## Ground-Based In Situ Measurements of Near-Surface Aerosol Mass Concentration over Anantapur: Heterogeneity in Source Impacts

B. S. K. REDDY<sup>1,3</sup>, K. R. KUMAR<sup>1,4</sup>, G. BALAKRISHNAIAH<sup>2</sup>, K. R. GOPAL<sup>\*1</sup>, R. R. REDDY<sup>1</sup>, V. SIVAKUMAR<sup>4</sup>, S. Md. ARAFATH<sup>1</sup>, A. P. LINGASWAMY<sup>1</sup>, S. PAVANKUMARI<sup>1</sup>, K. UMADEVI<sup>1</sup>, and Y. N. AHAMMED<sup>5</sup>

<sup>1</sup> Aerosol and Atmospheric Research Laboratory, Department of Physics, Sri Krishnadevaraya University, Anantapur 515003, Andhra Pradesh, India

<sup>2</sup> Institute of Environmental Engineering, National Chiao Tung University, Hsinchu, Taiwan
<sup>3</sup> Institute of Low Temperature Science, Hokkoido University, Sapporo 0600819, Japan
<sup>4</sup> School of Physics, University of KwaZulu-Natal, Durban 4000, South Africa
<sup>5</sup> Department of Physics, Yogi Vemana University, Kadapa 516003, India

(Received 11 November 2011; revised 9 March 2012)

#### ABSTRACT

Surface measurements of aerosol physical properties were made at Anantapur (14.62° N, 77.65° E, 331 m a.s.l), a semiarid rural site in India, during August 2008-July 2009. Measurements included the segregated sizes of aerosolsas as well as total mass concentration and size distributions of aerosols measured at low relative humidity (RH<75%) using a Quartz Crystal Microbalance (QCM) in the 25–0.05  $\mu m$  aerodynamic diameter range. The hourly average total surface aerosol mass concentration in a day varied from 15 to 70  $\mu g$  $m^{-3}$ , with a mean value of  $34.02\pm9.05~\mu g~m^{-3}$  for the entire study period. A clear diurnal pattern appeared in coarse, accumulation and nucleation-mode particle concentrations, with two local maxima occurring in early morning and late evening hours. The concentration of coarse-mode particles was high during the summer season, with a maximum concentration of  $11.81\pm0.98~\mu\mathrm{g}~\mathrm{m}^{-3}$  in the month of April, whereas accumulationmode concentration was observed to be high in the winter period contributed >68% to the total aerosol mass concentration. Accumulation aerosol mass fraction,  $A_{\rm f}$  (=  $M_{\rm a}/M_{\rm t}$ ) was highest during winter (mean value of  $A_{\rm f} \sim 0.80$ ) and lowest ( $A_{\rm f} \sim 0.64$ ) during the monsoon season. The regression analysis shows that both  $R_{\rm eff}$ and  $R_{\rm m}$  are dependent on coarse-mode aerosols. The relationship between the simultaneous measurements of daily mean aerosol optical depth at 500 nm (AOD<sub>500</sub>) and PM<sub>2.5</sub> mass concentration ([PM<sub>2.5</sub>]) shows that surface-level aerosol mass concentration increases with the increase in columnar aerosol optical depth over the observation period.

 $\textbf{Key words:} \ \ \text{aerosols mass concentration, size distribution, effective radius, backward trajectories}$ 

Citation: Reddy, B. S. K., and Coauthors, 2013: Ground-based in situ measurements of near-surface aerosol mass concentration over Anantapur: Heterogeneity in source impacts. Adv. Atmos. Sci., 30(1), 235–246, doi: 10.1007/s00376-012-1234-5.

### 1. Introduction

Presently, aerosols are recognized to be responsible for a significant impact on climate change affecting our environment. IPCC (2007) refers to aerosol radiative forcing, through direct, indirect, and semi-indirect effects. However, due to the strong inhomo-

geneity of aerosol sources and the high spatial and temporal variability of aerosol properties in the atmosphere, scientific understanding of this problem has remained incomplete. The variability and uncertainty in estimates of the effects of atmospheric aerosols on climate areassociated with particle size distribution. Physical and chemical composition of aerosols is gain-

<sup>\*</sup>Corresponding author: K. R. GOPAL, krgverma@yahoo.com

<sup>©</sup> China National Committee for International Association of Meteorology and Atmospheric Sciences (IAMAS), Institute of Atmospheric Physics (IAP) and Science Press and Springer-Verlag Berlin Heidelberg 2013

ing importance in studies related to global change, and long-term measurement of these parameters at different locations across the globe is important. Only by making measurements of near sources will scientists be able to make the link between source emissions and regional-scale air quality and climate impacts.

Important chemical transformations occur rapidly after emission. For a better understanding of the aerosol role in atmospheric processes, especially near the ground where most anthropogenic aerosols are concentrated, measurements in the near-surface layer of the atmosphere are desirable. Furthermore, when different layers of aerosols at different heights are present, uncertainties in column-integrated aerosol optical and physical parameters retrievals can be assessed, thanks to in situ observations. In situ instruments facilitate the collection of precise information about local trends that cannot be obtained through other means. This type of analysis was conducted by Calvello et al. (2009) over a semirural site in southwest Italy (Tito Scalo).

The importance and urgency of the regional characterization of atmospheric aerosols for reducing the current uncertainties and understanding climate implications at regional and global levels has been increasingly recognized (e.g., Charlson et al., 1992; Kaufman et al., 2002; Satheesh et al., 2006; Moorthy et al., 2009; Gogoi et al., 2011). Concerted efforts using groundbased networks as well as space-borne measurements, synergized with integrated field campaigns, have been accepted as the best means to characterize aerosols regionally and globally for input into climate models (Holben et al., 1998; Moorthy et al., 2008; Satheesh et al., 2009). Although columnar aerosol properties are generally considered for assessing the radiative impacts of aerosols, the role of near-surface aerosols is equally important, primarily because most of the aerosol sources are located on the Earth's surface. In the atmospheric boundary layer (ABL), these aerosols are generally well mixed chemically and physically and are distributed nearly homogeneously within the ABL by strong convective turbulences, especially in the tropics. Synoptic meteorology leads to long-range transport of these aerosols to total columnar loading over different geographic locations of India (e.g., Moorthy et al., 2003; Pillai and Moorthy, 2004; Kumar et al., 2009b; Satheesh et al., 2009).

In addition to their columnar contributions, near-surface aerosols also pose a risk for respiratory morbidity and cardiopulmonary mortality (Carlton et al., 1999). The finer particles [particle diameter  $(D_{\rm p}) < 2.5~\mu{\rm m},~{\rm PM}_{2.5}$ ], having longer atmospheric residence time, are easily inhaled deep into the respiratory system (Pillai et al., 2002). In addition, the particles in the accumulation-mode (0.1  $\mu{\rm m} < r < 1~\mu{\rm m}$ ) con-

tribute predominantly to visibility degradation and radiative interactions (Berico et al., 1997; Pillai et al., 2002). Despite their significance, data regarding the characterization of near-surface aerosols and their contribution to columnar properties is sparse over the South Asian region and India in general and over the southeastern part of India in particular. This region is unique because of its distinct topography; it is a rain shadow region, deprived of both the monsoons (southwest and northeast monsoons) and droughts due to distinct seasonal variations and source impacts. As such, we investigated the total and size-segregated near-surface aerosol mass concentrations using a Quartz Crystal Microbalance (QCM) cascade impactor over Anantapur (13.62°N, 77.65°E, 331 m a.s.l), in the southeastern part of India, during the period from August 2008 to July 2009, to infer the seasonal changes in the broad homogeneity in aerosol size distribution.

In this study, surface measurements of aerosol physical properties at Anantapur, India, were measured during August 2008–July 2009 are presented. Measurements included size-segregated as well as total mass concentration of aerosols and aerosol size distributions. Also presented are the meteorological parameters that influence these aerosol properties prevailing over this region: wind speed, direction, ambient air temperature, and relative humidity (RH). The origins of some aerosol episodes were investigated to identify the main source regions responsible for these special events. Monthly and diurnal variations of these parameters and the influence of meteorological conditions were also analyzed and are explained here.

#### 2. Experimental site and general meteorology

Atmospheric measurements were conducted at a semiarid rural site in the Department of Physics, Sri Krishnadevaraya University (SKU) campus; Anantapur (13.62°N, 77.65°E, 331 m a.s.l, Fig. 1). The location of Anantapur on the peninsular Indian subcontinent is shown in the inset. Anantapur represents a very dry, semiarid, rain shadow, and continental region of Andhra Pradesh, which is a nonindustrialized, medium-sized city with a population of 400 000 to 600 000 (considering the whole city area). Within a 50-km radius, this region is surrounded by a number of cement plants, lime kilns, and slab-polishing and brick-making units. These industries, the national highways (NH 44 and NH 205), and the town area are situated in the north to southwest side of the sampling site (Fig. 1). The study area is located a short distance from two major capital cities, it is  $\sim 200$  km from Bangalore and 400 km to Hyderabad.

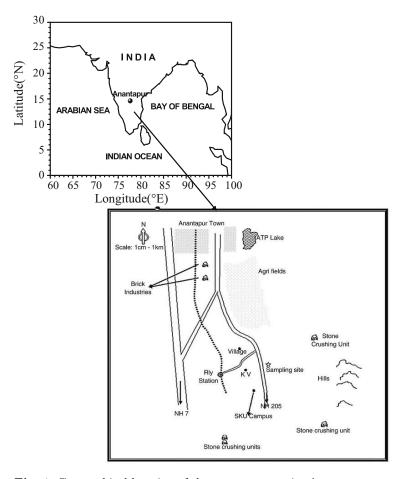


Fig. 1. Geographical location of the measurement site Anantapur over the Indian subcontinent and adjoining surrounding oceanic regions.

The experimental site is located in the southern part of the city and is close to the highway that surrounds the city. Local aerosol sources are mainly the heavy traffic (mainly diesel vehicles), together with the resuspension of materials on the ground. The old part of the city has rather narrow streets responsible for heavy traffic in some areas of the city, especially at rush hours. During the winter, domestic heating (mainly wood, charcoal, and diesel central heating) represents an additional important source of anthropogenic aerosols. On the other hand, forest fires and transported dust represent an additional source of aerosol particulates (Badarinath et al., 2009, 2010). The bowl-like topography of Anantapur favors wintertime inversions and low wind speeds. This, in combination with pollutant emissions mainly from traffic, can lead to a large accumulation of particulate aerosols and thus to high particle loads in the study area, which can cause environmental and human health problems.

The continental conditions prevailing at this site are responsible for large seasonal temperature differences, providing hot summers (March–May) and cool winters (December–February). Most rainfall oc-

curs during the monsoon (June–September) and postmonsoon (October and November) seasons. The annual rainfall of the district is 550 mm, less than the adjoining Rayalaseema region and other parts of Andhra Pradesh. The normal rainfall during the southwest monsoon period is 400 mm, which contributes >60%–70% of the total annual rainfall, whereas, normal rainfall is only  $\sim$ 150 mm for the northeast monsoon period, which contributes 30%–40% of annual rainfall. The remaining months of summer are warm and dry, with normal daily maximum temperature ranging between  $29^{\circ}$ C and  $42^{\circ}$ C. The winter months are cooler; the temperature falls  $\sim$ 20°C and fair weather conditions prevail, with low southeasterly winds at 4–5 m s<sup>-1</sup>, with moderate relative humidity (RH).

#### 3. Instruments and measurements

Measurements of mass size distribution of aerosols have been made regularly using a Quartz Crystal Microbalance (QCM) cascade impactor of model PC-2 (California Measurements Inc., Sierra Madre, CA). The total mass concentration  $(M_t)$  was estimated from

the size segregated mass concentration  $(m_{ci})$  using

$$M_{\rm t} = \sum_{i=1}^{10} m_{\rm c}i$$

The principle of operation of the QCM, precautions, data deduction, and error report have been explained in detail in several earlier papers (e.g., Pillai and Moorthy, 2001; Moorthy et al., 2003, Kumar et al., 2009b). The QCM at Anantapur has been regularly operated since May 2007, as part of the Aerosol Radiative Forcing over India (ARFI) project of Indian Space Research Organization's Geosphere Biosphere Programme (ISRO-GBP). The instrument has operated from the top of the building of height  $\sim 12$  m from the ground level and during periods of RH < 80%. Below 80% RH, the instrument is not very susceptible to moisture (Reddy et al., 2007); it requires stable RH during the sampling period, because of the affinity of quartz substrate to moisture. Also, the loss of absorbed water from water soluble particles by evaporation could occur during the collection of particles under low-pressure conditions (Pillai and Moorthy, 2001; Kumar et al., 2009b; Reddy et al., 2011). The dual unsealed crystal approach used in the QCM reduces the thermal and humidity effects to a minimum. The optimum sampling time is taken as that time which is sufficient to impart a frequency change of 20 to 30 Hz with a tolerance of -5 to +10 Hz (Madhavan et al., 2008). So the typical sampling duration was 5-6 minutes. Aerosol samples were collected once per week (i.e., 4–5 days per month) on Wednesdays, at hourly intervals hourly from 0800 to 1800 LST (local standard time) and continuously for a period of 24 h on the first Wednesday of the month at hourly intervals, depending on RH conditions. A total of  $\sim 700$ independent observations of aerosol mass concentration samples were collected during the study period and were used to examine the aerosol size distribution over the study area.

This instrument provides real-time measurements in 10 size bins, with 50% cutoff diameters: >25, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1, and 0.05  $\mu$ m for stages 1 to 10, respectively (see Table 1 of Kumar et al., 2009b). Its pump aspirates the ambient air at a flow rate of 0.24 L min<sup>-1</sup>. The QCM provides mass concentration of the particles collected in each stage ( $m_{ci}$ ) as a function of particle diameter assuming a value of 2000 kg m<sup>-3</sup> for  $\rho$ . Accordingly, it yields mass concentration in 10 size bins; the 50% cutoff diameter and the geometric mean diameter of these bins was taken from Pillai and Moorthy (2001). Stage 1 collects all particles with diameter >25  $\mu$ m; hence, no mean diameter is assigned to that stage.

The wind speed and direction play a major role in

determining the type of aerosol present over a location as they help in bringing aerosols from neighboring region to the measurement site. The high wind speeds also facilitate rapid transport of nascent continental aerosols, which leads to the formation of new submicron aerosols over the arid environments (which provide ample RH) through secondary (i.e., gas to particle) production (Pillai and Moorthy, 2001; Madhavan et al., 2008). Daily mean wind speed, wind direction, air temperature, RH, and total rainfall data were obtained from automatic weather stations (AWS) established at the observation site. Monthly mean prevailing meteorological conditions during the study period are shown in Fig. 2. The mean wind speed reached a maximum of  $\sim 2.7 \text{ m s}^{-1}$  in the month of July and minimum of  $\sim 1.0 \text{ m s}^{-1}$  during January (followed closely

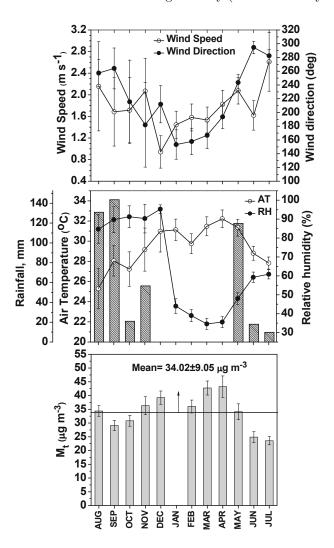


Fig. 2. Monthly mean variations of meteorological parameters (top and middle panels) and total aerosol mass concentration ( $M_{\rm t}$ , bottom panel) at Anantapur during the study period.

February:  $1.2 \text{ m s}^{-1}$ ). Most winds were southwesterly during the entire observation period. Monthly mean maximum temperature of ~35°C was observed in April, and a maximum RH of  $\sim 76.7\%$  was observed during July (Fig. 2, top panel). Monthly mean minimum temperature of  $\sim 27.8^{\circ}$ C was observed in July, and a minimum RH of  $\sim 31.6\%$  was observed during April (Fig. 2, middle panel). The total annual rainfall for the study period across the measurement site was 493.5 mm, of which 319.5 mm of rainfall was received during the southwest monsoon, which contributed  $\sim 64.7\%$  of the total rainfall for the year. Source regions responsible for special aerosol events can be detected by performing a backward trajectory analyses. To characterize the transport pathways of air-masses before they arrive at the study location and to detect the aerosol source regions responsible of some aerosol episodes in the measurement area, seven-day backward trajectories ending at 2300 UTC at Anantapur were derived at three different altitudes (500 m, 1500 m, and 2500 m above ground level) using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://www.arl.noaa.gov/ready/hysplit4.html; Draxler and Rolph, 2012).

#### 4. Results and discussion

### 4.1 Monthly variations in aerosol mass concentration and other retrieved parameters

Monthly average variation of total aerosol mass concentration  $(M_t)$  during the study period from August 2008 to July 2009 is shown in Fig. 2 (bottom panel), where the points indicate the monthly means and the vertical bars through them indicate the standard errors. The hourly average  $M_{\rm t}$  in a day varied from 15  $\mu g$  m<sup>-3</sup> to 70  $\mu g$  m<sup>-3</sup>, with a mean value of  $34.02\pm9.05~\mu g~m^{-3}$  for the entire study period. The annual variation is attributed mainly to the contrasting prevailing synoptic meteorological conditions at Anantapur (Fig. 2). It clearly depicts that the total mass concentration of near-surface aerosols has been observed to be maximum (44.87 μg m<sup>-3</sup>) during the summer (in the month of April), associated with increased seasonal solar heating. Due to the scanty rainfall during the summer season and the consequent longer residence time of aerosols, the higher thermal convections during summer led to deepening of ABL, resulting in heating the land and lifting the loose soil. This trend, combined with increased wind speeds, contributed to the enhancement in  $M_{\rm t}$ . In addition, the prominence of local anthropogenic domestic activities, such as the burning of wood, straw, for cooking and the burning of residual agricultural crop waste in nearby

agricultural fields, etc., also contribute significantly to accumulation-mode aerosols during this period, which contribute more to the total mass concentration. On the other hand, the highest rainfall (~67% of the annual total) occurs during the monsoon season, causing significant wash out of suspended particles (Vinoj et al., 2004; Gogoi et al., 2008), leading to the lower aerosol mass concentrations. The effect is also due to reduction in the continental features conducive to aerosol generation by shifting the air-mass. During winter, lower thermal convection leads to a shallower ABL, which, along with the stronger winds, enhances the ventilation coefficient, resulting in a gradual dilution of the surface concentration.

To observe the relative change in the aerosol mass size distribution, we divided the size segregated data into three modes: nucleation- (aerodynamic diameter  $<0.1 \mu m$ ), accumulation- (aerodynamic diameter  $0.1-1.0 \mu m$ ), and coarse- (aerodynamic diameter >1 μm) modes for the study period. Diurnal variations of aerosol mass concentration are very important in understanding the role of mesoscale atmospheric processes and the effect of local human activities. The diurnal variation of the ABL height and its structure is known to influence surface aerosol concentrations (Moorthy et al., 2003; Nair et al., 2007). Figure 3 shows a similar hourly mean diurnal variation of coarse-mode  $(M_c)$ , accumulation-mode  $(M_a)$ , and nucleation-mode  $(M_n)$  aerosol mass loading at the observation site for the entire study period. It shows a broad primary peak in the late night/early morning hours (0400-0600 LST) and then a secondary peak in the morning just after the sunrise (0800–0900 LST), followed by a sharp nocturnal peak around 2100 LST.

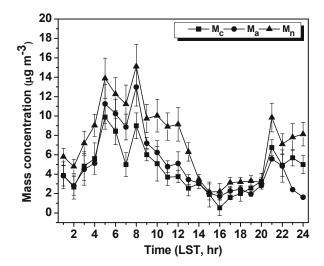


Fig. 3. Hourly average diurnal variation of size segregated aerosol mass concentrations for the measurement period.

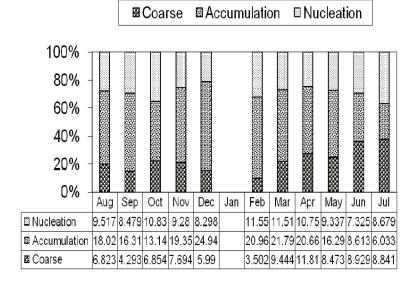


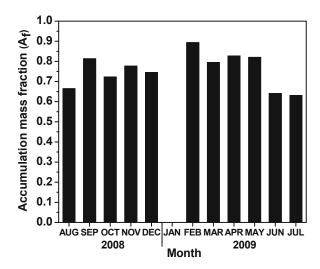
Fig. 4. Relative contribution of different modes (nucleation, accumulation and coarse) of particles to the total mass concentration for the observation period.

During 1200-2000 LST, all modes of concentration  $(M_{\rm c},\ M_{\rm a},\ {\rm and}\ M_{\rm n})$  remain more or less steady, with minimum values of 1  $\mu$ g m<sup>-3</sup>, 2.35  $\mu$ g m<sup>-3</sup>, and 3.59  $\mu$ g m<sup>-3</sup>, respectively, followed by a gradual increase until 2200 LST. Enhancement in aerosol mass concentration in the morning and evening hours has been attributed to the increase in vehicular traffic and related human activities in the study area (Latha and Badarinath, 2005). As night advances due to drastic reduction in the anthropogenic and rural activities lead to reduction in aerosol generation. In the early morning hours aerosols closer to the surface are lost by sedimentation, which results in a decrease in aerosol concentrations. The solar heating of land during the day increases convective activity, leading to an increase in ABL height (Stull, 1988). This increases the ventilation coefficient, resulting in faster dispersion of aerosols and resulting in lower aerosol concentrations during evening hours. The variability in aerosol mass concentration shows an inverse relationship with boundary layer height. Similar observations have been reported by Kumar et al. (2009b) from the same site and by Parameswaran et al. (1998) and Pillai and Moorthy (2001) across the urban coastal region of Trivandrum, India.

Figure 4 gives a clear illustration of the relative contribution of different modes (nucleation, accumulation, and coarse) of particles to the total mass concentration for the entire sampling period. Particle mass during the winterseason is dominated by fine-mode particles contributing 68% of the total mass concentration, and during the summer season it is dominated by coarse-mode particles contributing 32% of the total

concentration. The corresponding monthly variation is shown in the data table of Fig. 4. The monthly variability is nicely produced in the accumulationmode aerosol, while the variation is not as prominent in the nucleation- and coarse-mode aerosols. The maximum mass concentration in the accumulationmode recorded during two months (December 2008) and February 2009) was reflected in the nucleationmode while the coarse-mode mass concentration was evident in April 2009. However, the accumulationmode shows high concentrations for all months of the study period. On the other hand, nucleation-mode aerosol abruptly increased in March 2009. This indicates decoupling of columnar integrated features from the near-surface features as seen in the QCM mass distributions (Niranjan et al., 2008). The bars diagram in Fig. 5 show the annual variability of the accumulation aerosol mass fraction  $(A_f)$  from the in situ measurements using QCM for all of the sampling days. In general, the accumulation-mode (i.e., fine) particles are believed to be mostly of anthropogenic origin (Moorthy et al., 2005); therefore, we estimated the accumulation-mode mass fraction  $A_{\rm f} (= M_{\rm a}/M_{\rm t})$ . The figure reveals that the accumulation-mode aerosols contributed >68% to the total aerosol mass concentration throughout the year. The highest levels occurred during winter (mean  $A_{\rm f} \sim 0.80$ ) and the lowest during monsoon season ( $A_{\rm f} \sim 0.64$ ).

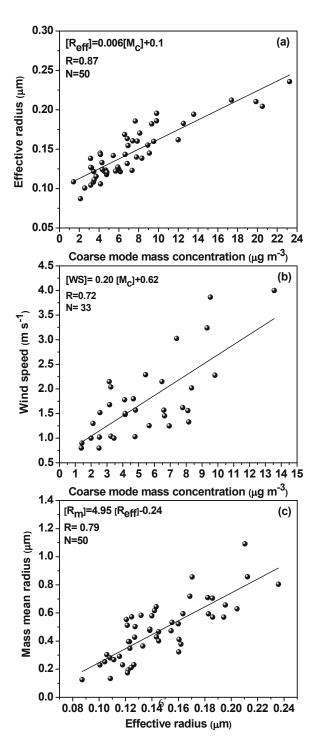
The effective radius  $(R_{\rm eff})$  is a useful measure of average aerosol particle size in poly-dispersive aerosol population. To see the association between effective radius and wind speed and coarse-mode mass concen-



**Fig. 5.** Monthly variation of accumulation aerosol mass fraction  $(A_f)$  averaged for all sampling days during the study period.

tration, the surface wind data was collected from an AWS at the observation site for the days on which QCM observations were made. For each individual measurement obtained from QCM,  $R_{\text{eff}}$  was estimated. These estimates were averaged to yield one value for each day, the mean value of atleast 10 independent estimates of  $R_{\text{eff}}$  during different times of that day. Figure 6 shows the scatter plots of coarse-mode aerosol mass concentration  $(M_c)$  with effective radius  $(R_{\text{eff}})$ and wind speed (WS) (Figs. 6a and b) for the study period. A strong correlation between coarse-mode aerosols and effective radius yields a correlation coefficient of 0.87, which is statistically significant. A good correlation between coarse-mode aerosols and leading wind speed (correlation coefficient 0.72), was also calculated. These correlations indicate that the induction of coarse-mode aerosols lead to a change in the size distribution and hence an increase in the effective radius. Moreover, Fig. 6c shows a steady correlation between effective radius and mass mean radius, with a correlation coefficient of 0.79.

Clearly, the nature of variations of  $R_{\rm eff}$  and  $R_{\rm m}$  are quite similar, and changes are associated with air-mass type. Both  $R_{\rm eff}$  and  $R_{\rm m}$  were high during the winter, indicating a relative abundance of coarse-mode particles at the observation site, whereas in the summer season,  $R_{\rm eff}$  and  $R_{\rm m}$  were low, indicating a profusion of fine-mode aerosol (Pillai and Moorthy, 2001). Figure 6 clearly shows that both  $R_{\rm eff}$  and  $R_{\rm m}$  are more dependent on coarse-mode aerosols. Effective radius  $(R_{\rm eff})$  and surface area both are important to study the effect of the change in the size distribution on the optical properties of aerosols. The mean diurnal varia-



**Fig. 6.** Scatter plots between coarse mode mass concentration  $(M_c)$  with (a) effective radius  $(R_{\rm eff})$  and (b) wind speed (WS) (c) effective radius  $(R_{\rm eff})$  and mass mean radius  $(M_{\rm m})$  during August 2008–July 2009.

tion of  $R_{\text{eff}}$  and surface area are shown in Fig. 7; they demonstrate nearly opposite natures. The lower values of  $R_{\text{eff}}$  indicate the abundance of smaller particles,

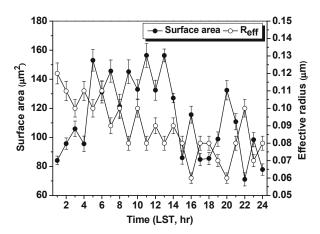
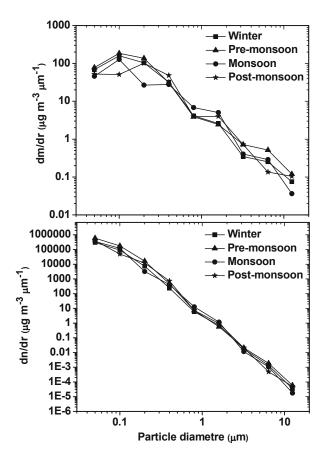


Fig. 7. Hourly average diurnal variation of surface area and effective radius for the study period.

and high surface area is primarily due to their large concentration. Both of these parameters produce a significant scattering effect.  $R_{\rm eff}$  is higher, indicating a relative dominance of coarse aerosols and a reduction in the surface area.

#### 4.2 Aerosol size distributions

OCM observations were further used to obtain the number and mass size distributions for the nearsurface aerosols. The size distribution of aerosols at any location is mainly governed by the relative strengths of different production and removal mechanism (Jaenicke, 1993). Several authors (Bates et al., 1998; Ramachandran and Jayaraman, 2002; Koponen et al., 2003; Ganguly et al., 2005) have shown that the size distribution of tropospheric aerosols is a combination of many lognormal distributions, where different models represent different production sources. Seasonal variation of aerosol mass size distribution and number size distribution over the study area are shown in Fig. 8, top and bottom panels, respectively. Clearly, the aerosol size distributions show a multimodal distribution over the measurement site. mass size distribution basically has three modes: a fine-mode  $(r < 0.15 \mu m)$ , an accumulation-mode  $(0.15 \mu m)$  $\mu m < r < 1.5 \mu m$ ), and a coarse-mode ( $r > 1.5 \mu m$ ). The fine- and accumulation-mode aerosols dominated during the winter and the monsoon season, whereas coarse-mode particles were observed during the summer (Latha and Badarinath, 2005). As the air-mass changed to a continental flow, more accumulation and coarse aerosols were added, and the increased rainfall led to a rapid reduction in the land-based natural sources (Kumar et al., 2009b). Thus, the fine-mode largely decreased, while the accumulation and coarsemodes were not much affected as they reloaded. Thus,

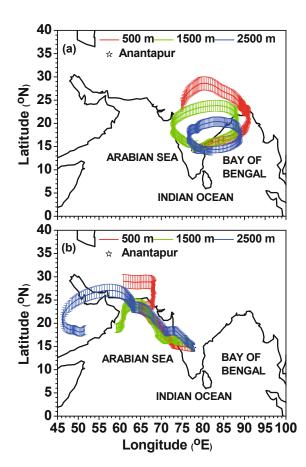


**Fig. 8.** Seasonal variation of mass (dm/dr) and number (dn/dr) size distributions as a function of diameter obtained from QCM.

the accumulation-mode particles attained relatively higher dominance during the monsoon season and the size distribution remained nearly monotonic (Moorthy et al., 2003). During the post-monsoon season, as the air-mass changed, the fine-mode grew, and the coarse-mode decreased slowly. Aerosol number size distribution derived from the QCM system showed a multimodal size distribution over the observation site, which is in good agreement with the existence of tri-modal size distributions with similar modes, as reported by Parameswaran et al. (1999).

#### 4.3 Influence of air-mass trajectories

Air-mass back trajectories are important to identify the source regions from where the pollutants originated and were subsequently transported to the observation sites. It is equally important to determine the heights from which aerosols descended, making the surface as well as columnar measurements. Because the residence time of aerosols is ~seven days in the lower atmosphere, seven-day backward trajectories were performed. The backward trajectories trace in



**Fig. 9.** Seven-day backward trajectories of airmasses pathways for two months (a) December, 2008 and (b) April, 2009 at different altitudes derived from HYSPLIT.

time the origin of air parcels and the paths they traveled at different heights before reaching the measurement site. In Fig. 9, the mean seven-day backward trajectories were drawn for the two months, namely December 2008 and April 2009, at three different altitudes (500 m, 1500 m, and 2500 m) over the measurement site, indicating the dominance of coarse and fine-modes, respectively. In December 2008, the trajectory loop with low wind speed, which had no defined path and usually meandered over urban areas and its surrounding rural areas, spent most of the time in the mainland of central part of India while passing over the Bay of Bengal before arriving at the sampling site, leading to an increase in accumulationmode aerosols at all levels. During April 2009, the trajectories arrived at the measurement site from the African/Arabian deserts traversing over the Arabian Sea might contribute more bigger aerosols and spend most of the time in the Arabian Sea carrying sea salt aerosols (i.e., coarse-mode particles) toward the sampling site. Therefore, the air-masses passing over industrial and urban areas were associated with maximum anthropogenic aerosols from air pollution might contribute to fine mode aerosols, while the air-masses passing coming from the African/Arabian countries across the Arabian Sea usually dominated, with high concentrations of coarse-mode aerosols.

# 4.4 Diurnal and monthly variations of total aerosol number density

The mean diurnal variation of total aerosol number density  $(N_T)$  for the entire sampling period is shown in Fig. 10. In the early morning hours, a broad peak was observed at 0500-0700 LST, and another broad peak just after local sunrise was observed at 0700-0900 LST. These peaks were due to local vehicular traffic, whereas the sharp peak was due to daily local anthropogenic activities such as burning of biomass used for domestic cooking, room heating, etc., and also due to the fumigation effect in the ABL, which has been widely discussed in the context of dispersion of pollutants (Stull, 1998). The total aerosol number density remained high during night until morning up to 1000 LST and decreased to a low value during noon hours ( $\sim 1500$  LST). In general, the nighttime aerosol density was 2-3 times larger than that during noon hours. Regarding the major anthropogenic sources of aerosols, the measurement site is surrounded by nearby villages, industries, and agriculture fields (clearly shown in Fig. 1) that include biomass burning from fields, burning of wood and other biomass used for domestic cooking and warming the interior, etc., and vehicular exhaust. Agriculture field work and dust raised by the vehicular traffic form other major sources of aerosols. Near the observation site, vehicular transport is moderate in the early morning hours, peaking around 0900-1100 LST and 1700-1900 LST. Burning of wood and biomass fuels for domestic cooking and

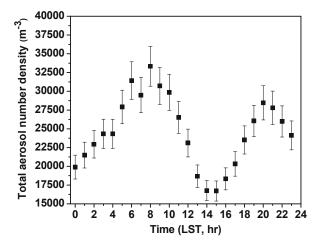


Fig. 10. Mean diurnal variation of total aerosol number density  $(N_{\rm T})$  for the study period.

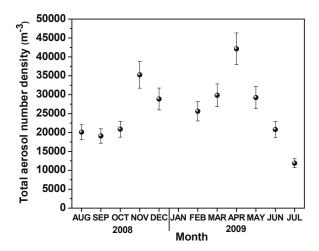
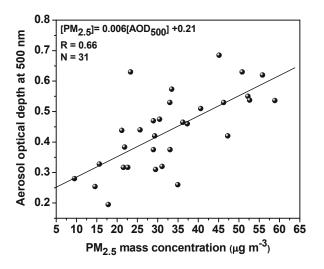


Fig. 11. Monthly mean variation of total aerosol number density  $(N_T)$  during the sampling period.



**Fig. 12.** Correlation between the daily mean aerosol optical depth at 500 nm and  $PM_{2.5}$  mass concentration ( $[PM_{2.5}]$ ) over the observation site.

the heating contribute the night time aerosol emissions in this region. In addition, the evening traffic may partly contribute to the high aerosol number density during night time. Figure 11 shows monthly mean variation of aerosol number density represented by solid circles. The total aerosol number density  $(N_{\rm T})$  was high during the month of April and low during July, with an average value of 25792.22±8452.67 m $^{-3}$ .

# 4.5 Relationship between daily mean $PM_{2.5}$ and aerosol optical depth

To examine the relationship between the  $PM_{2.5}$  mass concentrations ( $[PM_{2.5}]$ ) and aerosol optical depths (AOD) at 500 nm (AOD<sub>500</sub>),  $PM_{2.5}$  mass con-

centrations (aerosol particles with aerodynamic diameter <2.5 µm) were calculated for each sample obtained by using QCM. These fine-mode particles have longer residence times in the atmosphere, and they can cause lung and respiratory diseases. AOD measurements were made using a multi-wavelength solar radiometer (MWR, for more details please refer to Moorthy et al., 1988; Kumar et al., 2009a) on all clear-sky days simultaneously with QCM measurements for the same study period. In this study, only days when both AODs and mass concentrations were available were consid-The AODs increased with increase in PM<sub>2.5</sub> mass concentrations over the observation site. The straight-line fit yielded a non-zero intercept value of 0.21 and a correlation coefficient of 0.66, indicating a better agreement between the surface and columnar measurements.

#### 5. Summary and conclusions

In this study, the aerosol physical properties in a semiarid rural station of Anantapur, from August 2008 to July 2009 were investigated. The following major conclusions were derived by analyzing aerosol mass concentration, effective radius, mass mean radius and aerosol size distributions in the  $0.05-25-\mu m$  aerodynamic diameter range:

- (1) The near-surface aerosol characteristics over Anantapur showed distinct seasonal changes with annual high in total mass concentration ( $M_{\rm t} \sim 45~\mu {\rm g}~{\rm m}^{-3}$ ) in April and low ( $M_{\rm t} \sim 25~\mu {\rm g}~{\rm m}^{-3}$ ) in June, with a mean value of  $34.02\pm 9.04~\mu {\rm g}~{\rm m}^{-3}$ . The accumulation-mode aerosol mass concentration ( $M_{\rm a}$ ) exhibits variation similar to that of total mass concentration of aerosols ( $M_{\rm t}$ ), which was observed to be high during winter contributing 68% to  $M_{\rm t}$ .
- (2)  $R_{\rm eff}$  is high indicative of relative dominance of coarse aerosols and reduction in the surface area, which produces significant scattering effect. Aerosol size distributions derived from QCM particle analyzer showed a multi-modal size distribution at the observation site.
- (3) The history of air-mass trajectories in the month of December 2008 shows that long-range transport of aerosols circulate from the mainland of central India contribute more to the fine aerosols, whereas the distant transport of dust (i.e., coarse aerosols) from the Arabian/African deserts traversing the Arabian Sea leads to an abundance of bigger particles. This transport was quite noticeable during April 2009.
- (4) The relationship between the simultaneous measurements of daily mean aerosol optical depth at 500 nm and  $PM_{2.5}$  mass concentration shows that the near-surface aerosol mass concentration increased with

the increase in columnar aerosol optical depth over the observation period.

Acknowledgements. The authors indebted to the Indian Space Research Organization (ISRO), Bangalore for conducting this work through its Geosphere Biosphere Programme (GBP) under Aerosol Radiative Forcing over India (ARFI) project. We would like to acknowledge Dr. P. P. N. Rao, Programme Director, ISRO-GBP, Bangalore, Dr. K. Krishna Moorthy, Project Director and Dr. S. Suresh Babu, Project Manager, Space Physics Laboratory (SPL), Vikram Sarabhai Space Centre (VSSC), Trivandrum for their support and constant encourage-The back trajectories used in this publication were produced with HYSPLIT from the NOAA ARL website: http://www.arl.noaa.gov/ready/hysplit4.html. One of the authors (RRR) thanks University Grants Commission (UGC), New Delhi for providing Basic Science Research (BSR) faculty fellowship during this time when part of the work has been carried out. We are grateful to the Journal Editor and the two anonymous reviewers for their constructive and useful comments which improve the scientific content of the original paper.

#### REFERENCES

- Badarinath, K. V. S., S. K. Kharol, and A. R. Sharma, 2009: Long-range transport of aerosols from agriculture crop residue burning in Indo-Gangetic Plains— A study using Lidar, ground measurement and satellite data. *Journal of Atmospheric and Solar Terres*trial Physics, 71, 112–120.
- Badarinath, K. V. S., S. K. Kharol, D. G. Kaskaoutis, A. R. Sharma, V. Ramaswamy, and H. D. Kambezidis, 2010: Long range transport of dust aerosols over the Arabian Sea and Indian Ocean—A case study using the satellite data and ground based measurements. *Global and Planetary Change*, 72, 164–181.
- Bates, T. S. and Coauthors, 1998: Process controlling the distribution of aerosol particles in the lower marine boundary layer during the first Aerosol Characterization Experiment (ACE 1). *J. Geophys. Res.*, **103** (D13), 16369–16383.
- Berico, M., A. Luciani, and M. Formignani, 1997: Atmospheric aerosol in urban area measurements of TSP and PM10 standards and pulmonary deposition assessments. Atmos. Environ., 31, 3659–3665.
- Calvello, M., F. Esposito, G. Pavese, and C. Serio, 2009: Physical and optical properties of atmospheric aerosol by in-situ and radiometric measurements. Atmospheric Chemistry and Physics, 9, 25565–25597.
- Carlton, A. G., B. J Turpin, W. Johnson, B. T. Burkley, M. Simcik, S. J. Elsenreich, and R. J. Porcja, 1999: Microanalysis methods for characterization of personal aerosol exposures. Aerosol Science and Technology, 31, 66–80.
- Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J.

- A. Coakley, J. E. Hansen, and D. J. Hoffman, 1992: Climate forcing by anthropogenic aerosols. *Science*, **255**, 423–430.
- Draxler, R. R., and G. D. Rolph, 2012: HYS-PLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model access via NOAA ARL READY Website., NOAA Air Resource Laboratory, Silver Spring, MD. [Available online at http://ready.arl.noaa.gov/HYSPLIT.php]
- Ganguly, D., A. Jayaraman, H. Gadhavi, and T. A. Rajesh, 2005: Features in wavelength dependence of aerosol absorption observed over central India. *Geophys. Res. Lett.*, **32**, L13821, doi: 10.1029/2005GL023023.
- Gogoi, M. M., P. K. Bhuyan, and K. K. Moorthy, 2008: Estimation of the effect of long range transport on seasonal variation of aerosols over northeastern India. Annales Geophysicae, 26, 1–13.
- Gogoi, M. M., B. Pathak, K. K. Moorthy, P. K. Bhuyan, S. S. Babu, K. Bhuyan, and G. Kalita, 2011: Multiyear investigations of near surface and columnar aerosols over Dibrugarh, northeastern location of India: Heterogeneity in source impacts. Atmos. Environ., 45, 1714–1724.
- Holben, B. N., and Coauthors, 1998: AERONET-A federated instrument network and data archive for aerosol characterization. *Remote Sens. Environ.*, **66**, 1–16.
- IPCC, 2007: Climate Change 2007: The Physical Science Basis. Working Group/Contribution to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, New York, 996pp.
- Jaenicke, R., 1993: Tropospheric aerosols. *International Geophysics Series*, **54**, 1–31.
- Kaufman, Y. J., D. Tanre, B. N. Holben, S. Mattoo, L. A. Remer, T. F. Eck, J. Vaughan, and B. Chatenet, 2002: Aerosol radiative impact on spectral solar flux at the surface, derived from principal-plane sky measurements. J. Atmos. Sci., 59(3), 635–646.
- Koponen, I. K., A. Virkkula, R. Hillamo, V. M. Kerminen, and M. Kalumal, 2003: Number size distributions and concentrations of continental summer aerosols in Queen Maud Land, Antarctica. J. Geophys. Res., 108(D18), 4587, doi: 10.1029/2003JD003614.
- Kumar, K. R., K. Narasimhulu, R. R. Reddy, K. R. Gopal, L. S. S. Reddy, G. Balakrishnaiah, K. K. Moorthy, and S. S. Babu, 2009a: Temporal and spectral characteristics of aerosol optical depths in a semi-arid region of southern India. Science of the Total Environment, 407, 2673–2688.
- Kumar, K. R., K. Narasimhulu, R. R. Reddy, K. R. Gopal, L. S. S. Reddy, G. Balakrishnaiah, K. K. Moorthy, and S. S. Babu, 2009b: Size segregated mass concentration and size distribution of near surface aerosols over a tropical Indian semi arid station, Anantapur: Impact of long range transport. Science of the Total Environment, 407, 5589-5604.
- Latha, K. M., and K. V. S. Badarinath, 2005: Spectral so-

- lar attenuation due to aerosol loading over an urban area in India. Atmospheric Research, **75**(4), 257–266.
- Madhavan, B. L., K. Niranjan, V. Sreekanth, M. M. Sarin, and A. K. Sudheer, 2008: Aerosol characterization during the summer monsoon period over a tropical coastal Indian station, Visakhapatnam. J. Geophys. Res., 113, D21208, doi: 10.1029/2008JD010272.
- Moorthy, K. K., P. R. Nair, and B. V. K. Moorthy, 1988: A study on aerosol optical depth at a coastal station, Trivandrum. *Indian Journal of Radio and Space Physics*, 17, 16–22.
- Moorthy, K. K., P. S. Pillai, and S. S. Babu, 2003: Influence of changes in the prevailing synoptic conditions on the response of aerosol characteristics to land and sea breeze circulations at a coastal station. *Bound.-Layer Meteor.*, **108**, 145–161.
- Moorthy, K. K., and Coauthors, 2005: Wintertime spatial characteristics of boundary layer aerosols over peninsular India. *J. Geophys. Res.*, **110**, D08207, doi: 10.1029/2004JD005520.
- Moorthy, K. K., S. K. Satheesh, S. S. Babu, and C. B. S. Dutt, 2008: Integrated campaign for aerosols, gases and radiation budget (ICARB): An overview. *Journal of Earth System Sciences*, **117**(S1), 243–262.
- Moorthy, K. K., V. S. Nair, S. S. Babu, and S. K. Satheesh, 2009: Spatial and vertical heterogeneities in aerosol properties over oceanic regions around India: Implications for radiative forcing. *Quart. J. Roy. Meteor. Soc.*, **135**, 2131–2145.
- Nair, V. S., and Coauthors, 2007: Wintertime aerosol characteristics over the Indo-Gangetic plain (IGP): impacts of local boundary layer processes and longrange transport. J. Geophys. Res., 112, D13205, doi: 10.1029/2006JD008099.
- Niranjan, K., V. Srikanth, B. L. Madavan, T. Anjana devi, and B. Spandana, 2008: Temporal characteristics of aerosol physical properties at Visakhapatnam on the east-coast of India during I-CARB— Signatures of transport onto Bay of Bengal. *Journal* of Earth System Sciences, 117, 421–427.
- Parameswaran, K., R. Rajan, G. Vijaykumar, K. Rajeev, K. K. Moorthy, P. R. Nair, and S. K. Satheesh, 1998: Seasonal and long-term variations of aerosol content in the atmospheric mixing region at a tropical station on the Arabian Sea-Coast. *Journal of Atmospheric* and Solar Terrestrial Physics, 60, 17–25.
- Parameswaran, K., P. R. Nair, R. Rajan, and M. V. Ramana, 1999: Aerosol loading over Coastal and Marine environments in the Indian Ocean region during winter season. *Current Science*, 76, 947–955.

- Pillai, P. S., and K. K. Moorthy, 2001: Aerosol mass-size distributions at a tropical coastal environment: Response to mesoscale and synoptic processes. Atmos. Environ., 35, 4099–4112.
- Pillai, P. S., and K. K. Moorthy, 2004: Size distribution of near-surface aerosols and its relation to the columnar aerosol optical depths. *Annales Geophysicae*, 22, 3347–3351.
- Pillai, P. S., S. S. Babu, and K. K. Moorthy, 2002: A study of PM, PM10 and PM2.5 concentration at a tropical coastal station. Atmospheric Research, 61, 149–167.
- Ramachandran, S., and A. Jayaraman, 2002: Premonsoon aerosol mass loadings and size distributions over the Arabian Sea and tropical Indian Ocean. J. Geophys. Res., 107(D24), 4738, doi: 10.1029/2002JD002386.
- Reddy, R. R., K. Rama Gopal, K. Narasimhulu, L. Siva Sankara Reddy, and K. Raghavendra Kumar, 2007: Aerosol size distribution variation in Anantapur (14.62°N, 77.65°E) semi-arid zone and its impact on aerosol effective radius. Aerosol and Air Quality Research, 7, 550–562.
- Reddy, B. S. K., and Coauthors, 2011: Aerosol climatology over an urban site, Tirupati (India) derived from columnar and surface measurements: First time results obtained from a 30-day campaign. *Journal of Atmospheric and Solar Terrestrial Physics*, 73, 1727–1738.
- Satheesh, S. K., V. Vinoj, and K. K. Moorthy, 2006: Vertical distribution of aerosols overan urban continental site in India inferred using a micro pulse lidar. *Geophys. Res. Lett.*, 33, L20816, doi: 10.1029/2006GL027729.
- Satheesh, S. K., and Coauthors, 2009: Vertical structure and horizontal gradients of aerosolextinction coefficients over coastal India inferred from airborne lidar measurements during the integrated campaign for aerosol, gases and radiationbudget (ICARB) field campaign. J. Geophys. Res., 114, D05204, doi: 10.1029/2008JD011033.
- Stull, R. B., 1998: An Introduction to Boundary Layer Meteorology. Kluwer Academic Publishers, Dordrecht, 666pp.
- Vinoj, V., S. S. Babu, S. K. Satheesh, K. K. Moorthy, and Y. J. Kaufman, 2004: Radiative forcing by aerosols over Bay of Bengal region derived from ship-borne measurements and MODIS optical depths. J. Geophys. Res., 109, D05203, doi: 10.1029/2003JD004329.