Aerosol opto-physical properties: Temporal variation, aerosol type
discrimination and source identification

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Abstract

Atmospheric aerosol characterization experiments were conducted, for the first time, over AC-
Ahmednagar, a semi-urban location in south-west India, using a multi-spectral solar radiometer
from January 2016 to May 2018. The MODIS/Terra retrieved Level-2 daily swath AOD$_{550\ nm}$
data sets during 2011-2021 were also used to infer long-term behaviour of columnar aerosols at
AC-Ahmednagar. The daily-averaged, Microtops II Sun photometer measured AOD$_{500\ nm}$ and
AOD$_{1020\ nm}$ and MODIS retrieved AOD$_{550\ nm}$ reveal a discernible wide spectrum of variability in
their magnitudes. Magnitude-wise, AODs at both the wavelengths depict an increasing trend of
about 0.05 year$^{-1}$ (AOD$_{500\ nm}$) and 0.03 year$^{-1}$ (AOD$_{1020\ nm}$). However, MODIS AOD$_{550\ nm}$
illustrates statistically insignificant increasing trend of about 0.007 year$^{-1}$. Microtops II Sun
photometer derived Angstrom exponent (AE) depicts a noticeable day-to-day variability with
respect to its overall mean value (1.06 ± 0.30) indicating the presence of diverse-sized aerosols.
The AOD$_{500\ nm}$ against AE$_{440-870\ nm}$ contour density map analysis qualitatively characterizes
different aerosol types prevalent in the atmospheric column. The weighted PSCF (WPSCF)
analysis for AOD performed on a seasonal basis reveals the presence of possible contribution of
long-range transported plumes of varied aerosols on different scales and types from a variety of
emission sources from different regions.

**Keywords:** Aerosol optical depth, Ångström exponent, MODIS, fine-mode aerosols, coarse-
mode aerosols, coagulation.

1 INTRODUCTION

The tiny particles (at a size of $10^{-2}$ µm – $10^2$ µm), which are present in the suspended form in the
atmospheric column, collectively termed as the aerosols, play a crucial role in the functioning of
the Earth–atmosphere system as a whole (Ng et al., 2017). However, a substantial number of
uncertainties prevail regarding their accurate and quantifiable determination techniques within this
system. These uncertainties, in general, are known to be related to the comprehensive groups of
aerosol sources and consequent sinks, aerosol types, and aerosol transformational mechanisms.
The aerosol type is governed by its generation source area which is found to be crucial in deciding
its singular and integrated physical characteristics. For example, soot (black carbon) aerosols
formed through natural as well as anthropogenic combustion processes are mostly submicron-
sized with low valued single-scattering albedo and thereby high absorptivity in the optical region
of the electromagnetic spectrum. Nearer to the source regions, these aerosols are fine-mode
particles (size $< 1$µm) with high columnar concentrations. The aerosol ‘sources and sinks’ exhibit
wide variability and distribution over the globe so that their physical, optical, radiative, and
chemical properties display diverse variations with different geographic localities. Aerosols
generated through a variety of natural and anthropogenic activities get mixed and as a
consequence of this mixing process, each aerosol particle becomes a composite of dissimilar
chemical constituents determining its complex refractive index. Globally, the natural aerosol
sources are more important than the anthropogenic sources; however, on a regional scale, the anthropogenically produced aerosols are more important (Ramanathan et al., 2001). Aerosols formed at one location may get advected to long distances by the prevalent wind systems via different pathways at different atmospheric levels above ground and subsequently producing effects at remote locations away from the aerosol generation source (Pawar et al., 2015; Kolhe et al., 2018).

The AOD is known to be the main physical parameter linked to the columnar integrated optical activity of aerosols. This is often retrieved through the measurements of extinction of sunlight at different wavelengths from the ground-based observations of direct beam solar irradiance (Pawar et al., 2012; Aher et al., 2014; Varpe et al., 2018) and also from the space-based instruments through the modeling of scattered radiation (Kahn et al., 2005; Sayer et al., 2012; Toledano et al., 2012; Levy et al., 2013; Li et al., 2014). Also, AOD is the most comprehensive variable to determine the atmospheric aerosol loading and radiative forcing retrievals. It is defined as the extinction of direct solar irradiance traversing through the atmosphere via scattering (angular redistribution of energy) and absorption (conversion of energy into either heat or photochemical change) phenomena due to atmospheric aerosols. AOD observation at different wavelengths is a valuable tool to infer aerosol size distribution as well as its spatio-temporal variation. Over the last few decades, a steady increase in scientific interest in the assessment of the aerosol opto-physical, chemical, and radiative properties has identified pivotal role of the atmospheric aerosols in the determination/modification of the Earth–atmosphere radiation balance and hydrological cycle through their direct and indirect effects (Augustine et al., 2008; Varpe et al., 2018). Mostly, the global warming phenomena have been attributed to enhanced greenhouse gas (GHG) emissions (Solomon et al., 2009). At the same time, recent studies reveal that
anthropogenic aerosols influence climate through variations in the global radiation budget (Ramanathan et al., 2001; Bellouin et al., 2005; Sanap and Pandithurai, 2015). Albeit, there are effects on Earth–atmosphere energy budget, aerosols also affect the climate by modulating the hydrological cycle (Kvalevåg et al., 2013; Allen et al., 2014).

The present study investigates the prevalent column-integrated aerosol properties using AOD retrieved from the ground-based Microtops II Sun photometer measurements for the first time from a semi-urban observational site, AC-Ahmednagar, situated in Maharashtra state, India. The Microtops II Sun photometer retrieved AOD are also supplemented with MODIS/Terra derived AOD$_{550}$ nm (MOD04_3K_C6.1 (collection 6.1)) for the period January 2011–December 2021. The main objectives of the present investigation are: (i) The temporal (daily and seasonal) of the Microtops II Sun photometer derived AOD and the MODIS/Terra retrieved AOD$_{550}$ nm to delineate aerosol properties and to know overall columnar content of atmospheric aerosols at the site AC-Ahmednagar (ii) To study the spectral variability of Microtops II Sun photometer derived AOD, hence to estimate the Angstrom wavelength exponent ($\alpha$), a particle size indicator, to judge the presence of different sized aerosols at the observational site. (iii) Seasonal discrimination of dominant aerosol-types based on the constructed contour density map by the 2-dimensional binning of AOD$_{500}$ nm versus AE$_{440-870}$ nm (iv) Comparison/evaluation of MODIS/Terra algorithm retrieved AOD$_{550}$ nm (MOD04_3K_C6.1 (collection 6.1)) against Microtops II Sun photometer measured AOD.

2 SITE AND METEOROLOGY

Topographically, the study location, AC-Ahmednagar (19.09°N, 74.74°E, 649 m AMSL) is situated at about 120 km towards north-east of Pune at the confluence of three state and one
national highways (about 250 m from the sampling site) making it susceptible to the emissions from heavy traffic/vehicular density. Geographically, Ahmednagar is the largest district of Maharashtra and about 65% of the total population derives their livelihood through agricultural activity. Based on this, the post-harvesting activity of sugarcane waste burning is predominant in post-monsoon and winter months during the year. At AC-Ahmednagar, the meteorological parameters like temperature (T) and relative humidity (RH) are measured on an hourly basis by employing a weather monitor (Envirotech, Model WM 271) during the period January 2016 – May 2018. Fig. 1 displays a typical seasonal diurnal variation pattern of T with maxima and minima observed, respectively, during noon to late afternoon and early morning at AC-Ahmednagar. The diurnal variability observed in RH, however, depicts inverse correlation with diurnal variation in T. Seasonally, at AC-Ahmednagar (Fig. 1), mean values of temperature (T\text{Max}/T\text{Min}) were seen to be 38.1 ± 2.5/23.4 ± 3.0°C (pre-monsoon), 31.7 ± 1.5/17.9 ± 3.1°C (post-monsoon), 30.8 ± 2.9/23.2 ± 1.3°C (monsoon) and 31.8 ± 2.2/15.9 ± 2.2°C (winter) while RH varied from 14.5 ± 5.5 % (pre-monsoon) to 86.9 ± 7.4 % (monsoon). As is revealed from the current meteorological data and the long term (1982–2012) data, the climate of Ahmednagar appears to be local steppe in nature with an average annual temperature of about 26.6°C depicting a variation of 10.4°C throughout the year (Kolhe et al., 2018, 2019; http://en.climate-data.org/asia/india/maharashtra/ahmednagar-2808/). At Ahmednagar, the total rainfall during the year is about 561 mm with the maximum rainfall occurring in September (152 mm).

3 DATA, INSTRUMENTATION, AND ANALYSIS TECHNIQUE

AOD measurements at five discrete wavelengths in the spectral region, covering from visible to near IR range were carried out from morning till evening at the time resolution of 30 minutes on 137 clear sky cloudless days during 2016–2018 from the observational site AC-Ahmednagar.
Due to operational constraints on Microtops II Sun photometer, it was operated only on clear sky days, particularly during winter and pre-monsoon seasons. The Microtops II Sun photometer measures the intensity of the direct solar irradiance at 440, 500, 675, 870, and 1020 nm wavelengths having a full width at half maxima (FWHM) band pass of ~10 nm. The measured direct solar flux values at these five narrow band wavelengths were used to estimate the total optical depth of the atmosphere (TOD) using Lambert-Beer-Bouger law and the corresponding filter calibration constant. The aerosol optical depth (AOD) was then calculated by correcting TOD for the wavelength-dependent modeled optical depths due to molecular scattering (i.e. the Rayleigh scattering) and gaseous absorption (Devara et al., 1996; Aher and Agashe, 1996, 1998; Aher et al., 2014; Pawar et al., 2012; Kolhe et al., 2016). Since AOD is a small difference between TOD and the optical depth due to Rayleigh scattering, its’ accuracy is found to be sensitive to small uncertainties in the calibration procedure followed rather than the selected algorithms for the modeled components. The prime source of uncertainty in the sun photometric AOD measurements is the use of inaccurately determined calibration constant which is the wavelength-dependent extraterrestrial solar flux (V₀(λ), a sun photometer detector output often retrieved through Langley plot technique (Schmid and Wehrli, 1995; Kazadzis et al., 2018). The relative uncertainty in the calibration constant of the Microtops II Sun photometer is found to be < ± 0.5% for measurements at different wavelengths in the UV-A to near-IR wavelength range for air mass one (for overhead sun). This yields of ~ 0.005–0.03 error in the optical depth. The overall uncertainty in error in AOD measurements is ± 0.03. Generally, such conventional sun/sky-photometers/radiometers are regularly calibrated once/twice in a year using in-situ observations under full clear-sky conditions at high-altitude sites similar to Mauna Loa (Ningombam et al., 2014; Dumka et al., 2020). In the present work, before its deployment in the
field for AOD measurements at site AC-Ahmednagar, the Microtops II sun photometer was calibrated at a high altitude (AMSL ~1500m), pristine site Mt. Sinhgad situated towards the north-west of Pune (Supplementary file).

The retrieval of AOD at each wavelength as outlined above yields spectral variation of AOD from which Ångström parameters viz., Ångström exponent (AE) and Ångström turbidity coefficient ($\beta$) are determined from the Ångström empirical formula (Eq. (1)) by following linear regression technique (Pawar et al., 2012; More et al., 2013; Kolhe et al., 2016).

$$\text{AOD}_\lambda = \beta \lambda^{-AE} \quad (1)$$

Here, $\beta$ is the extinction coefficient corresponding to AOD at 1000 nm and is indicative of aerosol loading in the atmosphere. AE is an approximate measure of the size of aerosols contributing to AOD, defined as the ratio of the columnar number density of fine-to-coarse-mode aerosols which may vary from 4 to 0 or less (i.e. negative values) (Iqbal, 1983). Thus, when the size of aerosol particles is small of the order of air molecules (sub-micron, fine-mode), AE should approach 4 while it should tend to become 0 or even negative for super-micron (coarse-mode) aerosol particles. AODs, thus measured and the estimated Ångström parameters are used to study spectral and temporal characteristics of aerosols at AC-Ahmednagar.

The Microtops II Sun photometer measured AOD data are supplemented with MODIS sensor (onboard Terra satellite) derived Level-2 daily swath AOD data sets at a spatial resolution of 3 km (MOD04_3K_C6.1 (collection 6.1), AOD 550 nm) to know the overall columnar content of atmospheric aerosols over the observing location nearly throughout the year and also over long-term basis (2011-2021). The data is further analysed to compare/evaluate MODIS/Terra algorithm retrieved AOD$_{550\text{ nm}}$ with those measured by Microtops II Sun photometer at a site
AC-Ahmednagar. Differences between the two retrievals may be caused by the different characteristics of the sensors, such as different imaging times, sensor lifetimes, and sensor degradation (Sayer et al., 2014; Gupta et al., 2018; Sharma et al., 2021).

4 RESULTS AND DISCUSSION

4.1 Temporal variation of aerosol optical and microphysical properties

4.1.1 Microtops II Sun photometer

Fig. 2(a, b, c) depicts the variability of the daily-averaged $\text{AOD}_{500 \text{ nm}}$, $\text{AOD}_{1020 \text{ nm}}$, Ångström parameters, AE and $\beta$ (derived based on AOD spectral curve as explained in Section 3) and the standard deviation associated with each of these parameters observed in winter and pre-monsoon seasons during January 2016–May 2018. The daily-averaged $\text{AOD}_{500 \text{ nm}}$ and $\text{AOD}_{1020 \text{ nm}}$ respectively reveal a discernible wide spectrum of variability in their magnitudes in the range 0.13 (February 2017) – 0.80 (December 2017) and 0.06 (Feb 2017) –0.47 (May 2018) over the study period (Fig. 2(a)). Similarly, the Ångström parameter, $\beta$ varies between 0.05 (Feb 2017) and 0.48 (May 2018) for the same period of observation (Fig. 2 (b)). Further, the standard deviations observed in daily $\text{AOD}_{500 \text{ nm}}$, $\text{AOD}_{1020 \text{ nm}}$, AE and $\beta$ values are found to be of the order of 0.002–0.138, 0.001–0.153, 0.006–0.215 and 0.001–0.115 respectively (Fig. 2 (c)). As is seen in Fig. 2, the lines parallel to the abscissa represent the overall parameter mean labeled by the corresponding value for the period under study. The respective mean values for $\text{AOD}_{500 \text{ nm}}$, $\text{AOD}_{1020 \text{ nm}}$ and $\beta$ are found to be $0.42 \pm 0.14$, $0.18 \pm 0.06$ and $0.19 \pm 0.06$. Apart from the average scenario represented by the overall means of these parameters, it is essential to look at the daily-averaged $\text{AOD}_{500 \text{ nm}}$, $\text{AOD}_{1020 \text{ nm}}$, and $\beta$ variability during each observing month/year. To gauge the prevalent temporal variability in the daily-averaged $\text{AOD}_{500 \text{ nm}}$, $\text{AOD}_{1020 \text{ nm}}$, and $\beta$ parameters, the data plotted in Fig. 2 is subjected to descriptive statistical analysis which
encompasses the determination of mean, median, minimum, and maximum in the data points. The resulting statistical metrics are displayed in Table 1 from which it is seen that the magnitudes of mean, median, minimum, and maximum values of AOD$_{500 \text{ nm}}$, AOD$_{1020 \text{ nm}}$, and $\beta$ differ from year to year during the period under study. The observing period-wise mean (median) values for AOD$_{500 \text{ nm}}$ and AOD$_{1020 \text{ nm}}$ can be noticed to be $0.38 \pm 0.12$ (0.38) and $0.17 \pm 0.05$ (0.16) during 2016 (Jan–May), $0.42 \pm 0.11$ (0.40) and $0.18 \pm 0.05$ (0.18) during 2017 (Jan–May and Dec), while during 2018 (Jan–May) the respective values are $0.47 \pm 0.17$ (0.43) and $0.22 \pm 0.08$ (0.20). Magnitude-wise, AODs at both wavelengths depict an increasing trend of about 0.05 year$^{-1}$ (AOD$_{500 \text{ nm}}$) and 0.03 year$^{-1}$ (AOD$_{1020 \text{ nm}}$) during the period of observation despite AOD$_{500 \text{ nm}}$ being almost 2-fold higher as compared to AOD$_{1020 \text{ nm}}$. The difference in AOD magnitudes at 500 and 1020 nm wavelengths can be attributed to the extinction efficiencies of the prevalent aerosol ensembles of different sizes active at each wavelength as per the Mie scattering theory (Quenzel 1970; Varpe et al., 2018). The Ångström turbidity coefficient, $\beta$, more or less follows the same trend of variation as is observed for AOD$_{1020 \text{ nm}}$. Further, the occurrence of minimum/maximum values of AOD$_{500 \text{ nm}}$, AOD$_{1020 \text{ nm}}$, and $\beta$ do not show a specific pattern as sometimes these are seen to occur in the same month/season. Also, the year-to-year maximum-to-minimum ratio for AOD$_{500 \text{ nm}}$, AOD$_{1020 \text{ nm}}$, and $\beta$ is found to be 3.61, 3.82, and 3.95 during 2016 (Jan–May), 6.31, 5.89, and 6.15 during 2017 (Jan–May and Dec) while ratios are 4.26, 4.67 and 4.58 during 2018 (Jan–May) (Fig. 2(a, b); Table 1).

Fig. 2(b) also displays the temporal variability of daily-averaged AE. It is interesting to note that AE depicts prominent day-to-day variability with respect to the line parallel to the abscissa representing AE mean value ($1.06 \pm 0.30$). Superimposed with this mean AE line, there exists a large downward and upward varying tendency in AE magnitudes with Min and Max
values of 0.36 (March 2017) and 1.52 (January 2018) respectively indicating the presence of
diverse sized aerosols prevalent over the observation site AC-Ahmednagar (Kolhe et al., 2016;
Kokkalis et al., 2018; Varpe et al., 2018). The occurrence/presence of the diverse-sized aerosols
over Ahmednagar is further substantiated through the frequency analysis of AE values plotted in
Fig. 2(b) for the observation period during 2016–2018. During 2016 (Jan–May), on 15% of
occasions AE is found to be <1 while it appears to be >1 on 85% occasions indicating the
dominance of sub-micron fine-mode aerosols as against super-micron coarse-mode aerosols in
the atmosphere over Ahmednagar. AE variability scenario over Ahmednagar during 2017 (Jan–
May, Dec) and 2018 (Jan–May) is quite different as compared to that observed during 2016.
During 2017 and 2018, on 42% and 53% occasions AE emerges to be <1 while it is >1 on 58%
and 47% occasions respectively. These observations reveal that there is an upward enhancement
in super-micron coarse-mode aerosols in relation to sub-micron fine-mode aerosols which is also
evident from the rise in AOD during 2017 and 2018 (Table 1).

Several researchers (Xin et al., 2007; Soni et al., 2011; Kolhe et al., 2016; Patel et al.,
2017; Singh et al., 2020; Dahal et al., 2021) have observed large temporal variability in AOD
measurements over other locations (Table 2). From Table 2, it is seen that the magnitudes of
AOD$_{500 \text{ nm}}$ and AE over suburban locations viz. Xianghe and Shenyang (Xin et al., 2007) are
comparable to those observed at the current semi-urban site, AC-Ahmednagar. Although, AOD
$_{500 \text{ nm}}$ at the sites Pune (0.53 ± 0.13, Kolhe et al., 2016), Birtamode (0.68 ± 0.39, Dahal et al.,
2022) and Shanghai (0.64 ± 0.25, Xin et al., 2007) are higher as compared to those observed at
AC-Ahmednagar (0.42 ± 0.14), the AE values (AC-Ahmednagar = 1.06 ± 0.30; Pune = 1.05 ±
0.27; Birtamode = 1.09 ± 0.13; and Shanghai = 1.08 ± 0.24) are somewhat comparable. The
Microtops II Sun photometer measured mean AOD$_{500 \text{ nm}}$, AE$_{340-870 \text{ nm}}$ and $\beta$ are found to be 0.78
± 0.32, 0.78 ± 0.28, and 0.45 ± 0.21, respectively, at a site located in national capital Delhi 
(28.38°N, 77.10°E, 235 m AMSL) during January 2007– December 2008 (Soni et al., 2011). 
Although, the period of observation is different, at AC-Ahmednagar, the observed mean AOD$_{500}$ 
value is smaller while mean AE magnitude is higher as compared to those observed at Delhi 
(Soni et al., 2011) and Gorakhpur (Singh et al., 2020). However, at Desalpar (Patel et al., 2017) 
AE magnitude is observed to be ~1.53 times smaller with nearly same AOD$_{500}$ as compared to 
that observed at AC-Ahmednagar. These observations point towards the presence of probably 
different types of atmospheric aerosols with diverse size distributions and columnar content. The 
higher concentration of local pollutants, mainly contributed by the vehicular emission (~80%), 
industrial effluents (~12%), and household activities (8%) in the city could be the plausible 
reason for the occurrence of high AOD values observed over Delhi (Singh et al., 2010). The 
large variability in both AOD$_{500}$ and AE shows a variety of aerosol types over the region due 
to the advection of air masses of different characteristics as well as origin. The higher AOD$_{500}$ 
and AE values observed during late-autumn and winter season may be attributed to the advection 
of anthropogenic pollution at the study location (Kaskaoutis et al., 2014).

The current AOD measurements and the observed temporal variations in AOD$_{500}$, 
AOD$_{1020}$, AE and β at a semi-urban site, AC-Ahmednagar depict a different characteristic 
feature as compared with the studies mentioned above. Thus, the high day-to-day variability 
noticed in AOD$_{500}$, AOD$_{1020}$ and β as well as AE over AC-Ahmednagar could be assigned 
with seasonal nature of local aerosol sources such as heavy and light vehicular traffic, domestic 
activities with variety of aerosol types and local meteorology (viz. T, RH, WS, WD and ABL 
dynamics) (Kolhe et al., 2018; 2019). In addition to these local sources, the aerosol loading at 
AC-Ahmednagar is also influenced by the influx produced due to the advection of air masses of
diverse origin and characteristics on account of long-range transport (Guha et al., 2015; Pawar et al., 2015).

4.1.2 MODIS

Fig. 3 shows the (a) day-to-day (b) monthly (c) interannual variability and (d) frequency distribution of all-quality (QA = All) AOD$_{550}$ nm daily retrievals (average value of 5×5 pixels (~225 km$^2$)) during the period January 2011 to December 2021 (for 2842 days) over the study location AC-Ahmednagar. Just like Microtops II Sun photometer measured AOD$_{500}$ nm and AOD$_{1020}$ nm, MODIS/Terra retrieved AOD$_{550}$ nm also exhibits noticeable day-to-day/monthly variability to a larger extent as is evidenced by the large separation between minimum and maximum AOD values (Figs. 3(a, b)). From Fig. 3 (b), the monthly averaged lowest AOD$_{550}$ nm value is observed during January 2011 (0.27 ± 0.18) and highest during August 2017 (1.32 ±0.18). Over other years also AOD$_{550}$ nm magnitudes are found to be quite high (> 0.35 or more). Despite large difference amongst maximum-to-minimum AOD$_{550}$ nm values, the overall observation period (eleven year, for 2842 days) mean for the MODIS-TERRA retrieved AOD$_{550}$ nm is found to be 0.49 ± 0.24. These data reveal the occurrence of significantly high AOD over AC-Ahmednagar as compared to those reported at global scale (0.126) and over other continents (North America = 0.98; Africa = 0.168; Asia = 0.182) (Mhawish et al., 2017). These data at site AC-Ahmednagar indicates the dominance of moderate-to-massive aerosol burden. Further, the variability is indicative of aerosol types mainly attributed to variations in particulate source strength over monthly/seasonal scale. The long-term aerosol climatology i.e. the deseasonalized AOD time series for the site AC-Ahmednagar is presented in Fig. 3(c). The figure also depicts an annual rate of change in AOD (slope = 0.007 and Pearson’s Correlation, r (0.51). Again, as is observed in case of Microtops II Sun photometer measured AOD$_{500}$ nm/AOD$_{1020}$ nm, values,
MODIS AOD\textsubscript{550 nm}, illustrates statistically insignificant increasing trend of about 0.007 year\textsuperscript{-1} during the long-term MODIS AOD retrievals over AC-Ahmednagar. This trend is comparable to that observed by Kumar et al., (2018) (0.002 year\textsuperscript{-1}) for the individual sites located at upper (Karachi, Multan, Lahore), central (Delhi, Kanpur, Varanasi, Patna) and lower IGP (Kolkata, Dhaka) in south Asia. Further, the estimated trend (0.007 year\textsuperscript{-1}) is also equal to that reported by Moorthy et al. (2013) at northern Indian stations for the period, 2001-2011, using AOD data measured by employing Multiwavelength spectroradiometer. Also, the present AOD trend appears to be significantly lower than that of 0.01–0.04 year\textsuperscript{-1}, as reported by Dey and Di Girolamo (2011) for the IGP region using AOD obtained from MISR for the period 2000-2010.

The observed day-to-day variability in MODIS/Terra AOD\textsubscript{550 nm} could be further explored through the frequency distribution analysis of the data portrayed in Fig. 3(a) and the obtained results are displayed in Fig. 3(d). Principally, the range of AOD occurrence frequency for each distribution seems to be different during these years. Examination of Fig. 3 reveals that the AOD\textsubscript{550 nm} has the maximum frequency of occurrence in the range 0.3–0.7 with occurrence frequencies lying in the range 18.7% to 43.7%. Statistically, the increasing AOD\textsubscript{550 nm} trend during 2011–2021 could be partly ascribed to the differing frequencies of occurrences during these years. As discussed earlier, day-to-day variability observed in MODIS/Terra retrieved AOD\textsubscript{550 nm} substantiates the Microtops II Sun photometer AOD measurements at AC-Ahmednagar in the sense that the moderate-to-massive aerosol load over AC-Ahmednagar.

4.2 Seasonal variation of AOD
The individual Microtops II Sun photometer measured AOD$_{500 \text{ nm}}$ and MODIS/Terra retrieved AOD$_{550 \text{ nm}}$ plotted in Figs. 2(a) and 3(a) are grouped into seasons (without discriminating them in the year) to construct box-whisker diagrams together with the associated binned data of the daily averaged Microtops II Sun photometer measured AOD$_{500 \text{ nm}}$ (red color) and MODIS AOD$_{550 \text{ nm}}$ (blue color) during Jan 2016–May 2018 over AC-Ahmednagar and the results are shown in Fig. 4. As seen in Fig. 4, the hollow squares in respective box-whiskers correspond to the average value of the sampled data and whiskers show $\pm 1\sigma$ standard deviations. Further, boxes in Fig. 4 denote 25 percentile (lower line), median (notched portion), and 75 percentiles (top line) of the observed AODs. The minimum value of AOD for each season is indicated by the downward hollow triangle while the upward solid triangle represents the maximum AOD value. Further, from Fig. 4, it can be noticed that the mean, median, minimum, maximum, 25%, and 75% percentiles of AOD depict strong seasonality. In Fig. 4, seasonal mean and number of days (N) used in the estimation of a seasonal average AOD are also indicated which differ significantly for Microtops II Sun photometer (Fig. 4 (a)) and MODIS (Fig. 4 (b)) AOD data sets.

At AC-Ahmednagar, the seasonal MODIS/Terra retrieved mean AOD$_{550 \text{ nm}}$ (Fig. 4(b)) is found to be high in monsoon season (0.61 ± 0.20) and low in the winter season (0.46 ± 0.25) while for pre-monsoon (0.48 ± 0.21) and post-monsoon (0.50 ± 0.25) seasons, AOD$_{550 \text{ nm}}$ values significantly differ in magnitude along with other statistical parameters. Whereas, for Microtops II Sun photometer, AOD$_{500 \text{ nm}}$ (Fig. 4(a)) is slightly higher in winter (0.43 ± 0.17) with respect to that observed during pre-monsoon season (0.42 ± 0.13). The AERONET Sun-Sky AOD measurements at observation site Desalpar in western India also indicate the similar seasonal AOD$_{500 \text{ nm}}$ variability wherein AOD$_{500 \text{ nm}}$ is high in monsoon (0.61 ± 0.34) and low in winter (0.31 ± 0.14) with nearly the same AOD$_{500 \text{ nm}}$ during pre-monsoon (0.45 ± 0.30) and post-
monsoon (0.47 ± 0.26) seasons (Patel et al., 2017). The existence of high MODIS/Terra AOD\textsubscript{550 nm} during monsoon season could be assigned to the effect of cloud contamination and relative humidity leading to the hygroscopic growth of atmospheric aerosols giving rise to high AOD during this season (Ramachandran and Cherian, 2008; Chew et al., 2011; Huang et al., 2012; Indira et al., 2013; Chelani, 2015; Varpe et al., 2018). The Microtops II Sun photometer measured AOD\textsubscript{500 nm} during winter season is slightly higher than AOD in pre-monsoon which may be ascribed to the higher surface wind activity which may be responsible for lifting-up of soil-dust aerosols from dry lands and long-range transported dust from the arid regions (Aher et al., 2014).

4.3 Seasonal discrimination of the aerosol types

In the present work, the aerosol type characterization is performed with the help of contour density map analysis for winter and pre-monsoon seasons. The contour density map utilizes the relationship between AOD\textsubscript{500 nm} and AE\textsubscript{440-870 nm} as a consequence of their observed strong wavelength dependence. These 2-D binning patterns of AOD\textsubscript{500 nm} versus AE\textsubscript{440-870 nm} have been employed at several sites for diverse aerosol types (e.g., desert dust, anthropogenic aerosols, biomass smoke, etc.) (Masmoudi et al., 2003; Ogunjobi et al., 2004; Kim et al., 2004; Eck et al., 2010). Moreover, the scheme of the contour density map is dependent on the sensitivity of AOD and AE to different microphysical aerosol properties (Kalapureddy et al., 2009; Kaskaoutis et al., 2011). The AOD\textsubscript{500 nm} versus AE\textsubscript{440-870 nm} contour diagrams qualitatively specify the amount (viz., the columnar content) and dimension (viz., size) of the aerosols. In these contour maps, the rectangular regions represent urban/industrial (UI), mixed type (MT), clean maritime (CM), and desert dust (DD) type aerosols respectively. The boundaries of these regions correspond to the chosen threshold values of AOD\textsubscript{500 nm} and AE\textsubscript{440-870 nm}. 

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At the observing site, AOD$_{500\,\text{nm}}$ values lie in the range 0.1–0.9 and 0.2–0.8 while AE$_{440-870\,\text{nm}}$ fall in the range 0.8 – 1.5 and 0.3 – 1.6 respectively in winter and pre-monsoon seasons. To accommodate these wide-ranging AOD$_{500\,\text{nm}}$ and AE$_{440-870\,\text{nm}}$ magnitudes, the threshold values used by Pace et al. (2006) have been appropriately modified. Accordingly, AOD$_{500\,\text{nm}} < 0.2$ with AE$_{440-870\,\text{nm}} < 1.3$ correspond to CM type aerosols while AOD$_{500\,\text{nm}} > 0.2$ and AE$_{440-870\,\text{nm}} > 1.0$ characterize the UI type aerosols. Further, AOD$_{500\,\text{nm}} > 0.25$ associated with AE$_{440-870\,\text{nm}} < 0.7$ are suggestive of DD type aerosols advected from oceanic regions. Finally, for the remaining gaps, it is problematic to categorize aerosol types, and as such these are treated as MT type by considering various aerosol-mixing mechanisms like coagulation, condensation, humidification, and gas-to-particle conversion.

Fig. 5(a, b) displays the results of this analysis respectively for winter and pre-monsoon seasons during 2016–2018 at the observing site AC-Ahmednagar. For AOD$_{500\,\text{nm}} > 0.3$, the AE$_{440-870\,\text{nm}}$ magnitudes are higher (> 0.8) during winter season (Fig. 5(a)). It can be noticed that the AOD$_{500\,\text{nm}}$ and AE$_{440-870\,\text{nm}}$ pair with boundary values of 0.4–0.6 and 1.1–1.4 occupy a large contour density map region. The AE$_{440-870\,\text{nm}}$ values of these magnitudes are suggestive of fine-mode aerosol size distribution ensembles (Eck et al., 2005). This underlines UI/anthropogenic type aerosol field in the vertical column of the atmosphere over the observing site. This is found to be considerably more than the MT, CM, and DD aerosol types during the winter season. The MT type aerosols show their signature in aerosol columnar load over AC-Ahmednagar even though much less as compared to UI type aerosols. During the pre-monsoon season (Fig. 5(b)), however, the maximum contour density map region as is observed during winter gets transformed into MT type aerosols with AOD$_{500\,\text{nm}}$ and AE$_{440-870\,\text{nm}}$ pair values of 0.3–0.5 and 0.7–1.1 respectively. Additionally, the prominent presence of UI and DD type aerosols is also
seen during this season at AC-Ahmednagar. On the other hand, during the pre-monsoon season, the CM type aerosol contribution appears to be negligible. In aerosol discrimination analysis over Pune, Kolhe et al. (2016) and Vijayakumar et al. (2014) have also observed that the aerosol ensemble consists of UI, CM, DD, and MT aerosol types which significantly differ during winter and pre-monsoon seasons.

4.4 Transport pathways and source apportionment

To identify different sources of emissions and directional pathways of various aerosols present in the air column over AC-Ahmednagar, 5-day air mass trajectories up to 500 m above the ground level are compiled using HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph 2003). Air mass back trajectories provide valuable information about emission sources and their modes of transport to the measurement sites. To effectively define potential aerosol source regions, a well-received statistical analysis, i.e., the potential source contribution function (PSCF) is used (Ding et al. 2017; Ming et al. 2017; Vaishya et al. 2017; Zheng et al. 2017; Tiwari et al., 2018). The PSCF values for the grid cells are calculated by using Eq. (2)

\[ PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \]  

Here, \( m_{ij} \) and \( n_{ij} \) represents the total number of trajectory endpoints which are passing through and falls within the grid \((i, j)\), while \( i \) and \( j \) respectively represent the longitude and latitude. The PSCF's high values reflect the potential contributions to the concentration of pollutants in the receptor site, while the trajectory that passes through these cells represents key transport pathways that lead to high pollution loads at the reception area. To reduce the uncertainty in the
potential source contribution function (PSCF) values for cells with small $n_{ij}$ values, the PSCF values were multiplied by weighting function ($W_{ij}$). The $W_{ij}$ reduces the PSCF values when the total number of the endpoints in a particular cell was less than about three times the average value of the endpoints per each cell. The weighting function $W_{ij}$ is given by:

$$W_{ij} = \begin{cases} 
1.00 & 80 < n_{ij} \\
0.70 & 20 < n_{ij} \leq 80 \\
0.42 & 10 < n_{ij} \leq 20 \\
0.05 & n_{ij} \leq 10
\end{cases}$$

The weighted PSCF (WPSCF) analysis for AOD is performed on a seasonal basis (Fig. 6) to distinguish air mass trajectories and the possible aerosol hot-spot areas during the different seasons. The colored scale represents the contribution of the aerosol emission sources.

During winter, the air masses mostly arrive from north-western and eastern directions as well as from the central Indian region (covering a large part of the Indo Gangetic Plains region). The long-range transported air-mass plumes may be originated from south-west Asia, which, however, are less dusty due to the inactivity of dust in these areas during the winter. During the pre-monsoon season, the maximum contributions are from the western direction thus suggesting a mixture of dust with contaminant and oceanic aerosols. Additionally, long-range transported dust plumes originated from the Middle East, Arabia, and southwest Asia are also favored during this season. The observing site may also be influenced by marine air masses mainly from the Arabian Sea during monsoon season and in a much-limited scale for long-range transported dust plumes from southwest Asia. On the other hand, during post-monsoon season, the aerosol sources over AC-Ahmednagar are significantly influenced by north-western India, where mostly paddy crop residue burning occurs in this season. The WPSCF analysis highlights the different
emission sources for the seasonally changed aerosol types that are accumulated over the AC-
Ahmednagar throughout the year.

5 CONCLUSIONS

The Microtops II Sun photometer has been operated, for the first time, from a semi-urban
location, AC-Ahmednagar, Maharashtra State, to make AOD measurements. The MODIS/Terra
algorithm retrieved AOD$_{550\ nm}$ data during 2011-2021 is also used in conjunction with the
Microtops II measured AODs to investigate the characteristics of atmospheric aerosols over the
observing location. The major findings of the study include:

- The daily-averaged Microtops II measured AOD$_{500\ nm}$, AOD$_{1020\ nm}$ and MODIS/Terra
  retrieved AOD$_{550\ nm}$ reveal a discernible wide spectrum of variability in their magnitudes.
  Magnitude wise, AODs at both wavelengths depict an increasing trend of about 0.05
  year$^{-1}$ (AOD$_{500\ nm}$) and 0.03 year$^{-1}$ (AOD$_{1020\ nm}$) during the period of observation.
  MODIS AOD$_{550\ nm}$, however, illustrates a statistically insignificant increasing trend of
  about 0.007 year$^{-1}$ for the study period 2011-2021.

- Microtops II derived AE depicts conspicuous day-to-day variability with respect to its
  mean value (1.06 ± 0.30) indicating the presence of diverse-sized aerosols prevalent over
  the observation site AC-Ahmednagar.

- During winter UI/anthropogenic type aerosol field in the vertical column of the
  atmosphere predominate over the observing site AC-Ahmednagar. However, in the pre-
monsoon season, the maximum contour density map region observed during winter gets transformed into MT type aerosols in addition to the prominent presence of UI and DD type aerosols.

- The seasonal weighted PSCF (WPSCF) analysis for AOD reveals the presence of possible contribution of long-range transported air masses during winter and pre-monsoon seasons on different scales and types from a variety of emission sources.

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REFERENCES


Draxler, R.R., Rolph, G.D. (2003). HYSPLIT (Hybrid single-particle Lagrangian integrated trajectory) model. NOAA Air Resources Laboratory, Silver Spring


total aerosol load over the Bay of Bengal during winter season. Atmos. Chem. Phys. 11, 7097–7117. Doi:10.5194/acp-11-7097-2011.


Table 1. Statistical metrics of the Microtops II measured optical and microphysical parameters (AOD$_{500 \text{ nm}}$, AOD$_{1020 \text{ nm}}$, AE and β) for the period of measurements 2016–2018 at observing location AC-Ahmednagar.

<table>
<thead>
<tr>
<th>Year</th>
<th>Mean</th>
<th>Standard deviation (±1σ)</th>
<th>Median</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>AOD$_{500 \text{ nm}}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2016(Jan–May)</td>
<td>0.38</td>
<td>0.12</td>
<td>0.38</td>
<td>0.19</td>
<td>0.69</td>
</tr>
<tr>
<td>2017(Jan–May, Dec)</td>
<td>0.41</td>
<td>0.14</td>
<td>0.40</td>
<td>0.13</td>
<td>0.79</td>
</tr>
<tr>
<td>2018(Jan–May)</td>
<td>0.47</td>
<td>0.16</td>
<td>0.43</td>
<td>0.19</td>
<td>0.79</td>
</tr>
<tr>
<td>AOD$_{1020 \text{ nm}}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2016(Jan–May)</td>
<td>0.17</td>
<td>0.05</td>
<td>0.16</td>
<td>0.08</td>
<td>0.30</td>
</tr>
<tr>
<td>2017(Jan–May, Dec)</td>
<td>0.18</td>
<td>0.05</td>
<td>0.18</td>
<td>0.05</td>
<td>0.32</td>
</tr>
<tr>
<td>2018(Jan–May)</td>
<td>0.22</td>
<td>0.07</td>
<td>0.20</td>
<td>0.10</td>
<td>0.47</td>
</tr>
<tr>
<td>AE</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2016(Jan–May)</td>
<td>1.12</td>
<td>0.23</td>
<td>1.16</td>
<td>0.43</td>
<td>1.45</td>
</tr>
<tr>
<td>2017(Jan–May, Dec)</td>
<td>1.06</td>
<td>0.27</td>
<td>1.10</td>
<td>0.36</td>
<td>1.44</td>
</tr>
<tr>
<td>2018(Jan–May)</td>
<td>1.03</td>
<td>0.35</td>
<td>0.94</td>
<td>0.46</td>
<td>1.52</td>
</tr>
<tr>
<td>β</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2016(Jan–May)</td>
<td>0.17</td>
<td>0.06</td>
<td>0.17</td>
<td>0.08</td>
<td>0.32</td>
</tr>
<tr>
<td>2017(Jan–May, Dec)</td>
<td>0.19</td>
<td>0.05</td>
<td>0.19</td>
<td>0.05</td>
<td>0.32</td>
</tr>
<tr>
<td>2018(Jan–May)</td>
<td>0.23</td>
<td>0.08</td>
<td>0.21</td>
<td>0.11</td>
<td>0.48</td>
</tr>
</tbody>
</table>
Table 2. Comparision of AOD$_{500 \text{ nm}}$ and AE observed at AC-Ahmednagar with those observed at other locations

<table>
<thead>
<tr>
<th>Location</th>
<th>Background</th>
<th>Study period</th>
<th>AOD$_{500 \text{ nm}}$</th>
<th>AE</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ahmednagar</td>
<td>Semi-urban</td>
<td>2016–2018</td>
<td>0.42 ± 0.14</td>
<td>1.06 ± 0.30</td>
<td>Present study</td>
</tr>
<tr>
<td>Pune</td>
<td>Urban</td>
<td>2008–2015</td>
<td>0.53 ± 0.13</td>
<td>1.05 ± 0.27</td>
<td>Kolhe et al., 2016</td>
</tr>
<tr>
<td>Delhi</td>
<td>Urban</td>
<td>2007–2008</td>
<td>0.78 ± 0.32</td>
<td>0.78 ± 0.28</td>
<td>Soni et al., 2011</td>
</tr>
<tr>
<td>Gorakhpur</td>
<td>Sub-tropical</td>
<td>2014–2018</td>
<td>0.65 ± 0.27</td>
<td>0.92 ± 0.27</td>
<td>Singh et al., 2020</td>
</tr>
<tr>
<td>Desalpar</td>
<td>Rural</td>
<td>2014–2015</td>
<td>0.43 ± 0.26</td>
<td>0.69 ± 0.39</td>
<td>Patel et al., 2017</td>
</tr>
<tr>
<td>Birtamode</td>
<td>Urban</td>
<td>2018–2019</td>
<td>0.68 ± 0.39</td>
<td>1.09 ± 0.13</td>
<td>Dahal et al., 2022</td>
</tr>
<tr>
<td>Shanghai</td>
<td>Urban</td>
<td>2004–2005</td>
<td>0.64 ± 0.25</td>
<td>1.08 ± 0.24</td>
<td>Xin et al., 2007</td>
</tr>
<tr>
<td>Xianghe</td>
<td>Suburban</td>
<td>2004–2005</td>
<td>0.44 ± 0.32</td>
<td>1.12 ± 0.44</td>
<td>Xin et al., 2007</td>
</tr>
<tr>
<td>Shenyang</td>
<td>Suburban</td>
<td>2004–2005</td>
<td>0.46 ± 0.23</td>
<td>1.11 ± 0.32</td>
<td>Xin et al., 2007</td>
</tr>
</tbody>
</table>
Fig. 1. The average diurnal variations of temperature (T) and relative humidity (RH) measured at site AC-Ahmednagar during the period January 2016–May 2018.
Fig. 2. The variability of the daily-averaged AOD$_{500}$ nm and AOD$_{1020}$ nm (a), Ångström parameters, AE and β, derived on the basis of AOD spectral curve (b), and the corresponding standard deviations of these parameters (c) observed during January 2016–May 2018.
Fig. 3. The temporal variability of the daily retrieved MODIS AOD\textsubscript{550 nm} (a, b, and c) and its frequency distribution analysis (d) for the study period January 2011–December 2021.
Fig. 4. The box-whisker diagrams together with the associated binned data of the daily averaged (a) Microtops II Sun photometer measured AOD$_{500}$ nm (red color) and (b) MODIS AOD$_{550}$ nm (blue color) over AC-Ahmednagar during January 2016–May 2018.
Fig. 5. Contour density maps of the AOD$_{500\ nm}$ versus AE$_{440-870\ nm}$ in (a) winter (b) pre-monsoon seasons for the period 2016-2018.
Fig. 6. Weighted Potential Source Contribution Function (WPSCF) analysis plots of HYSPLIT 5-days back air mass trajectories at AC-Ahmednagar during (a) winter, (b) pre-monsoon, (c) monsoon, and (d) post-monsoon seasons.