Aerosol measurements over India: Comparison of MAX-DOAS measurements with ground-based (AERONET) and satellite-based (MODIS) data

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Abstract:

In this study, we present ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) measurements of aerosols from two urban locations (Delhi and Pune) and one high-altitude rural location (Mahabaleshwar) in India. We utilized a profile retrieval algorithm to estimate the vertical profiles and vertical column densities of aerosols (AOD) using MAX-DOAS measurements of the oxygen dimer (O_4) at ultraviolet and visible spectral regions. Our results from Pune show good agreement with the Aerosol Robotic Network (AERONET) observations at both UV and visible wavelengths, with correlation coefficients of 0.70 and 0.71, respectively. The MAX-DOAS retrieved AOD and the MODIS satellite-based AOD product at 360 nm for Pune and Mahabaleshwar showed similar variations with correlation coefficients of 0.70 and 0.74, respectively. However, over Delhi, the correlation was poor at 0.54. At 477 nm, all three locations had a moderately positive correlation between the MAX-DOAS and MODIS AODs, with statistically significant correlation coefficients of 0.66, 0.61, and 0.60, respectively. Over the high pollution regions, especially over Delhi, the MODIS AOD was systematically higher than MAX-DOAS AOD.

The vertical profiles of aerosol extinction coefficients (AEC) show that aerosols were concentrated within the boundary layer (below 2 km) in Pune and Delhi, with the highest AEC values observed up to 500 meters. Compared to urban locations, the AEC values over Mahabaleshwar are much smaller; however, the average AEC profile suggests that the air in Mahabaleshwar is homogenous through the boundary layer. MAX-DOAS was also effective at observing high aerosol events, which compared well with ground-based samplers. Overall, this study offers insights into the aerosol distribution and vertical profiles in various regions of India and demonstrates the usefulness of MAX-DOAS in aerosol characterization in India.

Keywords: AOD, MAX-DOAS, MODIS, AERONET, radiative forcing.
1.0 INTRODUCTION:

Aerosols in the atmosphere are solid or liquid particles that can greatly affect air quality, climate, and human health (World Health Organization, 2021). Aerosols can directly affect the Earth's system by absorbing and scattering solar radiation, altering the radiation balance, and influencing the climate (Carslaw et al., 2010; Ian and Mihalis, 2013; Seinfeld et al., 1998; Tomasi et al., 2017). They can also influence cloud formation, precipitation, reflectivity, and lifetime, resulting in an indirect effect that can either cool or warm the Earth's surface. Understanding these effects is crucial for precise climate predictions and the development of effective climate change policies (Alizadeh-Choobari, 2018; Alizadeh-Choobari and Gharaylou, 2017).

Atmospheric aerosols have natural (dust, sea salt, volcanoes, fires) and human-made (fossil fuels, industry, mining, biomass burning) sources (Carslaw et al., 2010; Ian and Mihalis, 2013; Levin and Cotton, 2009). The sources and types of atmospheric aerosols can vary depending on location, weather conditions, and human activities (Tomasi et al., 2017). The exploration of these sources is crucial for developing effective strategies to mitigate air pollution and its associated health and environmental consequences. Several efforts have been made recently for the identification of the sources and impacts on the atmospheric air quality (Robert J. Griffin, 2013; Sabetghadam et al., 2021, 2018). Understanding the sources and behavior of atmospheric aerosols is important for developing effective air pollution control strategies and environmental monitoring (Ian and Mihalis, 2013; Seinfeld et al., 1998).

Aerosols' impact on the radiative budget depends on their optical properties, columnar density, and vertical distribution, which are the main source of uncertainty in estimating aerosol radiative forcing (Choi and Chung, 2014; Ian and Mihalis, 2013; Mishra et al., 2015; Molero et al., 2021). Understanding atmospheric aerosols is important for developing
effective air pollution control strategies and environmental monitoring (Ian and Mihalis, 2013; Seinfeld et al., 1998; Tiwary and Williams, 2018). The South Asian summer monsoon can be influenced by both natural and anthropogenic aerosols and trace gases, leading to significant impacts (Bollasina et al., 2008; Lau et al., 2006; Wild et al., 2005). Understanding the vertical structure, and spatial and seasonal extent of aerosol distribution is crucial to determine the climatic impact of aerosols and regional air quality in India (Brooks et al., 2019; Wild et al., 2005). However, there is a lack of understanding of the vertical distribution of aerosols, leading to uncertainty in their distribution. Numerous investigations have focused on aerosol optical depth (AOD) over India, but studies on the vertical distribution of aerosols are limited. Efforts have been made to measure the vertical distribution of aerosols using ground-based lidars (Radhakrishnan et al., 2017; Raj et al., 2008; Satheesh et al., 2006; Devara et al., 2008; Devera et al., 2008; Ernest Raj et al., 1996; Niranjan et al., 2007; Parameswaran et al., 1991; Tiwari et al., 2003), spaceborne lidar (Babu and Sivaprasad, 2014; Kalluri et al., 2017; Mehta et al., 2021; Ratnam et al., 2021), aircraft (Gogoï et al., 2019; Padmakumari et al., 2013; Tripathi et al., 2005), high-altitude balloons (Babu et al., 2011; Jayaraman, 1991; Ramachandran and Jayaraman, 2003), rocket-borne photometers (Jayaraman et al., 1987; Jayaraman and Subbaraya, 1993, 1988) and ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) (Kumar et al., 2020). The reported observation in the Indian region carried out using satellite and lidar-based instruments posed a limitation in terms of their spatial and temporal coverage, result in significant gaps in aerosol extinction profile data. Therefore, the sustained long-term efforts over India are still lacking. MAX-DOAS is a cost-effective and user-friendly technique to measure global aerosols and trace gases by observing scattered sunlight from multiple viewing directions (Platt and Stutz, 2008; Wagner et al., 2004). It provides comprehensive information on aerosol properties and is widely used for detecting aerosols and retrieving data products such as AOD and vertical
profiles of aerosol extinction coefficients (AEC). These vertical profiles can also be used to retrieve the vertical distribution of trace gases (Frieß et al., 2019 and references therein). Studies conducted in Beijing, China (Clémer et al., 2010); Madrid, Spain (Wang et al., 2016); and Mexico City (Friedrich et al., 2019) have shown good agreement between the retrieved AOD values and those derived from co-located Aerosol Robotic Network (AERONET) sun-photometer measurements. The retrieved AOD value has also been widely used to validate satellite AOD estimates (Gielen et al., 2017; Irie et al., 2008; Schreier et al., 2016; Takashima et al., 2009; Tanvir et al., 2022; Wang et al., 2019). However, aerosol observations using MAX-DOAS in India have been rarely reported. Kumar et al. (2020) conducted a study in Mohali, India, over a period of four years and found good correlation with AERONET data. In addition, they evaluated satellite data and discovered a strong correlation with the observations obtained through satellite. However, they also indicated a slight overestimation by the MODIS with respect to the AOD retrieved from MAX-DOAS measurements.

Our study highlights the efficacy of the MAX-DOAS instrument in assessing aerosol parameters through a radiative transfer model across the Indian region. We utilized the MAX-DOAS technique in conjunction with the MAinz Profile Algorithm (MAPA) to measure aerosol optical properties such as AOD and vertical profiles of AEC over three locations in India (Pune, Delhi, and Mahabaleshwar) (Beirle et al., 2019). We perform a comparative analysis of AOD data products from satellite-based Moderate Resolution Imaging Spectroradiometer (MODIS) and ground-based Aerosol Robotic Network (AERONET) with the MAX-DOAS measurements. Discrepancies between the datasets are explored and discussed. We also examine the aerosol-induced atmospheric radiative forcing and heating using the Santa Barbara Discrete-ordinate Atmospheric Radiative Transfer model (SBDART) (Ricchiazzi et al., 1998).

2.0 DATA AND METHODOLOGY
2.1 Observation sites

Three locations in India, namely Pune and Mahabaleshwar in western India and Delhi in northern India (Fig. 1), were chosen to evaluate the applicability of the MAX-DOAS instrument in measuring aerosols in diverse environments.

2.1.1 Pune

Observations were conducted in Pune city, India (18.50° N, 73.5° E, 560 m AMSL) from January 2, 2018 to February 8, 2019 (Biswa and Mahajan, 2021). Pune is situated on the leeward side of the Western Ghats, approximately 120 km from the west coast of India, with the Bhosari and Pimpri-Chinchwad industrial complexes located about 15 km and 12 km away respectively to the northeast and northwest. The city center is approximately 7 km east-southeast from the measurement site. Due to the ongoing urbanization of the city, dust is the prime source of aerosols in Pune (Devara et al., 2005a), but the aerosols found in this region can be a mixture of soot-like, dust-type, water-soluble aerosols (Safai et al., 2010). Weather conditions drive the sources and types of atmospheric aerosols over Pune (Devara et al., 2005b). Like most of India, Pune experiences four meteorological seasons (winter: January-February, pre-monsoon: March–April–May, monsoon: June–July–August–September and post-monsoon: October–November–December) (Attri and Tyagi, 2010). The MAX-DOAS instrument was installed on the roof of the Prithvi hostel (18.54° N, 73.80° E; ~30 m above ground). The scanner pointed towards 355° with respect to the geometric north. Further details of the instrument are given in the next Section, 2.2.

2.1.2 Delhi

Observations in Delhi were carried out at the Indian Institute of Tropical Meteorology (IITM), Delhi campus (77.17° E, 28.63° N, ~216 m AMSL), India, from June 1, 2019 to September
Delhi, the capital of India, is one of the most densely populated cities in India. Delhi is surrounded by its satellite cities of Ghaziabad, Faridabad, Gurgaon, and Noida, making it one of the largest metropolitan regions in the world (United Nations, Department of Economic and Social Affairs, 2018). Delhi's prime pollution sources are transportation, manufacturing industries and thermal power plants (Tiwari et al., 2014). Similar to Pune, Delhi experiences season-wise aerosol distribution, with mostly human-made aerosols observed (Srivastava et al., 2012). In winter, pollution levels in Delhi enhance due to biomass and agricultural crop residue burning in the nearby rural area of Punjab and Haryana (Awasthi et al., 2011; Guttikunda and Gurjar, 2012). In pre-monsoon and post-monsoon seasons, natural aerosols (mostly dust) from the adjacent desert regions contribute to regional aerosol loading (Guttikunda and Gurjar, 2012). The aerosol loading is usually minimum during the monsoon season over Delhi due to the washout effect (Guttikunda and Gurjar, 2012). The sampling site is surrounded by small forests on three sides and a residential colony on the fourth. No major industrial installations or traffic intersections within 5 km radius. The MAX-DOAS instrument was installed on the rooftop of the IITM building at an altitude of ~10 m above ground. The instrumental azimuth angle (scanner pointing direction) is set to 240° with respect to the geometric north.

2.1.3 Mahabaleshwar

The Observations in Mahabaleshwar (17.92° N, 73.4° E ~1378 AMSL), India, were carried out at the High-Altitude Cloud Physics Laboratory (HACPL) from April 25 2018 to May 31 2018 (Biswas et al., 2021). HACPL is situated on a top mountain plateau. The base of the mountain is ~600 m above the mean sea level; therefore, the mountain plateau is almost 800 m above the neighboring area. Hence due to the plateau's elevation, the boundary layer remains below the HACPL site at night. During the day, the boundary layer gradually rises...
above the site. Mahabaleshwar is a small town situated in the mountainous Sahyadri range of the Western Ghats in western India, approximately 120 km southwest of Pune and 285 km southeast of Mumbai. The site is surrounded by dense vegetation and residential houses, hotels, and a rural market; hence the concentration of anthropogenic air pollution is low compared to urban locations. Most observed aerosol types over this region combine biogenic and anthropogenic emissions. The study by Leena et al., (2016) showed that the aerosol concentration is highly influenced by the long range airmass transport over this site. The MAX-DOAS instrument was installed on the roof of the HACPL building at an altitude of ~10 m above ground. The scanner unit was pointed towards the geometric north overlooking a forest canopy. The MAX-DOAS instrument used at Mahabaleshwar differs from the instrument used for Pune and Delhi. Further details of the instrument are given in Section 2.2.

2.2 MAX-DOAS measurement setup and spectral analysis.

This study utilized two sets of MAX-DOAS instruments. The MAX-DOAS instrument used in Pune and Delhi has two spectrometers operating in the ultraviolet (UV: 306.01-468.55 nm) and visible (441.95-583.27 nm) range, respectively. Both spectrometers have a full-width half-maximum resolution of 0.6 nm and a 100 µm slit. The second instrument used in Mahabaleshwar has two spectrometers with wavelength ranges, UV: 301.50–463.73 nm and VIS: 443.54–584.19 nm. In this instrument, both spectrometers have a full width half maximum resolution of 0.7 nm and 100 µm slit. Both the MAX-DOAS instruments have two parts, one scanner unit (that directs scattered sunlight towards the spectrometer) and one indoor unit containing two different spectrometers. The outdoor unit, comprised of a telescope, was used to measure scattered sunlight at different elevation angles, which is connected via a fiber bundle to the indoor unit housing the UV and visible spectrometers. Each elevation angle scan consists of sequential observations at eight angles (1°, 2°, 3°, 5°,
This study uses only measurements with SZA less than 75° for the profile retrievals to avoid any stratospheric influence. Every day after sunset, dark current, offset, and calibration spectra were recorded and later used during the analysis for spectral correction. Calibration spectra were recorded using an integrated mercury vapor lamp. Further details about the MAX-DOAS instruments can be found in our previous work by Biswas et al. (2019, 2021, 2021(b)). O₄ differential slant column densities (DSCDs) were retrieved using the QDOAS spectral fitting software (Danckaert, 2014) (https://uv-vis.aeronomie.be/software/QDOAS/ last access: 16/05/2021 12:00 UTC). Fig. 2 shows representative fits of O₄ at UV and visible from the Pune campaign. The spectral fitting settings, including the wavelength regions and cross-sections for each wavelength region, are given in Table 1. In addition to these settings, RMS filters applied to O₄ DSCDs, i.e., RMS greater than $1 \times 10^{-3}$, were removed. Fig. 3 shows the time series of O₄ DSCDs at 360 and 477 nm at different elevation angles over Pune, Delhi, and Mahabaleshwar, respectively.

2.3 Profile inversion

Vertical AEC profiles were retrieved from MAX-DOAS O₄ DSCDs by employing the MAPA (Beirle et al., 2019). A Monte Carlo approach is used to find an ensemble of best-matching parameters; column parameter (c) (i.e., AOD), height parameter (h), and shape parameter (s). This process consists of a forward model and an inversion algorithm (MAPA). The forward model used is the Monte Carlo Atmospheric Radiative Transfer Inversion Model (McArtim) (v1) (Deutschmann et al., 2011). The O₄ VCD is calculated from temperature and pressure profiles observed by radiosondes. Radiosonde data from Pune is obtained from an online repository (http://weather.uwyo.edu/upperair/sounding.html). For Delhi and Mahabaleshwar, radiosonde data was unavailable; therefore, a standard temperature and pressure profile was
used to calculate the O$_4$ VCD. For a detailed discussion of the inversion technique of the MAPA algorithm, please refer to (Beirle et al., 2019).

Some previous studies have reported a significant mismatch between modelled and measured O$_4$ DSCDs, which can be corrected using a scaling factor (0.8) applied to the measured O$_4$ absorption (Beirle et al., 2019; Clémer et al., 2010; Kumar et al., 2020; Wagner et al., 2019, 2010). However, in our study it was noticed that the AOD values retrieved using MAPA show good agreement with Aerosol Robotic Network (AERONET) measurements if no scaling factor is applied for O$_4$ and hence no scaling factor is applied. Similar to our study, few studies in the past have also found no need for a scaling factor to achieve agreement between measured and modelled O$_4$ DSCDs (Ortega et al., 2016; Peters et al., 2012; Schreier et al., 2016).

### 2.3.1 Flags/Threshold:

MAPA produces a set of profile parameters to assess the quality of the profile retrieval and help select valid DSCD sequences. These profile parameters were then passed through a quality check to ensure that erroneous results were filtered. We analyzed the year-long data from Pune and used it to establish standard thresholds for specific parameters. The precision of thresholds was checked by comparing the results with ground-based in situ measurements from the AERONET. Using AERONET observations, we adjusted the MAPA retrieval settings and set the flags thresholds accordingly. Below is a list of thresholds and the corresponding flags that were defined.

Elevation angles (EAS): When the solar zenith angle is small (<35°), the scattering angles are small. MAPA struggles to produce correct results in the UV wavelength region for such geometric conditions due to the uncertainty of the aerosol phase function. In this case, MAPA inversions are performed without considering the high elevation angles (EAS > 20°). This
Normalized RMS: Within MAPA, each elevation sequence yields the best match between the modelled DSCDs ($S_{mo}$) and the measured DSCDs ($S_{ms}$) sequence. Typically, the best match is defined in terms of the root mean square (RMS) $R$.

$$R = \sqrt{\frac{(S_{ma}-S_{ms})^2}{M}}$$  \hspace{1cm} (1)

Where $M$ is the number of EAS Generally, the $R$-value is proportional to the absolute column values. Therefore, just a threshold for $R$ would remove the valid sequences where the concentrations of the retrieved species are high. Hence, we consider the RMS normalized by the maximum DSCDs ($S_{max}$) in each scan:

$$R_n = \frac{R}{S_{max}}$$  \hspace{1cm} (2)

For aerosols, the $R_n$ thresholds ($R_n$ (UV EAS $1^\circ - 40^\circ$) = 0.075, $R_n$ (UV EAS $1^\circ - 10^\circ$) = 0.025, $R_n$ (VIS –EAS $1^\circ - 40^\circ$) = 0.2) were decided according to the match between the modeled and observed DSCDs. For trace gases, a threshold of $R_n$ (EAS $1^\circ - 40^\circ$) = 0.15, $R_n$ (EAS $1^\circ - 10^\circ$) = 0.10 was used.

Layer height: We used 1.5 km and 2.0 km as the height threshold for the Pune and Delhi regions. This assumption is quite reasonable as similar values of boundary layer height are also observed by previous studies over Pune and Delhi (Pal and Devara, 2012; Singh et al., 2010). Additionally, increasing the layer height more than the boundary layer reduces the correlation with AERONET and leads to unrealistic high AOD values. Due to the high-altitude topography of the Mahabaleshwar site, no height threshold limit was applied, as it was noticed that most sequences were filtered due to this parameter.
Cloud index: Clouds can influence MAX-DOAS observations of aerosols and trace gases and their subsequent profile inversion as they alter the light path and intensity (Platt and Stutz, 2008; Wagner et al., 2014). We used a cloud detection method using the MAX-DOAS measurements based on the measured radiance, color index (CI), and the retrieved O_4 absorption. The color index was calculated as the radiance ratio at two wavelengths, i.e., 320 nm and 440 nm (Enell et al., 2003; Sarkissian et al., 1994; Wagner et al., 2014). Subsequently, a cloud index (CI(5°)/CI(40°)) was calculated. A cloud index was adopted based on the color index introduced by Wagner et al. (2014) and the cloud screening method used by Sinreich et al. (2010). We compared the retrieved AOD and their corresponding RMS with the cloud index and observed that most of the estimated AOD values with low RMS are below the CL[5/40] value of 0.8. Hence, a threshold of 0.8 was set to filter clouds.

2.4 AERONET ground-based measurements.

AOD measurements made with a CIMEL CE 318 sun photometer, a component of the Pune-based (18.537°N 73.805°E ~595 AMSL) Aerosol Robotic NETwork (AERONET) station (Holben et al., 1998), were compared with the MAX-DOAS retrievals. We used level 2.0 (cloud-screened and quality assured data) to validate the retrieved AOD product. AERONET AOD data is available only over Pune among the three stations during the observation periods (https://aeronet.gsfc.nasa.gov/cgi-bin/draw_map_display_aod_v3). The AERONET database at Pune contains the AOD at 440, 675, 870, and 1020 nm wavelengths. For this study, we estimated AERONET AOD at 360 and 477 nm using the angstrom exponent (AE) between the standard spectral bands of the instrument (440 and 675 nm) according to Equation (3).

Using the flags mentioned above, we compared our filtered dataset with ground-based measurements from AERONET. The AOD at 360 and 477 nm were calculated using the
angstrom exponent (\(\alpha\)) between the standard spectral bands of the AERONET observations (440 and 675 nm).

\[
\alpha = -\frac{\log(\tau_{\lambda_1}/\tau_{\lambda_2})}{\log(\lambda_1/\lambda_2)} \tag{3}
\]

2.5 MODIS Satellite observations:

In this study, daily satellite measurements of aerosol products were obtained from MODIS Terra (MOD04_L2) and Aqua (MYD04_L2) at the spatial resolution of a 10 × 10 km pixel array (at nadir) (https://ladsweb.modaps.eosdis.nasa.gov/search/order/1/MODIS). MODIS sensors onboard the Terra and Aqua satellites are a part of NASA's Earth Science Data Systems (ESDS) Program. The MODIS Level 2 Atmospheric Aerosol Products offer complete global coverage of aerosol properties from the Dark Target (DT) and Deep Blue (DB) algorithms (Levy and Hsu, 2015). Additional information of MODIS satellite observations is available at http://dx.doi.org/10.5067/MODIS/MOD04_L2.006 (Terra) & http://dx.doi.org/10.5067/MODIS/MYD04_L2.006 (Aqua). The retrieved data set from both satellites were matched within a period of ± 15 min of the MAX-DOAS observations, centered at satellite overpass time. MODIS AOD in the wavelength range of 360 and 477 nm was calculated using estimated satellite AOD at 470 and 660 nm wavelengths by using the angstrom exponent. Considering that the MAX-DOAS path lengths, we compare MODIS observations from within 5 km of the measurement site, along the viewing direction.

3.0 RESULTS AND DISCUSSION.

3.1 AOD
Fig. 4 shows the time series of AOD retrieved at 360 and 477 nm in Pune, Delhi, and Mahabaleshwar. AOD over Pune exhibits a typical seasonal variation with higher values during winter and lower values during monsoon. The mean AOD at 360 nm for the entire measurement period was $0.60 \pm 0.41$, with the monthly means varying between 0.23 in July 2018 and 0.90 in January 2018, whereas the mean AOD at 477 nm for the entire measurement period was $0.32 \pm 0.28$, with the monthly means varying between 0.10 in July 2018 and 0.48 in January 2018. The seasonal cycle of aerosol at 360 nm is explicitly observed; in fact, the average concentration of AOD in winter ($0.78 \pm 0.46$) is almost double than AOD in pre-monsoon ($0.43 \pm 0.35$) and three times larger than that in monsoon ($0.26 \pm 0.18$). At 477 nm, AOD in winter ($0.45 \pm 0.26$) is larger than pre-monsoon ($0.28 \pm 0.22$) and monsoon season ($0.39 \pm 0.30$).

An aerosol discrimination method described by Sabetghadam et al. (2021) was used to classify aerosol types based on AOD at 550 nm and Angstrom Exponent (AE) from 360/477 nm. Monthly average AE values range between 0.46 and 1.43, implying a wide range of aerosol sizes over the region. The combination of seasonal AOD and AE analysis of different aerosol types shows the dominance of urban-type fine mode aerosols during winter (AOD = 0.41, AE = 1.28), mixed-type (bimodal) in pre-monsoon (AOD = 0.24, AE = 0.67), and aerosols indicating a marine influence in the monsoon season (AOD = 0.20, AE = 0.70) over the Pune. This implies that anthropogenic aerosols are a major contributor to total AOD over Pune. During the monsoon season (JJAS), AOD is low because of the washout effect and cleaner southwesterly winds from the Arabian Sea, but a considerable amount of data was excluded from June to September 2018 due to cloudy conditions. Pandithurai et al. (2007) found that submicron aerosols, which arise from burning fossil fuels and biofuels, were the primary source of aerosols during winter over Pune. The study also revealed that both pollution and dust influence aerosols in this region during pre-monsoon. Safai et al. (2007)
observed a similar seasonal variation over Pune in black carbon. The seasonal AE values (~0.67 in pre-monsoon and ~1.28 in winter) observed during the current study also support the dominance of fine mode particles in winter and the presence of a mixture of aerosols (bimodal) in pre-monsoon months over Pune.

The AOD values in Delhi exhibit a larger variation compared to Pune, particularly at 360 nm (Fig. 4). The mean AOD at 360 nm in Delhi was 1.02 ± 0.88, with the monthly means varying between 0.83 in August 2018 and 1.51 in September 2018, whereas the mean AOD at 477 nm was 0.42 ± 0.24 with the monthly means varying between 0.35 in August 2018 and 0.51 in July 2018.

The AOD at both 360 nm and 477 nm was higher at Delhi than at Pune, indicating higher pollution levels in Delhi. A mean value of 0.43 for the AOD and >2.0 for the AE over Delhi indicates the prevalence of urban/industrial fine mode particles during the pre-monsoon and monsoon seasons. Frequently, such elevated AOD levels are a consequence of the blending of regional anthropogenic aerosols, predominantly of urban origin, with mineral dust transported from desert areas at high altitudes during the pre-monsoon season (Pandithurai et al., 2008; Singh et al., 2010). Such events are common in the Indo-Gangetic plains of India. Delhi’s atmosphere is characterized by the omnipresence of dust throughout the year (Srivastava et al., 2014). Such Dust-storm events are observed frequently in the Indo-Gangetic plains of India during pre-monsoon months (Pandithurai et al., 2008; Vijayakumar et al., 2020). The high AOD values observed during monsoon and pre-monsoon months in our study could be the results of the high altitude (~6 km) aerosol layer over the region (Srivastava et al., 2014).

MAX-DOAS measurements at Mahabaleshwar were made during the pre-monsoon months (Fig. 4). The mean AOD at 360 and 477 nm at Mahabaleshwar was 0.99 ± 0.90 and 0.55 ±
0.58, respectively. Such high AOD values in summer are primarily driven by high solar radiation and temperature (Mukherjee et al., 2018). Most aerosols in the summer at Mahabaleshwar are secondary organic aerosols, making up a large fraction of tropospheric fine mode aerosol (Singla et al., 2019).

The mean AOD (0.42) and AE value (> 1.9) support the dominance of urban fine mode particles over the site during the study period. Aerosols of this type mainly originate from a wide variety of natural sources, such as wildfires and anthropogenic sources, like the combustion of fossil fuel and biomass burning (Leena et al., 2016). Contrary to our study, Leena et al., 2016 also showed that during pre-monsoon months dust types could be predominant, which may have originated with long range transport of dust, especially from the desert region in Oman. Other aerosols with larger diameter sizes can be observed that may have been transported with continental air masses (Leena et al., 2016). It should be noted that profile retrievals during dust storms are not dependable considering the multiple scattering effects and hence, the divergence from earlier studies concerning high values at Mahabaleshwar might stem from the adoption of MAPA algorithm and flags to effectively eliminate inaccurate profile retrievals.

3.2 Intercomparison with AERONET

Fig. 5 shows the intercomparison of retrieved AOD from the MAX-DOAS and sun photometer (AERONET) measurements for Pune. A moderate positive correlation between MAX-DOAS and AERONET measurements for both wavelengths is observed (r = 0.70 at 360 nm with p < 0.001, slope = 0.79, intercept = -0.03 and r = 0.71 at 477 nm with p < 0.001, slope = 0.92, intercept = -0.05). Nevertheless, there is a noticeable underestimation in the MAX-DOAS measurements, particularly at 360 nm during the pre-monsoon period. The slope at 360 nm was 0.76 with an intercept -0.03 and at 477 nm it was 0.92 with an intercept -
0.05, indicating that MAPA agrees better with AERONET at 477 nm than at 360 nm. The sampling differences can explain the underestimation of the retrieved AOD by MAX-DOAS. The sensitivity of the MAX-DOAS instrument is highest near the ground but drops to virtually zero above ~4 km, i.e. the different vertical extents of the atmosphere observed by the MAX-DOAS and AERONET (Kumar et al., 2020; Wang et al., 2016; Wittrock et al., 2004). This limited sensitivity increases with increasing aerosol loads (Tian et al., 2021). This difference between MAX-DOAS and AERONET (i.e. occasional occurrences of high AOD) is particularly relevant for dust events when the air mass can transport dust at higher altitudes, especially during the monsoon and pre-monsoon season (Aher et al., 2014; Vijayakumar et al., 2020). The second difference is that the sun photometer samples the atmosphere directly overhead from the measurement site, while the MAX-DOAS samples the atmosphere along paths across different elevation angles, resulting in geographical averaging. Similar underestimations by MAX-DOAS during dust events have been observed around other parts of the world (Davis et al., 2020; Gratsea et al., 2021; Wang et al., 2016).

### 3.3 Intercomparison with MODIS

We compared the MAX-DOAS retrieved AOD with the MODIS satellite-based AOD product over Pune, Delhi, and Mahabaleshwar at 360 and 477 nm, respectively (Fig. 6 and 7). At 360 nm, the correlation coefficients are 0.80, 0.29 and 0.62 for Pune, Delhi and Mahabaleshwar, respectively. The p-value for all three stations is less than 0.01, indicating that the correlation is statistically significant. The slope values are 1.12, 0.61, and 0.63 in Pune, Delhi and Mahabaleshwar, respectively. The relatively better agreement between MAX-DOAS retrieved AOD and MODIS satellite-based AOD products in Pune and Mahabaleshwar suggests that the MODIS product is reliable at these sites.
For AOD at 477 nm, the correlation for Pune, Delhi and Mahabaleshwar is observed to be 0.62, 0.61 and 0.65, respectively. The p-value for all three stations is less than 0.01, indicating that the correlation is statistically significant. The slope values are 0.37, 0.30, and 0.37 in Pune, Delhi and Mahabaleshwar, respectively. Overall, the analysis suggests a moderately positive correlation with statistical significance between the MAX-DOAS and MODIS AODs for all three locations.

Although a moderately good correlation exists between MAX-DOAS and MODIS, the mismatch between their AOD values, especially in polluted areas like Delhi, cannot be overlooked. Note that in this study, we applied linear regression to determine the comparability of measurements obtained from MAX-DOAS and MODIS. In Delhi, we observed that the slope is consistently low, and the intercept is high. If we average around the MODIS overpass time, the mean MAX-DOAS AOD at 360 nm for the measurement period over Pune, Delhi, and Mahabaleshwar was 0.83 ± 0.53, 1.31 ± 0.71, and 0.66 ± 0.29, respectively, compared to the MODIS AOD of 0.76 ± 0.37, 1.29 ± 0.33, and 0.69 ± 0.34. Additionally, the mean MAX-DOAS AOD at 477 nm for the measurement period over Pune, Delhi, and Mahabaleshwar was 0.36 ± 0.15, 0.47 ± 0.33, and 0.32 ± 0.13, respectively, compared to the MODIS AOD of 0.49 ± 0.25, 0.95 ± 0.32, and 0.45 ± 0.22. Fig. 8 depicts the frequency distributions of delta AOD, which is the difference between the MODIS and MAPA-derived AOD over Pune, Delhi, and Mahabaleshwar. At 360 nm, MODIS records slightly lower values in Pune and Mahabaleshwar, but at 477 nm, it reports relatively higher values particularly over Delhi. Intercomparison of MAX-DOAS, aerosol observations with satellite observations, are rarely done in India. According to a study by Kumar et al., 2020, there was a good agreement between the MODIS data product and the MAX-DOAS observations at Mohali, India. However, the study also found that the AOD obtained through MODIS were consistently higher than those obtained through MAX-DOAS. This is similar to
our observations, where at Pune, where we have the largest dataset for a thorough comparison, MODIS shows higher values than the MAX-DOAS (Fig. 7).

The reason for the differences in MODIS and MAX-DOAS observations is possibly due to the assumptions about aerosol properties such as phase function and single scattering albedo in retrieving data from MODIS, which determine the relationship between the observed backscattered radiance and the retrieved AOD. Therefore, more study on the validation of aerosol retrieval from satellites is needed. Due to the limited data, a comprehensive comparison with satellite estimates is not possible, especially at Delhi and Mahabaleshwar. This study emphasizes the need for extended, concurrent observations to effectively validate satellite aerosol retrievals.

### 3.4 Aerosol Extinction coefficient (AEC) vertical profiles

A few examples of comparison of the MAX-DOAS observed and MAPA-modelled \( \text{O}_4 \) DSCDs at 360 and 477 nm, along with the corresponding AEC vertical profiles for a few sequences over Pune, Delhi, and Mahabaleshwar are shown in Fig. 9, respectively. Most of the aerosol profiles over Pune and Delhi obtained from the forward model have shown similar shapes, implying that a large fraction of the atmospheric aerosols is in the boundary layer's lower layer (< 1 km). However, in the Mahabaleshwar, due to the plateau's elevations and different boundary layer mechanisms than Pune and Delhi, an elevated layer of aerosol up to 2 km from the surface was observed.

The seasonal and monthly mean vertical profiles of AEC at 360 and 477 nm at all the sites are shown in Fig. 10. Note that the AEC is always greater than or equal to zero, and the error bars ranging to negative values result from high scatter for not normally distributed values. Higher AEC is observed close to the surface (< 1 km) at all the sites during all seasons. At Pune, the highest AEC is observed during winter, followed by post-monsoon and pre-
monsoon, and the lowest values are observed during the monsoon season. At 360 nm, values increase from the surface to peak at about 200 m during all seasons. However, at 477 nm, such an elevated peak is not seen with the maximum observed near the ground. The surface AEC at 477 nm is around four times higher than at 360 nm. There is a significant difference between the 360 nm and 477 nm vertical profiles. The difference might be because of the different horizontal sensitivity ranges for both wavelengths due to the different probability for Rayleigh and aerosol scattering. Thus, the possible presence of a horizontal gradient will affect both wavelengths differently. However, after using data with identical timestamps, a similar vertical profile is observed in both wavelength regions except for the winter months. Due to the low number of common data points, long-term simultaneous observations are needed to compare the UV and visible regions precisely.

There are few studies in the past where the vertical profile of aerosols has been studied over Pune using ground-based instruments (Devara et al., 1994; Pal and Devara, 2012; Devara, 1995; Devara et al., 2008, 2003; Tiwari et al., 2003). Study by Devara, 1995 indicates that the maximum concentration of air pollutants in Pune was found to be high in the height range 500–1000 m. Their study indicates that the maximum concentration of air pollutants is in the surface layer. Devara et al., 1994 also indicated that the observed aerosol layer at the high altitude over Pune comes from long-range transport. Study by Tiwari et al. (2003) examined the seasonal variations in AEC, revealing that aerosols are predominantly concentrated within the lower atmospheric layers. This matches our observations, demonstrating an elevated aerosol content during the winter compared to the pre-monsoon period. Furthermore, the study illustrated a substantial reduction in aerosol content during the monsoon season, similar to our results.

Over Delhi, the AEC at 360 nm and 477 nm are greater than in Pune during the same season (Fig. 10), which is expected considering the higher sources in Delhi (CPCB, 2012; Gani et al.,
2019; Rao et al., 2016). As stated above, in the monsoon, the aerosol content in the atmosphere is primarily dominated by dust at high altitudes (~6 km) due to long-range transport. However, at the surface level, anthropogenic activities (such as road construction activities, industrial dust, etc.) play a key role in contributing to the high AEC values. Fig. 10 shows the elevated aerosol layer at 360 nm from the surface to ~1 km, where most aerosols are confined. At 477 nm, maximum aerosol content is observed at the surface; above that, the aerosol content decreases exponentially. Study by Komppula et al. (2012) observed similar profiles from lidar observations over Delhi. They also noted a high concentration of aerosols near the surface, followed by an exponential decline between 1 to 3 km. Using the satellite-based lidar data, Srivastava et al. (2014) observed a similar profile shape over Delhi. In their study during the monsoon month of July, they also observed a dusty layer above an altitude of 6 km. Srivastava et al. (2014) showed that regional activities primarily drive high aerosol concentrations at the surface layer.

During the same season, the profiles of Mahabaleshwar and Pune are similar, although Pune has a higher loading in the boundary layer. The shapes of the profiles at 360 and 477 nm are similar, indicating homogeneity of air in Mahabaleshwar through the boundary layer, dominated by a combination of fine and coarse modes. Fig. 10 clearly shows an elevated layer of aerosols at ~500 m above the surface. Mahabaleshwar has a significant abundance of carbonaceous aerosols, particularly during the summer season, owing to its high altitude and tropical location surrounded by forests and vegetation (Leena et al., 2017; Buchunde et al., 2022). However, dust transport dominates high altitude aerosol layers (Leena et al., 2016).

Unfortunately, there have been no lidar-based observations conducted at Mahabaleshwar which would provide a basis for comparison with our own observations. To understand the seasonal and temporal variation of aerosol's vertical profile, long-term ground-based observations are needed.
3.5 Elevated pollution case study

There was an event of elevated values of surface AEC between 10:00 AM to 02:00 PM on June 9, 2019 (Fig. 11) at Delhi that was observed in the MAX-DOAS retrievals. Fig. 11 shows a rise in the AOD values on June 9, 2019, peaking at 3.5 around 12:30 PM, compared to the average value of 1.02 during the measurement period. The examples of corresponding retrieved AEC vertical profile show a large aerosol loading extending up to approximately 500 meters, with a sharp decrease in aerosol concentration above this altitude. Fig. 11 (lower panel) shows an example of retrieved AEC profiles during the elevated values of AEC, along with a typical profile before the event.

To validate these observations, in situ measurements of PM$_{2.5}$ and PM$_{10}$ were obtained from ground-based samplers located at Pusa, Delhi, which is located close to the measurement site and is run by the Indian Central Pollution Control Board (CPCB) [https://cpcb.nic.in/real-time-air-quality-data/]. The PM$_{2.5}$ and PM$_{10}$ concentrations showed an increase during the same period, and the MAX-DOAS was able to capture the high pollution event (Fig. 11). The sudden increase in PM$_{2.5}$ and PM$_{10}$ is most probably due to local biomass burning event (Beig et al., 2020; Lalchandani et al., 2022). The air quality monitoring station at Pusa also shows an increase in carbon monoxide during the same period. The other air quality monitoring stations around Delhi located further from the measurement site did not show an increase in particulate matter, suggesting that it was a localized event.

This case study demonstrates the effectiveness of the MAX-DOAS instrument in detecting high levels of aerosol loading at the surface and hence can be used to make continuous measurements of aerosols, complementing ground-based stations by providing further information about the vertical profiles.

3.6 Radiative Implications
The aerosol radiation forcing (ARF) at a specific layer in the atmosphere can be defined as the disparity between the net fluxes (downward minus upward) with and without the presence of aerosols at that layer. We used the SBDART model for estimating the ARF and the heating rate (HR) of the atmosphere in the short-wave (0.25–4.0 µm) region (Ricchiazzi et al., 1998). The radiative transfer computations over Pune, Delhi, and Mahabaleshwar are performed for clear sky conditions using the observed AOD 550 nm. The AOD at 550 nm was calculated using the AE from the measurements.

The ARF and HR of the atmosphere over Pune persist at more than 20 Wm⁻² and 0.3 K day⁻¹, respectively, throughout the year, except for the monsoon season. Due to low aerosol content in monsoon season, the ARF and HR of the atmosphere over Pune reduce to ~15 W m⁻² and ~0.2 K day⁻¹, respectively. The monthly variation of ARF and HR is mainly governed by the weighted mean of AOD, which varied mostly between 0.2 and 0.6. Similar ARF and HR values over Pune were observed by (Kumar and Devara, 2012).

Over Delhi, the ARF and HR of the atmosphere persist more than 25–30 Wm⁻² and 0.25–0.3 K day⁻¹, respectively, throughout the study period. Despite the monsoon season, high ARF and HR values over Delhi compared to Pune's monsoon time measurements could be the result of high aerosol loading. Kumar et al. (2016) studied the ARF enhancement during extreme aerosol and clean days over Delhi. Their observed ARF value of the atmosphere is quite similar to our study. They concluded that high ARF and HR values over Delhi are primarily associated with dust.

On the other hand, the average ARF and HR over Mahabaleshwar during pre-monsoon were between 35 W m⁻² and ~0.34 K Day⁻¹, respectively. Understanding aerosol properties and their effect on the regional atmosphere is limited. Therefore, long term observations are needed to understand the seasonal variation in ARF over high-altitude stations.
In Delhi, the atmospheric ARF is higher than those in Pune because of higher aerosol loading during the monsoon season. However, despite the low AOD values in Mahabaleshwar, ARF is higher than in Delhi. Potential reasons for this could be the geographic positions of the study sites. Along with the prevailing meteorological conditions, the type of dominated aerosol and their continuous sources could play a key role in monitoring the radiative forcing over the regions (Buchunde et al., 2022; Kumar et al., 2016; Kumar and Devara, 2012; Surendran et al., 2013).

4.0 CONCLUSIONS

This paper presents the retrieval of AEC vertical profiles and resulting AOD from MAX-DOAS measurements from regionally representative urban (Delhi and Pune) and high-altitude rural (Mahabaleshwar) sites. This study employed the MAPA algorithm to retrieve AEC profiles in the lower troposphere by utilizing O$_4$ absorption in the UV (360 nm) and visible (477 nm) wavelength bands at multiple elevation angles as input. The retrieved AOD over Pune shows a typical seasonal pattern with high values in winter and low values in the monsoon season. Delhi shows the highest values of AOD and AEC, which is indicative of larger sources, while Mahabaleshwar shows the lowest values. The vertical profiles of AEC over Pune and Delhi show that a major fraction of the aerosol concentration is close to the Earth. Over Pune, high AEC is observed during the winter due to increased aerosol loading, followed by post-monsoon, pre-monsoon and monsoon months. Compared to Pune, high AEC is observed over Delhi in the monsoon season. Such high values in Delhi are expected, considering the higher sources in Delhi. Over Mahabaleshwar, the aerosol concentration is quite low compared to Pune and Delhi. However, fine mode aerosol which usually originates from wildfire or fossil fuel burning, seem to dominate the aerosol concentration at Mahabaleshwar. Comparison of MAX-DOAS observations with AERONET shows a good agreement with significant correlation coefficients, with an r of 0.70 at 360 nm and 0.71 at
477 nm. A comparison with MODIS satellite estimations shows that AOD at UV correlates well with MODIS at Pune and Mahabaleshwar but not in Delhi. The efficiency of MAX-DOAS in detecting high pollution events was also shown using a case study over Delhi.

Our study suggests that long-term and continuous measurements are necessary to understand the temporal and seasonal fluctuations of aerosols in the atmosphere. Given that India is one of the fastest developing countries in the world, it is also one of the largest hotspots for anthropogenic emissions. Therefore, long-term observations of the prime pollutants in the atmosphere will be crucial for future studies. The study shows that the MAX-DOAS instrument holds the potential to provide an effective approach for sustained long-term measurements of aerosol optical properties.

ACKNOWLEDGMENTS:

Indian Institute of Tropical Meteorology is supported by the Ministry of Earth Sciences, Government of India.
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Figure 1: MAX-DOAS observation locations over the Indian region. The three sites (urban: Pune and Delhi; rural: Mahabaleshwar) used in this study are shown along with major cities nearby. The arrow shows the viewing direction of the MAX-DOAS spectrometer.
Figure 2: DOAS fits for O₄ (in UV and visible wavelength range), O₄ (351-390 nm): 20th April 2018 03:25 hr (UTC); SZA: 52.9°; Elevation angle (EA): 3.0°; DSCD: 2.6 ± 0.03 × 10⁴³ molecules² cm⁻⁵; RMS: 4.2 × 10⁻⁴ (top panel). O₄ (460-490 nm): 19th April 2018 07:53 hr; SZA: 14°; EA: 3.0°; DSCD: 3.6 × ± 0.06 10⁴³ molecules² cm⁻⁵; RMS: 7.8 × 10⁻⁴. (bottom panel).
Figure 3: DSCDs of O$_4$, over at 360 nm and 477 nm over Pune, Delhi and Mahabaleshwar. The data are color-coded according to elevation angles.
Figure 4: Timeseries of AOD along with the uncertainty at 360 nm and 477 nm over Pune, Delhi, and Mahabaleshwar.
Figure 5: Intercomparison of time-series of AOD at 360 nm and 477 nm retrieved from MAX-DOAS measurements with AERONET data over Pune.
Figure 6: Intercomparison of time-series of AOD at 360 nm retrieved from MAX-DOAS measurements with MODIS terra data over Pune, Delhi and Mahabaleshwar.
Figure 7: Intercomparison of time-series of AOD at 477 nm retrieved from MAX-DOAS measurements with MODIS terra data over Pune, Delhi and Mahabaleshwar.
Figure 8: Frequency distributions of delta AOD (UV and VIS) i.e. the difference between MODIS and MAPA-derived AOD over Pune, Delhi and Mahabaleshwar.
Figure 9. Observed and modelled DSCDs and retrieved vertical profiles for O₄ at UV (column A and B) and visible wavelength (column C and D) from Pune (top panel), Delhi (middle panel) and Mahabaleshwar (bottom panel) campaign.
Figure 10: Seasonal mean aerosol extinction profiles and standard deviations at 360 nm and 477 nm over Pune, Delhi and Mahabaleshwar. Note that the altitude scale (y-axis) is adjusted to visualize the profile shape. Aerosol extinction is always greater than equal to zero. The error bars ranging to negative values, result from high scatter for not normally distributed values.
Figure 11: Case study of 9 June 2019 from Delhi. Top panel: Timeseries [IST] of AOD, surface aerosol extinction (AEC), PM 2.5 and PM 10 from 8 June 2019 to 10 June 2019. Bottom panel: examples of vertical profile of AEC, the red profile was observed during the high pollution event and blue profile is representative of a typical AEC vertical profile.