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ORIGINAL PAPER

Spatiotemporal variability of optical properties of aerosols over the Indo-Gangetic Plain during 2011–2015

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Abstract: The present study reports daily, monthly, and seasonal variations of aerosol optical depth (AOD) using aerosol robotic network (AERONET), MICROTOPS-II Sunphotometer, and Moderate Resolution Imaging Spectroradiometer (MODIS) data over three most polluted cities such as Kanpur, Varanasi, and Jaipur in the Indo-Gangetic Plain during 2011–2015. The monthly mean variations of AOD and Angstrom Exponent were also presented and compared with the meteorological parameters for all three cities. The concentrations of AODs in Kanpur and Varanasi were observed higher than in Jaipur throughout the study period. Overall maximum correlation (R^2 = 0.74) was found in between AERONET and MODIS AOD in the winter season over Kanpur during 2011–2015, whereas the lowest correlation (R^2 = 0.41) was found in between MICROTOPS-II Sunphotometer and MODIS AOD over Varanasi for the same period in the pre-monsoon season. Spectral variations of AODs at five different wavelengths, i.e., 380, 440, 500, 675, and 870 nm, were derived using AERONET for Jaipur and Kanpur and using MICROTOPS-II Sunphotometer for Varanasi during 2011–2015 inferred clearly that increasing wavelength provides decreasing AODs. The Hybrid Single-Particle Lagrangian Integrated Trajectories model was used for the backward trajectories analysis to assess the sources of large aerosol loadings over Jaipur, Kanpur, and Varanasi cities during some typical days.

Keywords: AERONET; MODIS; MICROTOPS-II Sunphotometer; AOD; HYSPLIT

1. Introduction

Aerosols are fine solid or liquid particles suspended in the atmosphere which arises both from direct emission and from the gas-to-particle conversion of vapor precursors [1–3]. The interaction of aerosols with incoming solar radiation shapes the prime source of its direct effect on climate [4], whereas aerosol-cloud indirect effects are cooling and warming of the land surface, cloud formation, change in wintertime average rainfall patterns, evaporation, etc. [5]. Indo-Gangetic Plain (IGP) is surrounded by very rich sources for natural and anthropogenic aerosols which has a high potential impact on regional and global climate [6]. Natural sources of aerosols are wind-borne dust, sea spray, volcanoes, etc., whereas anthropogenic sources of aerosols are: biomass burning, transportation, burning of fossil fuels, etc. [4, 7]. In the last decades, rapid growth in population and urbanization has led to a significant rise in transportation and industrialization, resulting in a steep rise in pollution levels [8–10]. Because of high extinction values present over the Indo-Gangetic Plain (IGP), it firmly impacts the local atmosphere [11]. The climatic and environmental impacts of aerosols are serious issues worldwide. Atmospheric aerosols derived from a variety of natural and manmade emission sources are responsible to influence air quality, human health, and radiation budget [4, 12–15]. Moreover, aerosol concentrations adversely affect visibility [16], agricultural crop production [17], and biological systems [18].

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Aerosol optical depth (AOD) is one of the noteworthy optical properties for presenting aerosols, and it is used in the radiative transfer calculation. National Aeronautics and Space Administration (NASA) has an arrangement of ground-based aerosols observing system under the Aerosol Robotic Network (AERONET) Program, in which programmed sun/sky radiometers are positioned at different places far and wide. An AERONET observing system was begun to work at first for the routine measurements of aerosols in the northern part of India at Kanpur over IGP in the year 2001. A few studies have approved the Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol products with the ground-based AOD measurements acquired from AERONET and MICROTOPS-II Sunphotometers [19, 20]. An examination and assessment of MODIS aerosol products with AERONET information were also made over IGP at Kanpur [6, 21]. An acceptable agreement between AERONET and MODIS AOD has indicated that the interpretation of satellite datasets is suitable for aerosol monitoring. The aerosols and surface properties retrieval jointly by AERONET and satellite perceptions with surface reflectance was also analyzed by Sinyuk et al. [22]. Ground-based MICROTOP-II Sunphotometer AOD information was validated with the satellite-based level 3 MODIS information during the year 2011 [23]. Even though satellites are proved to be a good tool to comprehend the expansive spatiotemporal qualities of aerosols and related impacts from worldwide to neighborhood scales, they cannot give a top to bottom perspective on airborne particles on a nearby scale and pose higher vulnerabilities when contrasted with the ground-based instruments. MODIS onboard TERRA and AQUA satellites are acknowledged as the most extensively validated sensors for aerosol properties retrieval [24, 25]. Anyway, comprehension of the atmospheric aerosol's behavior over the IGP is a great challenge since various parameters oversee the aerosols' dynamics and impact on the climatic change [26].

The significant reason for air, water, and land pollution over IGP is the large-scale uncontrolled industrial and urban expansions. IGP is a complex geo-climatic region that shows a high assorted variety in aerosol loading and optical properties [27, 28]. The IGP is profoundly influenced by mineral dust moved from the northwestern deserts, for example the Thar Desert, the dry western locale of Pakistan, the Arabian Peninsula, and Afghanistan at the pre-monsoon months [29]. Conversely, during post-monsoon and winter, the whole northern part is influenced by smoke and haze and by emissions from agricultural crop residues burning and waste material [30]. It is difficult and challenging to retrieve satellite aerosol properties due to seasonal variations over IGP [25]. Therefore, there is a need for aerosol research over IGP for a long time period to study the climatology aspects.

In the present study, ground-based AERONET has been used to investigate the aerosol variability over a 5-year period (2011–2015) for two stations over IGP, namely Jaipur and Kanpur. Simultaneously, the MICROTOPS-II Sunphotometer AOD data were also collected over Varanasi situated in the middle part of IGP. Satellite-derived MODIS TERRA level 3 AOD products have been utilized for the examined stations and corresponded with ground observed AOD information. The correlation between in situ and satellite-based observations was made to understand the better relation between AODs during different seasons. However, Angstrom Exponent (AE) values were collected to know the size of the particles present over the different sites. Spectral variations of AODs at five different wavelengths such as 380, 440, 500, 675, and 870 nm were collected using AERONET for Jaipur and Kanpur and using MICROTOPS-II Sunphotometer for Varanasi during 2011–2015. Meteorological parameters were collected and discussed to know the contrast in aerosol characteristics in terms of air temperature and relative humidity (RH). The Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPLIT) model was used for the backward trajectories analysis to assess the sources of large aerosol loadings over Jaipur, Kanpur, and Varanasi cities during some typical high aerosol loading days.

2. Study sites

Three major polluted cities Kanpur, Varanasi, and Jaipur situated over IGP in northern India are selected as study regions. Jaipur city faces serious dust storms and sandstorms in addition to other polluting atmospheric conditions during the pre-monsoon (March–June) season. Varanasi is located over the middle IGP and experiences a humid subtropical climate with extreme summer, intense rainfall during monsoon, and cold weather during the winter season [2, 27]. Kanpur is one of the highly polluted megacities in Asia, situated in the central part of the IGP. IGP is also known as the North Indian River plain which is an enormous and rich plain incorporating the majority of northern and eastern India. Practically 20% of the land territory and around 40% of food production of India are secured by IGP [23]. IGP is delimited by the Brahmaputra ridge in the east, the Aravalli mountains, the Thar Desert and the Arabian Sea in the west, the Himalaya in the north, and the Vindhyas and the Satpura extend in the south. The plain's population density is very high due to fertile soil for farming as well as the presence of numerous small and large rivers. This region has traditionally been responsible for the food security of India. The food security of India and other countries in South Asia is, however, now at a risk due to the continuously increasing population. The IGP is unequally found to be affected by high AOD values throughout the year [6, 31]. The dry dust particles conveyed via air during pre-monsoon (March–June) months over the sites from the Thar Desert are delivering continuous dust storms and make a dry climate, while the winds are blowing from the eastern part of the Ganga river basin conveys moisture during monsoon (July–August) months. During post-monsoon (September–October) and winter (November–February) seasons, the entire area is dominated by aerosols originated from anthropogenic sources stacked by local and northerly winds. The western disturbances during the winter season load the region with extreme fog and haze [32].

3. Experimental details and methodology

3.1. In situ measurements

Aerosol optical properties have shown a high spatial as well as a temporal variation of aerosols over IGP. In the present study, aerosols optical properties have been studied over Jaipur (26.90° N, 75.90° E), Kanpur (26.40° N, 80.40° E), and Varanasi (25.27° N, 82.99° E) over the IGP region. The ground-based observations by CIMEL sun/sky radiometer are conveyed at Jaipur and Kanpur over the IGP region under the AERONET program of NASA, USA. AERONET is a ground established remote sensing framework connected with well-calibrated Sunphotometers and radiometers [19, 33]. AERONET provides constant cloud-screened perceptions of spectral aerosol AOD, perceptible water, and inversion aerosol products in assorted aerosol regimes. Smirnov et al. [34] developed a procedure of an inversion computation for the aerosol properties. Sky radiance makes available direct, calibrated measurements of spectral AOD (normally at the wavelength of 440, 670, 870, and 1020 nm) with an accuracy of $\pm$ 0.015. Spectral measurements of sun and sky radiance are calibrated and screened for cloud-free conditions [35]. Optical properties of aerosol particles over Jaipur and Kanpur have been studied using level 3 AERONET daily data. AOD of the desired period for Varanasi station has been procured using the MICROTOPS-II Sunphotometer instrument. MICROTOPS-II is a valuable instrument for measuring column ozone, column water vapor, and AOD for climatological purposes and unique research studies in a wide scope of areas.

3.1.1. AERONET measurements

AERONET is a worldwide ground-based Sunphotometer network that gives aerosol optical properties by consistent calibration, cloud-screening, and retrieval strategies for all stations. AERONET AOD (https://aeronet.gsfc.nasa.gov/) data from 2011 to 2015 were taken and analyzed for Jaipur and Kanpur stations in the present study. AERONET provides AOD at several wavelengths between 340 and 1640 nm with $\sim \pm 0.01$ accuracy at wavelengths > 440 nm and $\sim \pm 0.02$ at shorter wavelengths [19]. AERONET aerosol products, like AE based on the 440 nm and the 870 nm wavelength, were used to determine the dominant aerosol type [34]. This information constitutes a high-quality, ground-based aerosol climatology, has been generally utilized for aerosol assessment and validation of model simulation and remote sensing applications [36, 37]. The Indian subcontinent is one of the highest populated, industrialized, and developing regions where aerosol not only affects the climate and environment but also human health and society.

3.1.2. MICROTOPS-II Sunphotometer measurements

MICROTOPS-II Sunphotometer instrument is produced by the Solar Light Company, Philadelphia, USA, which works on the principle of extension of solar radiation intensity at a definite wavelength. This instrument has minimal expense and its calibration and performance were introduced by Morys et al. [38]. MICROTOPS-II Sunphotometer instrument is manufactured for the assessment of AOD concentrations, and it is very easy to carry/operate [39]. Sunphotometer measures sun-powered radiance in five spectral wavebands from which it automatically derives AOD. This is a five-channel (380, 440, 500, 675, and 870 nm), handheld Sunphotometer that can be arranged to gauge all-out ozone, all-out water vapor, or aerosol optical thickness at different wavelengths. Likewise, at 940 nm wavelength, a channel is utilized for perceptible water vapor substances present over the region. The MICRTOPS-II Sunphotometer was used for the AOD sampling at each clear sky day by taking proper care throughout the whole years of interest. Three values simultaneously were taken using MICROTOPS-II Sunphotometer measurements during every observation to minimize the errors. The smaller values are utilized in analysis, and higher values are disposed of because smaller values relate to the most exact sun pointing [40]. The proper notebook was prepared to record the cloud condition and sunshine at every measurement.

3.2. Satellite measurements

MODIS is a facility instrument provided by NASA and managed by NASA's Goddard Space Flight Centre in Greenbelt, Maryland. It was built by Hughes Corporation's Santa Barbara Remote Sensing in Santa Barbara, California. MODIS performs close to worldwide day-by-day observations of aerosols. Satellite inferred column AOD and also it is a cost-effective approach to monitor and study aerosols distribution and impacts over an extensive period of time. MODIS inferred AOD is appropriate for such type of study because of its revisit cycle of 1–2 days. MODIS sensor has a wide spectral range, high spatial resolution, and near-daily global coverage to observe and monitor the Earth and subsequent changes. Seven of 36 channels (0.47–2.13 μm) are utilized to retrieve aerosol properties over cloud and surface screened regions. Over vegetated land, MODIS recovers AOD at three visible channels with high precision of $\pm$ 0.05 [36, 41]. Most recently, a deep blue algorithm has been implemented to retrieve aerosols over bright deserts on an operational basis, with an estimated accuracy of 20–30%. Due to the more noteworthy simplicity of the ocean surface, MODIS has the one of a kind ability to retrieve not only AOD with great accuracy, yet also quantitative aerosol size parameters [36]. MODIS onboard TERRA satellite level 3 AOD (550 nm) and Angstrom Exponent (AE) data for $1^\circ \times 1^\circ$ grid (http://giovanni.gsfc.nasa.gov/giovanni/) were carried out during 2011–2015 for aerosol study and its correlation with ground observed data.

Meteorological parameters such as temperature and RH were included to know the impact of these parameters on the characteristics of the aerosol (AOD and AE). The time series monthly surface air temperature product (mean_M2IMNXLFO_5_12_4_TLML) by MERRA-2 model at 0.5×0.625 resolutions was collected for further analysis. The downloaded temperature data were obtained in K and converted to °C for the proper association with RH and optical properties (AOD and AE). The monthly mean RH at surface product from Atmospheric Infrared Sounder (AIRS) at daytime/ascending (mean_AIRS3STM_006_RelHumSurf_A) and nighttime/descending (mean_AIRS3STM_006_RelHumSurf_D) were collected (http://giovanni.gsfc.nasa.gov/giovanni/). The average values of RH were used for further analysis with temperature and optical properties.

4. Results and discussion

The aerosol loadings in Kanpur, Jaipur, and Varanasi regions are found to increase considerably in the recent past. Kanpur is one of the main industrial areas of Uttar Pradesh having major sources of aerosols accumulation over the region. However, Jaipur is largely influenced by coarse particles such as dust and sea salt in the pre-monsoon months coming from the Thar Desert and the Arabia Peninsula. Anthropogenic activities (fuel/coal combustion, vehicular dust, and biomass/wood burning) over Varanasi are the major sources of aerosols [42], even though the aerosols and the precursor gases in the region have a threat to human health. The study regions (Jaipur, Kanpur, and Varanasi) over IGP with sufficient industrial activity and urbanization coupled with transportation and biomass/wood burning have a continuous layer of air containing enhanced concentrations of aerosol particles.

Figure 1 shows seasonal variations of mean AOD for the period 2011–2015 derived from MODIS TERRA over Indian regions. It clearly shows from the figure that aerosol loading is the highest over the IGP region compared to other parts. It can also be seen from the ranges of AOD which are shown by different colors of the AOD loadings over Kanpur and Varanasi are higher as compared to Jaipur. The maximum AOD was indicated by a purple color while the red color indicates the lowest AOD. Post-monsoon, winter, and pre-monsoon seasons are showing higher AOD concentrations compared to monsoon season. In monsoon season, concentrations of AOD are very less due to washing out by the rainfall. The urban and industrial activities as well as bio-fuel burning are the major sources of anthropogenic aerosols over these cities. The concentrations of industrial and urban pollution are maximum in the Northern Hemisphere, where most of the planet's population and therefore urban and industrial activities are located. During pre-monsoon months due to hot days, humid, and stagnant summer days, the air quality over urban areas often reaches unhealthy levels, and aerosols are primary sources [43].

4.1. Daily variation of AOD over IGP during 2011–2015

Figure 2 represents the daily variations of AERONET, MODIS AOD_{550} over (a) Jaipur, (b) Kanpur, and MICROTOPS-II, MODIS AOD_{550} over (c) Varanasi during 2011–2015. The results reveal a low-to-medium variation in AERONET and MODIS AOD for Jaipur (range of 0.07–1.81, mean: 0.42; range of 0.05–1.96, mean: 0.26, respectively), Kanpur (range of 0.08–2.13, mean: 0.68; range of 0.06–2.26, mean: 0.64, respectively), and MICROTOPS-II and MODIS AOD in Varanasi (range of 0.18–2.14, mean: 0.75; range of 0.06–2.32, mean 0.66, respectively). Nearly 59% and 64% of the days AERONET and MODIS AOD values are found below the yearly mean value line (solid black and red), respectively, over Jaipur. The dotted lines are representing the standard deviations of

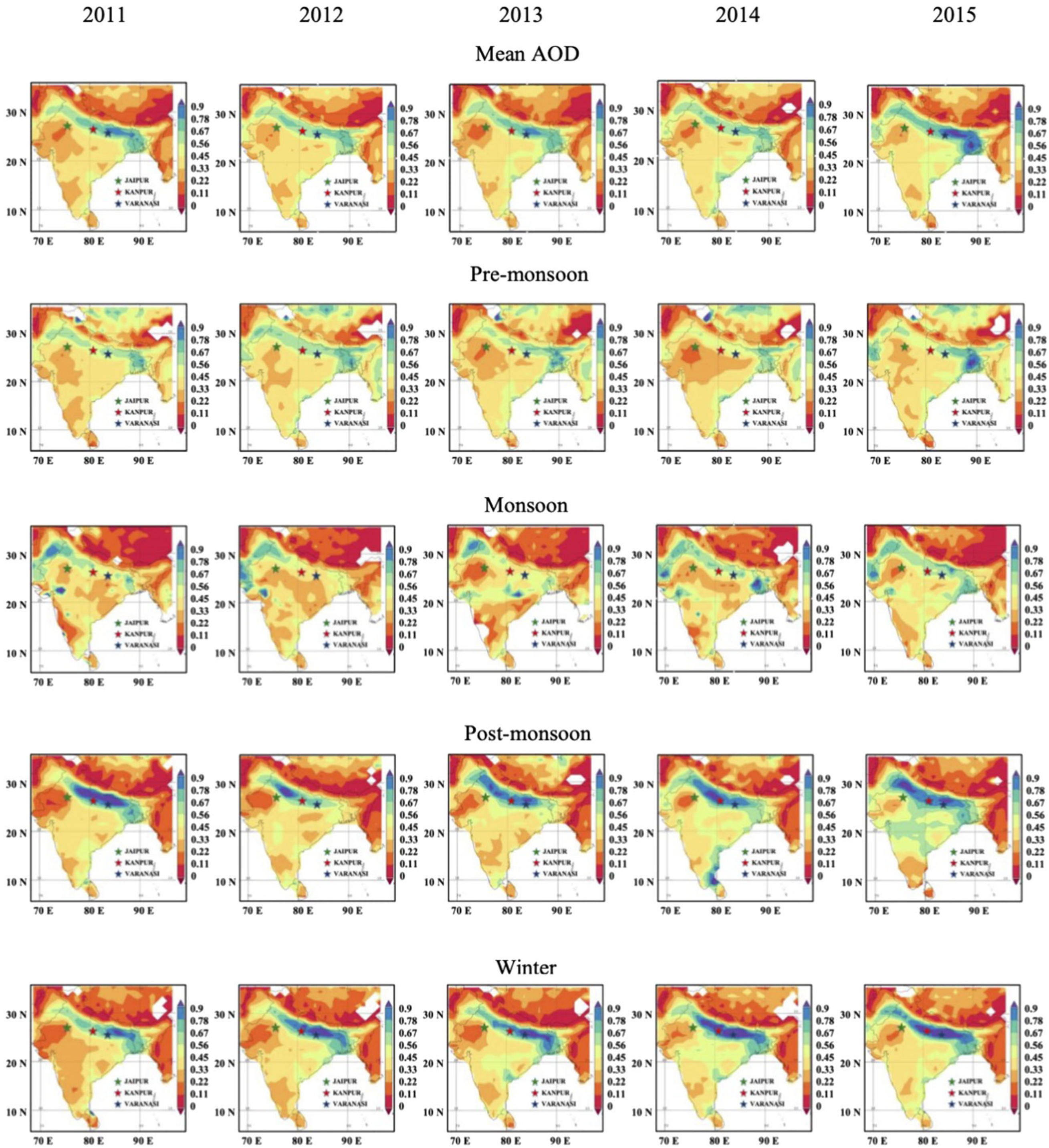

Fig. 1 Spatio-seasonal variations of mean MODIS AOD_{550} over Indian regions for the periods 2011–2015

AODs from the yearly mean value line observed by AERONET and MODIS. AODs > 1.25 are observed nearly 1% of the days, for both AERONET and MODIS AOD (Fig. 2a). During pre-monsoon months, the higher loading of AOD was indicated in May and June month observed by AERONET and MODIS over Jaipur. High AOD over IGP during pre-monsoon months can be attributed to dust-storm events and consequent aerosol loading from far source regions, Arabia peninsula [29]. The low-pressure system resulted in the enhancement of moisture content in the boundary layer, which is afterward replaced by a high-pressure system leading to clear sky conditions and weak wind flow during winter and post-monsoon. The temperature also attains its minimum value

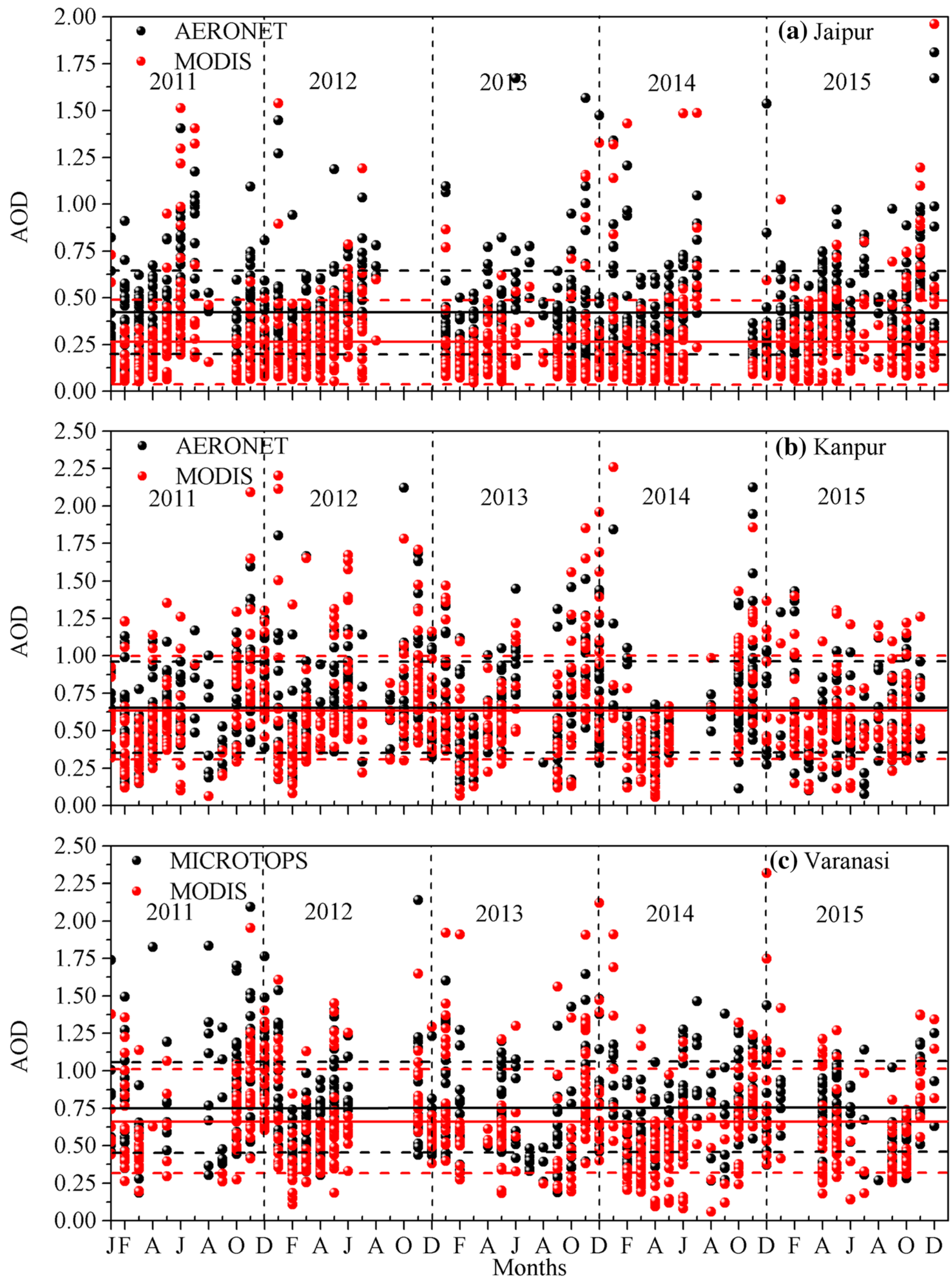

Fig. 2 Daily variations of AERONET, MODIS AOD_{550} over **a** Jaipur, **b** Kanpur, and **c** MICROTOPS-II, MODIS AOD_{550} over Varanasi during 2011–2015

and making conditions ideal for the accumulation of pollutants within the boundary layer and often results in fog formation over IGP [44, 45].

Kanpur is one of the highest aerosol loading regions over IGP, due to the excessive anthropogenic emissions. The aerosol loading over IGP in Kanpur has been found continuously increasing [6]. As we know Kanpur is one of North India's main commercial and industrial centers, it is famous for leather and textile goods which are major sources of aerosols. In Kanpur, AERONET and MODIS AOD yearly mean values are more or less the same (AERONET AOD mean: 0.65 and MODIS AOD mean: 0.64) as depicted in the figure with solid black and red line. Nearly 56% and 54% of the days AODs are below the yearly mean line for AERONET and MODIS AOD, respectively, in the study period. Only 4–6% of the days AODs > 1.25 are observed over Kanpur (Fig. 2b). In comparison with Jaipur, the very high concentrations of AOD are indicating the higher aerosols loading over Kanpur. However, the results of aerosols loading over Varanasi were found comparable to Kanpur and higher than Jaipur. The aerosols loading was found very high in January, May, June, October, November, and December indicated by higher concentrations of AOD. However, the

AOD values during the February, March, and April months of pre-monsoon were also found higher than Jaipur pre-monsoon months. This region also experiences high levels of biomass and fossil fuel burning [46].

In the region of Varanasi, nearly 56% of the days MICROTOPS-II and MODIS AOD values are below the yearly mean value line. Approximately 6–7% of the days AODs > 1.25 were observed for both types of datasets (Fig. 2c). For Varanasi, the AOD loading is enhanced during pre-monsoon days, post-monsoon days, and winter days, while a decrease is seen in the monsoon days. A possible explanation for this could be the recent changes in land use and land cover during monsoon days. A huge amount of AOD and large fluctuation in the depth of AODs were observed during the years 2011–2014. Very low concentrations of AOD during monsoon days indicate about some fractions of aerosols are washed away with precipitation during the monsoon days, thus lowering the AOD during this period [23]. Varanasi is credited to substantial traffic and vehicular exhausts, industrial emissions, coal and biomass burning, and constructional works.

4.2. Seasonal correlation between AERONET/MICROTOPS-II and MODIS AOD over the IGP

For intercomparison of ground-based AOD data with satellite data, the seasonal correlation was done for all three stations of Jaipur, Kanpur, and Varanasi. Figure 3 shows the correlation analysis of MODIS AOD_{550} and AERONET/MICROTOPS-II AOD_{550} at ground monitoring stations Jaipur, Kanpur, and Varanasi during the (a) pre-monsoon, (b) monsoon, (c) post-monsoon, and (d) winter seasons. Seasonal correlation is a single value that helps us to interpret the degree of relation between two variables. For Jaipur and Kanpur, daily AERONET AOD_{550} data has been plotted against average level 3 MODIS AOD_{550} data during 2011–2015. However, daily MICROTOPS-II Sunphotometer AOD_{550} data have been plotted against daily MODIS AOD_{550} data over Varanasi. The number of data points depends on the commonly available data for the same wavelengths of satellite and ground observation [1].

The pre-monsoon season correlation between AERONET/MICROTOPS-II and MODIS AOD was shown for Jaipur, Kanpur, and Varanasi during 2011–2015 (Fig. 3a). Pre-monsoon season has shown a good correlation (R^2 = 0.57) between AERONET and MODIS AOD over Kanpur. While overall poor correlation (R^2 = 0.41) was observed in the pre-monsoon season between MICROTOPS-II and MODIS AOD over Varanasi was lowest in comparison with the monsoon, post-monsoon, and winter seasons AOD. This means there is a poor level of agreement between ground-based and satellite-based AODs for the pre-monsoon season. It may be due to the large difference in the size of the particles present over the site during pre-monsoon season.

Figure 3(b) shows the monsoon season correlation between AERONET/MICROTOPS-II and MODIS AOD for Jaipur, Kanpur, and Varanasi during 2011–2015. Overall good correlation (R^2 = 0.62) between MICROTOPS-II and MODIS AOD was observed over Varanasi during the monsoon period. However, comparable weak correlations between AERONET and MODIS AODs were observed for Jaipur (R^2 = 0.48) and Kanpur (R^2 = 0.46) in the monsoon season. The several changes in land use and land cover during monsoon season may be the cause of the poor correlation between in situ and satellite measured AODs over the regions. Some fractions of aerosols are washed-out with precipitation, thus lowering the AOD during this period over the region.

Figure 3(c) shows the post-monsoon season correlation between AERONET/MICROTOPS-II and MODIS AOD for Jaipur, Kanpur, and Varanasi during 2011–2015. The correlation between AERONET and MODIS AOD over Jaipur (R^2 = 0.62) and Kanpur (R^2 = 0.61) during the post-monsoon season was found comparable. However, R^2 = 0.54 was found between MICROTOPS-II and MODIS AOD over Varanasi. The winter season correlation between AERONET/MICROTOPS-II and MODIS AOD for Jaipur, Kanpur, and Varanasi is shown during 2011–2015 (Fig. 3d). The overall best correlation between AERONET and MODIS AOD over Kanpur (R^2 = 0.74) was indicating the presence of similar size of particles observed between in situ and satellite measurements over the site. Thus, we can also say there is the least inconsistency between ground and satellite data for this season. A fairly good correlation between in situ and satellite observations was found over Jaipur (R^2 = 0.68) and Varanasi (R^2 = 0.62) in the winter season. A good correlation helps us to validate satellite data. Prasad and Singh [6] have demonstrated a correlation of 0.47 throughout the winter season and a correlation of 0.29 during the pre-monsoon season in the period of 2000–2005. Similarly, for the period 2005–2009, Kumar et al. [1] have shown an AERONET-MODIS AOD correlation of 0.68 for the winter season and 0.49 for the pre-monsoon season. A better correlation between AERONET and MODIS AOD was observed over Kanpur (R^2 = 0.74) during the winter season in the period 2011–2015 in the present study. During pre-monsoon, the correlations were observed over Jaipur (R^2 = 0.51) and Kanpur (R^2 = 0.57) also indicates better results than the previously explained studies.

The overall correlations between in situ and satellite measurements during the winter season were highest among all seasons. This means there is a good level of agreement between ground-based and satellite-based AOD measurements for the winter season. During pre-monsoon

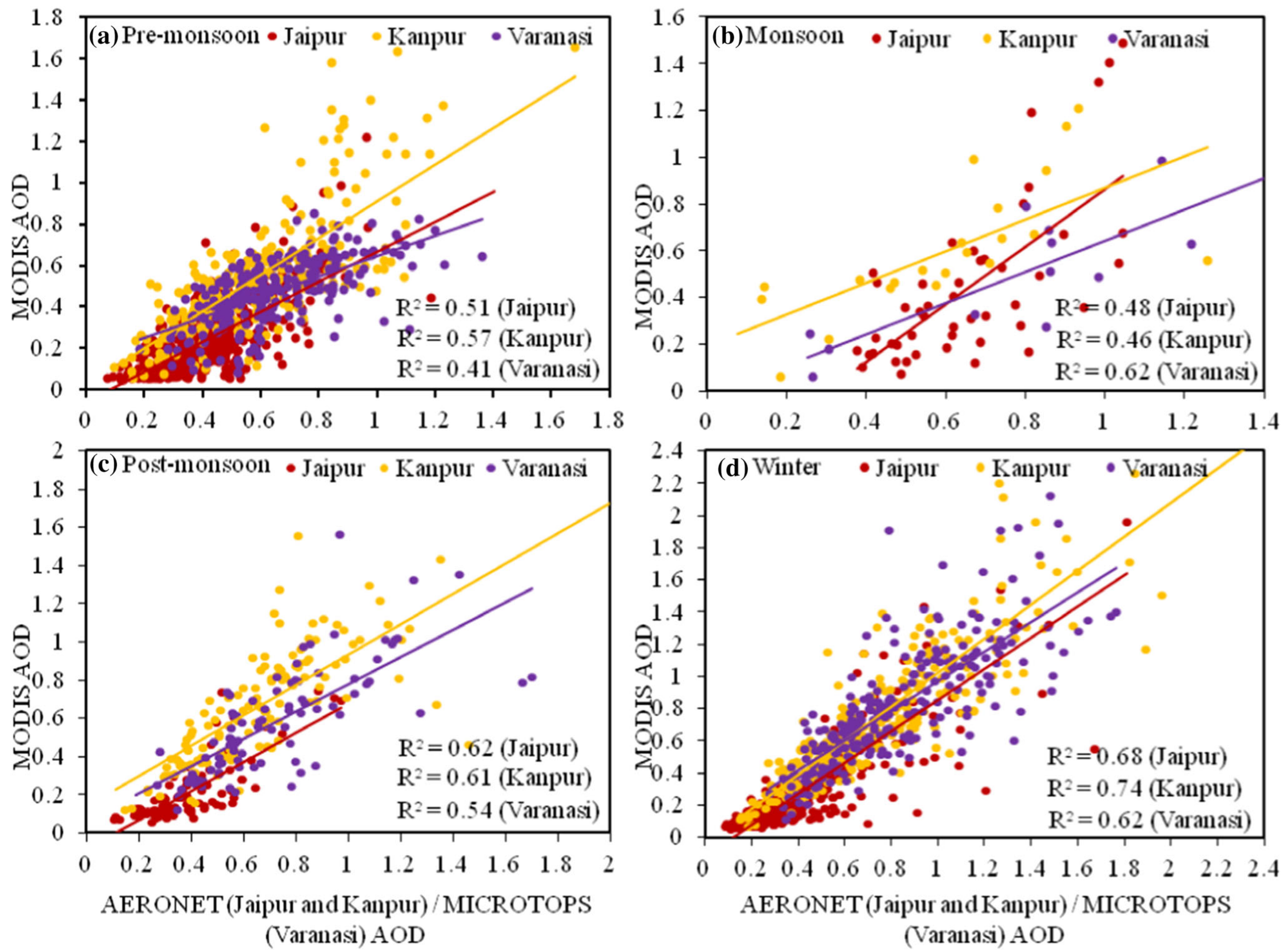

Fig. 3 Correlation of MODIS AOD_{550} and AERONET/MICROTOPS-II AOD_{550} at ground monitoring stations Jaipur, Kanpur and Varanasi during the **a** pre-monsoon, **b** monsoon, **c** post-monsoon, and **d** winter seasons

season, the poor correlation over Varanasi ($R^2 = 0.41$) between MICROTOPS-II and MODIS AOD demonstrates dissimilarity between the particles present over the site. Varanasi is influenced annually by long-range transported mineral dust from the western dry and desert regions of Africa, Arabia, and Thar Desert of Rajasthan at the pre-monsoon time. However, moderate to dense fog during the winters are major sources of pollution. Though the region is largely agriculture-based, high anthropogenic aerosol loading due to industry, biomass burning, and thermal power plants exists [47–49]. Aerosols can also threaten the safety of aviation by reducing visibility by almost one-third of the original. These aerosols are highly responsible for the change of time and location of traditional rainfall patterns.

4.3. Spectral variation of AOD at different wavelengths

The spectral dependence of AODs at ultraviolet–visible wavelengths specifies the significance of the fine-mode aerosols in the scattering processes mainly during the post-monsoon and winter seasons [50]. Figure 4 shows the spectral variation of AOD at five different wavelengths 380, 440, 500, 675, and 870 nm derived from in situ measurements using AERONET for Jaipur and Kanpur and MICROTOPS-II Sunphotometer for Varanasi during 2011–2015. It can be inferred clearly from the graphs, on increasing wavelengths, AOD values were decreased. The irradiance scattering at lower wavelengths is enhanced by the fine-mode particles related to higher concentrations. So along these lines, the AOD values are recorded higher at lower wavelengths. The similar commitments of AOD at relatively larger wavelengths are seen at coarse-mode

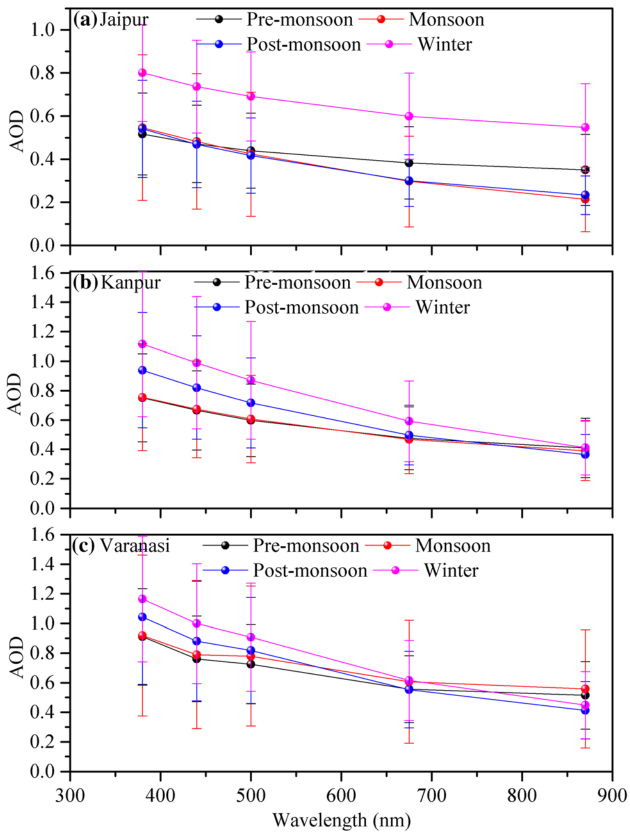

Fig. 4 Variation of the seasonal spectral means of AOD over **a** Jaipur, **b** Kanpur, and **c** Varanasi during 2011–2015

particles accessible to the locales [51, 52]. Desert dust and sea salt particles are the primary examples of mineral-based aerosols. They absorb certain wavelengths of sunlight and reflect others. Figure 4 clearly reflects the spectral variation of AODs at shorter wavelengths (380, 440, 500 nm) is different than longer wavelengths (675 and 870 nm) at Jaipur, Kanpur, and Varanasi.

It is noticed that magnitude of seasonal AODs is higher for Kanpur (0.36–1.12) and Varanasi (0.41–1.16), as compared to Jaipur (0.21–0.80) for the spectral wavelengths 380, 440, 500, 675, and 870 nm. This shows the strength of coarse-mode aerosols because of the condensation growth and coagulation mechanism of submicron aerosols, which are progressively proficient in producing larger aerosols. Larger aerosols are helpful in cloud nucleation. A large spectral variation in AOD was observed in each season with larger AOD values at shorter wavelengths and smaller values at longer wavelengths. The spectral variability at different short wavelengths (380, 440, 500 nm) and long wavelengths (675 and 870 nm) and the gradient among them propose asymmetric seasonal variations of AODs. It implies that the impact of seasonal changes in the dominance of fine or coarse-mode aerosols. It was observed that at shorter wavelengths, the variation in AOD values was more intense compared to the AODs at longer wavelengths. It might be because of more variation in light scattering due to changes in fine-mode aerosols at shorter wavelengths [51].

4.4. Monthly variations of optical aerosol properties and its association with meteorological parameters

Figure 5 shows the monthly variation of AOD, AE, temperature (°C), and RH (%) over Jaipur, Kanpur, and Varanasi during 2011–2015. The monthly variation of AOD with maxima during pre-monsoon (May–June), post-monsoon (October), and winter (November–January) was shown during 2011–2015 (Fig. 5a). This is a very similar pattern to those found over Kanpur [26] and Varanasi [27]. The maximum AOD during pre-monsoon months is attributed to the influence from dust aerosols approaching from the western regions [23], while high AOD in post-monsoon/winter months is associated with crop residue and biomass burning, especially for years with extreme biomass smoke emissions [7]. The AE values were higher in January while these AE values were found decreasing from February till June month, and then from July, these values were found increasing till December month (Fig. 5b). The maximum observed monthly mean AE values were found over Jaipur (November 2013; 1.38), Kanpur (August 2013; 1.45), and Varanasi (November 2013; 1.30). The minimum monthly mean AE values are observed in the pre-monsoon (February–June) months, which suggest the dominance of coarse-mode aerosols with a concurrent mixture of urban emissions, while maximum AOD values in winter months associated with maximum AE values are indicating the dominance of the fine smoke aerosols over the site [32]. The figure represents that AOD and AE have anti-correlation to each other, i.e., lower AOD has higher AE and vice versa. It can be seen that AOD increases in pre-monsoon months for all locations. This may be due to the hygroscopic growth of particles because in June, due to the beginning of monsoon season, air masses coming from the Bay of Bengal contain high water vapor content, which increases the size of aerosol particles. However, AE shows the opposite trend. Eck et al. [53] have reported that higher AE values suggest the dominance of small-sized particles and vice versa for coarse-sized particles.

In this respect, the monthly mean temperature has shown an almost overlapping pattern during 2011–2015 over Jaipur, Kanpur, and Varanasi (Fig. 5d). The minimum temperature occurs in January and increases continuously until May or June months and then slightly decreases during monsoon and post-monsoon reaching a minimum in winter months. During the study period of 2011–2015, the minimum monthly mean temperature of 14.87 °C (January 2013) and RH of 25.79% (May 2013) while the maximum

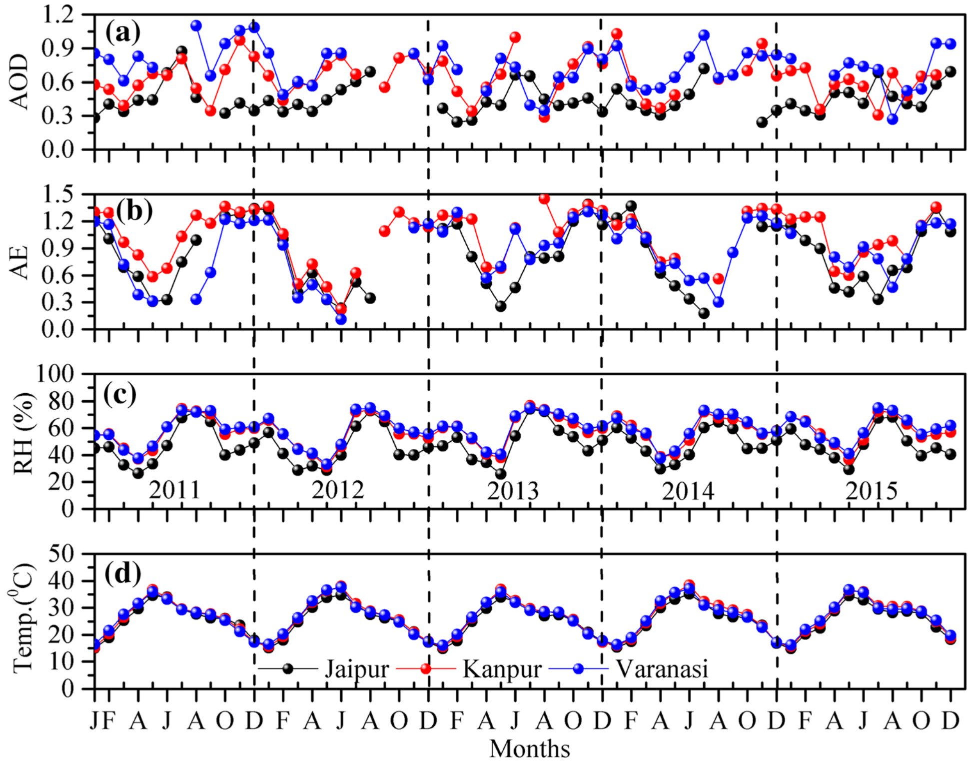

Fig. 5 Variation of the monthly mean of **a** AOD, **b** AE, **c** RH (%), and **d** temperature (°C) over Jaipur, Kanpur, and Varanasi during 2011–2015

monthly mean temperature of 35.07 °C (June 2014) and RH of 74.25% (July 2013) were observed at Jaipur. However, in Kanpur the minimum monthly mean temperature of 15.32 °C (January 2011) and RH of 31.31% (May 2012) while the maximum monthly mean temperature of 38.42 °C (June 2014) and RH of 76.56% (July 2013) were observed during 2011–2015. In Varanasi, the minimum monthly mean temperature of 16.01 °C (January 2013) and RH of 33.22% (May 2012) while the maximum monthly mean temperature of 37.54 °C (June 2012) and RH of 74.94% (July 2013) were observed during 2011–2015. It is analyzed from the study that temperature and RH have shown always inverse relation during 2011–2015. The monthly mean AE and temperature have shown an almost inverse relationship, while monthly mean RH has shown a direct relationship with monthly mean AE values.

4.5. Backward trajectories analysis using HYSPLIT Model

The air mass backward trajectories are very essential to recognize the origin point of aerosol sources and transport pathways reach up to observation sites. These backward trajectories show the different long-range transport pathways in terms of altitude and distance. The back trajectories during pre-monsoon, monsoon, post-monsoon, and winter seasons were analyzed over Jaipur, Kanpur, and Varanasi based on National Oceanic and Atmospheric Administration HYSPLIT model (https://www.ready.noaa.gov/HYSPLIT.php) [54]. The change in seasonal wind patterns has the potential to carry out various types of aerosols from different regions to the observation sites. For instance, sea salt from marine, dust from Desert, and anthropogenic aerosols originated from biomass/urban–industrial sources.

The plots of 5 days air mass back trajectories during (a) pre-monsoon, (b) monsoon, (c) post-monsoon, and (d) winter seasons at some higher aerosol loading days of 2013 over Jaipur, Kanpur, and Varanasi are shown in Fig. 6. The air mass seems to be transported from far source regions like the Arabia peninsula, the Sahara Desert, and the Thar Desert during pre-monsoon season over Jaipur, Kanpur, and Varanasi (Fig. 6a). The almost similar sources of aerosols were reported over IGP by Gautam et al. [29]. Tiwari et al. [55] also reported the air masses dominated over Jaipur are polluted dust particles mainly originated from the Middle East and the Thar Desert regions. Jaipur is largely affected by coarser particles such as dust and sea salt, which is increasingly articulated with longer pathways and huge latitudinal variability as pre-monsoon months advance [31].

Kanpur was found mostly dominated by organic carbon enriched type of aerosols generated from the local sources such as fossil fuels and industrial area [55]. Critical urban zones produce huge concentrations of aerosols because of industrial activity, vehicular emissions, etc. Anyway,

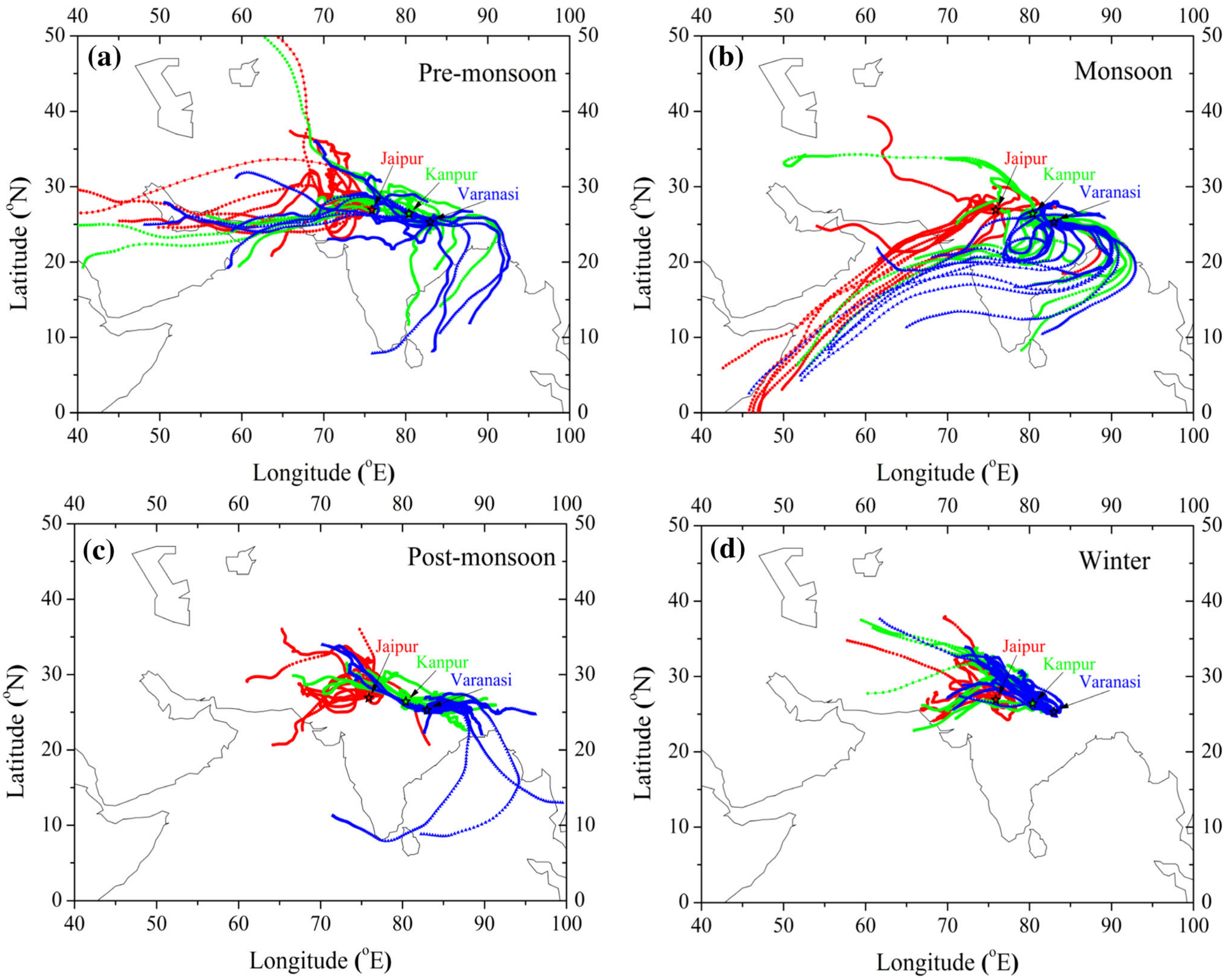

Fig. 6 Air mass back trajectories during **a** pre-monsoon, **b** monsoon, **c** post-monsoon, and **d** winter seasons at some higher aerosol loading days of year 2013

industrial and urban activities are the greatest source of anthropogenic aerosols. The concentration of industrial and urban pollution is greatest in the northern hemisphere, where most of the planet's population and therefore urban and industrial activity is located. In an earlier study over the IGP, Srivastava et al. [56] have revealed that air masses during the pre-monsoon period are mixed with the air masses that previously influenced Kanpur, which is additionally the situation seen in the current study during May and June. Varanasi is generally influenced by the long-range transported aerosols, with moderately enormous latitudinal variability. Some of the fires are usually produced for agricultural purposes to clear natural vegetation away for cropland and to maintain native plant communities in natural areas. The long-range transported dust plumes are from Arabian countries and southwest Asia, while some of the aerosols are arising from the Bay of Bengal as Sea salt over Varanasi. Tiwari et al. [57] have also shown that long-range transported dust plumes from Arabia, the Middle East, and southwest Asia were favored during the pre-monsoon season.

Sea spray is the primary source of aerosols which is mostly composed of sodium chloride (salt) during monsoon season. While during monsoon season marine air masses carry mostly sea salt from the Arabian Sea and the Bay of Bengal, which was accumulated over Jaipur, Kanpur, and Varanasi (Fig. 6b). During the post-monsoon season, mostly aerosols over Jaipur, Kanpur, and Varanasi are accumulated from the transported aerosols due to biomass burning from Punjab, Haryana, and Pakistan (Fig. 6c), while some of these aerosols are originated from Rajasthan and anthropogenic activities over Jaipur. Over Kanpur, some aerosols are arising from the Eastern part of India. Varanasi was also affected by some of the aerosols arising from the Bay of Bengal as Sea salt. Kaskaoutis et al. [7] have observed that Varanasi was mostly influenced by the aerosols originated from northwestern India because significant paddy crop residue burning occurs during post-monsoon season. During the winter season, the aerosols are accumulated mainly from the local/anthropogenic activities in Jaipur, Kanpur, and Varanasi (Fig. 6d). However, some of the aerosols are transported from Iran, Pakistan, and Afghanistan countries over Jaipur and Kanpur. During the winter season over Varanasi, vehicular dust and biomass/wood burning by local peoples are also the major concerns of aerosols. During the winter season, most pollutants were accumulated from anthropogenic activities [58].

5. Conclusions

The IGP has shown great interest in research because of its unique topography and also region is highly dominated by urban and industrial pollutants. The present atmospheric pollutants over IGP demonstrate significant daily, monthly, and seasonal variability of anthropogenic factors mixed with natural sources (mostly dust). In the present study, the variability of atmospheric aerosols has been studied over three locations (Jaipur, Kanpur, and Varanasi) over IGP during 2011–2015 using ground-based (AERONET/MICROTOPS-II Sunphotometer) as well as satellite MODIS TERRA measurements. The following outputs were concluded from the current research as:

- Daily variations indicate the enhancement of AOD loading during pre-monsoon, post-monsoon, and winter days, while lower AOD values were observed during monsoon days over all three sites during 2011–2015.
- Monthly variation trends show that AOD and AE have anti-correlation to each other to some extent, i.e., lower AOD has higher AE and vice versa. Anti-correlation between temperature and RH was found always over Jaipur, Kanpur, and Varanasi during 2011–2015.
- Seasonal fractions of AOD over sites show maximum AODs during winter and post-monsoon seasons. This is due to increased biomass burning and other anthropogenic activities. However, transported biomass burning from Pakistan, Haryana, and Punjab is the major cause of pollution.
- The correlation analysis for all three stations over IGP showed a good correlation between in situ and satellite measured AOD. Winter season showed the best correlations for all three stations, having R^2 values of 0.68, 0.74, and 0.62 for Jaipur, Kanpur, and Varanasi, respectively.
- Wavelengths are seen to be inversely related to AOD values. Kanpur and Varanasi are seen to have higher AODs as compared to Jaipur for the same wavelength.
- During pre-monsoon season, Middle East countries including the Sahara Desert, Arabia Peninsula, and the Thar Desert in India have been seen mostly to be significant sources of transported mineral dust and pollutants over IGP. During post-monsoon season biomass burning from Punjab, Haryana, Pakistan while local/anthropogenic activities during the winter season are the major sources of the aerosols over the sites. Marine sources from the Arabian Sea and the Bay of Bengal are the sources of pollutants during monsoon season.

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