1 **Influence of natural and anthropogenic emissions on aerosol optical properties over**
2 **a tropical urban site – a study using sky radiometer and satellite data**

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16 **Abstract**

17 Atmospheric aerosols are important aspects of climate research due to their impact on
18 radiative forcing. In the present study, the aerosol optical depth (AOD), the Ångström
19 exponent (α) and the single scattering albedo (SSA) over the urban region of Hyderabad,
20 India, were examined using Sun/Sky radiometer measurements during January –
21 December, 2008. AOD showed higher values on certain Julian days coinciding with the
22 occurrence of wintertime dust storm events in the Gulf Region and biomass burning due
23 to forest fires over Indian Region. The AOD values during wintertime dust event are
24 about ~55% higher than those on normal days. The SSAs show positive and negative
25 trends in α ($R^2 = 0.71$) and black carbon (BC) aerosols ($R^2 = 0.44$), respectively. The
26 aerosol size distribution shows a bimodal pattern with fine (~0.15 μ m) and coarse (~>7
27 μ m) mode during January –December, 2008. The MODIS AOD showed positive
28 correlation with sky radiometer-derived AOD values ($R^2 = 0.68$).
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30 **Keywords:** AOD, SSA, forest fires, Dust storms, black carbon
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1. Introduction

In the current assessment and prediction of global climate change, atmospheric aerosol concentrations and their optical properties are one of the largest sources of uncertainty (Hansen et al., 2000). Aerosol interactions with solar radiations results in both direct radiative forcing and indirect effects on clouds involving droplet properties, cloud dynamics and lifetime. Direct radiative forcing from aerosols is primarily a function of aerosol concentration in the total atmospheric column (aerosol optical depth), particle size distributions, and aerosol absorption properties (Eck et al., 2005). An accurate knowledge of the spatial and temporal extent of aerosol concentrations and properties is necessary for understanding the role of aerosols on climate (Hatzianastassiou et al., 2005). In this regard, several extensive observations on the optical properties of atmospheric aerosols were made using Sun/Sky radiometer at global scale (Liu et al., 2008; Cazorla et al., 2008; Kim et al., 2007; Arai and Liang, 2007; Cheng et al., 2006; Pandithurai et al., 2007). The best strategy for characterizing aerosols is to integrate measurements from different satellite sensors and ground-based measurements and models. Aerosols produced at one location can even reach far-off regions owing to prevailing transport mechanisms. Anthropogenic activities such as biomass burning, industrial emissions and vehicular combustion can influence the characteristics and distribution of aerosols. Several attempts are being made all over the world to characterize aerosols on regional basis and to form realistic and comprehensive models of aerosol parameters. Several experiments to characterize atmospheric aerosols were conducted over mid-latitudes and/or oceanic environments (Nair et al., 2004a; Ramanathan et al., 2001), while only few studies have been reported from tropical continental sites where spatial inhomogeneties are more pronounced.

Mineral dust is considered as one of the major contributors to the aerosol loading in the troposphere influencing the seasonal variability of the aerosol optical properties and the local radiative forcing (Durant et al., 2009; Liu et al., 2003). Dust storms have considerable impacts on climate variability, nutrient dynamics and biogeochemical cycles of oceans, soil characteristics, and ambient air quality. Satellite and surface observations have shown that the global sources of the atmospheric dusts are the arid and semi-arid desert regions contributing to the long-range transport of dust particles lifted by strong surface winds ($>5 \text{ m s}^{-1}$) (Prospero et al., 2002; Washington et al., 2003).

1 The present study describes the columnar aerosol properties over the urban region of
2 Hyderabad, India, using ground-based sun/sky radiometer measurements, satellite data
3 and **on** global three-dimensional aerosol transport model.

4 5 **2. Study Area**

6 The study area of Hyderabad, India, is located between 17° 10' and 17° 50' N latitude and
7 78° 10' and 78° 50' E longitude. Hyderabad is the fifth largest city of India, highly
8 urbanized with population of 5,751,780 inhabitants (census 2001). The city is influenced
9 not only by vehicular pollutants, but also by industrial as well as biomass-burning
10 emissions. The climate of the region is semi-arid with a total rainfall of ~700 mm
11 occurring mostly during the monsoon season in the June-October period. The climatology
12 of the area experiences four dominant seasons each year, winter (December-February),
13 pre-monsoon (March-May), monsoon (June-September), and post - monsoon (October-
14 November). The measurements reported in this work were carried out under clear-sky
15 conditions in the premises of the National Remote Sensing Centre (NRSC) campus
16 located at Balanagar (17° 28' N and 78° 26' E) well within the urban center of
17 Hyderabad.

18 19 **3. Datasets and Methodology**

20 Extensive aerosol observations were carried out at the National Remote Sensing Centre,
21 Hyderabad, India, using a Prede sun/sky radiometer (POM-01L, Prede Incorporated,
22 Japan) during January – December, 2008. This instrument is widely used in the SKYNET
23 aerosol radiation network (<http://atmos.cr.chiba-u.ac.jp/aerosol/skyNET>) for sun/sky
24 observations, which can be used to retrieve aerosol optical depth and Ångström exponent.
25 This instrument makes measurements of both direct and diffuse sky radiances at pre-
26 defined scattering angles at regular intervals. The sun/sky radiometer was operated from
27 January – December, 2008 on clear-sky days at 10-min intervals. The sun/sky radiance
28 data were analysed with the radiative transfer model SkyRad Pack version 4.2 (Nakajima
29 et al., 1996; <http://www.ccsr.u-tokyo.ac.jp/~clastr/>) to retrieve aerosol optical depth
30 (AOD), single scattering albedo (SSA), Ångström exponent (α) and volume size
31 distribution. AOD is calculated on the basis of the Beer-Lambert-Bouger law with
32 correction for Rayleigh scattering, the change of Sun-Earth distance, and ozone optical
33 depth by considering the average columnar Ozone value of 300 DU. The sky radiometer
34 was operated in disc scan mode to estimate the solid view angles at different wavelengths
35 as part of a calibration procedure recommended by the manufacturer. Detailed calibration

and data reduction procedures for this instrument are described elsewhere (Nakajima et al., 1996; Pandithurai et al., 2007).

The Ångström exponent was derived from the spectral dependence of AOD (τ_a) observed at 400, 500, 675, 870 and 1020-nm wavelengths using Ångström's formula (Ångström, 1964):

$$\tau_a(\lambda) = \beta \lambda^{-\alpha}$$

where τ_a is the estimated AOD at the wavelength λ , β is the Ångström's turbidity coefficient, which equals AOD at $\lambda = 1$ m, and α is the Ångström exponent.

Continuous and near-real-time measurements of black carbon (BC) aerosol mass concentration were carried out using an aethalometer; model AE-21 of Magee Scientific, USA (Borak et al., 2003). The instrument takes ambient air from an altitude of 3 m above the ground using its inlet tube and pump. The BC mass concentration is estimated by measuring the change in the transmittance of a quartz filter tape, on to which the particles impinge. The instrument was operated at a time base of 5 min, round the clock with a flow rate of 2.6 LPM. The instrument has been factory-calibrated and errors in the measurements are $\pm 2\%$.

In addition, MODIS active fire locations products from web fire maps (<http://maps.geog.umd.edu/activefire.html/>) were used for the study period. MODIS Fire detection is performed using a contextual algorithm (Giglio et al., 2003) that exploits the strong emission of mid-infrared radiation from fires. The algorithm uses brightness temperatures derived from the MODIS 4- and 11- μ m channels. The description of MODIS fire products generation is given elsewhere (Giglio et al., 2003). NOAA HYSPLIT model runs and meteorological fields from NCEP reanalysis data were used to study the air mass trajectories over the region (Draxler and Rolph, 2003).

4. Results and Discussion

Figure – 1 (a – c) shows the variation of daily mean AOD, SSA at 500 nm and α with standard deviation of the daily averaged value derived from sky radiometer during January – December, 2008. There are some gaps in the time series due to prevailing

atmospheric conditions, which did not provide enough clear-sky days for measurements. The AOD values showed an increase on certain Julian days (i.e., 33, 49-52, 56 and 68), as shown in **Figure – 1a**. The high AOD values during Julian days 49-52, 56 are associated with biomass-burning activity over the Indian region as shown in MODIS fire images of **Figure – 6**. The AOD during wintertime dust event (Julian day 33) was about 50-55% higher than the optical depth obtained on normal days. SSA is a useful quantitative parameter for estimating radiative properties of aerosols (Lewis et al., 2008). The SSA values at 500 nm varied between 0.9-1.0 over the region during the study period as shown in **Figure – 2b**. The SSA values derived from PREDE skyradiometer at different wavelengths are summarized in **Figure – 3**. The day-to-day variations observed in SSA were related to meteorological conditions, variations in air mass trajectory, relative humidity, temperature inversions, biomass burning episodes and dust particles driven by high winds over the region. The lower values of SSA on certain days are mainly due to presence of absorbing aerosols over the region. The gradual decrease in α values were observed on certain days during February – March due to the dominance of coarse mode particles associated with wintertime dust storm events over the region (**Figure – 1f**). α is a good indicator of aerosol-size distribution and its variation. The magnitude of α is determined by the fraction ratio of fine and coarse modes. The α values are small ($\sim < 1.0$) during the dominance of coarse mode particles (Eck et al., 2005).

Figure – 4 (a – b) shows the scatter plots of SSA at a wavelength of 500 nm (SSA_{500}) vs α and BC aerosol mass concentration over the study area. α showed linear relation ($R^2 = 0.71$) with SSA_{500} suggesting that higher SSA_{500} values are associated with fine-mode particles. SSA_{500} showed negative correlation with BC aerosol mass concentration ($R^2 = 0.44$) (**Figure – 4b**) suggesting that low SSA_{500} values were due to absorbing aerosols over the region. The instantaneous BC values were observed to be higher than 40 g/m^3 during the biomass-burning period; BC values less than 40 g/m^3 are considered in **Figure – 4b**. Earlier studies based on aircraft measurements over the study site suggested an elevated layer of BC aerosols up to 2 km height during winter period (Krishna Moorthy et al., 2004). In urban regions, there is a possibility of elevated aerosol layers occurring due to heat island formation and related convective activity.

Terra and Aqua MODIS (Moderate Resolution Imaging Spectroradiometer) true color composite images were analyzed to study the intense dust aerosol loading over the

1 Arabian Sea on 2nd February and 8th March, 2008 (Figures – 5 (a & b)). Wintertime dust-
2 storms events over Asian region are the main source of dust aerosol loading in to the
3 atmosphere and these dust aerosols are transported at long distances under favorable wind
4 conditions (Kim et al., 2008). It is evident from Figure – 5 that the entire Gulf region and
5 Arabian Sea were under influence of dust aerosols on 2nd February and 8th March, 2008.
6 These dust aerosols were transported downwind and affected the atmospheric aerosol
7 properties over the measurement site as shown in Figure – 1. Five-day back trajectories
8 from NOAA HYSPLIT model were analyzed to study the role of long-range transport
9 over the urban region of Hyderabad during dust storm and biomass-burning events over
10 the region. Figure – 6 shows the 5-day backward trajectories for 2nd February (Julian day
11 33) and 8th March, 2008 (Julian day 68), respectively. The back trajectories suggest that
12 air mass from north-western region having dust storms and biomass burning due to forest
13 fires was advected over the study site resulting in increased columnar aerosol mass
14 concentration. The increase in columnar loading of absorbing aerosols resulted in
15 decrease in SSA values shown in Figure – 2. Earlier studies over the region reported
16 long-range transport of aerosols from biomass burning and dust storm over the region
17 (Kharol et al., 2006; Badarinath et al., 2007a & b).

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19 Figure – 8 and 9 shows the sky-radiometer-derived monthly and seasonal variations in
20 aerosol volume-size distribution during 2008. The aerosol volume-size distribution has
21 bimodal structure with one peak at about 0.15 μm showing a maximum value in February
22 due to anthropogenic emissions and biomass burning due to forest fires. The second peak
23 with a radius of about 7 μm in winter is due to coarse-mode particles over the region due
24 to dust events as shown in Figure – 5. The coarse-mode aerosol volume-size distribution
25 derived from inversion of PREDE skyradiometer are reported to differ from CIMEL
26 skyradiometer (Che et al., 2008); the possible discrepancies due to the SKYRAD pack are
27 under further investigated.

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29 Sky radiometer AOD at 500-nm wavelength is converted to 550 nm (AOD_{550}) using
30 Ångström's formula for comparing it with Terra MODIS derived AOD at 550 nm.
31 (Ångström, 1964). Figure – 10 shows the scatter plot of Terra MODIS AOD_{550} and sky
32 radiometer-derived AOD_{550} suggesting a correlation ($R^2 = 0.68$).and the deviation has
33 been attributed to spatial resolution of MODIS sensor besides other governing factors.
34 Similar observation over Hong Kong ($R^2 = 0.625$) was reported in the literature (Cheng et
35 al., 2006).

5. Conclusions

Aerosol optical depth (AOD), Ångström exponent (α), single scattering albedo (SSA) and volume-size distributions were investigated using satellite data and sky radiometer data at the urban region of Hyderabad, India, during January-December, 2008. The results of study suggested that –

- α showed positive correlation with SSA, suggesting that increase in fine mode aerosol loading from anthropogenic sources results in lower SSA values over the study region. The variations in SSA suggests absorbing aerosol loading over the region associated with biomass burning and dust loading during pre-monsoon months.
- Increase in black carbon aerosol mass concentration due to vehicular emissions coupled with emissions from forest fires and dust storms results in decreased SSA values over the region on certain Julian days.
- Sky radiometer derived AOD showed good correlation ($R^2 = 0.68$) with MODIS-derived AOD values.
- Dust outbreaks from the Persian Gulf region and biomass burning due to forest fires play a vital role in modifying atmospheric aerosol properties over the study site. The aerosol properties from ground based measurements and global aerosol transport model are useful for estimating aerosol radiative forcing as more spatial locations on observations may reduce the uncertainties.

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Figure captions

Figure – 1 (a-e) Variations of sky radiometer-derived AOD at different wavelengths (f) Ångström exponent (α) (The symbols indicates mean and standard deviation.) during 2008.

Figure – 2 As in Fig.1, but for SSA.

Figure-3 Variation of SSA at different wavelengths in 2008.

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Figure – 4 (a –b) Scatter plot of (a) Ångström exponent vs SSA₅₀₀ and (b) BC vs SSA₅₀₀.

Figure – 5 (a) True Color Composites of Terra MODIS and (b) Aqua MODIS on 2nd February and 8th March, 2008 showing intense dust aerosol loading over the Arabian Sea.

Figure – 6 NOAA HYSPLIT 5-day backward trajectories ending at Hyderabad, India on 2nd February and 8th March, 2008

Figure – 7 MODIS active fires composite showing increased occurrence of fires during Julian days 41 – 70

Figure – 8 (a-f) Monthly average of aerosol volume size distribution during 2008.

Figure – 9 (a-d) Seasonal variations of aerosol volume size distribution during 2008.

Figure – 10 Scatter plot of Terra MODIS aerosol optical depth at 550 nm (AOD₅₅₀) vs Sky radiometer AOD₅₅₀.

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Figure 4

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NOAA HYSPLIT MODEL
Backward trajectories ending at 00 UTC 02 Feb 08
GDAS Meteorological Data

NOAA HYSPLIT MODEL
Backward trajectories ending at 00 UTC 08 Mar 08
GDAS Meteorological Data

Figure – 6

Figure – 7

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Figure – 8

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