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Estimation of particulate matter from satellite- and ground-based observations over Hyderabad, India

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Long-term trends in surface-level particulate matter of dynamic diameter ≤2 μm (PM_2) in regard to air quality observations over Greater Hyderabad Region (GHR), India are estimated by the synergy of ground-based measurements and satellite observations during the period 2001–2013 (satellite) and July 2009–Dec 2013 (ground-based). Terra Moderate Resolution Imaging Spectroradiometer (MODIS)-derived aerosol optical thickness (AOT) (MODIS-AOTs) was validated against that measured from Microtops-II Sunphotometer (MTS) AOTs (MTS-AOTs) and then utilized to estimate surface-level PM_2 concentrations over GHR using regression analysis between MODIS-AOTs, MTS-AOTs, and measured PM_2. In general, the MODIS-estimated PM_2 concentrations fell within the uncertainty of the measurements, thus allowing the estimate of PM_2 from MODIS, although in some cases they differed significantly due to vertical heterogeneity in aerosol distribution and the presence of distinct elevated aerosol layers of different origin and characteristics. Furthermore, significant spatial and temporal heterogeneity in the AOT and PM_2 estimates is observed in urban environments, especially during the pre-monsoon and monsoon seasons, which reduces the accuracy of the PM_2 estimates from MODIS. The estimates of PM_2 using MTS or MODIS-AOT exhibit a root mean square deference (RMSD) of about 8–16% against measured PM_2 on a seasonal basis. Furthermore, a tendency of increasing PM_2 concentrations is observed, which however is difficult to quantify for urban areas due to uncertainties in PM_2 estimations and gaps in the data set. Examination of surface and columnar aerosol concentrations, along with meteorological parameters from radiosonde observations on certain days, reveals that changes in local emissions and boundary-layer dynamics, and the presence or arrival of distinct aerosol plumes aloft, are major concerns in the accurate estimation of PM_2 from MODIS, while the large spatial distribution of aerosol and pollutants in the urban environment makes such estimates a considerable challenge.

1. Introduction

Due to the dramatic increase in population, industrialization, and energy demands, atmospheric aerosols and pollutant emissions have gradually been increasing over

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South Asia (Dey and di Girolamo 2011; Kaskaoutis et al. 2011a). Long-term satellite observations over India show that aerosol load and pollutants are higher over the densely populated Indo-Gangetic Plains (IGP) (Dey and di Girolamo 2010), major urban centres (Ramachandran and Cherian 2008), as well as around power plants (Prasad, Singh, and Kafatos 2011; Shaiganfar et al. 2011). Over Hyderabad in particular, previous studies showed high levels of aerosols, particulate matter, black carbon (BC), and air pollutants (NOx, SO_2, organics, tropospheric O_3, etc.), rendering it as one of the most polluted cities in India (e.g. Beegum et al. 2009; Dumka et al. 2013; Gummeneni et al. 2011; Swamy et al. 2012). Aerosols over Hyderabad are mostly composed of fine particles containing a complex mixture of sulfate, soot, and smoke (organic and inorganic substances), which is further deteriorated by long-range transport of dust and sea-salt particles (Badrinath et al. 2007, 2010; Kaskaoutis et al. 2009; Sinha et al. 2012, 2013a). Both monitoring and forecasting of particulate matter concentrations over major urban centres has therefore received significant attention among scientists, policy-makers, health regulatory agencies, and the media (Dey et al. 2012; Singh and Kaskaoutis 2014). Based on air quality standards and human mortality, the US Environmental Protection Agency has defined threshold values of 12–15 μg m^{-3} (primary–secondary) and 35 μg m^{-3} for the annual and 24-hour $PM_{2.5}$ averages, respectively, as the air quality standard (EPA: http://www.epa.gov/air/criteria.html). The Central Pollution Control Board of India has also recently set these standards as 40 and 60 μg m^{-3} for annual and daily averages, respectively (http://cpcb.nic.in).

Traditionally, ground-based instruments have been used to measure the mass concentration of $PM_{2.5}$; these, however, represent point measurements without being capable of providing the spatial distribution of particulate matter. On the other hand, satellite remote sensing, with its promising spatial coverage, offers a new dimension in the study and monitoring of air quality from space (Gupta et al. 2006, 2013; Lee et al. 2011). Satellite remote sensing is an innovative and potential technique for monitoring and estimation of particulate matter air quality from space at various spatial and temporal resolutions. Several studies have suggested various techniques to estimate the surface-level $PM_{2.5}$ mass concentration using aerosol optical thickness (AOT) retrieval from polar (Engel-Cox et al. 2004; Liu, Franklin, and Koutrakis 2007; Gupta and Christopher 2009a, 2009b; Van Donkelaar et al. 2010) and geostationary (Paciorek et al. 2008) orbits. The most common technique uses a linear regression between measured AOT and surface $PM_{2.5}$ (μg m^{-3}), based on the assumption that AOT and $PM_{2.5}$ are linearly related. However, the variety of aerosol sources and long-range transport, along with variable meteorological conditions and mixing-state (external and internal) processes, strongly modify aerosol microphysical and optical properties and perturb the AOT–$PM_{2.5}$ relationship. Furthermore, the accuracy of $PM_{2.5}$ estimation from satellites depends on the accuracy of AOT retrieval, which is a function of aerosol type and surface reflectance (Kahn et al. 2010; Levy et al. 2010), vertical aerosol homogeneity, and the atmospheric dynamics within the boundary layer. The Moderate Resolution Imaging Spectroradiometer (MODIS) on board the EOS Terra and Aqua satellites provides almost daily AOT retrievals over cloud/snow/ice-free global regions at a spatial resolution of 10 km × 10 km, and has been mostly used for $PM_{2.5}$ monitoring (Hoff and Christopher 2009). MODIS has recently released a 3 km × 3 km aerosol product, which is very useful over urban cities to evaluate and monitor small-scale gradients in pollution.

The present work aims at estimating surface PM_2 levels over an urban environment in GHR, India using the synergy of satellite (MODIS) and ground-based (Microtops-II (MTS) and quartz crystal microbalance (QCM)) measurements, and also highlights the limitations of satellite estimation of particulate matter from space. MODIS-derived aerosol optical thickness (MODIS-AOT)s was compared to Microtops-II Sunphotometer (MTS) AOT (MTS-AOT)s for the validation of satellite retrievals. Correlations of measured PM_2 concentrations with MTS- and MODIS-AOTs are used to establish relationships among them, and a statistical approach for PM_2 estimation and mapping over GHR during the period 2001–2010 was carried out. Very few studies have provided satellite-derived particulate matter estimates for India (Gupta et al. 2006; Dey et al. 2012). The present work is the first of its kind to perform PM_2 estimation over GHR using a MODIS data set, and is also aimed at providing PM_2 spatio-temporal variation over the past decade in an urban environment.

2. Study location and aerosol climatology

The ground-based measurements were carried out at the premises of the Tata Institute of Fundamental Research, Balloon Facility (TIFR-BF) Campus (17°28′N, 78°34′E), located around 15 km from the urban centre of Hyderabad, at the northeastern edge of the city (Figure 1(*b*)). GHR has a population of over 6 million and is considered one of the most aerosol-laden urban centres in India, with large concentrations of BC and anthropogenic aerosols (Beegum et al. 2009). In general, high AOT (mostly coarse mode) dominates

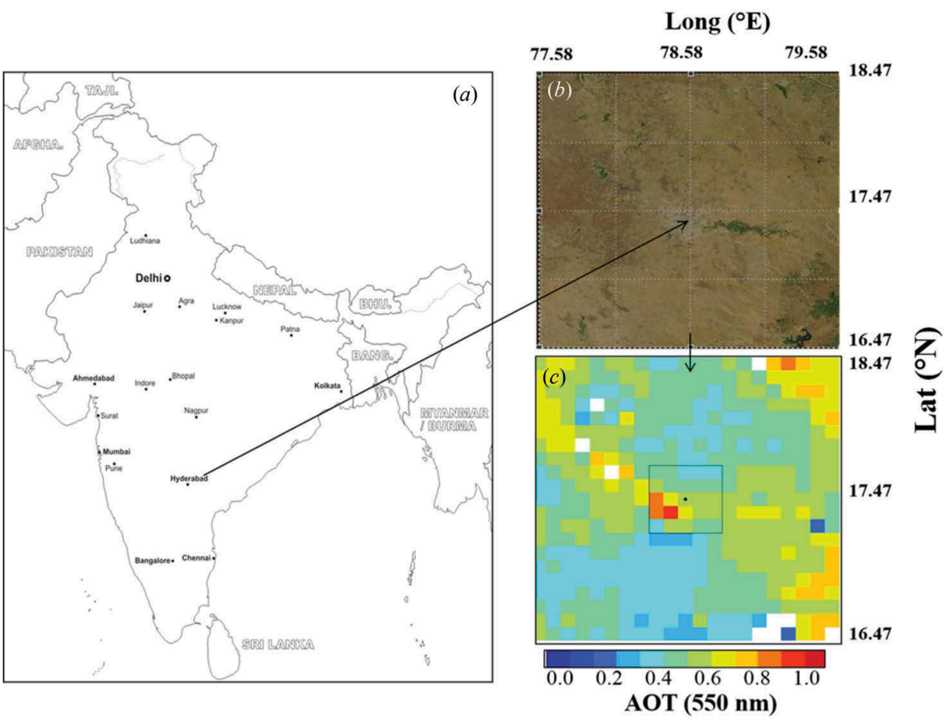

Figure 1. Terrain of Peninsular India (*a*) and site map of the measurement location at Hyderabad (*b*). The coloured map represents 10 years' (2001–2010) mean AOT_{550} from Terra-MODIS with 2° × 2° spatial domain centred at the measurement site (*c*). The AOT_{550} values are shown on the colour scale. The smaller domain within the map denotes an area of 50 km × 50 km covering the Hyderabad urban area.

during the pre-monsoon (March–May) and monsoon (June–September) seasons, while in the post-monsoon (October–November) and winter (December–February), fine mode and steep AOT spectra are the prominent features over Hyderabad (Kaskaoutis et al. 2009; Sinha et al. 2012, 2013a). Ten-year (2001–2010) mean quality-controlled (Level 2; 10 km × 10 km) AOT (550 nm) retrieval from MODIS around GHR is shown in Figure 1(*c*). The inset image corresponds to MODIS averaged AOT during the period 2001–2010 covering a 2° × 2° spatial domain centred at the study location. The small square at the centre of the inset corresponds to urban Hyderabad. MODIS-AOT exhibits a wide range, with higher values (AOT > 0.8) over the urban centre. Kharol et al. (2011) reported much higher AOT values over urban Hyderabad (Level 2, 10 km × 10 km data set) than those found when the surrounding environs are included in the spatial domain (Level 3, 1° × 1° data set), suggesting large spatial heterogeneity in aerosol distribution. Similarly, Gupta et al. (2013) found significant heterogeneity in aerosol loading around Lahore, Pakistan, strongly influenced by local emission rates, micro-meteorological factors controlling the atmospheric circulation over an urban environment, and the location of heavy industries. Most of the industrial zones are situated in eastern, northeastern, and northwestern Hyderabad, and may contribute to the high aerosol loading observed from MODIS over these areas. Southern Hyderabad is mostly forested area with less industrial activity and sparse habitat, and is associated with relatively low MODIS-AOT values. These findings reveal the necessity for satellite retrieval at high spatial resolution and accuracy above urban areas, where pronounced spatio-temporal variations, changes in emission rates, and specific meteorological air parcels (cells) render aerosol distribution very heterogeneous.

3. Data and methodology

3.1. Ground-based measurement of aerosols

The entire series of ground-based measurements and GPS radiosonde observations were carried out at TIFR-BF during the period July 2009–December 2013, using the instruments described below. A QCM cascade impactor (model PC-2HX, California Measurements Inc., USA) was used for size-segregated total aerosol mass concentration measurements. The instrument deploys 10 stages with 50% efficiency cut-off radii at 10, 5.6, 3.0, 2.0, 1.0, 0.5, 0.3, 0.2, 0.1, and 0.05 μm. For the scope of the present work, the sum of the six stages of diameter <2.0 μm (i.e. cut-off radius <1 μm) was used to estimate total particle concentrations, referred to hereafter as PM_2. The instrument samples the ambient air through a community inlet at a constant flow rate of 2 l per minute and temporal resolution of 30 minutes during daytime (i.e. from around 9:00 to 17:00 IST (IST = UTC+5:30)). The measurements are restricted to atmospheric conditions with relative humidity (RH) <75% in order to avoid aerosol growth via humidification. RH was recording continuously at the weather station site and the QCM measurements were stopped when RH exceeded 75%. Ramachandran and Jayaraman (2002) estimated a maximum overall uncertainty of about 25% for aerosol mass measurement for all the stages of QCM, which is much lower (around 10%) at high mass concentrations. In the present study, 30 minute QCM retrievals were averaged on an hourly, daily, and seasonal basis. Especially for the comparison between PM_2 and MODIS-AOTs, the QCM measurements were averaged at a time interval of ±30 minutes of the satellite overpass. The composite aerosol mass concentration (total mass: M_T) obtained from QCM measurements was used in combination with an aethalometer (model AE-42, Magee Scientific), which measured BC mass concentration, in order to calculate the BC mass fraction (F_{BC}). The mass concentrations measured at 880 nm are considered as standard for BC measurements (Sinha et al. 2013b). Detailed descriptions of BC measurements and uncertainty analysis are presented elsewhere (Sinha et al. 2013b and references therein).

The spectral AOT measurements were carried out under cloudless conditions from July 2009 to December 2013 using a well-calibrated portable Microtops-II (MTS) model (Solar Light Company, USA). The MTS measures direct-beam irradiance at seven wavelengths, six of which (380, 440, 500, 675, 870, and 1020 nm) are used for aerosol retrieval. Aerosol retrieval was derived from instantaneous solar flux measurements based on the internal calibration of MTS. The field of view (FOV) of the instrument is 2.5°, while the full width at half-maximum (FWHM) band width at each AOT channel is 2.4 ± 0.4 nm. Spectral AOT was measured every 30 minutes, from around 7:00 to 17:30 IST under clear-sky conditions. Inter-comparison (at wavelength 500 nm) was regularly carried out with a multi-wavelength radiometer (Sinha et al. 2012) available on-site, and data corrected for any offset. The typical error in AOT measurement from MTS is about 0.03 (Morys et al. 2001; Porter et al. 2001). MTS has been extensively used for aerosol studies over Hyderabad (Sinha et al. 2012, 2013a, 2013b). The perturbed irradiance measurements due to possible cloud contamination caused by cirrus clouds undetected by the human eye or due to inaccuracies in the targeting of the sun disk were excluded if the AOT and Ångström exponent (α) values exceeded two standard deviations from the daily mean value. In addition, for accuracy in spectral AOT retrieval, the method proposed by Kaskaoutis et al. (2011b) was applied in the present data set using the relation $a_2 - a_1 = \alpha$ (a_1 and a_2 are the constant terms in the second-order ln(AOT) *vs*. ln(λ) curve), excluding the data points that showed a large scatter (i.e. $R^2 < 0.97$ in the second-order polynomial fit).

3.2. Vertical measurement of aerosol profiles

A portable micro-pulse lidar (MPL) system operating at TIFR-BF was also used in the current study, for measurement of the vertical profiles of the aerosol extinction coefficient (β) on specific days, in order to examine the boundary-layer and free-troposphere dynamics that are related to surface particulate matter and columnar AOT. The MPL operated at 532 nm after sunset under cloudless conditions, providing vertical profiles of the aerosol extinction coefficient during the period April 2009–March 2010. The transmitted energy is 10 μJ, with a pulse repetition frequency of 2.5 kHz. The MPL employs a diode-pumped Nd-YAG laser system, a co-axial transceiver for transmitting laser pulses and detection of photons, a dedicated data acquisition system, and a computer control and interface system. More details about the instrumentation and method of analysis are discussed elsewhere (Kumar 2006; Sinha et al. 2013b).

Radiosonde measurements obtained from the University of Wyoming (*http://weather.uwyo.edu/upperair/sounding.html*) for Hyderabad station (15 km from the observation site) were also used to monitor the vertical atmospheric structure using the profiles of temperature, pressure, RH, wind speed (WS), and wind direction (WD). The uncertainty in temperature measurements is ± 0.3°C, while the accuracy for RH varies from ±2% near the surface to ±15–20% between 5 and 15 km. Thermodynamic profiles of potential temperature θ (K), RH (%), WS (m s^{-1}), and WD (°) were examined on specific days, along with the vertical profiles of the aerosol extinction coefficient in order to reconcile the influence of atmospheric thermodynamics on aerosol particle distribution and vertical profiles over Hyderabad.

3.3. Satellite retrievals

In this study, Level 2 C005.1 AOT_{550} retrievals at a spatial resolution of 10 km × 10 km from MODIS were used during the period 2001–2013, centred over Hyderabad (see Figure 1(*c*)). The MODIS sensor on board the Terra satellite passes over the equator from north to south at 10:30 LT (local time). The wide swath of 2330 km provides an early-global coverage of the Earth's surface in 1–2 days in the spectral range 0.41–15.00 µm, with a spatial resolution ranging between 250 and 1000 m. MODIS-AOT retrievals were performed by means of the dark target algorithm, and therefore, retrievals over bright surfaces (such as desert and bright urban areas) are biased (Levy, Remer, and Dubovik 2007). The expected uncertainty in MODIS-AOTs over land is 0.05 ± 0.15 × AOT, while several global validation studies over Aerosol Robotic Network (AERONET) locations revealed that about 72% of MODIS-AOT retrievals were within the expected uncertainty (Levy et al. 2010). MODIS-AOT was correlated with co-located data from MTS, and measured PM_2 concentrations obtained at ±30 minutes of MODIS overpass for the estimation of PM_2 over Hyderabad.

4. Results and discussion

4.1. Inter-comparison of sun photometer and MODIS-AOT

An inter-comparison between MTS-AOT_{550} and MODIS-AOT_{550} was carried out over Hyderabad during the period July 2009–December 2013 (Figure 2). The MTS-AOT_{550} values were calculated using the interpolation method (AOT_{550} = AOT_{500} $(500/550)^{\alpha}$ using Ångström's exponent α, which was calculated from AOT values at 500 and 675 nm ($\alpha = -\ln(AOT_{500}/AOT_{675})/\ln(500/675)$). The MTS-$AOT_{550}$ values are associated with errors well below 5%, due to the high accuracy of spectral AOT measurements. The hourly mean MTS-AOT_{550} values within ±30 minutes of the satellite overpasses were used for comparison with MODIS-AOT_{550} following the spatial and temporal co-location procedure described by Ichoku et al. (2002). Level 2 (10 km × 10 km) MODIS-AOTs with the highest quality (QA = 3) centred over GHR were utilized for the inter-comparison analysis. The correlation coefficient between MTS-AOT_{550} and MODIS-AOT from the linear regression was found to be 0.81, which lies within the uncertainties ±(0.05 ± 15% AOT), found in the correlation between MODIS and AERONET AOTs (Levy et al. 2010). Figure 2 shows that MODIS-AOTs are consistently lower than MTS-AOTs, with a mean bias of about −0.1 or 24% of the MTS-AOT_{550}, which is comparable to those obtained by various other validation studies over India and globally (e.g. Jethva, Satheesh, and Srinivasan 2007; Prasad and Singh 2007; Levy et al. 2010). Despite the limited number of observations for each year that satisfy the spatial and temporal selection criteria, the results show a satisfactory correlation, which is much better than those reported for Hyderabad by Kharol et al. (2011) using either Level 2 or Level 3 MODIS data. Kharol et al. (2011) found significant deviations between MODIS and Microtops-II Sunphotometer AOTs, along with poor correlations between Level 2 or Level 3 MODIS-AOTs, plausibly due to the spatial resolution and the degree of spatial AOT heterogeneity around GHR (see Figure 1(*c*)). The general agreement between MODIS retrievals and sun photometer AOTs indicates the potential capability of MODIS-based aerosol monitoring to reproduce aerosol distribution over and around GHR.

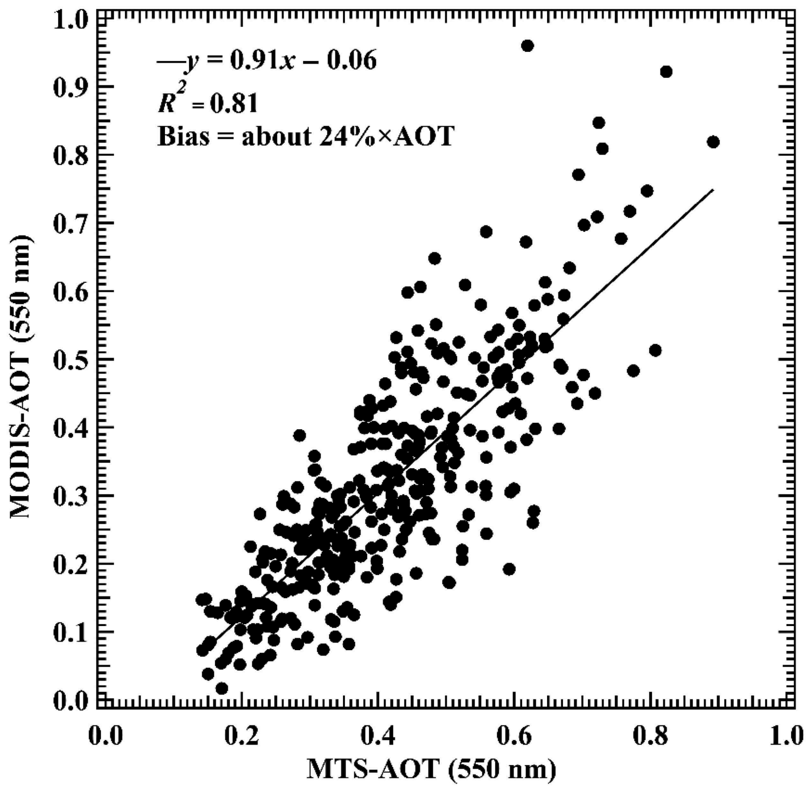

Figure 2. Inter-comparison between daily mean Microtops Sunphotometer AOT_{550} (MTS-AOT) and Terra-MODIS-derived AOT_{550} (MODIS-AOT) for co-located measurement over Hyderabad during July 2009–December 2013.

4.2. *Temporal variation of near-surface and column aerosols*

Figure 3(*a–b*) shows the daily mean variation of AOT_{550} values from both MTS and MODIS, as well as the F_{BC} over Hyderabad during the period July 2009–December 2013. A pronounced daily and annual AOT_{550} variability can be seen, with maximum values (0.5–0.9) during April–June and minimum (around 0.2) during the monsoon season. The significant variations in AOT and F_{BC} time series are attributed to the influence of anthropogenic emissions, boundary-layer dynamics, seasonally changed meteorological conditions, and long-range transport of various aerosol types and sources (Kaskaoutis et al. 2011a; Sinha et al. 2013a; b). The results show a negligible trend in MODIS-AOT_{550} and MTS-AOT_{550} over Hyderabad during the observational period, in contrast to the increasing trend observed from both Terra and Aqua MODIS over the city during the period October 2002–May 2008 (Kharol et al. 2011). Furthermore, Moorthy et al. (2013) found an annual average increase in AOT_{500} of 0.01 (3.63%) over central India.

F_{BC} (Figure 3*b*) exhibits large daily and seasonal variation, with higher values during post-monsoon and winter which are highly associated with the lower mixing-layer height during the cold period of the year (Sinha et al. 2013b). The daily mean variations in MTS-AOT_{550} and F_{BC} are generally consistent, except for the pre-monsoon season when the columnar AOT is strongly influenced by elevated aerosol layers

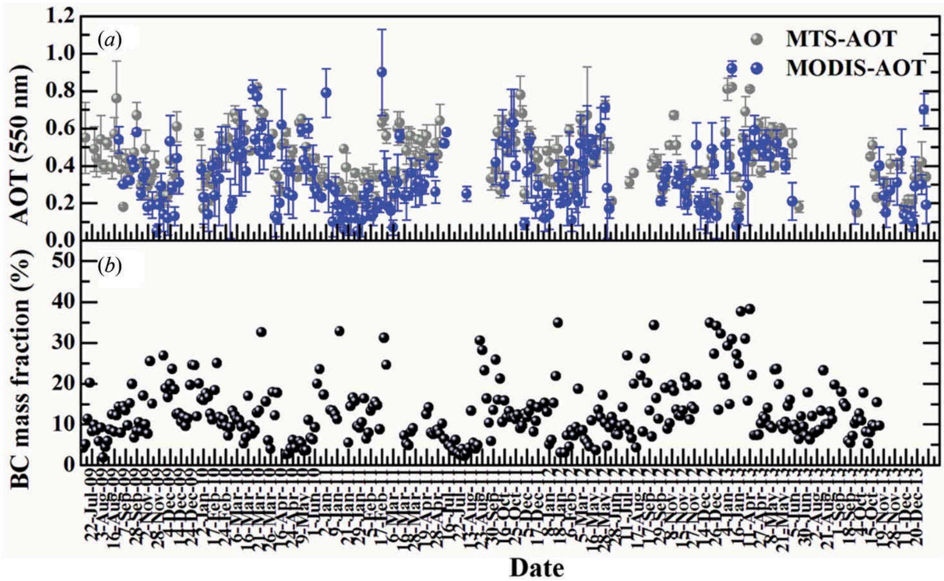

Figure 3. (*a*) Temporal variation in Microtops Sunphotometer AOT_{550} (MTS-AOT), Terra-MODIS AOT_{550} (MODIS-AOT), and (*b*) BC mass fraction over Hyderabad during the period July 2009–December 2013. Both AOTs and BC mass fractions represent daily mean values, whereas MODIS-AOT_{550} values are instantaneous observations corresponding to the same period of observation. There were no sufficient measurements of BC mass concentrations during November and December 2013, and BC mass fraction was not estimated for these months.

associated with long-range transport of dust and a deeper boundary layer, along with strong convective eddies (Sinha et al. 2013b). During late post-monsoon and winter seasons, the shallower boundary layer, frequent temperature inversions, calm winds, and a dry and stable atmosphere inhibit the vertical mixing of aerosols (Sinha et al. 2013b). During the pre-monsoon, the MTS-AOT_{550} values are at their highest levels (0.9) while the surface F_{BC} is less affected, exhibiting values comparable to those found in the post-monsoon and winter. This poses a serious concern in regard to the suitability of near-surface aerosol measurements for the characterization of columnar properties, especially when a significant amount of aerosols is present at elevated layers. In general, the relationship between surface and columnar aerosol properties can vary substantially even within a day, due to variations in the source regions, emission rates, aerosol transportation at different altitudes, as well as the presence of multiple structured aerosol layers aloft. Moreover, boundary-layer dynamics and changes in the mixing-layer height that control the dilution processes may influence the amount and variability of near-surface aerosol concentrations, with negligible effects on the column (Sinha et al. 2013b). Therefore, selected days exhibiting inconsistency in the diurnal variation between PM_2 and AOT over Hyderabad are examined in Section 4.3.1, along with MPL and radiosonde profiles.

4.3. PM2–AOT relationship

The scatter plots between co-located measurements of PM_2 mass concentrations and AOT_{550} from MTS and MODIS on a seasonal basis are shown in Figure 4. The results

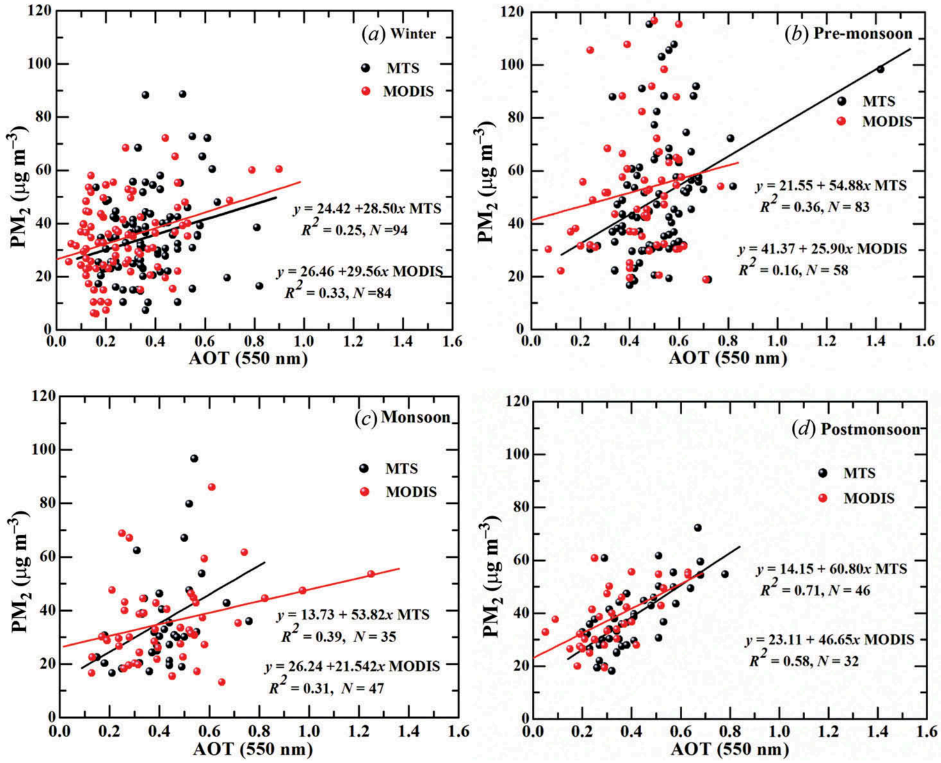

Figure 4. Scatter plot between surface PM_2 and AOT_{550} obtained from Microtops Sunphotometer (MTS) and Terra-MODIS (MODIS) for four typical seasons over Hyderabad for July 2009-–December 2013.

show moderate to low correlation between surface PM_2 and columnar AOTs, with correlation coefficients of 0.25(0.33), 0.36(0.16), 0.39(0.31), and 0.71(0.58) for MTS (MODIS) in winter, pre-monsoon, monsoon and post-monsoon, respectively. Despite the greater degree of scatter, the correlations were found to be statistically significant at the 95% confidence level ($p < 0.05$). The computed correlations are considered appropriate for an accurate estimation of PM_2 from MODIS over GHR, but this approach can be performed with relative accuracy in the post-monsoon. Physical and chemical aerosol properties are important parameters in the characterization of aerosol types and sources that control the scattering and absorption processes in the atmosphere and affect AOT and its spectral variation (Valenzuela et al. 2015). Thus, changes in the physico-chemical characteristics of near-surface and columnar aerosols may also affect the PM_2–AOT relationship. However, chemical composition measurements for a better assessment of the PM_2–AOT relationship are not available (except for BC) at the study location.

Despite the high F_{BC} (Figure 3(*b*)), along with the low mixing layer height (Sinha et al. 2013b), the relatively frequent occurrence of water-insoluble aerosol components (comprising mostly coarse-mode particles; results from the Optical Properties of Aerosols and Clouds (OPAC) model; Sinha et al. 2013a) and transported dust in winter, although only occasionally over Hyderabad (Sinha et al. 2012), could likely moderate the covariance of PM and AOT (Figure 4(*a*)). The PM_2–AOT relationship exhibits greater scatter in the pre-monsoon and monsoon seasons due to the increased presence of elevated dust plumes transported from arid regions in northwestern India and Arabia (Badarinath et al., 2007b; Sinha et al. 2012), despite high F_{BC} (Figure 3(*b*)). Furthermore, Sinha et al. (2013b) revealed the presence of multiple aerosol plumes at 2–3.5 km during the monsoon, which further lowers the correlation between PM_2 and AOT (Figure 4(*c*)), as these do not seem to significantly affect near-surface aerosols. The aerosol extinction coefficient profiles from MPL over Hyderabad (Sinha et al. 2013b), showing the confinement of aerosols within the lower boundary layer during the post-monsoon and increased aerosol loading aloft during the pre-monsoon and monsoon, are also reflected in the PM_2–AOT correlation (Figure 4(*b*) and (*c*)). However, even in the monsoon, elevated aerosol plumes (2–3.5 km) – mostly comprising biomass-burning smoke and anthropogenic pollution from northern and northwestern India (Kaskaoutis et al. 2014) – are transported over Hyderabad on certain occasions (Badrinath, Kharol, and Sharma 2009), thus lowering the relationship between near-surface and columnar aerosols. Sinha et al. (2013a) showed the enhanced presence of fine-mode aerosols, either recently emitted locally or transported from northern India, which may have substantially contributed to PM_2 at the surface during the post-monsoon over Hyderabad. This, along with shallower mixing layer height, stable atmospheric conditions causing accumulation of aerosols near the surface, and high F_{BC} (Figure 3(*b*)), results in an excellent correlation between particulate matter and AOT during the post-monsoon season (Figure 4(*d*)). Previous studies (Liu et al. 2005; Gupta and Christopher 2009a) have shown that the inclusion of local meteorology in the regression analysis can improve surface particulate matter estimation, since in several cases variations in surface aerosol loading do not correspond to columnar ones and vice versa. In this regard, three representative days associated with negative correlations between surface PM_2 and columnar AOT are examined in Section 4.3.1, in order to further understand such anti-correlations.

4.3.1. Case studies of PM_2–AOT relations

As discussed above, the diurnal variation among near-surface PM_2 mass concentration, F_{BC} (see Figure 3(*b*)), and columnar AOT may be significant and even follows a reverse relationship, thus rendering PM_2 monitoring from space a real challenge, and also it limits monitoring of PM_2 from space in such cases. This reverse relationship is mainly due to the differences in local emission rates, boundary-layer dynamics, and the transport of aerosol plumes aloft. In order to determine the specific atmospheric and meteorological conditions associated with inverse diurnal AOT–PM_2 relationships, three individual cases are analysed. In all three cases, negative correlations between PM_2, M_{T}, and MTS-AOT_{550} are revealed, suggesting reverse diurnal variation of near-surface and columnar aerosols within an urban environment. Figure 5(*a–f*) shows the scatter plot between PM_2 and MTS-AOT_{550} observations obtained on an hourly basis during the same day (left panel) and the mean profiles of the normalized aerosol extinction coefficient (β; km^{-1}), θ (K), RH(%), WS (m s^{-1}), and WD (°) (right panel). The radiosonde profiles correspond to early morning (5:30 LT) observations, while the extinction coefficient corresponds to night-time MPL profiles. Sinha et al. (2013b) showed that MPL-derived columnar AOT is satisfactorily correlated with daytime MTS-AOTs over Hyderabad, indicating stable atmospheric conditions during the daytime. Furthermore, during the study no significant changes in atmospheric and/or meteorological conditions (i.e. sudden dust movement over the site or the arrival of a cold/warm front) were noted that might have affected the

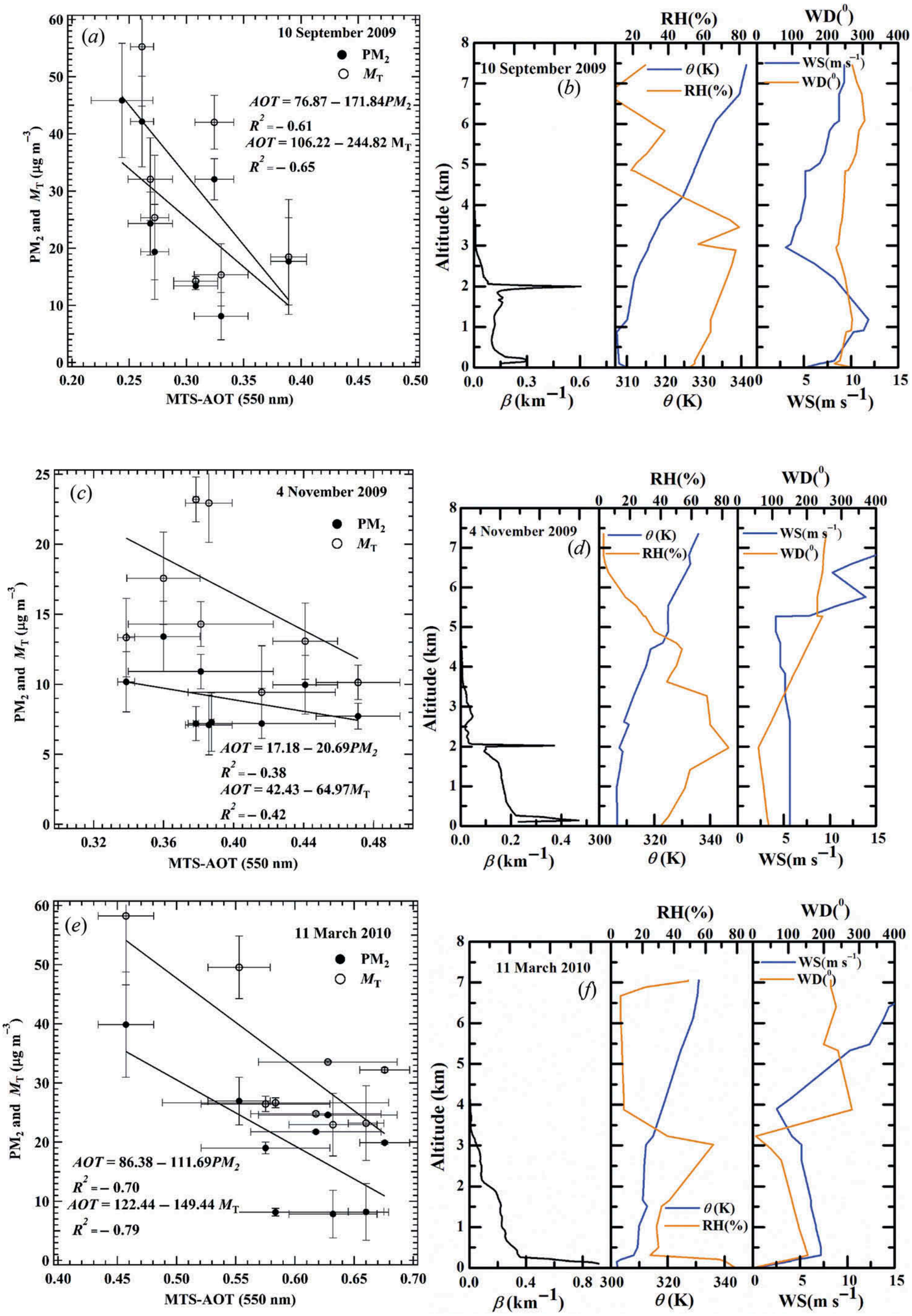

Figure 5. Scatter plot between PM_2, M_T, and Microtops Sunphotometer AOT_{550} (left panel) and vertical profiles of extinction coefficient along with meteorological parameters (right panel) for specific days over Hyderabad.

vertical distribution of aerosol and meteorological profiles in regard to day–night transition, and therefore the night-time aerosol profile is expected to represent the daily pattern.

10 September 2009

The observed negative correlations between PM_2 and MTS-AOT_{550} (R^2 = –0.61) and between M_T and MTS-AOT_{550} (R^2 = –0.65) (Figure 5(*a*)) on this particular day can be explained by the elevated aerosol layers, which contribute to columnar AOT but not significantly to PM_2 concentration at the surface. The aerosol extinction coefficient profile on this day (Figure 5(*b*)) reveals the presence of an elevated aerosol layer at around 2 km with a peak extinction coefficient of about 0.6, which is almost double that near the surface (about 0.3). The θ (K) profile shows a well-mixed layer up to 1.5 km and which is associated with high WS (>10 m s^{-1}). September corresponds to the end of the Indian summer monsoon. During this month, convective eddies are weaker and the vertical transport of aerosols take place only by vertical shear of horizontal wind, supported by strong winds >12 m s^{-1} at 1 km. A large increase in θ (K) at altitudes above 1.5 km suggests a convectively stable sub-adiabatic atmospheric condition. During the monsoon, the air circulation pattern over Hyderabad is mostly driven by the strong southwesterly winds while the upper levels of the boundary layer are associated with strong turbulent mixing and, therefore, lofting of aerosols (Sinha et al. 2013b). Analysis of aerosol optical properties, in conjunction with air mass back-trajectories, indicated that the elevated aerosol layers contain a significant fraction of coarse-mode particles with a mixture of dust and marine aerosols (Sinha et al. 2013b). Furthermore, boundary-layer dynamics affecting mostly surface PM_2 concentrations result in uplift of aerosols (lower PM_2 values) during the hot mid-day hours.

4 November 2009

The aerosol extinction coefficient profile on 4 November 2009 (Figure 5(*d*)) shows a high aerosol concentration within the first 200 m, and also a well-mixed aerosol layer of high extinction (0.2 km^{-1}) up to 2 km. During this period, extensive agricultural biomass burning occurred in northwestern India, transporting smoke aerosols on northerly winds over Hyderabad (Badrinath, Kharol, and Sharma 2009; Kaskaoutis et al. 2014). The vertical profile of θ (K) from the ground to around 2.0 km on that day suggests a convectively stable and well-mixed atmospheric layer, while the large increase in θ (K) above 2 km suggests convectively stable sub-adiabatic atmospheric conditions. This inversion in θ (K) profile corresponds to progressive change in WD from northeast to south-southwest at about 2.5 km, indicating the advection of different aerosol masses that may have affected the diurnal evolution of columnar aerosol properties. The vertical RH profile shows a steady increase from the surface up to around 2.5 km, and a progressive decrease aloft. Despite the shallower boundary layer on this day, the elevated aerosol layers from various sources can substantially change the diurnal evolution between MTS-AOT_{550} and PM_2, leading to a negative correlation of R^2 = –0.38 and –0.42 between MTS-AOT_{550} and PM_2, and M_T and MTS-AOT_{550}, respectively (see Figure 5(*c*)).

11 March 2010

In this case, we observed a strong negative correlation (R^2 = –0.70 and –0.79), respectively, between PM_2 and MTS-AOT_{550} and M_T and MTS-AOT_{550} (Figure 5(*e*)), due to reverse diurnal variation in near-surface and columnar aerosols. On this day, a very high aerosol extinction coefficient was observed near the surface, which decreased drastically up to 0.5 km (Figure 5(*f*)). Due to high surface insolation and enhanced convection, aerosols in the pre-monsoon can rise up to about 2–4 km over Hyderabad, contributing to much greater extinction in the middle troposphere (Sinha et al. 2013b). During the hot pre-monsoon season, excess aerosols may be associated with an increase in secondary aerosol formation and transformation processes facilitated by photochemical production, due to both high solar insolation and the diurnal cycling of substances between the gas and particle phases (Sinha et al. 2013a). The mixing of fine-mode aerosols from anthropogenic emissions and forest fires, with transported dust plumes, is the basic atmospheric feature over Hyderabad during the pre-monsoon season. The vertical profile of θ (K) shows an unstable atmosphere from 1.0 to around 3.5 km. Marked changes in RH, WS, and WD are also observed, which strongly modify the vertical mixing of aerosols. The vertical wind shear and convective instability conditions near the surface are stronger during the pre-monsoon season, leading to increased vertical mixing (Sinha et al. 2013b). RH ranges from about 30 to 80% below 1 km and decreases from about 65 to 8% above 3 km, indicating the presence of dry air at this atmospheric level and convective outflow of boundary-layer air masses. During the pre-monsoon, PM_2 is mostly composed of fine-mode anthropogenic aerosols due to urban emissions, while the columnar AOT contains a significant coarse component due to the predominance of dusty air masses aloft (Kaskaoutis et al. 2009; Sinha et al. 2012). Thus, the correlation between PM_2 and AOT may be weak or even negative on certain days during the pre-monsoon when air masses of different origin co-exist in the vertical.

In summary, the particulate matter–AOT correlation depends on several factors, such as 1) diurnal variation in the boundary-layer height that affects surface aerosol concentrations; 2) changes in vertical mixing caused by instability of the atmosphere; 3) wind shear; 4) variations in the optical properties, chemical composition, and size distribution of near-surface and columnar aerosols; and 5) modification of vertical aerosol profiles and the presence of intense aerosol layers aloft.

4.4. Assessment of surface particulate matter from columnar AOT

The estimation of surface PM_2 mass concentration from AOT measurements is performed using an empirical relationship based on the linear regression between PM_2 and AOT (MTS and MODIS) as:

$$PM_2 = m \times (\mathrm{AOT}) + c, \tag{1}$$

where m and c represent the slope and intercept of the linear regression, respectively.

Figure 6(*a–d*) summarizes the results of the measured and estimated PM_2 mass concentrations over Hyderabad on a seasonal basis. Equation 1 was used as a seasonal basis for PM_2 estimates due to different correlations between PM_2 and AOT (Figure 4), leading to different slope and intercept values for both MTS- and MODIS-AOT correlations. Table 1 summarizes the various statistical parameters obtained from the comparison between measured and estimated PM_2 for each season. The measured PM_2 corresponds to the hour-averaged concentrations within ±30 minutes of the satellite overpass, while MTS-AOTs correspond to the same time interval. The results show that the measured PM_2 values are in general agreement with those estimated using MTS- and MODIS-AOT, with differences up to ±10–20% in the vast majority of cases. However, large deviations (up to 100%) from the measured PM_2 are seen in some cases, especially during the pre-monsoon and monsoon seasons, when the widest variability in PM_2 concentrations is

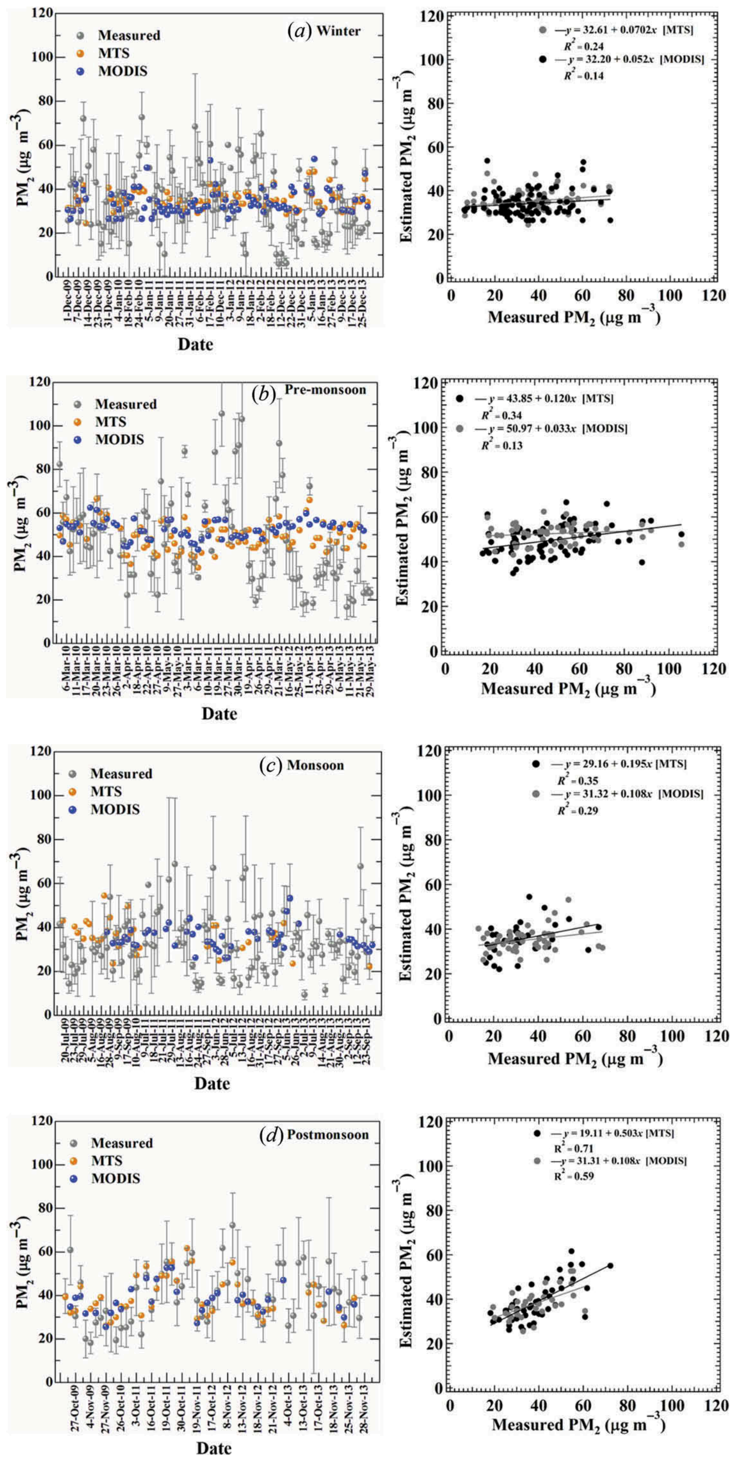

Figure 6. Time series of co-located hourly average of measured and estimated PM_2 from MTS- and MODIS-AOTs within ±30 minutes of satellite overpass. The Microtops Sunphotometer AOT_{550} (MTS) is co-located with PM_2 with respect to Terra-MODIS overpass. The vertical bars correspond to ±1σ standard deviation from the daily mean PM_2 concentration. The right-hand panel shows the scatter plot of measured and estimated PM_2 over Hyderabad for July 2009–December 2013.

Table 1. Statistical parameters obtained from the comparison between measured and estimated PM_2 for each season.

	MTS-AOT (PM_2 measured – PM_2 estimated)						MODIS-AOT (PM_2 measured – PM_2 estimated)					
	Mean (μg m^{-3})	Range (μg m^{-3})	RMSD (μg m^{-3})	NMB	MFB	MFE	Mean (μg m^{-3})	Range (μg m^{-3})	RMSD (μg m^{-3})	NMB	MFB	MFE
Winter	−1.40	−3.89 to 34.62	11.58	−0.024	−0.11	0.36	0.62	−37.20 to 46.25	14.46	−0.021	−0.06	0.37
Pre-monsoon	−1.51	−42.31 to 53.54	14.96	−0.02	−0.11	0.32	−3.10	−55.33 to 57.92	16.98	−0.01	−0.10	0.33
Monsoon	−2.81	−21.53 to 31.72	12.32	−0.03	−0.18	0.33	−0.71	−27.03 to 37.12	12.93	−0.03	−0.12	0.30
Post-monsoon	−0.03	−15.63 to 28.82	8.68	−0.0005	−0.03	0.18	0.003	−17.21 to 26.07	8.92	−0.00004	−0.03	0.20

RMSD = root mean square difference; NMB = normalized mean bias; MFB = mean fraction bias; MFE = mean fraction error.

observed. The differences between measured and estimated PM_2 concentrations were found to be low, ranging from −3.10 to 0.62 μg m^{-3} (Table 1). In several cases, the PM_2 estimates deviate significantly from the measured values, whether using MTS- or MODIS-AOT (Table 1). For all months and seasons, the differences are greater for MODIS-AOT usage, thus indicating more uncertainty in PM_2 monitoring from MODIS. The negative normalized mean bias (NMB) and mean fraction bias (MFB) values (see Table 1) indicate an underestimation of the PM_2 estimates, with either MTS- or MODIS-AOT, for all seasons over Hyderabad. The relatively lower values of root mean square difference (RMSD) (8.68 and 8.92 μg m^{-3}) and other statistical parameters, such as NMB (−0.0005 and 0.00004), MFB (−0.03 and −0.03), and mean fraction error (MFE) (0.18 and 0.20) for MTS- and MODIS-AOT, respectively, for the post-monsoon season suggest better accuracy in PM_2 monitoring during this season from MTS- and MODIS-AOTs. The PM_2 *vs*. AOT scatter plot is associated with higher values of correlation ($R^2 = 0.71$ (MTS) and 0.59 (MODIS)) (see Figure 6(*d*); right panels) in this season, while large scatter is observed for the other seasons (Figure 6; right panels).

The estimation of PM_2 from AOT measurements relies on the assumption that these parameters are linearly related. However, the AOT-PM_2 relationships showed large scatter, especially for extreme (low and high) values of both PM_2 and AOT (Figure 4). Thus, on certain days with large aerosol concentrations near the ground – due to either higher emission rates or increased AOT due to transported aerosol plumes aloft – changes in near-surface aerosol do not correspond to respective changes in columnar, and vice versa. On these days, the PM_2 estimates vary significantly from the measured ones (Figure 6). This is especially true around urban locations with large heterogeneities in aerosol emissions and land-use/land-cover changes (Kharol et al. 2011; Gupta et al. 2013), rendering space-estimated PM mapping a real challenge. However, such an attempt was made for the first time around Hyderabad city and in the next section we provide the spatial distribution of estimated PM_2 around GHR during the period 2001–2010.

4.5. Spatial distribution of particulate matter

The relationships between PM_2 and AOT analysed above are based on ground measurements over a point location at Hyderabad, while several previous studies (Wang and Christopher 2003; Gupta and Christopher 2009a; Gupta et al. 2013) have shown variability in AOT–PM relationship as a function of time and space around urban locations. Due to the absence of a dense, ground-based network around the study area, it is impossible to achieve the spatial distribution of PM_2 based only on ground measurements, and therefore we utilized satellite data to provide spatial PM_2 distribution for the period 2001–2010, assuming that the AOT–PM_2 relationship (Equation 1) is stable in space and time (season) for all pixels around GHR. However, such conditions are unlikely around urban centres with significant heterogeneity in their anthropogenic emission sources (industry, power plants, heavy traffic, etc.). The assumption of homogeneity between columnar AOT and surface PM_2 over the entire area probably biases the results due to spatial and vertical aerosol heterogeneity around the urban area. Thus, the uncertainty in performing these estimations is maximized, particularly during times of greater spatio-temporal and vertical heterogeneity in aerosol distribution, i.e. during the pre-monsoon and monsoon when the PM_2–AOT relationship is biased significantly (Figure 4(*a*–*d*)). Furthermore, the PM_2–AOT correlation over TIFR was performed for the period July 2009–December 2013, and this may be different when applied to other years or more extended periods. Within these limitations, a first attempt has been made to carry out PM_2 mapping around GHR on a seasonal and annual basis for the period 2001–2010.

Figure 7(*a*, *b*) shows the seasonal and annual mean variation in spatial distribution PM_2 around GHR during the period 2001–2010 estimated using MODIS-AOT retrievals and seasonal PM_2–AOT relationships. The results show that PM_2 mass concentration around GHR ranges from around 10 to 30 μg m^{-3}, which is close to the mean measured values at Hyderabad (see Figure 6). MODIS-estimated mean PM_2 mass concentration over GHR shows a marked seasonal variability, with higher values in the pre-monsoon (16.8 ± 1.9 μg m^{-3}; seasonal mean ±1σ) and monsoon (15.8 ± 1.7 μg m^{-3}), followed by winter (12.4 ± 1.3 μg m^{-3}) and post-monsoon (10.9 ± 1.0 μg m^{-3}) (Figure 7(*a*)). A significant spatial and temporal heterogeneity in PM_2 values for an urban environment is revealed, even for the same season and between the seasons of different years. Higher PM_2 concentrations also associated with increased spatial heterogeneity are observed in the monsoon (mainly) and pre-monsoon seasons, while more homogeneous atmospheric conditions with lower PM_2 concentrations prevail in the post-monsoon and winter seasons. It should be noted that intra-annual variation (Figure 7(*b*)), especially in the monsoon, maybe also be affected by the changing monsoonal circulation, rainfall frequency, and intensity that affect aerosol concentrations over India (Padma Kumari et al. 2013; Kaskaoutis et al. 2012). Furthermore, a tendency of increasing PM_2 concentration is found for more recent years (2007 onwards) (Figure 7(*b*)), which is consistent with the increase in estimated $PM_{2.5}$ (>15 μg m^{-3}> found using Multi-angle Imaging Spectroradiometer (MISR) AOT between March 2000 and February 2010 over selected hotspot regions in India (Dey et al. 2012). This slightly increasing

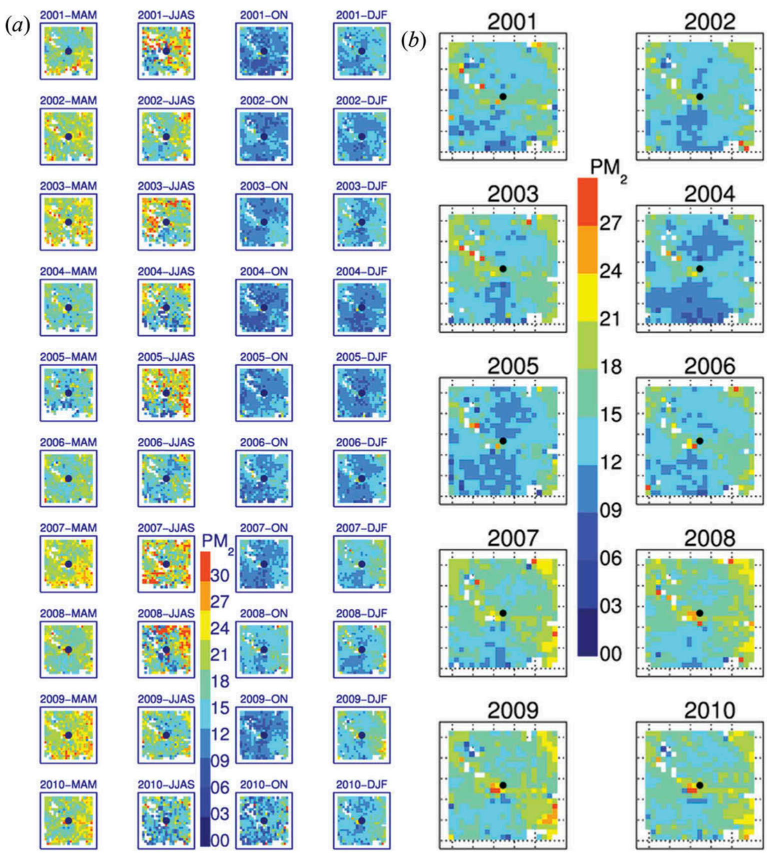

Figure 7. Seasonal (a) and annual mean (b) spatial distribution of MODIS-PM_2 around Greater Hyderabad region (GHR) for 2001–2010. The MODIS-PM_2 maps were generated using the relationship between Terra-MODIS-AOT and measured PM_2. For PM_2 estimation, different slope and intercept values were used for each year (as shown in Figure 4) using linear regression (Eq. 1).

trend in PM_2 concentrations over central India agrees with the aerosol build-up (3.63% over the last decade) reported by Moorthy et al. (2013), mostly attributed to increase in industrial activity, high-capacity coal-powered plants, and numbers of vehicles. However, the trend in regard to estimated PM_2 concentrations around GHR is difficult to quantify due to significant uncertainties in the estimations and gaps in the data set. Apart from the higher PM_2 concentrations in the northeastern area of the city (as explained above), the urban environment shows a tendency for higher PM_2 mass concentration as a result of increased anthropogenic activities, traffic, and industrial activity.

5. Conclusions

This study analysed columnar aerosol loading as measured using ground-based and space-borne instruments, as well as concentrations of surface particulate matter of dynamic diameter ≤2 μm (PM_2), over an urban location (Hyderabad, India), aiming to achieve the first-ever satellite-based PM_2 mapping over this region based on the relationship between PM_2 and AOT. This study also emphasizes the limitations of satellite monitoring of PM_2 from space. Satellite retrievals of Terra-MODIS-AOT (MODIS-AOT) were compared to Microtops-II Sunphotometer (MTS) AOT (MTS-AOT), and then correlated with measured PM_2 on a seasonal basis to estimate PM_2 concentrations. The key findings are now summarized.

1. The satellite observations were co-located with the MTS-AOTs at time intervals of ±30 minutes of the satellite overpass, and their comparison showed a satisfactory correlation with a coefficient of 0.81, indicating the potential capability of satellite-based aerosol monitoring to reproduce aerosol distribution particularly over regions where ground-based measurements are sparse.
2. In general, higher surface aerosol mass concentrations were associated with higher columnar AOT. The seasonal mean diurnal variation in MTS-AOT and PM_2 revealed a general consistency, although on certain days a negative correlation was found. This was attributed to distinct elevated aerosol layers of different origin and characteristics, such as dust plumes and/or long-range transported biomass-burning aerosols, as well as to the daily evolution of the mixing-layer height that controls the dilution processes and ground-level PM_2 concentrations.
3. The vertical profiles of the extinction coefficient obtained from micro-pulse lidar (MPL) measurements revealed the presence of distinct aerosol layers aloft associated with different sources of air masses in the vertical, which is also well supported by thermodynamic parameters. Thus, it is concluded that on certain occasions the correlation between surface particulate matter measurements and columnar AOT will be poor and the estimation of particulate matter concentrations from MODIS highly uncertain.
4. The differences between measured and estimated PM_2 mass concentrations, on average, were found to be very low (−1.40 to −0.03 μg m^{-3} for MTS-AOT and −3.10 to 0.62 μg m^{-3} for MODIS-AOT) for the four seasons considered in this study. However, in specific cases these differences were found to be very high, ranging from −42.3 to 53.5 μg m^{-3} in the pre-monsoon season, when the highest biases were observed. The RMSD between measured and estimated PM_2 – using either MTS- or MODIS-AOT – was found to be about 8–16% on a seasonal basis.
5. A significant seasonal, inter-annual spatial heterogeneity and an increasing tendency in estimated PM_2 concentration were revealed around GHR for the period

2001–2010. The urban environment showed a tendency for higher AOT and PM_2 mass concentrations compared with the surroundings, resulting from increased anthropogenic activities, traffic density, and industrial activity.

The uncertainties in satellite retrieval and the significant temporal and spatial variability of aerosols around an urban environment revealed that the estimation of particulate matter from space-borne instruments constitutes a real challenge in pollution-related studies. This is a first satellite-based attempt to map the spatial variability of PM_2 around GHR. Increased availability of ground-based measurements will offer the possibility of more detailed and comprehensive analysis to explore this technique over other Indian mega-cities in the future.

Key points

- Spatio-temporal distribution of particulate matter and AOT.
- Significant spatial heterogeneity and increasing trend in regard to particulate matter.
- Estimation of particulate matter from space is highly uncertain due to elevated aerosol layers.

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Disclosure statement

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