rather than temporal.

3.3. BC Radiative Forcing Estimate

[11] Aerosol radiative forcing is the change in the net flux either at the top of the atmosphere or at the surface with and without aerosols. In the current study, the

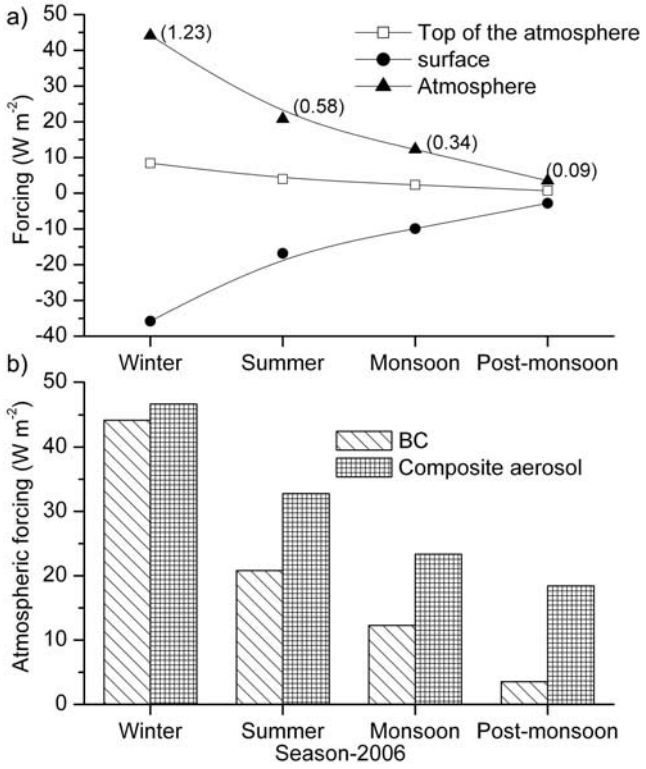

Figure 4. (a) Seasonal average shortwave BC radiative forcing. The values in parentheses indicate heating rates in Kelvins per day. (b) Atmospheric absorption due to BC and composite aerosol.

measured BC mass concentration was introduced as input into the OPAC model [*Hess et al.*, 1998] and the resultant optical effects in terms of the aerosol optical depth, phase function and single scattering albedo (SSA) due to the BC mass concentration alone were evaluated.

[12] The optical parameters thus derived from the above OPAC model were then fed to the Santa Barbara DISORT Atmospheric Radiative Transfer model (SBDART) developed by *Ricchiazzi et al.* [1998] to estimate the BC aerosol radiative forcing. As the location is a coastal one, the spectral albedo of the surface was taken as a combination of three basic surface types (ocean water, vegetation and sand) out of the available five types of SBDART. The shortwave aerosol forcing for all atmospheric layers calculated at 5° zenith interval are used to determine the diurnal averages. The diurnal averaged forcings for surface and top of the atmosphere are determined separately for all the seasons. The difference between the TOA and surface forcing is taken as atmospheric forcing. It represents the amount of energy trapped within the atmosphere by aerosols. The atmospheric heating rate was calculated following *Liou* [1980]

$$\frac{\partial T}{\partial t} = \frac{g}{c_p}\frac{\Delta F}{\Delta P}$$

where $\partial T/\partial t$ is the heating rate (s^{-1}), g is the acceleration due to gravity, *Cp* the specific heat capacity of air at constant pressure ($\sim$1006 J kg^{-1} K^{-1}) and P is the atmospheric pressure, respectively. However, to check the accuracy of this estimate, we have also computed the radiative forcing using composite aerosol mass concentrations measured with a collocated Quartz Crystal Microbalance with and without introducing the BC mass concentration and difference between the two is expected to give the BC forcing. It is observed that the results match with an accuracy of ±5%.

[13] Figure 4a shows the diurnally averaged BC aerosol forcings for all the seasons. The surface forcing during winter is as high as −35.78 W/m^2 and at the top of the atmosphere (TOA) is +8.4 W/m^2. The negative forcing values observed at the surface imply a net cooling effect and the positive sign for the TOA forcing arises due to the absorption by soot. The difference between the TOA and the surface gives the atmospheric forcing which is +44.178 W/m^2 indicating a net atmospheric absorption. This positive forcing represents a considerable amount of heating of the lower atmosphere and has been conjectured as potential factor causing global warming [*Jacobson*, 2001] during winter. As discussed in previous section the absorbing aerosol mass concentrations came down significantly as the seasons progressed from winter to post monsoon and hence the contributed AOD also came down which in turn resulted in the decline of all the forcing values for the subsequent seasons. They were −16.8 W/m^2 at surface, +3.99 W/m^2 at TOA during summer; −9.9 W/m^2 at surface, +2.36 W/m^2 at TOA during monsoon; −2.81 W/m^2 at surface, +0.7 W/m^2 during post monsoon. The corresponding values of heating rate in Kelvins per day were shown in Figure 4a (numbers in the parentheses) for each of the seasons. As it is usual convention to represent the forcing due to composite aerosol, to avoid confusion between BC forcing and composite aerosol forcing, the atmospheric absorption due to BC and composite aerosol are compared in Figure 4b, which indicates a 65% contribution of BC to composite aerosol forcing.

[14] The annual mean surface forcing at Visakhapatnam was −16.33 W/m^2, and the TOA forcing was +3.86 W/m^2 with an atmosphere forcing of +20.19 W/m^2. *Intergovernmental Panel on Climate Change* [2001] estimated the global mean clear sky radiative forcing due to BC was in between +0.4 to +0.8 W/m^2. The direct effect of positive atmospheric forcing can intensify low-level inversion, which slow down convection and in turn inhibits cloud formation. Large database over closely gridded stations are required in order to quantify the BC aerosol climatic effects over a particular region. Nevertheless in the present study we attempted to present the scenario of seasonal BC aerosol radiative forcing and the appended heating rates due to the presence of black carbon aerosol over a typical location on the east coast of India.

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