# Investigation on Seasonal Variations of Aerosol Properties and its Influence on Radiative Forcing over an Urban Location in Central India

3	Subin Jose <sup>1</sup> , Biswadip Gharai <sup>*1</sup> , K. Niranjan <sup>2</sup> and P.V.N Rao <sup>1</sup>
4	<sup>1</sup> Atmospheric and Climate Science Group, National Remote Sensing Centre.
5	<sup>2</sup> Department of Physics, Andhra University.
6	
7	*Corresponding author: Biswadip Gharai
8	Email:g.biswadip@gmail.com
9	

# 10 Abstract:

11 Aerosol plays an important role in modulating solar radiation, which are of great concern in perspective of regional climate change. The study analysed the physical and optical properties of 12 13 aerosols over an urban area and estimated radiative effect using three years in-situ data from 14 sunphotometer, aethalometer and nephelometer as input to radiative transfer model. Aerosols properties indicate the dominance of fine mode aerosols over the study area. However presence 15 16 of coarse mode aerosols is also found during pre-monsoon [March-April-May]. Daily mean 17 aerosol optical depth showed a minimum during winter [Dec-Jan-Feb] (0.45-0.52) and a maximum during pre-monsoon(0.6-0.7), while single scattering albedo ( $\omega$ ) attains its maximum 18 19 (0.78±0.05) in winter and minimum (0.67±0.06)during pre-monsoon and asymmetry factor 20 varied in the range between 0.48±0.02 to 0.53±0.04. Episodic events of dust storm and biomass 21 burning are identified by analyzing intrinsic aerosol optical properties like scattering Ångström 22 exponent (SAE) and absorption Angström exponent (AAE) during the study periods and it has 23 been observed that during dust storm events  $\omega$  is lower (~0.77) than that of during biomass 24 burning(~0.81). The aerosol direct radiative effect at top of the atmosphere during winter is -11.72  $\pm 3.5$  Wm<sup>-2</sup>, while during pre-monsoon; it is -5.5  $\pm 2.5$  Wm<sup>-2</sup>, which can be due to observed 25 lower values of  $\omega$  during pre-monsoon. A large positive enhancement of atmospheric effect of 26

~50.53Wm<sup>-2</sup> is observed during pre-monsoon compared to winter. Due to high aerosol loading in
 pre-monsoon, a twofold negative surface forcing is also observed in comparison to winter.

29 Keywords: Aerosol Optical Depth, Single Scattering Albedo and aerosol direct radiative effect.

30 **1. Introduction** 

31 Atmospheric aerosols and their impact on climate have gained considerable attention from 32 scientific community and policy makers during past recent years [IPCC, 2013]. Aerosols effects 33 on climate are mainly classified into two viz. aerosol direct radiative effect (ADRE) and indirect 34 radiative effect. ADRE [Charlson et al., 1992] is associated with scattering or absorbing of 35 incoming solar radiation by aerosols thereby, producing a negative or positive radiative effect at Top of the Atmosphere (TOA). Scattering aerosols like sea salt, sulphate etc. induce climate 36 37 cooling, while absorbing aerosols like black carbon (BC) induce a warming effect at TOA [Bond 38 et al., 2013]. Indirect radiative effect is the mechanism by which aerosols alter microphysical and 39 radiative properties of clouds, thereby influencing albedo, lifetime and precipitation of clouds 40 [Ramanathan et al., 2001]. Jacobson [2001] has pointed that radiative properties of aerosols 41 depends also on its mixing state, that is the degree to which chemical components occur as 42 independent particles (external mixing) as compared to a component mixture in each individual 43 particle (internal mixing). Mixing state of aerosols will be extremely important while considering 44 impact due to long range transported aerosols [Chinnam et al., 2006]. Global estimation of ADRE is found to be varied from -0.85 to +0.15 Wm<sup>-2</sup>with an uncertainty of 1 Wm<sup>-2</sup> [IPCC 45 2013].Uncertainty involved in ADRE estimation is due to ambiguities associated with sources, 46 47 lifetime, distribution etc. [Anderson et al., 2003].

48 Uncertainty associated with ADRE can be minimized to some extend by their accurate 49 characterization [Quinn et al., 1998]. Towards this, many international and national efforts have 50 initiated like Aerosol Robotic Network (AERONET) [Holben et al., 1998], SKYNET [Takamura 51 and Nakajima, 2004], Aerosol Radiative Forcing Over India Network (ARFI Net) [Moorthy et 52 al., 2009] etc. to list a few. In addition to this, past decade have viewed a number a campaigns 53 for regional aerosol characterization over water bodies like INDOEX [Ramanathan et al., 2001]; 54 ACE-1 [Bates et al., 1998] and also over distinct land masses like ACE-Asia [Huebert et al., 55 2003]; SAFARI-2000 [King et al., 2003] etc. These efforts have brought out diverse physical and optical properties of varied aerosol species, which in turn has helped to minimize their 56 57 uncertainty in determining their radiative effect.

58 Because of their high spatio-temporal heterogeneity it is speculated that aerosols may be 59 more capable of altering atmospheric and oceanic circulation, especially on regional scale, than 60 greenhouse gases [Ming and Ramaswamy., 2011]. Present study aims at better estimation of 61 ADRE over an urban location, Hyderabad in Central India using in-situ data of 2010-2012 as 62 input to radiative transfer model. Novelty of this study is that critical aerosol optical parameters 63 (viz. Aerosol Optical Depth ( $\tau$  or AOD), Single Scattering Albedo ( $\omega$  or SSA) and asymmetry 64 factor (g) etc.) required for estimation of ADRE are either measured or derived from in-situ 65 observations.

Section 2 of this paper describe about study area, data sets and methodology. Sensitivity analysis of estimated ADRE is discussed in section 3. In Section 4, seasonal variation of aerosol radiative properties and associated radaitive effect are discussed. Further aerosol episodic events (viz. dust storm and biomass burning) which had impact on local aerosol system are identified and analyzed. Conclusions are presented in Section 5.

# 71 **2. S** 72

74

### 2. Study area, data set and methodology

# 73 **2.1** Study area

Hyderabad is the capital of newly formed state Telangana in Central India, with a population 75 76 of more than 4 million (http://censusindia.gov.in). Altitude of the study site is ~557 m above 77 mean sea level. It has a hot semi-arid steppe climate with four dominant seasons; winter (Dec-78 Jan-Feb), pre-monsoon (Mar-Apr-May), monsoon (Jun-Jul-Aug-Sep) and post-monsoon (Oct-79 Nov). Meteorological parameters recorded at India Meteorological Department (IMD) 80 (www.imd.gov.in), Begumpet, Hyderabad over a period of 1997 - 2012 is analysed. Study 81 revealed that long term daily mean of maximum (T<sub>max</sub>=36.1°C) and minimum temperatures 82  $(T_{min}=17.54^{\circ}C)$  are similar to that was observed during the study period of 2010-2012 (T<sub>max</sub>=36.0°C and T<sub>min</sub> 17.33°C). However, analysis of diurnal variation of temperature recorded 83 during study period shows that T<sub>max</sub> and T<sub>min</sub> are ~42°C and ~16°C, respectively. Long term 84 85 (1997-2012) mean relative humidity varies between 40 - 80% with lowest observed during pre-86 monsoon and maximum during monsoon. Local wind speed varies typically from 5 - 15ms<sup>-1</sup>, with a maximum observed during monsoon. Long term (1957-2012) annual mean of rainfall over 87 Hyderabad is ~827mm, while annual rainfall during study period 2010–2012 is found to be 1192 88 89 mm, 625 mm and 778 mm, respectively. Aerosol measurements are carried out at the campus of National Remote Sensing Centre (NRSC) (17.47° N, 78.43° E) located in heart of the city. The 90 91 sampling site is near to state high way and main sources of aerosols are from vehicular emission, 92 local/long range transport of dust aerosols, biomass burning and industrial emissions [Jose et al., 93 2015b].

94 2.2 Data set and methodology

96 2.2.1 Aerosol optical depth

97

95

98 Microtops II (Solar lights, USA) is a hand held sun photometer that measures spectral 99 aerosol optical depth  $\tau$  at five narrow spectral bands centered at 380, 440,500,675 and 870nm 100 with a wavelength precession of  $\pm 1.5$ nm and a full width at half maximum (FWHM) band pass 101 of 10nm. Details about design, calibration and performance of Microtops are detailed in Morys et 102 al.[2001]. It makes use of Beer-Lamberts-Bouguer law for estimation of  $\tau$ . In general 103 uncertainties associated with ' $\tau$ ' measurements by Microtops is <0.02 for lower wavelength 104 bands and <0.01 for higher bands [Porter et al., 2001]. In the present study, data were collected 105 only during clear days with a sampling frequency of 30 min.

106 Spectral dependence of  $\tau$  can be expressed using well-known Ångströms empirical relation 107 [Ångström, 1964,] where,  $\tau_{\lambda}$  is the aerosol optical depth at  $\lambda$ , ' $\beta$ ' is the Ångström turbidity 108 coefficient which equals  $\tau$  at  $\lambda=1\mu$ m and ' $\alpha$ ' is the Ångström exponent. ' $\alpha$ ' provides information 109 about aerosol size distribution and Ångström turbidity coefficient ( $\beta$ ) gives an idea about 110 atmospheric condition [Eck et al., 1999]. Daily observed  $\tau$  and derived  $\alpha$  is then averaged to 111 obtain a daily mean, which forms basic data set for further analysis.

112

#### 2.2.2 Aerosol scattering coefficient

A calibrated integrating Nephelometer (TSI- 3563, USA) is used to measure scattering coefficient of aerosols during the study period. It measures aerosol scattering and back scattering coefficient at three different wavelengths viz. 450, 550 and 700nm with high sensitivity and proven accuracy [Anderson et al., 1996]. Its principle of operation is well documented in literatures [Anderson et al., 1996]. It is operated at a flow rate of 20 LPM with a data averaging time of 300 sec during study period. We followed Anderson and Orgen, [1998] to correct measurement biases due to non-angular idealities. Parameter g, which is the mean value of Cosine of scattering angle over the total solid angle weighted by the phase function is calculated using an empirical relation relating backscattering fraction (b) provided by Andrews et al. [2006]. Uncertainty associated with g calculated using the above method is generally less than 10% [Niranjan et al., 2011].

# 124 2.2.3 Estimation of aerosol absorption coefficient.

125 Aerosol absorption measurements are carried out using a seven channel Aethalometer (AE-126 31, Magee Scientific, USA) for the same study period. It measures light attenuation at seven 127 wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) through a quartz filter matrix in which 128 particles get deposited. It is operated at a flow rate of 3 LPM with a data averaging time of 5min. 129 Differences in light transmission through particle laden spot and blank portion of filter are 130 attributed to attenuation, which is directly proportional to raw absorption coefficient [Hansen et 131 al., 1984]. There are several literatures explaining methodologies for rectifying the above raw 132 absorption coefficient [Arnott et al., 2005; Weingartner et al., 2003]. In this study we adopted 133 methodology proposed by Arnott et al. [2005]. The expected uncertainties in the estimation of  $\sigma_{abs}$  using this methodology are generally less than 10%. 134

Simultaneous measurements on absorption and scattering are then used to obtain aerosol single scattering albedo at 550 nm ( $\omega_{550}$ ), which is defined as the ratio of scattering to extinction at 550 nm.

138

### 139 2.3 Radiative transfer model

In this study we used Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART)
code [Ricchiazzi et al., 1998] for analyzing radiative transfer in short wave (SW) range (0.25-4.0

µm). It is one of the widely used models to compute radiative transfer in both clear and cloudy
atmospheres. Radiative transfer equations for a vertically inhomogeneous, non -isothermal,
plane-parallel atmosphereare numerically integrated in SBDART using DISORT (Discrete
Ordinate Radiative Transfer) radiative transfer module developed by Stamnes et al. [1988].
Present SBDART code allows up to 65 atmospheric layers and 40 radiation streams (40 zenith
angles and 40 azimuthal modes).

148 The main input parameters like spectral values of  $\tau$ ,  $\omega$  and angular phase function of 149 scattered radiation for SBDART are taken from in-situ observations. We have used MODIS 150 derived land surface products (MCD43) on black-sky albedo and white-sky albedo to calculate 151 actual albedo over Hyderabad required for radiative transfer calculation [Satheesh et al., 2010]. 152 We also used tropical atmospheric profiles for running the model. With these inputs, we run SBDART with and without aerosols for all clear sky days during the study period with a 153 154 frequency of 1hour and for 24 hours period. ADRE at TOA and surface, are then estimated by 155 taking difference between instantaneous changes in net solar radiation for with and without 156 aerosols skies [Russell et al., 1999]. Atmospheric effect which implies the amount of energy 157 trapped within the atmosphere due to aerosols is calculated by taking difference between TOA 158 and surface effect. Negative values of ADRE indicate cooling of Earth-Atmosphere system and 159 positive values indicates warming. Overall uncertainty in estimated effect is due to small 160 deviations in simulations and uncertainties in surface parameters used as input to SBDART. 161 Ricchiazzi et al. [1998] reported the accuracy of SBDART is within 3% when compared with 162 precise ground observations in SW range.

In this study, we analysed ARDF for two seasons viz. pre-monsoon and winter as all the concurrent essential *in-situ* input data for RT model run were available for these seasons.

#### 165 **3. Sensitivity analysis of ADRE**

166 Uncertainties involved in estimation of ADRE utilizing in-situ data depends greatly on the 167 quality of data fed in to the radiative transfer model. Uncertainty in ADRE estimation can be 168 achieved following McComiskey et al.[2008]

169 
$$\Delta(ADRE) = \sqrt{\left(\frac{\partial ADRE}{\partial \tau} * \Delta \tau\right)^2 + \left(\frac{\partial ADRE}{\partial \omega} * \Delta \omega\right)^2 + \left(\frac{\partial ADRE}{\partial g} * \Delta g\right)^2}$$
(1)

170 where,  $\partial ADRE/\partial \tau$ ,  $\partial ADRE/\partial \omega$  and  $\partial ADRE/\partial g$  represents sensitivity of  $\tau$ ,  $\omega$  and g respectively. 171 Mean value of  $\tau$ ,  $\omega$  and g observed during study period are independently constrained while 172 performing sensitivity analysis of each.  $\Delta \tau$ ,  $\Delta \omega$  and  $\Delta g$  are the uncertainties associated with 173 measurements of  $\tau$ ,  $\omega$  and g. Calculated uncertainty values for each parameter are ±0.02, ±0.03, 174 ±0.05, respectively and estimated  $\Delta ADRE$  using Equation (1) is 2.4 Wm<sup>-2</sup>.

#### 175 **4. Results and Discussions**

#### 176 *4.1 Temporal variation of aerosol radiative properties over Hyderabad.*

177 Key optical parameters that are required to understand aerosol complex interaction with 178 radiation are  $\tau$ ,  $\omega$  and Phase function [Haywood and Shine, 1995]. The  $\tau$  is vertical integral of 179 aerosol extinction coefficient from Earth's surface to TOA. The  $\omega$  is a function of particle size, 180 shape and its refractive index. Higher magnitude of  $\omega$  is considered as an index for relative 181 dominance of scattering properties of aerosols with respect to its absorption. Angular distribution 182 of scattering radiation is represented by phase function. Since, estimation of phase function is 183 mathematically complex; Henyey-Greenstein (HG) phase function [Henyey and Greenstein, 184 1941] is an often-used approximation for phase functions to speed up calculations in some 185 radiative transfer codes.

186 Monthly and seasonal variation of aerosol radiative properties ( $\tau$ ,  $\omega$ ,  $\alpha$  and g) at 550 nm over 187 Hyderabad is shown in Figure-1. Vertical bar represents standard deviation from mean. Figure 188 showed a significant seasonal variation of aerosol radiative properties during study period. Mean 189  $\tau_{550}$  showed a minimum (0.45-0.52) during winter and maximum (0.6-0.7) in pre-monsoon. Observed high values during pre-monsoon can be associated with high surface temperature 190 191 leading to aerosol lofting by convection coupled with lack of removal mechanism. Babu et al. 192 [2011] reported two elevated BC layers one at ~4.5 km, and another above 8 km over this region 193 using balloon experiment. Also dust storms and biomass burning are very frequent over Indian 194 region during this period [Gharai et al., 2013], and elevated layers of these aerosols can be 195 observed in locations away from source regions [Jose et al., 2015a]. Local re-suspended soil dust 196 also play a significant role in columnar aerosol loading during this period. One of the recent 197 studies [Babu et al., 2016] on vertical distribution of atmospheric aerosols over Indian mainland 198 have reported a clear enhancement in aerosol loading and its absorbing nature at lower free 199 troposphere (FT) levels (above the planetary boundary layer) during pre-monsoon compared to 200 winter. Model simulations of BC over Indian region [Kumar et al., 2015] also reported that BC 201 showed distinct but opposite seasonality in the lower troposphere (LT) and FT with BC showing 202 winter maximum and summer minimum in the LT and vice versa in the FT. Angström 203 parameter,  $\alpha$  (380-870nm), which represents spectral dependencies of AOD showed lowest 204 values ~0.56 during May-Jun (pre-monsoon) and maximum values >1.25 in Nov-Dec (post-205 monsoon to winter) during the study period. This large seasonality in  $\alpha$  could be due to 206 dominance of fine mode aerosols in winter and mixed with coarse mode aerosols in pre-monsoon months. Eck. et al. [2010] also reported similar seasonal variations of  $\alpha$ (440-870nm) over 207 208 Kanpur, India.

209 The  $\omega$  of aerosols during study period showed a maximum in winter and minimum in pre-210 monsoon. Observed mean values of  $\omega$  during pre-monsoon, and winter are 0.67±0.06, and 211  $0.78\pm0.05$ , respectively. This seasonal difference in  $\omega$  significantly affects TOA effect over this 212 region as a small difference in  $\omega$  can change the sign of TOA forcing [Hansen et al, 1997]. The  $\omega$ 213 observed over Hyderabad is comparable to those reported from other Indian urban cities, like 214 Ahmedabad [0.62-0.84], Delhi [0.63-0.72] and Vishakhapatnam [0.65-0.9] [Srivastava et al., 215 2011; Soni et al., 2010; Niranjan et al., 2011]. However, observed  $\omega$  over Hyderabad is lower 216 than those reported from rural locations [Montilla et al., 2011], which can be due to enhancement 217 in absorbing particles like BC over urban sites. Reported BC mass fraction to total suspended 218 particles (TSP) over Hyderabad varies from 5-15% [Sinha et al., 2013] which is quite high 219 compared to other sites of the country [Safai et al., 2013]. Pre-monsoon minimum in  $\omega$  can also 220 be associated with presence of more long range transported aerosols (biomass burning and dust) 221 in addition to local emissions, which are more absorbing in nature. Pandithurai et al. [2008] 222 reported a reduction in  $\omega$  from 0.84 to 0.74 over New Delhi from March to June, when dust 223 transports are more prominent.

Our analysis shows that during study period g varied in the range  $0.53\pm0.04$  in winter and 0.48±0.02 in pre-monsoon. The column averaged g values during pre-monsoon over other locations of the country are found to be  $0.44 \pm 0.05$  (Bhubaneshwar),  $0.48 \pm 0.03$  (Chennai),  $0.43 \pm 0.07$  (Trivandrum) and  $0.38 \pm 0.06$ (Goa), respectively [Ramachandran and Rajesh., 2006]. Sensitivity analysis of g with respect to TOA effect suggests that a 10% decrease in g corresponds to a 19% reduction in radiative effect at TOA and a 13 % reduction at the surface as reported by Niranjan et al. [2011].

231 4.2 Aerosol direct radiative effect (ADRE).

232 Daily ADRE is computed using SBDART with observed aerosol optical parameters as 233 primary inputs. Figure 2a shows averaged ADRE at TOA, Surface and Atmosphere during 234 winter and pre-monsoon seasons of the study period. Vertical lines on each bar represent the 235 respective standard deviation. It is observed from figure that aerosol radiative effect at TOA 236 during winter is  $-11.72 \pm 3.5$  Wm<sup>-2</sup>, while during pre-monsoon it becomes  $-5.5 \pm 2.5$  Wm<sup>-2</sup>. This 237 is largely due to seasonal variation of  $\omega$ , as it varies from 0.67±0.06 to 0.78±0.05 from pre-238 monsoon to winter. Figure 2b shows percentage occurrence of ADRE at TOA during winter and 239 pre-monsoon. Analysis reveals that during winter about 96% of the days ADRETOA are having 240 negative values indicating a cooling effect, while during pre-monsoon  $\sim 41\%$  of the days 241 ADRE<sub>TOA</sub> are having positive values. Corresponding effect at surface during winter and premonsoon are found to be -46.25±3.4 Wm<sup>-2</sup> and -90.56.4±2.4 Wm<sup>-2</sup>, respectively. Atmospheric 242 243 effect which represents energy trapped within the atmosphere is calculated as difference between 244 TOA and surface effect. Estimated atmospheric effect during winter and pre-monsoon over the 245 study period is found to be 34.52±6 Wm<sup>-2</sup> and 85.05±10 Wm<sup>-2</sup>, respectively. In the present study, heating rate is calculated following Liou et al. [2002]. Reported boundary layer height (BLH) 246 247 over this region is ~3.2 and 1.7 Km, respectively during pre-monsoon and winter [Mahalakshmi 248 et al., 2011]. Owing to maximum BLH of ~3 km, we considered pressure difference ( $\Delta P$ 249 =300hPa) between the surface and 3 km height for calculating heating rate. Heating rate during 250 winter and pre-monsoon are 1.12 and 2.75 Kday<sup>-1</sup>, respectively. However, realistic value of 251 heating rate may deviate as much as 30 to 50% depending on aerosol mixing height compared to 252 that estimated using a mean  $\Delta P$  of 300 hPa [Babu et al., 2007].

Aerosol radiative effect over any location is very intricately dependent on several parameters such as total column burden of aerosols, their vertical distribution in the atmosphere, 255 single scattering albedo, their size distribution, scattering phase functions, reflectance of 256 underlying surface, relative humidity in the atmosphere, solar insolation and many more. A 257 comparison study of ADRE reported from other sites of the country including oceanic regions 258 will be of great interest and also provides an insight into radiative impact caused by aerosols in 259 this part of the world. Figure 3 shows spatial distribution of reported atmospheric effect (Wm<sup>-2</sup>) 260 due to aerosol over Indian subcontinent and adjacent oceanic regions. It can be observed that during per-monsoon significantly high atmospheric effect (>30 Wm<sup>-2</sup>) is observed over Indian 261 262 land mass compared to winter. Oceanic regions surrounding Indian landmass especially Bay of 263 Bengal showed high atmospheric effect (~11Wm<sup>-2</sup>) in comparison to the Arabian Sea (~3Wm<sup>-2</sup>) 264 [Moorthy et al., 2009]. High atmospheric effect observed during pre-monsoon can have larger 265 implications on regional-scale dynamical process such as wind flow, convective activity and even precipitation patterns [Menon et al., 2002]. Ramanathan et al. [2001] have pointed out that 266 267 high atmospheric effect over Indian Ocean will reduce evaporation from ocean surface; thereby 268 reducing moisture inflow which in turn weakens the monsoon rainfall. On the contrary, Lau et al. [2006] and Lau and Kim [2006] have proposed the Elevated Heat Pump (EHP) hypothesis, 269 270 suggesting that desert dust, mixed with soot aerosols over northern India and the foothills of the 271 Himalayas, may cause enhanced heating in the middle/upper troposphere over southern slopes of 272 the Tibetan Plateau [Gautam et al., 2009] and in turn strengthened the land-sea gradient, thus 273 resulting in the advancement of the monsoon rainfall in early summer. From above discussions 274 it's obvious that aerosols undoubtedly have a role in climate change, whether it enhances or 275 offsets this change is still a matter of scientific debate and have to be assessed carefully with 276 more observational studies.

277 *4.3 Influence of long range transport.* 

278 In this section we tried to analyse the influence of long range transported aerosols over study 279 area. Previous studies [Badrinath et al., 2009; Jose et al., 2015a] have reported the presence of 280 long range transported biomass burning and dust aerosols during pre-monsoon over study area 281 under favorable metrological condition. These episodic events during study days are identified 282 by analyzing intrinsic aerosol optical properties like scattering Angström exponent (SAE) and 283 absorption Ångström exponent (AAE). Spectral dependence of aerosol scattering (absorption) 284 coefficient is quantified in terms of SAE (AAE) and is estimated by negative slope of scattering 285 (absorption) vs. wavelength in log-log plot. SAE gives an understanding of size distribution of 286 aerosol particles, while AAE reveals the source characteristics of absorbing aerosols. AAE ~1 287 indicates the dominance of fossil fuel aerosols, AAE  $\geq 2$  indicate the presence of dust and 288 biomass burning aerosols and AAE between 1 and 2 can be considered as a mixture of both 289 [Bergstorm et al., 2002]. SAE depends primarily on the particles' size and ranges from 4 290 (Rayleigh atmosphere) to 0 (large particles) [Valenzuela et al., 2014]. Analysis shows that mean 291 SAE and AAE values of aerosol particles over the study area during pre-monsoon are 1.56±0.27 292 and 1.19±0.16, respectively. These values represent a typical urban distribution where local 293 emission dominates other sources. Figure 4 shows the temporal variation of SAE and AAE 294 during pre-monsoon days during study the period, episodic events are identified and highlighted 295 in red circles for dust storm events and blue circles for biomass burning days. Each episodic 296 event is further confirmed with satellite observations. In order to demonstrate, satellite 297 observations during 22-23March, 2012 are investigated, where the regional atmosphere is 298 influenced by both long range transported dust and aerosols due to biomass burning. True color 299 image composite of event day (22-23 March 2012) as seen by MODIS is shown in Figure 5(a) 300 and the overall aerosol loading as observed by Terra MODIS is shown in figure 5(b). An intense

301 dust loading can be seen on the Persian Gulf region (North western side of study area). Jose et al. 302 [2015c] analysed the impact of this intense dust storm on SW radiation at TOA using satellite 303 measurements over the Arabian Sea (AS) and reported, a TOA SW aerosol radiative force efficiency of -39 Wm<sup>-2</sup> per unit AOD. MODIS derived active fire location [Gigilo et al., 2003] 304 305 are overlaid on the image, which clearly shows the presence of biomass burning over Central 306 India. Five days backward trajectory obtained from Hysplit model [Draxler et al., 2003] ending 307 over study area on the same day at different altitude (1, 2 and 4Km) overlaid on true color image 308 also indicates the possible transport of dust and biomass aerosols towards study site. During this 309 event an enhancement in  $\tau$  (0.5-0.7) and  $\omega$  (0.7-0.8) is observed over study site. An enhancement 310 in AAE during these days indicates more loading of biomass aerosols as the source region is in 311 close proximity to the study site. However a decrement in SAE from 1.5 to ~1 during the event 312 days also reveals the possible presence of coarser aerosols which could be due to presence of 313 dust aerosols. Estimated effect on 22March, 2012 at TOA, surface and atmosphere are -8.1, -314 91.51 and 83.4 Wm<sup>-2</sup>, respectively. Variations of aerosol optical properties and estimated effect 315 over the study area during similar episodic events occurred are given in Table1. Important 316 observations that can be drawn from these events are; during dust storm events  $\alpha$  values are 317 found lesser than those compared with biomass events. The  $\omega$  during majority of dust storm days 318 (except on 23March 2012 and 3May2012) are lower (~0.77) than biomass burning days (0.81). 319 Low value of  $\omega$  during dust storm prominent days over study area can be possibly due to 320 deposition of black carbon on dust particles [Satheesh et al., 2006]. Chemical analysis of dust 321 samples collected over Kanpur, India during dust events also reveals possible mixing of anthropogenic pollution and estimated  $\omega$  is found to be low (0.74) due to presence of black 322 323 carbon [Chinnam et al., 2006]. Our analysis revealed an enhancement of heating rate between 1.55 to 3.6 Kday<sup>-1</sup> occurred during dust dominant days, which can be attributed as enhanced absorption by possible mixing of dust with local atmosphere. In contrast to dust storm days, high  $\omega$  are observed during biomass burning days suggests an increase in organic carbon which is light scattering part in carbonaceous aerosols and thereby induce a cooling effect at TOA over the study area. Net atmospheric effect during biomass burning events varied from ~50-80 Wm<sup>-2</sup>; which corresponds to a heating rate of 1.5 to 2.7 Kday<sup>-1</sup>.

### **330 5.** Conclusions

331 Present study estimated ADRE over an urban location (Hyderabad) in Central India during
332 the study period 2010-2012. Critical aerosol optical parameters required for estimation of ADRE
333 are either measured or derived from *in-situ* observations. The salient findings are the following:

Aerosol radiative properties showed significant seasonal variation during the study
 period. Seasonal variation of microphysical aerosol optical properties (α, SAE) indicates-the
 dominance of fine mode aerosols over study area; also presence of coarse mode aerosols are
 identified during pre-monsoon months.

• Mean ' $\tau_{550}$ ' showed a minimum (0.45-0.52) during winter and maximum (0.6-0.7) in premonsoon. While,  $\omega$  of aerosols showed a maximum in winter (0.78±0.05) and minimum in pre-monsoon (0.67±0.06). The g varied in the range 0.48±0.02 to 0.53±0.04 during the study period.

Estimated radiative effect at TOA during winter is -11.72 ±3.5 Wm<sup>-2</sup>, while during pre monsoon, it is -5.5 ±2.5 Wm<sup>-2</sup>. Corresponding values at surface during winter and pre monsoon are -46.25±3.4 Wm<sup>-2</sup> and -90.56.4±2.4 Wm<sup>-2</sup>, respectively.

Atmospheric radiative effect during winter and pre-monsoon are 34.52±6 Wm<sup>-2</sup> and
 85.05±10 Wm<sup>-2</sup>, respectively. This corresponds to a heating rate of about 1.12 and 2.75
 Kday<sup>-1</sup>, respectively.

Investigation on identified dust storm and biomass burning events reveals that α values
 are lesser during dust storm than those compared with biomass events. Also during majority
 of dust storm days, ω are lower (~0.77) than biomass burning days (0.81).

• Estimated heating rate due to biomass burning and dust storm aerosols on local atmosphere are about 2.26±0.51 and 2.08±0.6 Kday<sup>-1</sup>, respectively.

353

# 354 Acknowledgement

355 The authors are thankful to Director NRSC, for his encouragement of this work. Authors

356 acknowledge ISRO-GBP ARFI programme for funding the study. First author is also thankful to

357 Mr. Suraj Reddy, FEG, NRSC for computational support. Authors acknowledge the MODIS

358 team and NOAA Air Resources Laboratory (ARL) for provision of satellite data and model data

359 used in this publication.

#### 360 **References**

- 361 Ångström, A., (1964). The parameters of atmospheric turbidity. Tellus 16,64–75.
- Anderson, T., & Covert, D. (1996). Performance characteristics of a high-sensitivity, three wavelength, total scatter/backscatter nephelometer. *Journal of Atmospheric and Oceanic Technology*, 13, 976–986. http://doi.org/10.1175/1520-0426(1996).
- Anderson, T. L., Charlson, R. J., Winker, D. M., Ogren, J. A., & Holmén, K. (2003). Mesoscale
   variations of tropospheric aerosols. Journal of the Atmospheric Sciences, 60(1), 119-136

Anderson, T., & Ogren, J. (1998). Determining aerosol radiative properties using the TSI 3563
integrating nephelometer. *Aerosol Science and Technology*, 29(1), 57–69.
http://doi.org/10.1080/02786829808965551.

- Andrews, E., Sheridan, P.J., Fiebig, M., McComiskey, A., Ogren, J.A., Arnott, P., Covert, D.
  Elleman, R., Gasparini, R., Collins, D., Jonsson, H., Schmid, B., Wang, J., (2006).
  Comparison of methods for deriving aerosol asymmetry parameter. Journal of Geophysical
  Research–Atmospheres 111, art. no. D05S04.
- Arnott, W., Hamasha, K., Moosmüller, & H., Sheridan, P. J., & Ogren, J. A. (2005). Towards
   aerosol light-absorption measurements with a 7-wavelength aethalometer: Evaluation with a
   photoacoustic instrument and 3-wavelength nephelometer. *Aerosol Science and Technology*,
   39(1), 17–29. http://doi.org/10.1080/027868290901972.
- Babu, S. S., Moorthy, K., & Satheesh, S. K. (2007). Temporal heterogeneity in aerosol
  characteristics and the resulting radiative impacts at a tropical coastal station-Part 2: Direct
  short wave radiative forcing. *Annales Geophysicae*, 25(11), 2309–2320.
- Babu, S. S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., ...
  Kumar, V. H. A. (2011). Free tropospheric black carbon aerosol measurements using high
  altitude balloon: Do BC layers build their own homes up in the atmosphere *Geophysical Research Letters*, 38(8), 1–6. http://doi.org/10.1029/2011GL046654.
- Babu, S. S., V. S. Nair, M. M. Gogoi, K. K. Moorthy, (2016), Seasonal variation of vertical
  distribution of aerosol single scattering albedo over Indian sub-continent: RAWEX aircraft
  observations, Atmospheric Environment, (in press), DOI:10.1016/j.atmosenv.2015.09.041.
- Badarinath, K. V. S., Kumar Kharol, S., & Rani Sharma, A. (2009). Long-range transport of
  aerosols from agriculture crop residue burning in Indo-Gangetic Plains-A study using
  LIDAR, ground measurements and satellite data. *Journal of Atmospheric and Solar- Terrestrial Physics*, 71(1), 112–120. http://doi.org/10.1016/j.jastp.2008.09.035.
- Bates, T. S., Huebert, B. J., Gras, J. L., Griffiths, F. B., & Durkee, P. A. (1998). International
  Global Atmospheric Chemistry (IGAC) project's first aerosol characterization experiment
  (ACE 1): Overview. *Journal of Geophysical Research: Atmospheres (1984–2012)*, *103*(D13), 16297–16318.
- 396 Bergstrom, R. W., Russell, P. B., & Hignett, P. (2002). Wavelength Dependence of the 397 Absorption of Black Carbon Particles: Predictions and Results from the TARFOX 398 Experiment and Implications for the Aerosol Single Scattering Albedo. 399 http://doi.org/10.1175/1520-0469(2002)059<0567.
- Bond, T., & Doherty, S. (2013). Bounding the role of black carbon in the climate system: A
  scientific assessment. *Ournal of Geophysical Research: Atmospheres, 118(11), 5380-5552.*
- Charlson, R. J. R., Schwartz, S. E., Hales, J. M., Cess, R. D., Coakley, jr J. A., Hansen, J. E., &
  Hofmann, D. J. (1992). Climate forcing by anthropogenic aerosols. *Science*, 255(5043),
  404 423–430.

- Chinnam, N., Dey, S., Tripathi, S. N., & Sharma, M. (2006). Dust events in Kanpur, northern
  India: Chemical evidence for source and implications to radiative forcing. *Geophysical Research Letters*, *33*(8), 1–4. http://doi.org/10.1029/2005GL025278.
- 408 Draxler, R. R., and G. D. Rolph. (2003). HYSPLIT (Hybrid Single-Particle Lagrangian
  409 Integrated Trajectory) Model. Silver Spring, MD: NOAA Air Resources Laboratory.
  410 http://ready.arl.noaa. gov/HYSPLIT.php.
- Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'neill, N. T., ... Kinne, S.
  (1999). Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols. *Journal of Geophysical Research: Atmospheres (1984–2012), 104*(D24), 31333–31349.
- 415 Eck, T. F., Holben, B. N., Sinyuk, A., Pinker, R.T., Golub, P., .... and Xia, X. (2010).
  416 Climatological aspects of the optical properties of fine/coarse mode aerosol mixtures.
  417 *Journal of Geophysical Research:* 115(D19205),doi:10.1029/2010JD014002.
- Gautam, R., Hsu, N. C., Lau, K. M., Tsay, S. C., & Kafatos, M. (2009). Enhanced pre-monsoon
  warming over the Himalayan-Gangetic region from 1979 to 2007. *Geophysical Research Letters*, *36*(7), 1–5. http://doi.org/10.1029/2009GL037641.
- Gharai, B., Jose, Subin., and Mahalakshmi, D. V, (2013). Monitoring intense dust storms over
  the Indian region using satellite data- a case study. International Journal of Remote sensing.
  34, 7038–7048.
- Giglio, L., Descloitres, J., Justice, C. O., & Kaufman, Y. J. (2003). An enhanced contextual fire
  detection algorithm for MODIS. *Remote Sensing of Environment*, 87(2-3), 273–282.
  http://doi.org/10.1016/S0034-4257(03)00184-6.
- Hansen, A., Rosen, H., & Novakov, T. (1984). The aethalometer—an instrument for the realtime measurement of optical absorption by aerosol particles. *Science of the Total Environment*.
- Hansen, J., Sato, M., & Ruedy, R. (1997). Radiative forcing and climate response. *Journal of Geophysical Research: Atmospheres*, *102*(D6), 6831–6864.
- Haywood, J., & Shine, K. (1995). The effect of anthropogenic sulfate and soot aerosol on the
  clear sky planetary radiation budget. *Geophysical Research Letters*, 22(5), 603–606.
  http://doi.org/10.1029/95GL00075.
- Henyey, L., & Greenstein, J. (1941). Diffuse radiation in the galaxy. *The Astrophysical Journal*,
  93, 70–83.
- Holben, B. N., Eck, T. F., Slutsker, I., Tanre, D., Buis, J. P., Setzer, A., ... Nakajima, T. (1998).
  AERONET—A federated instrument network and data archive for aerosol characterization. *Remote Sensing of Environment*, 66(1), 1–16.

- Huebert, B. J., Bates, T., Russell, P. B., Shi, G., Kim, Y. J., Kawamura, K., ... Nakajima, T.
  (2003). An overview of ACE-Asia: Strategies for quantifying the relationships between
  Asian aerosols and their climatic impacts. *Journal of Geophysical Research: Atmospheres*(1984–2012), 108(D23).
- IPCC, 2013: Climate Change (2013): The Physical Science Basis. Contribution of Working
   Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change.
- Jacobson, M. Z. (2001). Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. *Nature*, 409(6821), 695–7. http://doi.org/10.1038/35055518.
- Jose Subin, Biswadip Gharai, Y. Bhavanikumar and P.V.N Rao, (2015a). Radiative
  implication of a haze event over eastern India. Atmospheric Pollution Research 6 (2015)
  138-146.
- Jose Subin, K. Niranjan, Biswadip Gharai, and P.V.N Rao, (2015b). Characterisation of
  absorbing aerosols using ground and satellite data at an urban location,Hyderabad.
  Aerosol and Air Quality Research (accepted). doi: 10.4209/aaqr.2014.09.0220.
- Jose Subin and Biswadip Gharai. (2015). Satellite based Shortwave aerosol radaitive
   forcing during an intense duststorm over Arabian Sea. Atmospheric Science Letters
   (in press). DOI: 10.1002/asl.597.
- King, M. D., Platnick, S., Moeller, C. C., Revercomb, H. E., & Chu, D. A. (2003). Remote
  sensing of smoke, land, and clouds from the NASA ER-2 during SAFARI 2000. *Journal of Geophysical Research: Atmospheres (1984–2012), 108*(D13).
- Kumar, R., M. C. Barth, G. G. Pfister, V. S. Nair, S. D. Ghude, and N. Ojha. (2015). What
  controls the seasonal cycle of black carbon aerosols in India?, J. Geophys. Res. Atmos.,
  120, 2015. DOI:10.1002/2015JD023298.
- Lau, K., Kim, M., & Kim, K. (2006). Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau. *Climate Dynamics*, 26(7-8), 855–864.
  http://doi.org/10.1007/s00382-006-0114-z.
- Lau, K. M., & Kim, K. M. (2006). Observational relationships between aerosol and Asian
  monsoon rainfall, and circulation. *Geophysical Research Letters*, 33(21), 1–5.
  http://doi.org/10.1029/2006GL027546.
- 469 Liou, K. (2002). An introduction to atmospheric radiation (2nd ed.). Academic press.
- Mahalakshmi, D. V, Badarinath, K. V. S., & Naidu, C. V. (2011). Influence of boundary layer
  dynamics on pollutant concentrations over urban region A study using ground based
  measurements, 40(June), 147–152.

- McComiskey, A., Schwartz, S. E., Schmid, B., Guan, H., Lewis, E. R., Ricchiazzi, P., & Ogren,
  J. a. (2008). Direct aerosol forcing: Calculation from observables and sensitivities to inputs. *Journal of Geophysical Research: Atmospheres*, *113*(9), 1–16.
  http://doi.org/10.1029/2007JD009170.
- Menon, S., Hansen, J., Nazarenko, L., & Luo, Y. (2002). Climate effects of black carbon
  aerosols in China and India. *Science (New York, N.Y.)*, 297(5590), 2250–3.
  http://doi.org/10.1126/science.1075159.
- 480 Ming, Y., & Ramaswamy, V. (2011). A model investigation of Aerosol-Induced changes in
  481 tropical circulation. *Journal of Climate*, 24(19), 5125–5133.
  482 http://doi.org/10.1175/2011JCLI4108.1.
- Montilla, E., Mogo, S., Cachorro, V., Lopez, J., & de Frutos, a. (2011). Absorption, scattering
  and single scattering albedo of aerosols obtained from in situ measurements in the subarctic
  coastal region of Norway. *Atmospheric Chemistry and Physics Discussions*, *11*(1), 2161–
  2182. http://doi.org/10.5194/acpd-11-2161-2011.
- Moorthy, K. K., Nair, V. S., Babu, S. S., & Satheesh, S. K. (2009). Spatial and vertical
  heterogeneities in aerosol properties over oceanic regions around India: Implications for
  radiative forcing. *Quarterly Journal of the Royal Meteorological Society*, *135*(645), 2131–
  2145..
- Morys, M., Mims, F. M., Hagerup, S., Anderson, S. E., Baker, A., Kia, J., & Walkup, T. (2001).
  Design, calibration, and performance of MICROTOPS II handheld ozone monitor and Sun
  photometer. *Journal of Geophysical Research: Atmospheres (1984–2012), 106*(D13),
  14573–14582.
- 495 Niranjan, K., Spandana, B., Anjana Devi, T., Sreekanth, V., & Madhavan, B. L. (2011).
  496 Measurements of aerosol intensive properties over Visakhapatnam, India for 2007. *Annales*497 *Geophysicae*, 29(6), 973–985. http://doi.org/10.5194/angeo-29-973-2011.
- Pandithurai, G., Dipu, S., Dani, K. K., Tiwari, S., Bisht, D. S., Devara, P. C. S., & Pinker, R. T.
  (2008). Aerosol radiative forcing during dust events over New Delhi, India. *Journal of Geophysical Research*, *113*(D13), 1–13. http://doi.org/10.1029/2008JD009804.
- Porter, J. N., Miller, M., Pietras, C., & Motell, C. (2001). Ship-based Sun photometer
   measurements using Microtops Sun photometers. *Journal of Atmospheric and Oceanic Technology*, 18(5), 765–774.
- Quinn, P. K., Coffman, D. J., Kapustin, V. N., Bates, T. S., & Covert, D. S. (1998). Aerosol
  optical properties in the marine boundary layer during the First Aerosol Characterization
  Experiment (ACE 1) and the underlying chemical and physical aerosol properties. *Journal*of *Geophysical Research: Atmospheres (1984–2012), 103*(D13), 16547–16563.

- Ramachandran, S., & Rajesh, T. a. (2008). Asymmetry parameters in the lower troposphere derived from aircraft measurements of aerosol scattering coefficients over tropical India. *Journal of Geophysical Research: Atmospheres, 113*(16), 16212.
  http://doi.org/10.1029/2008JD009795.
- Ramanathan, V., Crutzen, P. J., Lelieveld, J., Mitra, A. P., Althausen, D., Anderson, J., ...
  Chung, C. E. (2001). Indian Ocean Experiment: An integrated analysis of the climate
  forcing and effects of the great Indo-Asian haze. *Journal of Geophysical Research: Atmospheres (1984–2012), 106*(D22), 28371–28398.
- Ricchiazzi, P., Yang, S., Gautier, C., & Sowle, D. (1998). SBDART: A Research and Teaching
  Software Tool for Plane-Parallel Radiative Transfer in the Earth's Atmosphere. *Bulletin of the American Meteorological Society*, 79(10), 2101–2114. http://doi.org/10.1175/15200477(1998)079<2101:SARATS>2.0.CO;2.
- Russell, P. B., Hobbs, P. V., & Stowe, L. L. (1999). Aerosol properties and radiative effects in
  the United States East Coast haze plume: An overview of the Tropospheric Aerosol
  Radiative Forcing Observational Experiment (TARFOX). *Journal of Geophysical Research*,
  104(D2), 2213. http://doi.org/10.1029/1998JD200028.
- Safai, P. D., Raju, M. P., Budhavant, K. B., Rao, P. S. P., & Devara, P. C. S. (2013). Long term
  studies on characteristics of black carbon aerosols over a tropical urban station Pune, India. *Atmospheric Research*, *132-133*, 173–184. http://doi.org/10.1016/j.atmosres.2013.05.002.
- Satheesh, S., Deepshikha, S., Srinivasan, J., & Kaufman, Y. J. (2006). Large dust absorption of
  infrared radiation over Afro-Asian regions: Evidence for anthropogenic impact. *Geoscience and Remote Sensing*, *IEEE Transactions*, *3*(3), 307–311.
  http://doi.org/10.1109/LGRS.2006.869988.
- Satheesh, S., Vinoj, V., & Krishna Moorthy, K. (2010). Radiative effects of aerosols at an urban
  location in southern India: Observations versus model. *Atmospheric Environment*, 44(2010),
  5295-5304. http://doi:10.1016/j.atmosenv.2010.07.020.
- Sinha, P. R., Dumka, U. C., Manchanda, R. K., Kaskaoutis, D. G., Sreenivasan, S., Krishna
  Moorthy, K., & Suresh Babu, S. (2013). Contrasting aerosol characteristics and radiative
  forcing over Hyderabad, India due to seasonal mesoscale and synoptic-scale processes. *Quarterly Journal of the Royal Meteorological Society*, *139*(671), 434–450.
  http://doi.org/10.1002/qj.1963.
- Soni, K., Singh, S., Bano, T., Tanwar, R. S., Nath, S., & Arya, B. C. (2010). Variations in single
  scattering albedo and Angstrom absorption exponent during different seasons at Delhi,
  India. *Atmospheric Environment*, 44(35), 4355–4363.
  http://doi.org/10.1016/j.atmosenv.2010.07.058.
- 543 Srivastava, R., Ramachandran, S., Rajesh, T. a., & Kedia, S. (2011). Aerosol radiative forcing 544 deduced from observations and models over an urban location and sensitivity to single

- 545
   scattering
   albedo.
   Atmospheric
   Environment,
   45(34),
   6163–6171.

   546
   http://doi.org/10.1016/j.atmosenv.2011.08.015.
   6163–6171.
   6163–6171.
- 547 Stamnes, K., Tsay, S., Wiscombe, W., & Jayaweera, K. (1988). Numerically stable algorithm for
  548 discrete-ordinate-method radiative transfer in multiple scattering and emitting layered
  549 media. *Applied Optics*, 27(12), 2502–2509. http://doi.org/10.1364/AO.27.002502.
- Takamura, T., & Nakajima, T. (2004). Overview of SKYNET and its activities. *Opt. Pura Apl*,
  37(3), 3303–3308.
- 552 Valenzuela, a., Olmo, F. J., Lyamani, H., Antón, M., Titos, G., Cazorla, a., & Alados-Arboledas, L. (2015). Aerosol scattering and absorption Angström exponents as indicators of dust and 553 554 dust-free days over Granada (Spain). Atmospheric Research, 154, 1–13. http://doi.org/10.1016/j.atmosres.2014.10.015. 555
- Weingartner, E., Saathoff, H., & Schnaiter, M. (2003). Absorption of light by soot particles:
  determination of the absorption coefficient by means of aethalometers. *Journal of Aerosol Science*, *34*(10), 1445–1463. http://doi.org/10.1016/S0021-8502(03)00359-8.