Seasonal variation of surface and vertical profile of aerosol properties over a tropical urban station Hyderabad, India

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[1] One year measurement of vertical profiles of volume backscatter and extinction coefficient, aerosol optical depth (AOD), mass concentration of black carbon (BC) and composite aerosol along with thermodynamic structure of the atmosphere has been carried out over an urban tropical location of Hyderabad(17.47°N, 78.58°E), India, during April 2009 to March 2010. The mean mixing layer height (MLH) exhibits large seasonality exceeding 4 km in pre-monsoon period whereas in winter it comes down to ~1.5 km with an annual mean value of 2.35 ± 1.02 km. Surface BC mass fraction (F_{BC}) shows marked seasonal variation from winter (13 ± 1.9%), pre-monsoon (8.19 ± 2.16%), monsoon (7.3 ± 1.8%) to post-monsoon (11.8 ± 0.18%). The profiles of volume backscatter and extinction coefficients reveal presence of elevated aerosol layers from 2 to 4 km and strong oscillations during pre-monsoon (March–May) and monsoon (June–September) seasons, respectively, while in post-monsoon (October–November) and winter (December–February), the aerosols are well within the lower boundary layer and also exhibit a drastic decrease with increasing altitude. These elevated aerosol layers and vertical distribution appear to be closely linked to the thermodynamic structure of the atmosphere. The aerosol optical properties in conjunction with air mass back trajectory analysis indicate that the observed elevated aerosol layers during pre-monsoon and monsoon could contain significant fraction of coarse mode particles with a mix of dust and marine aerosols. Further analysis reveals that the aerosols within atmospheric boundary layer (ABL) dominate the column aerosol loading with ABL-AOD contributing to ~77.7 ± 17.0%, with significant seasonal variation from winter (86.2 ± 13.1%), pre-monsoon (76.6 ± 12.8%), monsoon (54.2 ± 15.6%) to post-monsoon (80.8 ± 14.8%).

Seasonal variation of ABL-AOD and BC mass fraction follows similar pattern in the ABL indicating that BC may be an important contributor to the ABL aerosol loading.

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1. Introduction

[2] Over the last few decades, South Asia has witnessed a rapid increase in population, industrialization, and energy demands, and consequently two to sixfold increase in emission of atmospheric aerosols and other pollutants [Lawrence and Lelieveld, 2010 and references therein]. Atmospheric aerosol in the region is a major factor that contributes to regional atmospheric warming, disruption in regional precipitation patterns, intensification of storms, and melting of Himalayan glaciers and snow packs [Carrico et al., 2003; Ramanathan et al., 2007; Gautam et al., 2010]. The spatial and temporal inhomogeneities in surface, columnar, and vertical aerosol characteristics, due to the variety of the aerosol sources and sinks, the influence of special features of meteorology (for instance monsoon), mixing processes, and their intricate interaction with clouds in this region, have been imposing large uncertainties in understanding the regional as well as global climate change. This is particularly important for the Indian subcontinent with all its natural diversities, high population density, diverse living habits, and the growing industrialization and urbanization.

[3] A variety of aerosol sources and particle types exist in the Indian subcontinent, which have been receiving significant attention to understand and model their chemical composition and radiative impact at regional scales [Lawrence and Lelieveld, 2010]. Aerosols over the Indian subcontinent can be broadly classified into anthropogenic/industrial aerosols [composed of a large fraction of black carbon (BC), sulphate, nitrate, and ammonium], desert or mineral dust
(regionally produced or long-range transported), biomass burning (either from crop residue or forest fires), and sea salt (transported in to the mainland from the humid marine air masses in the summer monsoon). However, it can vary significantly in the local and regional scales. Systematic measurements have been carried out over several locations namely Dibrugarh (27.3°N, 94.5°E) [Pathak et al., 2010], Minicoy (8.3°N, 73.04°E) [Vinoj et al., 2010], Nainital (29.23°N, 79.41°E) [Dumka et al., 2011], Kathmandu (27.67°N, 85.31°E) [Ramana et al., 2004], Kullu Valley (31.9°N, 77.11°E) [Guleria et al., 2012], Delhi (28.63°N, 77.17°E) [Srivastava et al., 2012], Kanpur (26.47°N, 80.33°E) [Misra et al., 2012], Ahmedabad (23.03°N, 72.55°E) [Ganguly et al., 2006], Pune (18.53°N, 73.85°E) [Devara et al., 2008], Visakhapatnam (17.7°N, 83.3°E) [Niranjan et al., 2007, 2012], Bangalore (13.0°N, 77.0°E) [Babu et al., 2002], eastern coastal India and Bay of Bengal [Moorthy et al., 2009], and Maldives (5.0°N, 73.5°E) [Eck et al., 2001] to examine the aerosol particle properties. Results from these studies reflect the large variation in surface, columnar, and vertical aerosol particle characteristics specific to the region.

Despite the fact that the aerosol columnar properties and surface BC concentrations have been well examined over India and Southeast Asia [Lawrence and Liebelveld, 2010], the vertical aerosol profiles have not been monitored and documented well except for a few isolated studies [e.g., Campbell et al., 2012]. The aerosol vertical profiles over south Asia by means of lidars, aircrafts, or balloons have been carried out over the oceanic regions in the frameworks of extensive field campaigns, such as Indian Ocean Experiment (INDOEX) [Franke et al., 2001; Müller et al., 2003; Forêt et al., 2006], Integrated Campaign for Atmospheric Aerosols, Gases, and Radiation Budget (ICARB) [Babu et al., 2008; Satheesh et al., 2009], and Winter Integrated Campaign for Atmospheric Aerosols, Gases, and Radiation Budget (W-ICARB) [Sinha et al., 2011], while over Indian mainland there are only few investigations made during the last decade [e.g., Jayaraman et al., 1995; Moorthy et al., 2004; Tripathi et al., 2005; Satheesh et al., 2006; Gadhavi and Jayaraman 2006; Niranjan et al., 2007; Devara et al., 2008; Hegde et al., 2009; Rajeev et al., 2010, Mishra et al., 2010; Komppula et al., 2012; Mishra et al., 2012].

Over Hyderabad (17.47°N, 78.58°E), India, vertical profiles of aerosol particles have been investigated by Gadhavi and Jayaraman [2006] using in situ aircraft flights on campaign mode basis. Surface-based lidar profiles are reported for specific events such as long-range-transported desert dust [Badarinath et al., 2007, 2010] and biomass smoke from crop residue burning [Badarinath et al., 2009]. In the present work, as compared with previous ones, we analyze 1 year records of lidar profiles providing the seasonality of the vertical aerosol particle scattering distribution and investigate this presence in relation to mixing layer variation and long-range transport.

The understanding of the specific characteristics of near-surface and free-troposphere aerosol particles can be achieved by lidar profiling by examining extinction and backscatter coefficient, lidar ratio, depolarization ratio, and attenuated color ratio [e.g., Dulac and Chazette, 2003; Balis et al., 2004]. Each may be very different both over time and vertical distribution and may not always be resolvable from passive ground-based and satellite remote sensing based on their optical significance. Aerosol layers above the atmospheric mixing layer are mainly associated with long-range transport and may contribute significantly to the column aerosol optical depth (AOD) variations [Lin et al., 2007; Xu et al., 2008]. The vertical distribution of aerosols is thus very critical in understanding their effect on radiative forcing and climate [Meloni et al., 2005; Pelon et al., 2008; Koch and Del Genio, 2010; Liu et al., 2012]. Especially over the Indian subcontinent, aerosol particle distribution influences the heating stability and thermal structure of the atmosphere, which has vital implications on regional warming and in modification of thermal gradient between land and ocean and in cloud formation processes [Menon et al., 2002; Lau et al., 2006; Gautam et al., 2009, 2010; Kuhlmann and Quaas, 2010; Ramachandran and Kedia, 2010; Chakravarty et al., 2011].

The highly concurrent and comprehensive measurements of the aerosol particle properties are necessary for understanding the science of atmospheric pollution in the region and also for the development of mitigation solutions firmly grounded on science, which will lead to societal benefits. Multi-instrument measurements using micro-pulse lidar (MPL), sun photometer Microtops-II (MT), Aethalometer, quartz crystal microbalance (QCM), and radiosonde profiles were collected out over Hyderabad, India, during a 1 year period (April 2009 to March 2010). The seasonal variation in the aerosol vertical profiles as well as the contribution of BC aerosol to the boundary layer aerosol loadings and the association of all the above with thermodynamic structure of the atmosphere are the main highlights of the present study.

### 2. Instruments and Data Analysis

The instruments used in the present work (see Table 1) were operating at the National Balloon Facility (NBF) of Tata Institute of Fundamental Research (TIFR), located about 15 km from the center of Hyderabad, at the northeast edge of the city. Detailed descriptions about the site as well as the seasonal variability in meteorological parameters over Hyderabad can be found elsewhere [e.g., Kaskaoutis et al., 2009; Sinha et al., 2012a]. The primary instruments of focus for this analysis are as follows.

| Table 1. List of Measured Parameters and Instruments Used in the Present Study |
|---|---|---|---|---|
| S. No. | Parameters Measured | Instruments | Study Period |
| 1 | Extinction coefficient, backscatter coefficient, LR | Micro pulse lidar (532 nm) | April 2009 to March 2010 |
| 2 | AOD | Microtops-II sun photometer | April 2009 to March 2010 |
| 3 | Black Carbon mass concentration | Seven channel Aethalometer | April 2009 to March 2010 |
| 4 | Aerosol mass concentration | Quartz Crystal Microbalance | July 2009 to March 2010 |
| 5 | Vertical profile of meteorological parameters | Radiosonde (RS-RW-80) | April 2009 to March 2010 |
2.1. Micro-Pulse Lidar

[9] The light detection and ranging (lidar) system has the capability to profile structured layers of aerosol particles through the middle and upper troposphere and can also be used for identifying several dynamic parameters such as mixing layer height (MLH) and entrainment zone depth [Monà et al., 2012]. A portable lidar based on the Micro Pulse Lidar (MPL) technique [Spinhirne, 1993] was used in the present study operating in late-evening hours (after sunset) under cloudless condition on intermittent days during April 2009 to March 2010. The main operating characteristics of the MPL are summarized in Table 2. A Nd:YAG laser with second harmonic output at 532 nm is used as the light source. The transmitted energy is 10 μJ with a pulse repetition frequency of 2.5 kHz. The laser system ranging bin width was set at 200 ns corresponding to a vertical resolution of 30 m, and backscatter signal profiles were integrated into a time bin of 1 min and stored for subsequent data analysis.

[10] The power of the analog signal detected by lidar after elastic scattering due to presence of air molecules and aerosol particles simultaneously can be expressed as

\[ P(r) = P_L \left( \frac{\eta(\lambda)}{\beta(\lambda, r) \gamma(r)} \right) \frac{c^2 \lambda}{2} e^{-\frac{2}{c} \int \sigma(\lambda, r) dr} \]  

(1)

where \( P_L \) is the emitted laser power, \( \eta(\lambda) \) the area of receiver of the telescope, \( \beta(\lambda, r) \) the spectral transmission factor, \( \gamma(r) \) is the overlap correction factor between the field of view of the telescope and the laser beam, \( c \) the speed of light, \( f \) the laser pulse length, \( r \) the range, and \( \sigma(\lambda, r) \) and \( \beta(\lambda, r) \) are the range-dependent volume extinction and backscatter coefficient, respectively. Equation (1) contains two unknown atmospheric parameters and is related through lidar ratio \([i.e., \text{extinction-to-backscatter ratio (LR)}]\). It is a very critical task to determine the LR in order to solve the lidar equation. Although there are several other methods for solving the lidar equation and calculating extinction profile [Welton et al., 2001, Kovalev and Eichinger, 2004], the Fernald-Klett inversion method is a fully adequate approach for the purpose of the present study. The lidar equation derived after the Fernald-Klett inversion is given as

\[ \beta = \frac{\exp[-(S_{ref}) - S(r_0)]]}{\beta(r_{ref}) + \frac{2}{C} \int_{r_0}^{r_{ref}} dr \exp[-(S(r_0) - S(r_{ref}))]} \]  

(2)

where \( \beta(r_{ref}) \) is the boundary condition set on \( \beta(r) \) at the reference far-end range. The vertical profile of the backscattered signal is calculated by solving equation (4) expressed as

\[ S(r_{ref}) - S(r_0) = \ln[r_{ref}^2 P(r_{ref})] - \ln[r^2 P(r)] - \frac{3}{4\pi} \int_{r_0}^{r_{ref}} \beta(r) dr + \frac{2}{C} \int_{r_0}^{r_{ref}} \beta_s(r) dr \]  

(3)

where \( X \) is the normalized range-corrected signal (NRCS).

[11] The value of 0.035 for the constant \( C \) (inverse of assumed LR) is accounted to solve equation (4), which could be treated as an average value for rural, urban, and maritime aerosol [Kovalev, 1993]. The upper limit of \( r \) is chosen at 8 km, where the aerosol contribution is negligible relative to particles scattering random noise and the magnitude of clean-air signal [Leon et al., 2009]. The Rayleigh correction is applied to raw signal using measured pressure and temperature profiles from the radiosonde observations over the site.

[12] Although MPL features a co-axial optical geometry and the laser beam of the transmitter overlaps almost perfectly with the receiver’s field of view (FOV), a range still exists within which the plane of scattered radiation is not completely overlapped by the detector FOV. The imperfect overlap must to be corrected for the lower level aerosol particle properties [Campbell et al., 2002]. In order to overcome the complexity associated with overlap, this MPL was designed in such a way that the laser head and transmitting optics are mounted on the side of the receiver telescope and housed in a closed box to maintain a clean, thermally stable, and dry environment [Kumar, 2006]. The FOV of the receiver telescope is <400 μm, which is double than the divergence of the expanded laser beam (Table 2). With this arrangement, a complete overlap between the laser beam and the telescope FOV was achieved at every altitude above 150 m. This further justifies our choice of operation near sunset, since the wide FOV would lower signal to noise for profiles collected exclusively during bright daytime hours alone.

### 2.1.1. Estimation of Extinction Coefficient

[13] The lidar equation has to be solved to calculate the aerosol extinction coefficient with the arbitrary selection of LR. Measured AODs from sun photometer during daytime were used to constrain total signal transmission through the presumably surface-detached aerosol layers, which are assumed well mixed, constrain is then applied for adjusting the LR and get the best fit value of LR by an iterative

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### Table 2. Operating Characteristics of the Micro Pulse Lidar (MPL)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Transmitter</th>
<th>Receiver</th>
</tr>
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<tbody>
<tr>
<td>Pulse energy</td>
<td>10 μJ</td>
<td>Cassegrain</td>
</tr>
<tr>
<td>Pulse repetition rate</td>
<td>2.5 kHz</td>
<td>Diameter: 15 cm</td>
</tr>
<tr>
<td>Pulse duration</td>
<td>&lt;10 ns</td>
<td>Telescope F-ratio: 9</td>
</tr>
<tr>
<td>Beam divergence</td>
<td>&lt;1.5 mrad</td>
<td>Filter bandwidth: 0.5 nm</td>
</tr>
<tr>
<td>Divergence of the expanded laser</td>
<td>&lt;200 μrad</td>
<td>Field of view: &lt;400 μrad</td>
</tr>
<tr>
<td>Wavelength</td>
<td>532 nm</td>
<td>Quantum efficiency: &lt;10%</td>
</tr>
<tr>
<td>Polarization</td>
<td>Linear</td>
<td></td>
</tr>
<tr>
<td>Range resolution (height interval)</td>
<td>30 m</td>
<td></td>
</tr>
</tbody>
</table>
processes [Fernald, 1984]. Thus, in the absence of direct LR measurements, column-averaged LR values may be estimated by constraining the vertical integral of lidar-derived aerosol extinction coefficients with independent AOD measurements [Welton et al., 2000; Chazette, 2003; He et al., 2006]. The estimation of LR following this procedure requires the extinction profile up to the top of the atmosphere and concurrent measurements of columnar AOD from sun photometer, which may introduce bias in the present estimation when the distinct aerosol layers are aloft beyond the detection limit of MPL. We assume that the particle size distribution and the aerosol properties are homogenous throughout the atmospheric vertical column, so that the LR can be assumed constant in the vertical. Evaluation of the effect of this assumption is difficult, since it is impossible to know how much the aerosol properties change with altitude and, even if they were known, there are no analytical tools at this point to estimate the effect of the changes on the inversion [Lewandowski et al. 2010]. Kovalev [1995] showed that inappropriate LR values may alter the retrievals under inhomogeneous atmospheres. The uncertainty in the estimated LR values is ~10–15%, which is comparable to that which was reported by Franke et al. [2001]. It is shown that the effective lidar ratio is highly sensitive to the selection of reference signal \( S(r_0) \) and causes an important source of uncertainty in lidar data retrieval [Chazette, 2003]. The uncertainty in \( S(r_0) \) depends on signal-to-noise ratio at given altitude and on the possible occurrence of residual aerosol and clouds [Leon et al., 2009].

### 2.2. Sun Photometer

[Spectral AOD measurements have been performed systematically over Hyderabad during cloudless condition using portable Microtops sun photometer (MT) and ozonometer that measure the direct-beam irradiance at seven wavelengths; six (380, 440, 500, 675, 870, and 1020 nm) are used for aerosol retrievals. In the present study, MT-derived AODs were used for days corresponding with their night MPL observations. The aerosol retrievals are carried out from the instantaneous solar flux measurements using the instrument’s internal calibration. Typical errors in AOD measurements from MT are ~0.03 [Morris et al., 2001; Porter et al., 2001]. More details about the description of MT, errors, and uncertainties can be found elsewhere [e.g., Kaskaoutis et al., 2010 and references therein]. The corrections for molecular scattering, ozone, and/or water vapor absorption at specific wavelengths are taken care of internally and, therefore, the uncertainties due to these effects have negligible contribution to AOD.

### 2.3. Aethalometer

[Continuous real-time measurements of the mass concentration of BC aerosols were carried out using a portable seven-wavelength (370, 470, 520, 590, 660, 880, and 950 nm) Aethalometer (model AE-42, Magee Scientific). This instrument applies a semi-continuous optical absorption method to measure the attenuation of light by aerosol particles at selected wavelength channels. It aspirates ambient air at a standard flow rate of 5 L/min from an altitude of ~10 m above the ground level. The measurement sampling interval was kept at 5 min. Aerosol particles are continuously deposited on a quartz fiber filter that acts as a perfect diffuse scattering matrix with light absorbing particles embedded on it. The BC mass concentration is then determined from the change in transmittance through the quartz filter tape due to the particle deposition. Due to strong absorption of BC aerosol at 880 nm, the mass concentrations measured at this wavelength are considered as standard for BC measurements.

Filter-based absorption techniques for the measurement of BC aerosol particles encounter various systematic errors that need to be corrected [Bond et al., 1999a, 1999b]. The overestimation of BC mass concentration due to multiple scattering \((C)\) within the filter is partly compensated by higher particle loading called the shadowing effect \((R)\) in the filter, which decreases the optical path. Based on several experiments, Weingartner et al. [2003] found that the shadowing effect and multiple scattering factor are quite significant for pure soot particles, while being almost negligible for aged aerosols (mixture of several components). In the present study, the values of \(R=1\) and \(C=2.355, 2.656, 2.677, 2.733, 2.827, 2.933,\) and 2.925 were used for the seven wavelengths, respectively, following Schmid et al. [2006]. The maximum uncertainty for BC can reach 20% (ranging from ~40 to 60 ng m\(^{-3}\)), with higher percentage error for low mass concentrations [Moorthy et al., 2007].

### 2.4. Quartz Crystal Microbalance

[A quartz crystal microbalance (QCM) cascade impactor (model PC-2HX, California Measurements Inc., USA) was used for the aerosol mass concentration measurements. The instrument deploys 10 stages with 50% efficiency cutoff radii at 10, 5.6, 3.0, 2.0, 1.0, 0.5, 0.3, 0.2, 0.1, and 0.05 \(\mu m\). It samples ambient air at a constant flow rate of 2 slpm (standard liters per minute) and was operated every 30 min during day time. Before a measurement, filtered air is flushed in order to facilitate stability within the crystals. During each measurement, relative changes in frequency between the sampling and reference crystals are recorded as a function of time, and the composite aerosol mass concentration calculated by assuming a particle density of 2 \(g/cm^3\) for relative humidity conditions below 75%. The uncertainties in aerosol mass concentration are in the range of 10–20% at very low

<table>
<thead>
<tr>
<th>Month</th>
<th>Number of Days</th>
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<tbody>
<tr>
<td>Apr 2009</td>
<td>5</td>
</tr>
<tr>
<td>May 2009</td>
<td>2</td>
</tr>
<tr>
<td>Jun 2009</td>
<td>2</td>
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<tr>
<td>July 2009</td>
<td>2</td>
</tr>
<tr>
<td>Aug 2009</td>
<td>2</td>
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<tr>
<td>Sep 2009</td>
<td>2</td>
</tr>
<tr>
<td>Oct 2009</td>
<td>4</td>
</tr>
<tr>
<td>Nov 2009</td>
<td>7</td>
</tr>
<tr>
<td>Dec 2009</td>
<td>10</td>
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<tr>
<td>Jan 2010</td>
<td>5</td>
</tr>
<tr>
<td>Feb 2010</td>
<td>5</td>
</tr>
<tr>
<td>Mar 2010</td>
<td>9</td>
</tr>
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</table>

*Winter: December–February; pre-monsoon: March–May; monsoon: June–September; post-monsoon: October–November.*
mass concentrations (~10 $\mu g \text{ m}^{-3}$). The error reduces to <10% for high mass concentrations >30 $\mu g \text{ m}^{-3}$ [Nair et al., 2009]. QCM measurements were used in combination with Aethalometer-derived BC mass concentration in order to calculate the BC mass fraction during days of coincident MPL observations.

2.5. Radiosonde

Meteorological parameters including temperature, pressure, relative humidity (RH), wind speed, and direction, were profiled during the study period using RS80-15N GPS radiosondes. The uncertainty in temperature measurements is ±0.3°C, while it is ±0.5 hPa for pressure below 20 km. The accuracy of RH measurements depends upon ambient temperature as well as altitude, and it varies from ±2% near the surface to ±15–20% between 5 and 15 km [Manchanda et al., 2012]. In the present study, thermodynamic profiles [i.e., potential temperature ($\theta$), relative humidity (RH) and wind speed and direction] are examined along with the vertical profiles of lidar backscatter coefficient on specific days in order to reconcile the influence of thermodynamic structure on the aerosol particle distribution over Hyderabad. The data base for the determination of MLH (illustrated in Figure 1) is acquired from similar radiosonde observations during daytime at an unused Hyderabad airport (17.45°N, 78.47°E) that is ~10 km away from the present study site.

3. Results and Discussion

3.1. Black Carbon Aerosols and Mixing Layer Variations

This section analyzes boundary-layer variation and BC contribution to aerosol mass load near the surface. Figure 1 shows the monthly variation of MLH and F$_{BC}$ from April 2009 to March 2010. MLH was estimated using radiosonde measurements, considering the MLH as the height where potential temperature presents the sharpest gradient near surface (i.e., height of the elevated inversion which segregates the mixed layer from the stable layer above). F$_{BC}$, defined as the relative apportionment of BC to the composite aerosol mass concentration, is an index characterizing the source contribution of aerosol particles. F$_{BC}$ was calculated from the composite aerosol mass and BC mass concentrations using QCM and Aethalometer measurements, respectively. QCM measurements are not available before July 2009. Although the contribution of F$_{BC}$ is usually below 12%, it is an important parameter for the estimation of BC radiative forcing due to its potential radiative effects, which are comparable in magnitude to those of greenhouse gases [Jacobson, 2001].

The MLH exhibited large monthly variability from ~1.5 km during August to January to ~4 km during April to May with an annual mean value of 2.35 ± 1.02 km (Figure 1). Surface air temperature can reach up to 37°C in late pre-monsoon, decreases from monsoon to winter, and controls the annual pattern of MLH which in turn influences the vertical distribution of aerosol particles [Satheesh et al., 2006; He et al., 2006]. Large convection in the pre-monsoon period can reduce the local emissions at the surface and increase upper level aerosol particle concentrations. Similar to these findings, lidar profiles collected at Delhi reveal that, on an average, the aerosol layer exhibits highest thicknesses during pre-monsoon (~5.5 km), when surface heating and therefore, convective mixing is at its annual maximum [Komppula et al., 2012].

The F$_{BC}$ exhibits large annual variation and increases gradually from August to December (except for a small decrease in November), corresponding with a shallower mixing layer. A different pattern is observed from winter to pre-monsoon, presumably due to weakening of BC emissions, deeper boundary layer, and air masses that originated from different sectors. F$_{BC}$ varies from 5% to 15%, with mean values for winter, pre-monsoon, monsoon, and post-monsoon of 13 ± 1.9%, 8.19 ± 2.16%, 7.3 ± 1.8%, and 11.8 ± 0.18%, respectively. Satheesh et al. [1999] report much lower BC mixing ratio values of 6% over Kaashidhoo in Maldives (5.0°N, 73.5°E). Oceanic observations over Bay of Bengal (BoB) showed that F$_{BC}$ varied from 5.8 ± 0.6% (pre-monsoon) to 2.9 ± 1.15% (post-monsoon) [Sumanth et al., 2004]. Overall, the analysis shows that the F$_{BC}$ near the surface is larger during post-monsoon, especially in winter, and is highly associated with MLH and dynamics, thus likely playing a significant role in the structure of the vertical aerosol profiles.

3.2. Vertical Profiles of Extinction and Backscatter Coefficient

The seasonal mean vertical profiles of aerosol extinction coefficient over Hyderabad are shown in (Figure 2). In these profiles, data with low signal-to-noise ratio were removed following the methodology by Kim et al. [2007]; thus, we screened the cloud-contaminated data if the extinction coefficient is larger than 1.0 km$^{-1}$ in the boundary layer and >0.5 km$^{-1}$ for higher altitudes.

Contrasting features associated with noticeable seasonal variability of the aerosol extinction coefficient are observed from these data. The vertical profiles of aerosol extinction coefficient during pre-monsoon and monsoon seasons reveal a smoother decrease of the extinction coefficient with altitude, elevated, and structured aerosol layers from 2 to 4 km while a relatively thick particle layer near
the surface and a steep decrease with increasing altitude are observed during winter season. The low and thick layer near the surface during winter is most likely associated with the low MLH and highest BC concentrations (Figure 1). BC aerosol alone does not contribute solely to the aerosol extinction profile. However, vertical profiles of BC concentration during pre-plume and post-plume episodes show the enhanced presence of BC aerosols in the polluted plume and correspond with the trends observed in the composite aerosol particle concentration and size distribution over Hanimaadhoo (6.78°N, 73.18°E), Maldives [Corrigan et al., 2008]. Using high-altitude balloon-borne measurement, we reported elevated BC aerosol layers (similar structure seen in aerosol extinction profile) over Hyderabad during March 2010 [Babu et al., 2011].

[24] Aircraft measurements during February 2004 over Hyderabad showed a rapid decrease in BC concentration with altitude, reducing by a factor of 2 from the surface to 550 m. Above this, a subsequent gradual decrease was observed, with BC concentration decreasing from 1 to 0.8 μg m⁻³ from 550 to 2200 m [Moorthy et al., 2004]. This rapid decrease in aerosol extinction coefficient correlates with the lower surface inversion layer exhibiting weaker development near boundary layer compared to other seasons. This suggests that the surface extinction coefficient measured by lidar during winter comprised a relatively large BC fraction. The vertical structure of the volume

Figure 2. Mean seasonal variation of the vertical extinction coefficient (M m⁻¹) over Hyderabad during April 2009 to March 2010. The horizontal bars correspond to standard deviations from the seasonal mean. The values mentioned in the figures are the mean for each season at 1–3 km altitude and the parenthesis correspond to 10 and 90 percentiles for the same altitude range.
aerosol extinction coefficient is found to be similar to the measured vertical behavior of BC concentration for different seasons (as discussed above) over Hyderabad. However, this comparison can be treated as qualitative rather than quantitative due to different sampling periods. Extinction represents the combined effect of the composite aerosols.

[26] Similar seasonality of aerosol vertical profiles has been observed over India. Niranjan et al. [2006] report dense surface aerosol layers (up to ~2 km) in December 2004 over Kharagpur (22.31°N, 87.31°E), eastern India, while Ganguly et al. [2006] observed high extinction coefficient near the surface followed by a sharp decrease with increasing altitude during winter and post-monsoon seasons over Ahmedabad. Similar results were recently observed over Delhi [Komppula et al., 2012] and the foothills of Himalayas [Dumka et al., 2011] for the winter season. Komppula et al. [2012] found slightly higher aerosol concentrations in monsoonal periods, associated with larger aerosol particles (influence of transported dust), while the high extinction coefficient near the surface in post-monsoon and winter was supported by high BC mass concentrations. The mean extinction coefficient values for each season at 1–3 km altitude are also estimated (Figure 2) and found to be comparable (except pre-monsoon) with those reported over Delhi by Komppula et al. [2012].

3.2.1. Backscatter Coefficient

[27] The seasonal-mean profiles of backscatter coefficient shown as a function of altitude in Figure 3 are qualitatively similar to that found for the extinction coefficient, suggesting relative turbidity in the LR [Ackerman, 1998]. The annual variability of the aerosol vertical profiles is related to several parameters dealing with aerosol emissions, convection, boundary-layer dynamics, and long-range transport, along with the horizontal and vertical motion of the wind [Colette et al., 2008]. During winter season, the backscatter coefficient is at its highest values near the surface with a drastic decrease afterward due to weakening of thermal convection and turbulence. Anthropogenic aerosols and BC affect the study location in winter, associated with low wind speeds and stable atmospheric conditions causing accumulation of pollutants [Sinha et al., 2012a]. Some indications of increased values (aerosol plumes up to 4–5 km height) are observed in pre-monsoonal period which could be associated with dust plumes from northwestern directions [Badarinath et al., 2007]. The large differences in the seasonal profiles between near-surface aerosols, which nominally reflect local sources, and elevated aerosol layers, which are mainly transported from large distances, suggest vertical decoupling. The seasonal profiles of the backscatter coefficient over Hyderabad are similar to that was found over Delhi [Komppula et al., 2012]. However, the seasonal mean values of backscatter

Figure 3. Same as in Figure 2, but for backscatter coefficient (Mm⁻¹ sr⁻¹).
coefficient at 1–3 km altitude are found to be higher over Hyderabad for all seasons.

3.3. Comparison of Vertical and Columnar Aerosol Properties

[28] In this section, aerosol particle optical characteristics are analyzed from both MPL and MT observations. MT-derived aerosol particle properties correspond to daytime-averaged values for those days coinciding with MPL observations. The aerosol particle profiles from MPL have been divided into the atmospheric boundary layer (ABL) and free troposphere (FT) by determining MLH. MLH is an essential parameter for estimating the contribution of boundary layer and free tropospheric aerosols to the columnar AOD. An approach of the gradient method to detect MLH is shown in Figure 4, which illustrates the vertical profile of normalized range corrected lidar signal (NRCS) (left), its first derivative (middle), and potential temperature (right) for 1 January 2010 at Hyderabad. To estimate the contribution of boundary layer and free tropospheric aerosols, MLH is determined by detecting the first significant negative gradient of the NRCS that results from the strong decrease in aerosol backscatter due to lower particle concentration [Flamant et al., 1997; Amiridis et al., 2005; Baars et al., 2008]. Following this method, we estimate the AOD within and above the boundary layer by integrating the extinction coefficient profiles for altitudes below and above the MLH, considered as boundary layer and free troposphere contributions (i.e., ABL-AOD and FT-AOD) respectively.

[29] Figure 5 shows the daily variation of MT AOD, z (380–870 nm), LR, and fractional MPL AOD for the ABL and FT, respectively, at Hyderabad during April 2009 to March 2010.

March 2010. The high MT AOD (above 0.6) in pre-monsoon is attributed to the combined effects of a stagnant synoptic meteorological patterns, likely secondary aerosol formation through gas-to-particle conversion due to increased solar radiation and transported dust and biomass-burning plumes [Kaskaoutis et al., 2009; Sinha et al., 2012b]. Aerosol particles may also be under the influence of hygroscopic growth in the humid monsoon season, resulting in larger particle size and efficient scatterers [Reid et al., 1999]. High z values are observed in winter (1.34 ± 0.26), while they are lower in pre-monsoon and mainly in monsoon (Table 4). High z values (1.30 ± 0.17) are also observed in post-monsoon, usually associated with air masses coming from regions where significant biomass burning occurs. The ABL-AOD maximizes in the pre-monsoon and occasionally in monsoon and winter, while the FT-AOD is above 0.2 in pre-monsoon and monsoon seasons and much lower during the rest of the year. This is attributed to the stronger convection, buoyancy, and variations in MLH as well as to the long-range transport of naturally produced aerosol particles.

[30] The estimated LR using the iterative process ranges from 22 to 90, exhibiting a mean value of 42.5 ± 13.5. The LR is strongly related to aerosol particle size, type, shape and composition, refraction index, and also the wavelength (and or instruments) used for its determination [He et al., 2006; Barnaba et al., 2007; Müller et al., 2007]. Large LR values are typically associated with a scattering efficiency between spheres and spheroids, since the non-spherical particles exhibit large decrease of the backscatter coefficient compared to the spherical ones and consequently larger LR values [Mishchenko et al., 1997]. Higher LR values are observed in post-monsoon and winter most likely to be associated with absorbing anthropogenic

Figure 4. Vertical profile of normalized range corrected signal (NRCS) (left), its first derivative (middle) and potential temperature (right) for one typical day on 1 January, 2010 over Hyderabad. The atmospheric aerosol particles show the different vertical structure in the atmospheric boundary layer (ABL) and in the free troposphere (FT). Vertical profile of θ (K) is obtained from concurrent meteorological profiling during lidar observation.

Figure 5. Daily variation of the sun photometer MT-AOD, Angstrom exponent (z), lidar ratio (LR), and MPL-derived AOD in ABL and FT for all the days of lidar observations over Hyderabad during April 2009 to March 2010.
aerosols, while in monsoon they are significantly lower (Table 4), indicating influence of marine and or dust aerosols over the site. However, on specific days during monsoon, LR values are >40 sr, close to those reported for non-spherical dust particles [Barnaba et al., 2004; Pavese et al., 2009].

Komppula et al. [2012] report LR values below 40 sr (at 532 nm) during winter and pre-monsoon over Delhi, indicating the presence of cleaner air with relatively more aged aerosols. Similar to the present study, the highest LR values over Delhi are also found in post-monsoon (60 sr) associated with high AOD (at 532 nm) during winter and pre-monsoon over Delhi, indicating the influence of the marine boundary layer decreasing the LR to ~30 sr.

Franke et al. [2003] found that the vast majority of aerosols were below 532 nm for well-aged polluted air were found in the range 40–45 sr over Maldives during INDOEX [Müller et al., 2001], while freshly emitted continental polluted plumes exhibited higher values (45–75 sr). In contrast, clean marine atmospheres presented lower LR values (20–30 Sr). Chen et al. [2009] report monthly mean LR (532 nm) >55 sr over Maldives during INDOEX at heights above 1 km, associated with small absorbing particles when the air masses traversed the eastern and northeastern India. The influence of the marine boundary layer decreased the LR to ~30 sr. Liu et al. [2006] also found similar results about the contribution of the near-surface aerosols to columnar AOD, as they reported 60–80% contribution for winter and 30–40% for summer. Furthermore, Franke et al. [2003] report a 30–60% contribution of free-tropospheric aerosol layers over Maldives during INDOEX. Similarly, Liu et al. [2012] found that the vast majority of aerosols were below 18 (July 2003) to 44 sr (March 2004) in Hong Kong [He et al., 2006]. Anderson et al. [2003] calculated LR values of 50 ± 5 sr for fine-mode dominated samples and 46 ± 8 sr for coarse-mode dominated samples at low RH during the ACE-Asia. Literature survey of various LR values estimated at different environments over the globe is presented in He et al. [2006] and Zhang et al. [2010], which are close to our findings in the vast majority of the cases.

The daily variation of the ABL and FT AODs is depicted in more detail in Figure 6. The data show that the ABL aerosols contribute from a minimum of 43.0% to a maximum of 79.9% to the total AOD. The mean ABL contribution is 77.7 ± 17.0%, corresponding to a mean ABL-AOD and FT-AOD of 0.38 ± 0.20 and 0.12 ± 0.11, respectively (Table 4). On a seasonal basis, the contribution of ABL-AOD to the total AOD was found to be 86.2 ± 13.1%, 76.6 ± 12.8%, 54.2 ± 15.6%, and 80.8 ± 14.8% for winter, pre-monsoon, monsoon, and post-monsoon, respectively. This seasonal variation is similar to that found for FBC, indicating that BC aerosols play an important role in the magnitude of ABL-AOD over Hyderabad.

Satheesh et al. [2006] also found similar results about the contribution of the near-surface aerosols to columnar AOD, as they reported 60–80% contribution for winter and 30–40% for summer. Furthermore, Franke et al. [2003] report a 30–60% contribution of free-tropospheric aerosol layers over Maldives during INDOEX. Similarly, Liu et al. [2012] found that the vast majority of aerosols were below 18 (July 2003) to 44 sr (March 2004) in Hong Kong [He et al., 2006]. Anderson et al. [2003] calculated LR values of 50 ± 5 sr for fine-mode dominated samples and 46 ± 8 sr for coarse-mode dominated samples at low RH during the ACE-Asia. Literature survey of various LR values estimated at different environments over the globe is presented in He et al. [2006] and Zhang et al. [2010], which are close to our findings in the vast majority of the cases.

The mean LR was found to be 29.1 ± 4.0 sr for non-spherical dust particles [Barnaba et al., 2004; Pavese et al., 2009].

Table 4. Seasonal Averages for Aerosol Characteristics over Hyderabad During the period April 2009 to March 20101

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Winter</th>
<th>Pre-monsoon</th>
<th>Monsoon</th>
<th>Post-monsoon</th>
<th>Yearly</th>
</tr>
</thead>
<tbody>
<tr>
<td>MT AOD500</td>
<td>0.44 ± 0.13</td>
<td>0.65 ± 0.17</td>
<td>0.41 ± 0.11</td>
<td>0.40 ± 0.11</td>
<td>0.48 ± 0.17</td>
</tr>
<tr>
<td>530–870</td>
<td>1.34 ± 0.26</td>
<td>1.25 ± 0.51</td>
<td>0.62 ± 0.22</td>
<td>1.30 ± 0.17</td>
<td>1.20 ± 0.416</td>
</tr>
<tr>
<td>ABL AOD532</td>
<td>0.34 ± 0.12</td>
<td>0.56 ± 0.24</td>
<td>0.22 ± 0.12</td>
<td>0.28 ± 0.09</td>
<td>0.38 ± 0.206</td>
</tr>
<tr>
<td>FT AOD532</td>
<td>0.06 ± 0.07</td>
<td>0.19 ± 0.13</td>
<td>0.18 ± 0.08</td>
<td>0.07 ± 0.06</td>
<td>0.12 ± 0.11</td>
</tr>
<tr>
<td>LR</td>
<td>45.0 ± 17.9</td>
<td>40.9 ± 10.3</td>
<td>31.7 ± 9.1</td>
<td>46.1 ± 8.7</td>
<td>42.5 ± 13.5</td>
</tr>
</tbody>
</table>

1Each season has only few data, so the mean values must be interpreted carefully. The ABL and FT AOD532 were obtained from normalized lidar profiles.

Figure 6. Daily variation of MPL AOD532 for atmospheric mixing layer (ABL) and free troposphere (FT) over Hyderabad during April 2009 to March 2010. The mean ABL contribution (%) to the total AOD corresponding to a mean ABL-AOD and FT-AOD is also mentioned in the figure. For more details see the text and Table 4.
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3.4. Aerosol Sources and Transport

The combination of the air-mass trajectory analysis along with the vertical profile of aerosol particle scattering provide important information about the vertical structure, and sources for transported particles may be roughly interpreted from these data as well and the transport mechanisms of aerosol particles [Wang et al., 2010]. In order to study the relation between synoptic air masses and aerosol particle properties over Hyderabad, 5-day isotropic back trajectories at three altitudes 500, 1500, and 4000 m above ground level (assumed as the representative altitudes below, at, and above the MLH) are analyzed on specific days when MPL profiles are available (Figure 7) using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model [Draxler and Rolph, 2003]. It is found that air masses originate from five primary pathways, each exhibits with different characteristics within our measurements: (1) air masses originating from Africa-Arabia-Middle East, which are rich in coarse-mode aerosols; (2) air masses from southern Arabian Sea and northern Indian Ocean, featuring a significant maritime component; (3) air masses from IGP, which are mainly composed of anthropogenic aerosols; (4) air masses from BoB and Southeast Asia that are enriched in biomass burning during the dry season; and (5) local air masses dominated by urban-industrial aerosols. Similar source regions, transport pathways, and aerosol particle properties have been identified by air-mass trajectories at several locations in India [e.g., Pathak et al., 2012; Guleria et al., 2012; Kumar et al., 2012; Beegum et al., 2012].

3.5. Vertical Distribution of Aerosols and Thermodynamic Structure

Vertical profiles of aerosol particle scattering and thermodynamic structure of the atmosphere can be used to interpret the atmospheric layers [Corrigan et al., 2008]. MPL has the capability for resolving boundary layer evolution at relatively high spatial and temporal scales. Regular profiling of vertical profiles of temperature, RH, and wind speed and direction was carried out using radiosonde (RS-80) for days of MPL observations in order to study the (i) potential of the MPL technique for monitoring of thermodynamic structure of the atmosphere and (ii) distributions of aerosol particle scattering in the vertical scale and the impact of atmospheric stability [Martucci et al., 2007]. In this aspect, we present five cases (16 April, 4 November, 26 December 2009, 9 January, and 21 March 2010) corresponding to different seasons, weather conditions, synoptic scale wind pattern, and air mass advection, which are each depicted in Figures 8a–8e. The presence of stratuscumulus cloud at ~2 km altitude is seen for two cases: (i) 4 November 2009 and (ii) 9 January 2010. The lidar signal declined with altitude within the stratuscumulus cloud, from signal attenuation, which is a common limitation for profiling liquid water cloud layers with lidar.

Figure 8 (left column) shows the variation of aerosol particle vertical structure in terms of the normalized range-corrected signal (NRCS in Phe km$^{-2}$/µs per shot), while the right column depicts the time-averaged vertical profiles of backscatter coefficient BSC (in Mm$^{-1}$sr$^{-1}$), potential temperature $\theta$ (K), RH (%), and wind speed (ms$^{-1}$) and direction (°). This NRCS is obtained by correcting the lidar raw signal for after-pulse behavior and energy normalization correction following the methodologies described elsewhere [Campbell et al., 2002; Pelon et al., 2008]. The time-height distribution of NRCS shows strong dependence with altitude. It is fairly persistent up to ~1.5 and ~4 km for 9 January and 16 April, respectively, indicating shallower and deeper mixing layer heights. In general, the NRCS and BSC reveal that the aerosol vertical profiles are similar during nighttime.
hours of the same day, as interpreted from the negligible standard deviations.

[39] Nighttime MPL measurements on 16 April 2009 (Figure 8a) reveal the presence of a stable, well-mixed, and rather homogeneous aerosol layer within the ABL extending up to 4–5 km height, with an LR value of ~40 sr that is characteristic of a well-mixed aerosol layer with anthropogenic dominance. The air mass for this case is mainly of

Figure 7. Five-day air-mass back trajectories for the specific days shown in Figures 8 and 9 at 500, 1500, and 4000 m: (a) 16 April 2009, (b) 26 December 2009, (c) 09 January 2010, (d) 21 March 2010, (e) 04 November 2009, (f) 21 June 2009, (g) 23 July 2009, and (h) 15 September 2009. The backward air mass trajectory is obtained from the NOAA/NCEP HYSPLIT model.
continental origin (Figure 7a) likely incorporating dust and anthropogenic aerosols over the studied region given as AOD = 0.66 and $\alpha = 0.69$. The mean BSC profile shows a relatively rather homogeneous scattering layer from 1 to 4 km, while the $\theta$ profile is mostly adiabatic (~330K) till 5 km. Large increases in $\theta$ values with altitude above 5 km suggests a convectively stable sub-adiabatic atmospheric condition. RH is very low (about 25–30%) below 2.5 km, increasing up to 5 km in the elevated aerosol layers, and then rapidly decreasing above the inversion. Moderate west and northwesterly winds prevail during the lidar profiling.

Patterns differ noticeably during winter season. Aerosol particles are mostly confined near the surface, and their concentration is reduced drastically with altitude This is likely responsible for the frequent occurrence of low level haze and fog due to high humidities induced by nocturnal radiative cooling during winter over Hyderabad. Two characteristic winter days are shown in Figures 8b and 8c for 26
December 2009 and 9 January 2010, respectively. For both days, the lower air mass originates from the Indian mainland or Southeast Asia, associated mainly with low northeasterly winds and likely incorporating abundant aerosol loading during its transit. Although at 4000 m, the air parcel encountered oceanic regions and thus likely relatively clean air (Figures 7b and 7c). On 26 December, the BSC is high near the surface, reducing drastically through 1 km. For 9 January, the high BSC values extended up to 2 km. The BSC profile shows that most of aerosol particles are confined below 1.5 km in the night and early morning hours for the next day.

This variability in aerosol particle vertical distribution in winter season is compared with the thermodynamic structure of the atmosphere. Winter nights are characterized by elevated inversion layers, due to strong cooling at the surface and warming at upper levels [Tiwari et al., 2003]. Vertical profiles of $\theta$ and RH depict a strong inversion at 600 m and large gradient at ~2.5 km. Above this height, RH exhibits a sharp negative gradient decreasing from 60% to 5% on 26 December, indicating a warm drier atmosphere and stable stratified layer that inhibits the vertical mixing. Vertical profiles of wind speed and direction indicate strong winds (>5 m s$^{-1}$) suggesting the likely

Figure 8. Typical profiles of normalized range squared signal (left column: Phe km$^{-2}$/μm s per shot), backscatter coefficient (BSC) and concurrent radiosonde measured thermodynamic structure (right column: i.e., potential temperature ($\theta$), relative humidity (RH), and wind speed and direction) shown for comparison on specific days over Hyderabad during April 2009 to March 2010: (a) 16 April 2009, (b) 26 December 2009, (c) 09 January 2010, (d) 21 March 2010, and (e) 04 November 2009. The measured and estimated aerosol parameters (AOD, $\alpha$, and LR) are also indicated in each figure. Phe = photoelectrons, μj = microjoule, and μs = microsecond.
Figure 8. (continued)
advection of distinct air masses prevail above 1.5 km. BSC profiled on 9 January 2010 reflects several aerosol layers which each correlate well with the vertical variation of thermodynamic profiles. During winter season, lower atmospheric layers over Hyderabad are influenced by urban emissions, including BC aerosols. In general, the well-mixed layer is within the shallower boundary layer. Satheesh et al. [2006] report the presence of aerosol particle layers between 1.0 and 1.5 km during winter nighttime over Bangalore, but not during summer and post-monsoon. Although such layers are observed at almost all nights preceding sunny days in winter, similar features are not found in our study. Nevertheless, the data for 26 December 2009 reveal some indication of layers above a clearly defined shallow nocturnal layer of ~300–600 m thickness, but without the much scattering intensities from the cases over Bangalore.

On 21 March 2010 (Figure 8d), vertical profiles of BSC do not vary significantly during the nighttime. Aerosols are mainly confined within the lowest layer (<0.5 km) and gradually decrease up to 3 km. Vertical profiles of BSC decreases by a factor of 3 from the surface to 1 km, with large variability evident at higher altitudes. The near-surface aerosol load is higher than those observed in the previous cases discussed above. Mean MT AOD$_{500}$ is very high (0.9), associated with relatively large values of $z_{aero} \approx 1.34$. The lower level air mass originated from the east (Figure 7d), where extensive forest fires occurred in Southeast Asia during the measurement, as seen from MODIS observations (not shown). The transport of biomass-burning aerosols from Southeast Asia (back trajectory at 1500 m) may be responsible for the elevated layers up to 3 km, as observation is consistent with observed thermodynamic structure. The $\theta$ profile is adiabatic with a relatively strong capping inversion and well-mixed atmospheric layer extended from near the surface to 4 km. A convectively stable sub-adiabatic atmospheric condition is observed above this. RH is low, increasing up to the MLH and rapidly decreasing afterwards. The relatively calm wind (5–7 m s$^{-1}$) and backing winds suggest mixing of different air masses carrying mostly anthropogenic absorbing aerosol which is also reflected from estimated LR value of 60 sr (typical for polluted air mass). Franke et al. [2003] report higher LR values over the Maldives, corresponding to absorbing aerosol particle characteristics which showed relatively high values of $z_{aero} \approx 1.41 \pm 0.29$ and mean AOD$_{500}$ (0.43 ± 0.06). On this day, air masses at lower levels originated mostly from IGP and Southeast Asia (Figure 7e). Badarinath et al. [2009] report backscatter coefficient values at 532 nm as high as 6 Mm$^{-1}$ sr$^{-1}$ near the surface for an average day and 13 Mm$^{-1}$ sr$^{-1}$ for a polluted day with biomass-burning aerosols over Hyderabad. These values are close to those observed in our case.

3.6. Multiple Aerosol Layers in the Free Troposphere

Three case studies are shown to illustrate the presence of multiple structured elevated aerosol layers above MLH as well as plume-like aerosol characteristics at the edge of the mixing layer during monsoon season (Figures 9a–9c). For each day, the NRCS reveals the presence of multiple aerosol layers, the so-called “Monsoon Layers” [Manoj and Devara, 2011], as well as their oscillatory nature. The bottom and top height of these layers are found to be near 2.0 and 5.5 km, respectively. Parameswaran et al. [1997] report that such layers are most likely generated by accretion of aerosols in stable layers sandwiched between turbulent layers. Plume-like aerosol features just above the boundary layer (3–4 km) are visible on 21 June, 23 July, and 15 September (Figures 9a–9c). More specifically, the NRCS on 21 June (Figure 9a) reveals the presence of additional elevated aerosol layers at 3–4 km with thickness ranging from ~50 to 500 m above the well-defined boundary layer. A considerable variation in thickness and intensity is found during the 4 h period of MPL profiling. The BSC profile (Figure 9a) also reveals the structure of thin elevated aerosol layers that are comparable in magnitude with the boundary layer. Accurately assessing the origin of such layers is highly uncertain, since they are somewhat different than those observed during major dust storms [Liu et al., 2008b; Chen et al., 2010; Zhang et al., 2010; Satheesh et al., 2009; Müller et al., 2003; Ganguly et al., 2006; Niranjan et al., 2006, 2007; Satheesh et al., 2006, 2009; Rajeev et al., 2010; Mishra et al., 2010; Komppula et al., 2012] show that the aerosol plumes can rise up to 3–5 km, while some of the studies [e.g., Franke et al., 2003; Satheesh et al., 2006, 2009; Rajeev et al., 2010; Mishra et al., 2010] note multilayered aerosol particle structure.

Vertical profiles of RH exhibit large variation with strong gradients ranging from 53% to 86% at ~3 km and increasing from 3 to 4 km. Winds were mostly southwesterly (except for 15 September) exhibiting considerable fluctuation with altitude and intensity ranging from 5 to 25 m s$^{-1}$ at 3 km. During monsoon season, the upper levels of the boundary layer correspond with strong turbulent mixing, and thus lofting of aerosols upward. However, the thermodynamic structure evident does not support the mixing of aerosol from the surface as $\theta$ increases with altitude, indicating a stratified atmosphere. Further, the presence of elevated layers is not reflected in surface measurements of BC and composite mass concentration (not shown) indicating weak or almost no exchange across the mixing layer. Long-range transport is likely responsible for the observed layers. The back trajectories at all altitudes for all 3 days (Figures 7f–7h) show the air masses mainly originating from the west including the Arabian Peninsula and Arabian Sea, which suggest dust mixed with marine aerosol particles. This is consistent with observed optical aerosol particle characteristics which showed relatively low values of $z_{aero} \approx 0.84$, 0.76, and 0.69 and high values of AOD$_{500}$ (0.33, 0.46, and 0.43) and large contributions of the FT-AOD (56%, 56%, and 48%). The relatively low values of $z_{aero}$ indicate the presence of coarse-mode aerosol particles at elevated layers. The low LR values 29, 33, and 40 sr for 21 June, 23 July, and
Figure 9. Same as in Figure 8 but for typical days of lidar profiles during the monsoon season: (a) 21 June 2009, (b) 23 July 2009, and (c) 15 September 2009. Vertical profiles of NRCS reveal the multiple thin layers and plume-like aerosol structure.
15 September, respectively, indicate an enhanced presence of marine and or dust aerosols [Ackermann, 1998].

4. Summary and Conclusions

[46] In situ measurements of surface aerosol particle mass concentration (composite and BC), column integrated particle optical properties (aerosol optical depth), and vertically resolved aerosol scattering profiles (volume backscatter and extinction coefficient) along with thermodynamic structure [i.e., potential temperature (θ), relative humidity (RH), and wind speed and direction] are presented over Hyderabad, India for April 2009 to March 2010. The vertical aerosol profiles are found to be strongly dependent on diurnal and seasonal changes based on the regional and synoptic meteorology, origin and transport pathways of air masses, and the variation in mixing layer height (MLH). The major findings of the study can be summarized as follows:

1. Surface BC mass fraction (F_BC) shows marked seasonal variation from winter (December–February; 13 ± 1.9%), pre-monsoon (March–May; 8.19 ± 2.16%), monsoon (June–September; 7.3 ± 1.8%) to post monsoon (October–November; 11.8 ± 0.18%). The mixing-layer height (MLH) exhibited seasonal variability, exceeding 4 km in pre-monsoon and falling to 1.5 km in winter. The seasonal vertical distribution of aerosol particles correlates closely with this feature.

2. Micro pulse lidar (MPL) measurements reveal complex and variabilities of the aerosol particle vertical distribution within the mixing layer and free troposphere. The majority of aerosols are confined within the boundary layer (below 2 km). In some cases during pre-monsoon and monsoon seasons, they reach higher altitudes (~4 km). Within the boundary layer, aerosol particles are mostly of local origin corresponding with high F_BC, while they mainly originated from long-range transport in the free troposphere. Seasonal mean profiles reveal the presence of a relative thick aerosol layer near surface to 2 km, except for pre-monsoon and monsoon seasons when this aerosol layer extends to higher altitudes.

3. The vertical distribution of aerosols does not exhibit significant daily variation during post-monsoon and winter when air masses originate from the polluted Indo-Gangetic Plains (IGP). The variability in vertical profiles, as observed from the standard deviations of lidar backscatter, was larger in pre-monsoon when air masses of contrasting characteristics (dust particles, biomass burning, and marine aerosols) and various intensity influence the study region. Vertical lidar extinction and backscatter coefficient profiles reveal that the aerosol properties are significantly modified depending on season, MLH, thermodynamic structure and long-range transport.

4. The contribution of atmospheric boundary layer AOD to total columnar integrated AOD was found to be 86.2 ± 13.1%, 76.6 ± 12.8%, 54.2 ± 15.6%, and 80.8 ± 14.8% for winter, pre-monsoon, monsoon, and post-monsoon, respectively. ABL-AOD and BC mass fraction follow similar patterns in the ABL, indicating that BC may be an important contributor to the ABL aerosol loading.

5. Aerosol vertical profiles revealed multiple thin layers and plume-like aerosol characteristics on some days in monsoon season, the so-called “Monsoon Layers.” These layers are most likely generated by accretion of aerosols in stable layers that are sandwiched between turbulent layers [Parameswaran et al., 1997]. The observed aerosol particle layered structure is likely associated with the advection of distinct air masses of different origin through long range transport.

[47] This study illustrated the usefulness of combined analysis of vertical, columnar integrated aerosol particle optical properties along with thermodynamic structure of the atmosphere. It is seen that the temporal inhomogeneities in surface, columnar, and vertical aerosol particle characteristics due to the variety of the aerosol sources and sinks, the influence of dynamic and synoptic meteorology, and the mixing processes. This study also highlights the importance of the measurement of vertical profile of aerosol particle distribution with varying origin. A study on the local and regional variation of mixed-composite aerosols particle radiative forcing using radiative transfer model constrained by observation of aerosol particle properties, with case studies focusing at the episodes of anthropogenic pollution, biomass burning, dust, and marine aerosol transport needs to be carried out. Results from this study would provide significant motivation for future studies relating to their regional radiative and climate impact.

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