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Aerosol physical properties and Radiative forcing at the outflow region from the Indo-Gangetic plains during typical clear and hazy periods of wintertime

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[1] Results of a campaign mode measurements of column integrated aerosol optical depths, near surface mass concentrations and LIDAR profiles during winter 2004 at Kharagpur located at the vent of the out flow region from the Indo-Gangetic plains are presented with a view to characterize the atmospheric aerosol radiative forcing during hazy and clear sky conditions. The multispectral optical depths, surface mass concentrations and black carbon are high, while a decrease in Angstrom exponent value was observed during the hazy sky days. During hazy conditions, the LIDAR profiles show a decrease in the mixed layer height leading to confinement and subsidence of aerosols. Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model along with Optical Properties of Aerosols and Clouds (OPAC) are used to perform composite aerosol forcing estimation, incorporating LIDAR derived mixed layer heights. A considerable decrease in estimated single scattering albedo has been observed during hazy sky days. The atmospheric absorption during hazy sky days increased by 75% compared to clear days. Negative forcing was observed both at the top of the atmosphere and at the surface. The forcing estimates of the present study are compared with the similar studies over other locations in the Indo-Gangetic plains during the campaign period. **Citation:** Niranjan, K., V. Sreekanth, B. L. Madhavan, and K. Krishna Moorthy (2007), Aerosol physical properties and Radiative forcing at the outflow region from the Indo-Gangetic plains during typical clear and hazy periods of wintertime, *Geophys. Res. Lett.*, *34*, L19805, doi:10.1029/2007GL031224.

1. Introduction

[2] The effects of anthropogenic aerosols on climate constitute one of the biggest uncertainties in quantifying climate change due to large spatio temporal variability and the complex ways in which the aerosols impact climate. Transmission of solar radiation decreases exponentially with increasing Aerosol Optical Depth (AOD) and therefore, events of aerosol haze cause reduction in the spatial and temporal mean solar energy at the earth's surface and additional heating of the Planetary boundary layer. Recently *Ramanathan et al.* [2007] have found that atmospheric brown clouds enhanced lower atmospheric solar heating by about 50 per cent. However, current understanding on the physiochemical and optical properties of ambient aerosols associated with regional haze phenomenon is still fairly limited [*See et al.*, 2006]. Many Southeast Asian countries have been constantly plagued by recurring haze episodes during winter. More regionally in India, the southern edge of Himalayan region and the densely populated Indo-Gangetic plains are suffering with persistent aerosol haze as observed from satellite data [*Kaufman et al.*, 2002] resulting in solar radiative disturbance over substantial fraction of the region during dry winter monsoon period [*Ramanathan et al.*, 2001]. The haze consists of aerosols and particulate matter that have both direct effects like the reduction in the surface solar irradiance, which are restricted to regions beneath the haze layer and indirect effects that include cooling of land surface, increase in frequency and strength of thermal inversion that can trap more pollution, reduction in winter time average rainfall, reduction in evaporation etc. [*United Nations Environment Programme Center for Clouds and Chemistry Climate*, 2002]. In order to characterize the said aerosol haze, Indian Space Research Organisation (ISRO) conducted a major land campaign at eight stationary locations covering the entire Indo-Gangetic plains during December 2004. Various complementary instruments have been set up at these locations for systematic observations of aerosol physical and optical properties and their variations during clear, hazy and foggy periods. Characterization of aerosol parameters and their radiative effects during hazy and foggy conditions are equally important, as high humid conditions and fog droplets have the potential to alter the characteristics of aerosols by aqueous phase chemical reactions. Extensive measurements on aerosol optical and physical properties and aerosol vertical profiling using a Micro Pulse LIDAR have been made at Kharagpur (one of the eight identified sites) keeping in view the importance of aerosol characterization in the outflow regions from the Indo-Gangetic Plains. The measurements were made at the Regional Remote Sensing Service Centre (RRSSC) of the Indian Space Research organization, located on the Indian Institute of Technology campus, Kharagpur. From a forward trajectory analysis, *Niranjan et al.* [2006] found that the north Indian haze has the potential to pollute the oceanic regions surrounding sub-continent and southern parts of peninsular India, and as Kharagpur is located under the vent of the outflow region from the Indo-Gangetic plains, the current study assumes importance in the regional characterization of aerosol haze. Over Kharagpur, the first half of the campaign period was characterized by clear sky conditions and during most of

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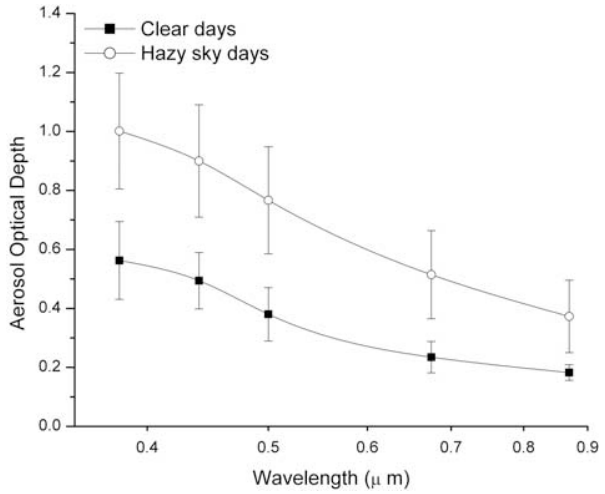

Figure 1. Mean Aerosol Optical Depth spectra for clear and hazy sky days.

the later part hazy sky conditions prevailed. The total period of observation is classified into clear and hazy periods based on the surface meteorological parameters and visibility. By and large clear days are characterized by 10 Km visibility, ~45% relative humidity (RH) and ~30°C ambient temperatures, while hazy sky days by less than 2–3 Km visibility, ~68% relative humidity and ~20°C ambient temperatures. All the data was screened for cloud free conditions. No events of fog were observed at Kharagpur during the campaign period.

2. Instrumentation and Data Acquisition Protocol

[3] The campaign was conducted during the whole month of December 2004 and the following measurements were made: i) aerosol spectral optical depth at 5 wavelengths viz., 380, 440, 500, 675 and 870 nm using a Microtops Sun Photometer (Solar Light Co, USA), with a Global Positioning System (GPS) receiver attached with the Photometer to provide information on the location, altitude and pressure, ii) near surface aerosol mass – size distributions using a ten channel Quartz Crystal Microbalance (QCM) Impactor (California Measurements Inc., USA), with aerodynamic cut-off diameters at 25, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1 and 0.05 μm respectively, (iii) near real time mass concentration of Black carbon (BC) using an Aethalometer (Magee Scientific, USA) and (iv) vertical profiles of aerosol back scatter intensity using a SESI Micro Pulse LIDAR (MPL) system at 532 nm, which employs an optical transceiver that acts as both transmitter and receiver (telescope) consisting of a pulsating Nd:YAG/Nd:YLF laser, Si-APD photon counting detector, signal processing unit and data processor (Multi Channel Scalar). The observations were started on the 2nd of December at Kharagpur and were continued till 27th of December 2004. The time resolution of AOD measurements was every half hour (day time only) and that of the QCM system was 1 hour, round the clock. The time resolution of BC mass concentration was 5 min, round the clock. The MPL was operated every evening from 17:00 hrs to 20:00 hrs. A total of 7 clear days and 14 hazy sky days data are considered for this study.

3. Results and Discussion

3.1. Aerosol Optical Depth and Surface Mass Concentration

[4] The simplest and in principle the most accurate and easy to measure aerosol parameter is the optical depth which is the single most comprehensive index to remotely assess the aerosol burden in the atmosphere from ground based measurements. Figure 1 shows the diurnal mean aerosol optical depth spectra using Microtops measurements for clear and hazy sky days. The vertical bars represent the $\pm 1\sigma$ deviation from the mean value. The AOD at 500 nm during hazy sky days (0.76) is double than that of clear days (0.38). A non-uniform increase in AODs has been taken place during hazy sky days when compared to clear days which resulted in the decrease in Angstrom size index α. As the RH during hazy sky days is around 68%, significant hygroscopic particle growth is expected [*Frank et al.*, 1998] which might be the reason for a decrease in α from 1.47 during clear days to 1.22 during hazy sky days. It may be noticed that though there is a decrease in the value of α, it has not decreased below 1.0 suggesting the domination of fine mode particle concentration. The increase in length of the error bars during hazy sky days suggests a high variability of AOD compared to that of clear days. During the same campaign over Delhi, *Ganguly et al.* [2006] reported a mean AOD value at 500 nm as 0.91 during hazy periods which is significantly higher compared to the present study as that location is a highly polluted and one of the mega cities of India.

[5] Figure 2a shows the mean surface aerosol mass in the nucleation/fine mode (geometric mean radius <0.1 μm), accumulation mode (geometric mean radius between 0.1 and 1 μm) and coarse mode (geometric mean radius >1 μm) evaluated from the QCM measurements and the surface BC mass during both clear and hazy conditions. The total mass concentrations were around 100 $\mu g/m^3$ and 225 $\mu g/m^3$ during clear and hazy sky days respectively. There is an overall increase in mass concentrations in all modes including black carbon aerosol during hazy sky days, but there was a significant increase in accumulation mode aerosol concentration during hazy sky days. During hazy conditions, the prevailing weather indicated higher humidity levels making conditions favorable for growth of hygroscopic aerosols as well as for the formation of new particles by condensation and nucleation by gas to particle conversion from the precursors emitted due to anthropogenic activity. According to *Intergovernmental Panel on Climate Change* [2001] the water uptake by aerosols is the key uncertain in aerosol direct effect. From the results of a comprehensive field campaign over Singapore between March 2001 and March 2002, *See et al.* [2006] reported that on hazy sky days, the time integrated mass concentration of PM2.5, ions, metals, EC and OC were a factor of 2 higher than on clear days. The black carbon aerosol mass concentrations has also increased from ~12 $\mu g/m^3$ to ~22 $\mu g/m^3$ from clear to hazy sky days, while *Ramachandran et al.* [2006] reported a marginal increase from ~1.6 $\mu g/m^3$ to ~2.3 $\mu g/m^3$ over a semi urban north Indian site Hisar. An

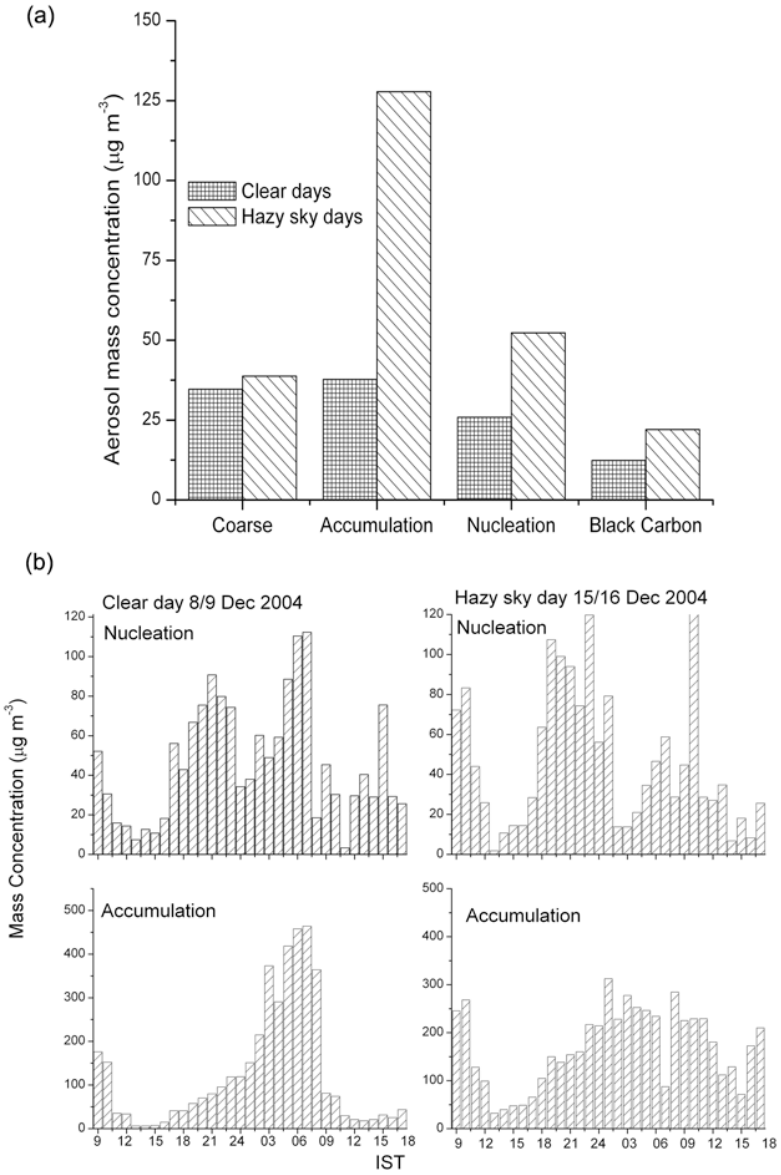

Figure 2. (a) Mean near surface aerosol mass concentration in the nucleation, accumulation and coarse mode along with BC mass concentration during clear and hazy sky days. (b) Diurnal variation of nucleation and accumulation mode aerosol mass concentrations during (left) clear days of 8–9 December 2004 and (right) hazy sky days of 15–16 December 2004.

increase in BC mass fractions to the total mass concentrations from 8% to ~12% has also been observed, which might partially be due to the increased anthropogenic activity such as fossil fuel, burning of wood etc. on the road side by the local people for generating heat during hazy sky days which are characterized by low ambient surface temperatures of ~20°C.

[6] In Figure 2b are shown the typical diurnal variations of the nucleation and accumulation mode aerosol mass concentrations on a typical clear (8/9 Dec. 2004 left column) and hazy (15/16 Dec. 2004 right column) days. The time axis starts at 0900 IST of the first day of observations (8th and 15th Dec. 2004 respectively) and were continued till the next evening 17:00 IST. A substantial increase in the near surface mass concentration in the accumulation mode was observed on the first hazy day, i.e., 15th December 2004 compared to the clear day concentrations on the 8th and 9th. Another important point to be noticed is that the surface mass concentrations increased further to as high as 150 μg m^{-3} on the second hazy day. A possible explanation for such an increase is given based on the aerosol radiative forcing in section 3.3.

3.2. LIDAR Measured Aerosol Backscatter Profiles

[7] Figure 3 shows typical mean vertical aerosol backscatter intensity profile during a clear and hazy day. The horizontal bars indicate the $\pm 1\sigma$ deviation of aerosol backscatter intensity. The ambient surface mean temperatures during hazy periods are quite low, inhibiting the effective aerosol convective activity compared to clear days, there by

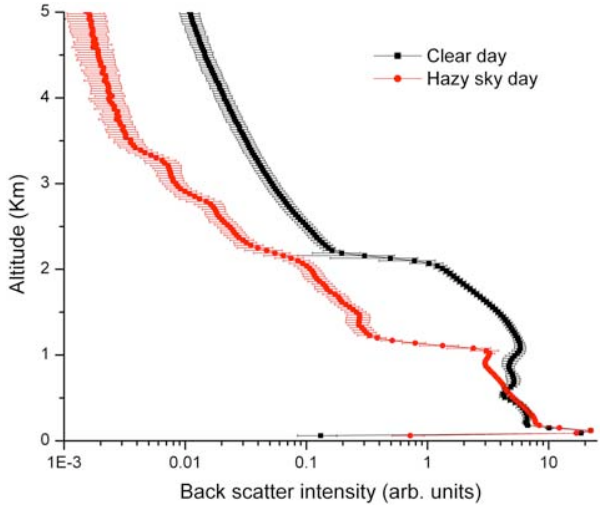

Figure 3. LIDAR mean vertical backscatter intensity profiles up to 5 Km for a typical clear and hazy day between 18:00 and 19:00 hours.

reducing the ventilation coefficient. As a result of it, strong low level inversions (capping) take place close to the Earth's surface which traps the locally produced aerosols within the surface layer causing an increase in surface extinction coefficient. Also the enhanced amount of humidity makes the particles grow and constrains its vertical extent gravitationally, which is reflected as an increase in the near surface accumulation mode aerosol concentration. During clear days significant backscatter has been observed up to 2.5 Km, while it is suppressed within an altitude of 1.5 Km during hazy sky days. The LIDAR derived mean mixed layer heights are 0.63 and 0.43 Km during clear and hazy conditions respectively. The backscatter intensity during hazy sky days is higher than that on clear days up to 0.5 Km as large particles scatter more compared to fine particles. Over Delhi, *Ganguly et al.* [2006] found that on hazy and foggy days, aerosol extinction becomes almost negligible above 0.5 Km from ground level while on clear days it asymptotically approaches zero value above this height.

3.3. Aerosol Radiative Forcing Estimate

[8] 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 OPAC (Optical Properties of Aerosols and Clouds) aerosol model developed by *Hess et al.* [1998] was used in estimating the optical properties of the composite aerosols. The measured optical depth spectrum was reconstructed by constraining the mass concentration of soot to the measured daytime mean value and varying the number concentration of other two components (water soluble and insoluble) which are chosen as chief components for the present location. The AOD's are reconstructed iteratively until the modeled values and measured values match within ±5% deviation. Principal optical parameters viz., the aerosol optical depth, single scattering albedo, and phase function are derived separately for clear and hazy sky days by incorporating LIDAR derived mean mixed layer heights and at 50% and 70% relative humidity respectively.

[9] The optical and radiative properties derived from the above model were then fed to the Santa Barbara DISORT Atmospheric Radiative Transfer model (SBDART) developed by *Ricchiazzi et al.* [1998] to derive the aerosol radiative forcing. The shortwave (0.2–4.0 μm) radiative forcing estimation was performed separately assuming the tropical atmosphere, with and without aerosol. As the location is a semi-urban and semi-arid type, the spectral albedo of the surface was taken as a combination of two basic surface types (vegetation and sand) out of the available five types of SBDART. The maximum excursion of the zenith angle over the location for the period of observation is computed using sun declination, hour angle and the equation of time from the standard astronomical ephemeris. The aerosol forcing for all atmospheric layers [*Hess et al.*, 1998] calculated at 5° zenith interval within the excursion range is used to determine the diurnal averages. The diurnal averaged forcing for surface and top of the atmosphere are determined and the difference between the TOA and surface forcing is taken as atmospheric forcing, which represents the amount of energy trapped within the atmosphere due to the presence of aerosols (Figure 4).

[10] During clear, cloud free sky conditions, the surface aerosol forcing is ~ -54 W/m^2, top of the atmosphere forcing is -4.53 W/m^2. The hazy sky days are characterized by large negative surface forcing value of ~ -85 W/m^2 as the ground reaching solar flux was further inhibited due to the increase in overall size of the particles, as large particles scatter solar radiation back to space more effectively when compared to the fine particles. Also the OPAC derived single scattering albedo value at 500 nm is relatively high (0.88) during hazy sky days than on clear days (0.8538) supporting the above observation. The energy trapped within the atmosphere due to the presence of aerosols is 50 W/m^2 and 76 W/m^2 respectively during clear and hazy sky days. The large positive atmospheric forcing value during hazy sky days is due to the combined effect of enhanced BC mass fraction, hygroscopic growth of aerosols and their confinement in the lower altitudes. This enhance-

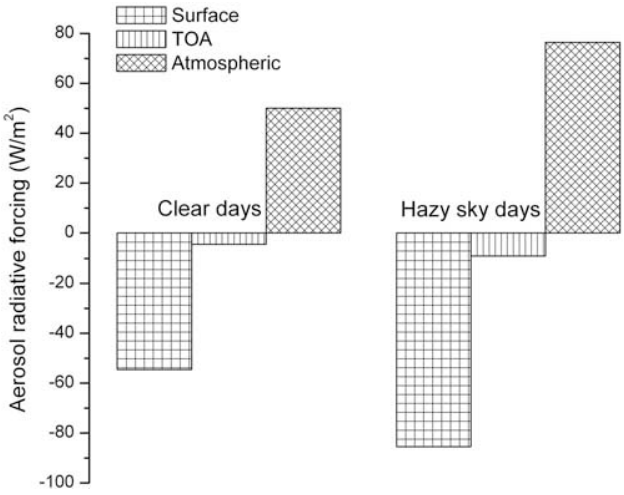

Figure 4. Composite aerosol radiative forcing for clear and hazy sky days.

Table 1. Parameters Representing the Aerosol Radiative Forcing at Different Locations in the Indo-Gangetic Plain During the Same Observational Campaign

Aerosol Radiative Forcing	Kharagpur (Semi-Urban/ Semi-Arid)		Hisar (Semi-Urban)		Delhi (Polluted North Indian City)	
	Clear	Hazy	Clear	Foggy	Clear	Hazy
Surface forcing, W/m^2	−54	−85	−21	−45	−51	−75
TOA forcing, W/m^2	−4.53	−9	−3	3	0.9	1.9
Atmospheric forcing, W/m^2	50	76	18	49	52	77

ment in heating up of lower atmosphere can further strengthen low level inversions during winter months restricting aerosols to be trapped near the earth's surface. This entire process may become a cyclic phenomenon and there is possibility for accumulation of more and more aerosols at the surface layer as soon as the phenomenon onsets, which is reflected in a further increase in the accumulation mode aerosol concentration on the second day (16 Dec. 2004) shown in Figure 2b. Also the aqueous phase chemical reactions during hazy and foggy days make the aerosol radiative effect more complex. Therefore, once a hazy event builds up, there is a probability for accretion of more amount of accumulation mode aerosol (result of hygroscopic growth) as the days progressed in the hazy episode as was observed during the later half of the campaign period. This extended duration of haze events break due to the inhibition of the progressive growth partly due to sedimentation and fall out mechanisms, break-up of the low-level capping by solar flux and when surface winds set in.

[11] The parameters representing the aerosol SW radiative forcing at Kharagpur are compared with values of similar studies from other locations in the Gangetic plains during the same observational period and are shown in Table 1, which show that the radiative forcing at Kharagpur (present study) is higher than that over Hisar [*Ramachandran et al.*, 2006] and is comparable to Delhi [*Ganguly et al.*, 2006].

4. Summary

[12] 1. During hazy sky days column AOD at 500 nm is as high as 0.76 compared to clear day value of 0.38. A decrease in the aerosol size index 'α' during hazy sky days indicated the relative dominance of coarse mode particles.

[13] 2. The near surface mean mass concentrations during clear days is ~100 μg/m^3, while during hazy sky days it is ~225 μg/m^3 due to subsidence and confinement of aerosols.

[14] 3. The LIDAR derived mean mixed layer height is about 0.63 Km and 0.43 Km during clear and hazy sky days respectively indicating a clear subsidence during hazy sky days.

[15] 4. The reduction in surface reaching solar flux due to the presence aerosols was ~54 W/m^2 during clear days and as high as ~85 W/m^2 under hazy conditions.

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