

# Quantification of aerosol type, and sources of aerosols over the Indo-Gangetic Plain



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## HIGHLIGHTS

- AODs exhibit prominent seasonal variation over Kanpur and Gandhi College in IGP.
- Coarse mode aerosol concentration is always higher over Kanpur than Gandhi College.
- Fine mode aerosols dominate IGP during winter and postmonsoon.
- Absorbing aerosols over IGP are Mostly BC, and Mixed BC and Dust.
- Enhanced absorption due to OC aerosols may be negligible over IGP.

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## ABSTRACT

Differences and similarities in aerosol characteristics, for the first time, over two environmentally distinct locations in Indo-Gangetic plain (IGP) – Kanpur (KPR) (urban location) and Gandhi College (GC) (rural site) are examined. Aerosol optical depths (AODs) exhibit pronounced seasonal variability with higher values during winter and premonsoon. Aerosol fine mode fraction (FMF) and Ångström exponent ( $\alpha$ ) are higher over GC than KPR indicating relatively higher fine mode aerosol concentration over GC. Higher FMF over GC is attributed to local biomass burning activities. Analysis of AOD spectra revealed that aerosol size distribution is dominated by wide range of fine mode fractions or mixture of modes during winter and postmonsoon, while during premonsoon and monsoon coarse mode aerosols are more abundant. Single scattering albedo (SSA) is lower over GC than KPR. SSA spectra reveals the abundance of fine mode (coarse mode) absorbing (scattering) aerosols during winter and postmonsoon (premonsoon and monsoon). Spectral SSA features reveal that OC contribution to enhanced absorption is negligible. Analysis shows that absorbing aerosols can be classified as Mostly Black Carbon (BC), and Mixed BC and Dust over IGP. Mixed BC and dust is always higher over KPR, while Mostly BC is higher over GC throughout the year. The amount of long range transported dust exhibits a gradient between KPR (higher) and GC (lower). Results on seasonally varying aerosol types, and absorbing aerosol types and their gradients over an aerosol hotspot are important to tune models and to reduce the uncertainty in radiative and climate impact of aerosols.

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## 1. Introduction

Indo-Gangetic plain (IGP) is one of the largest river basins in the world. It is densely populated (population density of 500–1000 or more per sq. km., based on 2001 census) and is one of the most

highly polluted regions in the world (Singh et al., 2004; Di Girolamo et al., 2004; Ramana et al., 2004; Tripathi et al., 2005; Dey and Di Girolamo, 2010; Srivastava et al., 2011). This region is bordered by the Himalayas to the north, Vindhyan Satpura ranges to the south, Thar desert and the Arabian Sea in the west and the Bay of Bengal in the east. Due to its unique topography and diverse aerosol emissions (e.g., dust, black carbon, nitrate, sulfate and organics) from densely populated and industrialized areas over IGP, this region has become a hot spot for environmental research. Several studies using ground based measurements and satellite data have shown

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persistent heavy aerosol loading over the IGP region throughout the year (Singh et al., 2004; Di Girolamo et al., 2004; Ramana et al., 2004; Prasad et al., 2006; Badarinath et al., 2009; Eck et al., 2010; Giles et al., 2011). High aerosol loading over this region not only results in poor air quality leading to adverse health effects (Jai Devi et al., 2009) but also perturbs the climate significantly (Bollasina et al., 2011, and references therein). During winter at several locations in the IGP, large aerosol concentrations near the surface as well as in the vertical column were reported due to lower temperature and shallow boundary layer (Singh et al., 2004; Tripathi et al., 2005; Prasad et al., 2006; Ramachandran et al., 2006; Tare et al., 2006; Eck et al., 2012; Choudhry et al., 2012). This feature is seen during winter because inversion layer remains at low altitudes due to lower temperature, and the development of boundary layer is weak which aids in trapping the particles within this layer (Komppula et al., 2012). In addition, formation of fog and visibility degradation is a consistent feature during winter over many locations in IGP, which occurs due to favorable meteorological conditions aided by high aerosol concentration over IGP (Ramachandran et al., 2006; Tare et al., 2006; Eck et al., 2012). IGP is highly influenced by mineral dust during premonsoon and sea salt during monsoon, while during postmonsoon fine mode aerosols dominate (Dey et al., 2004; Tare et al., 2006; Chinnam et al., 2006; Badarinath et al., 2009; Srivastava et al., 2012).

The present study has been conducted over two different locations in IGP namely, Kanpur (26.5°N, 80.2°E, 123 m above mean sea level (AMSL)) and Gandhi College (25.9°N, 84.1°E, 60 m AMSL) with distinct meteorological conditions, aerosol sources, and topography. Both Kanpur and Gandhi College are Aerosol Robotic Network (AERONET) (Holben et al., 1998) stations. Kanpur is an urban, industrial city with a population of more than 4 million and is located ~250 km east of the mega city, New Delhi (Fig. 1). Gandhi

College located in Ballia district of Uttar Pradesh, is a rural village location in Ganga basin, southeast of Kanpur. In rural locations, biofuel burning (burning of wood, dung cake and crop waste) is a predominant source of aerosols, while fossil fuel burning (coal, petrol and diesel oil) is the major aerosol source over urban regions in India (Habib et al., 2006; Rehman et al., 2011). The biomass burning emissions increase over Gandhi College during winter owing to space heating in addition to cooking activities (Choudhry et al., 2012). Therefore, atmosphere over Kanpur is expected to be dominated more by urban and industrial emissions (Giles et al., 2011; Kaul et al., 2011), while Gandhi College is bound to be influenced by a mixture of rural and urban aerosol emissions as it is situated downwind of major urban centers, such as Delhi, Lucknow and Kanpur (Fig. 1).

It should be noted that although there exist a number of studies over the IGP using AERONET and satellite (Moderate Resolution Imaging Spectroradiometer (MODIS) and Multiangle Imaging Spectroradiometer (MISR)) data, the present work is expected to advance our knowledge on aerosol characteristics over the IGP, because all the previously published studies (a) were conducted only over Kanpur (e.g., Singh et al., 2004; Eck et al., 2010; Srivastava et al., 2011; Eck et al., 2012) to name a few, and did not include Gandhi College, (b) were conducted over both Kanpur and Gandhi College, but only for a specific season (e.g., Dey et al., 2004; Chinnam et al., 2006; Giles et al., 2011; Srivastava et al., 2012), (c) focused on radiative effects of aerosols (Tripathi et al., 2005; Ramachandran and Kedia, 2012) and/or (d) discussed the spatial variations in aerosol characteristics using remote sensing (MODIS and MISR) data (Prasad and Singh, 2007; Dey and Di Girolamo, 2010).

In the present work, for the first time, we perform a comprehensive investigation on the seasonal variation in optical and



Fig. 1. The study locations, Kanpur and Gandhi College, are denoted by stars on the Google map of South Asia. Four metro cities in India are denoted by squares. Lucknow city is denoted by circle.

physical properties of aerosols using a relatively long term (2006–2010) coincident ground based measurements of aerosol properties with AERONET sun/sky radiometers over two distinctly different (urban vs. rural) environments to construct a climatology of aerosol features over the IGP. The present study attempts to delineate the seasonally varying contribution of natural (coarse mode) and anthropogenic (fine mode) aerosols emitted from various sources based on the spectral distribution of optical characteristics of aerosols. Results on spatiotemporal variability of aerosol size distribution, source regions, aerosol types, and their optical and physical characteristics over two distinctly different environments in the same region will be useful inputs to regional and global aerosol models when assessing regional radiative forcing and climate impacts.

## 2. Wind patterns and meteorological conditions

Surface level daily mean temperature, relative humidity (RH) and wind speed are obtained from National Center for Environmental Prediction (<http://www.cdc.noaa.gov/>) reanalysis at  $2.5^\circ \times 2.5^\circ$  resolution, and daily rainfall data are obtained from Tropical Rainfall Measuring Mission (<http://disc2.nascom.nasa.gov/Giovanni/tovas/>) at  $1^\circ \times 1^\circ$  resolution during 2006–2010. These data are further used to calculate the seasonal averages and given in Table 1. Both Kanpur and Gandhi College exhibit similar meteorological conditions (Table 1). During winter temperature and wind speed are lower, and the atmosphere is dry (low RH) over both the locations as compared to other seasons (Table 1). In pre-monsoon temperature and wind speed increase over both locations. RH is higher while temperature is lower over Gandhi College than Kanpur throughout the year. During monsoon RH is highest, and both the locations receive maximum rainfall (>95% of total annual rainfall). However, Gandhi College receives comparatively higher rainfall than Kanpur during the study period. During post-monsoon temperature, wind speed and RH reduce over both the locations. Depending on the prevailing meteorological condition over IGP, an year can be broadly divided into four distinct seasons, namely winter (December, January, February, DJF), premonsoon (March, April, May, MAM), monsoon (June, July, August, September, JJAS), and postmonsoon (October, November, ON) (Singh et al., 2004; Dey et al., 2004). Therefore, on the basis of meteorology, aerosol properties are grouped into four major seasons of winter, premonsoon, monsoon and postmonsoon, and presented.

### 2.1. Air backward trajectory analysis

Air back trajectory analysis has been conducted to identify the possible source regions and the transport pathways of pollutants before they reach the measurement locations. 5-day back

**Table 1**  
Seasonal mean surface level meteorological parameters over Kanpur and Gandhi College during 2006–2010 along with  $\pm 1\sigma$  variation from the mean.

Location	Seasons	Temperature (°C)	Relative humidity (%)	Wind speed (m s <sup>-1</sup> )	Rainfall (mm)
Kanpur	Winter	23.8 ± 2.9	32 ± 11	2.4 ± 1.3	24 ± 26
	Premonsoon	33.1 ± 3.8	27 ± 9	3.2 ± 1.3	31 ± 14
	Monsoon	29.3 ± 3.4	80 ± 21	2.9 ± 1.6	732 ± 197
	Postmonsoon	25.8 ± 1.6	57 ± 16	1.9 ± 1.1	46 ± 64
Gandhi College	Winter	22.7 ± 2.4	43 ± 12	2.1 ± 1.0	19 ± 16
	Premonsoon	29.8 ± 2.2	52 ± 17	2.2 ± 1.1	71 ± 45
	Monsoon	27.1 ± 1.5	92 ± 8	2.4 ± 1.1	1023 ± 255
	Postmonsoon	24.7 ± 1.6	73 ± 14	2.2 ± 1.1	41 ± 39

trajectories are calculated at 0600 Universal Time (UTC) for individual days during 2006–2010 over both locations using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) meteorological model (Draxler and Hess, 1998). Trajectories are calculated at four different heights of 500 m (mixed layer height), 1000 m (boundary layer height), 2500 m (above the boundary layer where the aerosols can be lifted by convection and transported over long distance), and 4000 m (free troposphere) for both the locations. Seasonal mean trajectories are calculated by taking the mean of individual trajectories corresponding to different seasons during 2006–2010. Trajectory analysis has been conducted at different heights as a function of season in order to be able to explain the variability of aerosol properties in total atmospheric column.

Fig. 2 shows the mean trajectories with  $\pm 1\sigma$  variation for each season during 2006–2010 over Kanpur and Gandhi College. During winter and postmonsoon, air masses are from west over both the locations at all the four altitude levels. However, trajectories are more local during postmonsoon than in winter. During these seasons, in addition to the local anthropogenic activities, advection from agriculture field waste burning is expected to contribute to the aerosol content (Badarinath et al., 2009). Back trajectory analysis suggests that aerosols can be of mixed type during winter and postmonsoon. During premonsoon trajectories are mainly westerly/northwesterly over Kanpur; while over Gandhi college trajectories at lower heights (up to 1000 m) are influenced by southeast part of India, and are from the west at higher heights. Trajectories originate from arid region of western India, Thar desert and Pakistan during premonsoon indicating transport of dust particles to the study locations (Ramachandran and Kedia, 2010; Giles et al., 2011). In addition, locally produced dust from the arid land (due to low RH and high wind speed) is expected to increase the concentration of coarse mode aerosols over the study locations. During monsoon, direction of winds change and are mainly from west and southwest direction; winds originate from the oceanic regions and travel through the continental India before reaching the study locations suggesting transport of mixed aerosol particles over IGP. This analysis on potential aerosol source regions will be useful while interpreting the seasonal variation in aerosol characteristics in terms of the aerosol types.

## 3. Data and analysis

### 3.1. Aerosol optical properties

In the present study, level 2.0 cloud screened and quality assured data of aerosol optical depth (AOD), fine mode fraction (FMF), single scattering albedo (SSA), extinction and absorption Ångström exponents, and sphericity fraction obtained from Aerosol Robotic Network (AERONET) (Holben et al., 1998) over Kanpur and Gandhi College during 2006–2010 are utilized. This time period is chosen owing to the coincident availability of data over both the locations. AERONET is a ground based Sun/sky radiometer which measures direct solar and diffuse sky radiance in the spectral range of 0.34–1.02  $\mu\text{m}$  (Holben et al., 1998). The field of view of the instrument is about  $1.2^\circ$  and it makes direct solar radiation measurements every 15 min. For direct solar measurement, triplet observations are made at each wavelength for calibration (using Langley technique) and to screen clouds. AERONET calculates extinction (absorption) Ångström exponents using information on spectral dependence of extinction (absorption) AOD as a function of wavelength (Russell et al., 2010); sphericity fraction is calculated as the fraction of spherical to spheroidal plus spherical particles (Giles et al., 2011). Details about the measurement protocol for AERONET, calibration techniques, methodology, data processing and quality



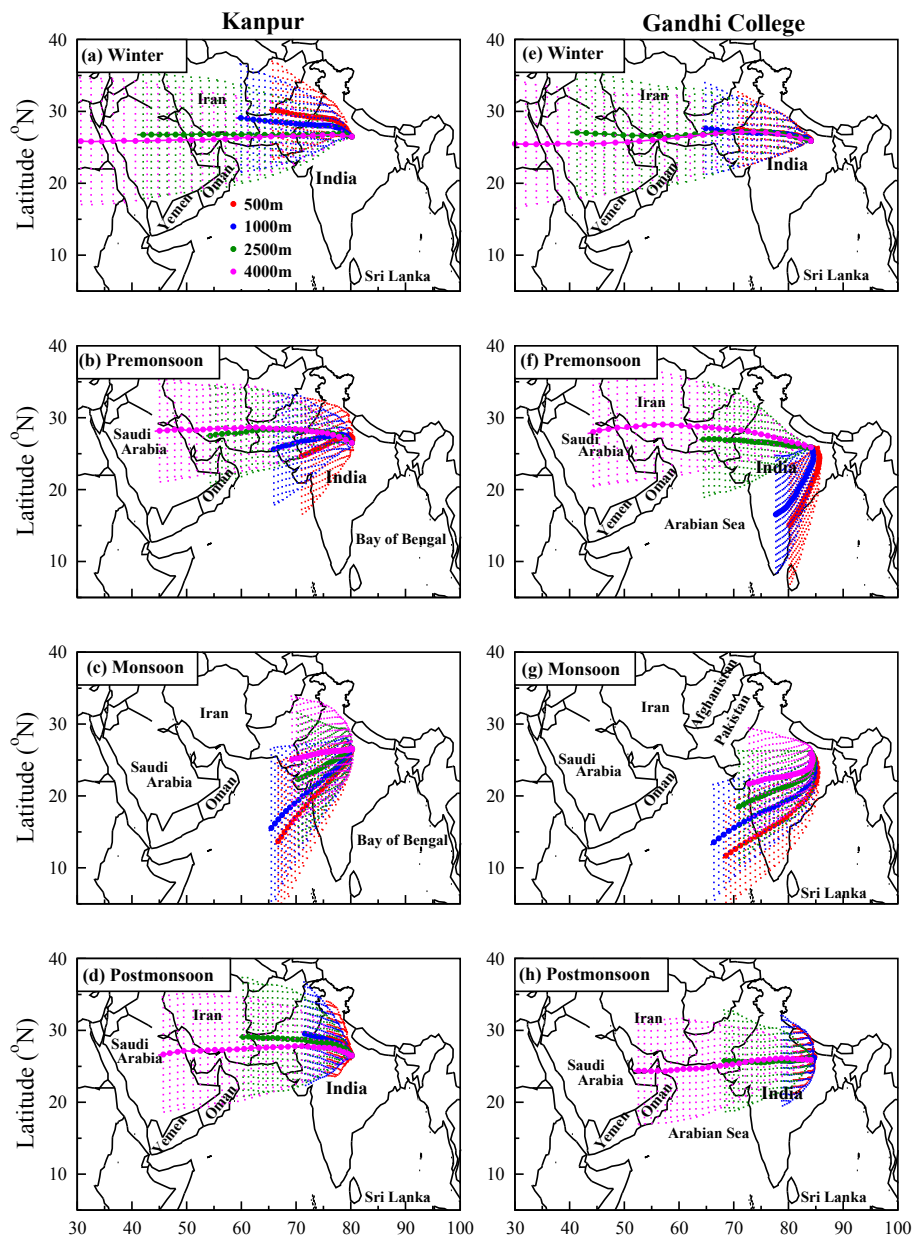


Fig. 2. Seasonal averages of 5-day air back trajectories at 500 m, 1000 m, 2500 m and 4000 m corresponding to 0600 UTC during 2006–2010 over Kanpur (a, b, c, d) and Gandhi College (e, f, g, h). Stippling represent variation from the mean pathway during each season for 2006–2010.

can be found in literature (Holben et al., 1998; Eck et al., 1999; Dubovik et al., 2000; O'Neill et al., 2003).

In the present work, direct solar measurements made at four spectral channels (0.44, 0.50, 0.675 and 0.87  $\mu\text{m}$ ), FMF at 0.5 and 0.675  $\mu\text{m}$ , and sky radiance measurements made at four spectral channels (0.44, 0.675, 0.87 and 1.02  $\mu\text{m}$ ) are utilized. FMF is calculated as the ratio of fine mode AOD (0.01–1.0  $\mu\text{m}$  radius range) to total AOD (0.01–10  $\mu\text{m}$  radius range) from sky radiance measurements. The uncertainty in AODs calculated using the direct solar radiation measurements is less than  $\pm 0.01$  for wavelength  $\geq 0.44$   $\mu\text{m}$  and is less than  $\pm 0.02$  for shorter wavelengths (Holben et al., 1998). The error in AERONET derived FMF is  $\sim 10\%$  (O'Neill et al., 2003). Error in SSA is  $\pm 0.03$  when AOD at 0.44  $\mu\text{m}$  is  $> 0.2$  and is higher (0.05–0.07) when AOD at 0.44  $\mu\text{m}$  is  $\leq 0.2$  (Dubovik et al., 2000). Aerosol properties corresponding to individual observation (at every 15 min time interval) and typically about 20

measurements/data points a day on each day during 2006–2010 are used to calculate the monthly/seasonal average and utilized in the present work. The total number of data points available for each season during 2006–2010 is given in Table 2. As the data sets used are not sufficiently long enough to examine the inter-annual variability in AODs in detail and/or the trends, in this study the monthly mean and seasonal variations in aerosol characteristics are averaged for the 5-year period and discussed.

Table 2

Number of data points over Kanpur and Gandhi College in different seasons during 2006–2010 available from AERONET site and utilized in the present study.

Location	Winter	Premonsoon	Monsoon	Postmonsoon
Kanpur	4648	10159	5143	4576
Gandhi College	867	7047	2733	1381

### 3.2. Ångström exponent and its derivative

Spectral variation in AODs can be represented by Ångström power law ( $\tau = \beta\lambda^{-\alpha}$ , where  $\tau$  is AOD,  $\lambda$  is the wavelength in  $\mu\text{m}$ , and  $\alpha$  and  $\beta$  are Ångström parameters).  $\alpha$  is calculated from the linear fit of  $\ln \tau$  versus  $\ln \lambda$  data and is known as the wavelength exponent.  $\beta$  is the turbidity coefficient which is equal to the AOD at  $1 \mu\text{m}$ . The  $\alpha$  value depends on aerosol size distribution and varies from 1 to 3 for fresh and aged smoke (Nicolae et al., 2013), and for urban aerosol particles, while  $\alpha$  is nearly zero for coarse mode aerosols such as dust and sea salt (Eck et al., 1999).  $\beta$  ranges between 0 and 1, and is an indicator of the amount of aerosols present in the atmospheric column. In the ambient atmosphere aerosol size distribution is multimodal with contribution from both fine and coarse mode aerosols. Therefore, Ångström power law can introduce significant errors in the determination of aerosol size distribution (Eck et al., 1999). Under these conditions deviation from linear behavior of  $\ln \tau$  versus  $\ln \lambda$  data occurs, which can be quantified by fitting a second order polynomial of the form.

$$\ln \tau = \alpha_2 (\ln \lambda)^2 + \alpha_1 \ln \lambda + \alpha_0, \quad (1)$$

where  $\alpha_0$ ,  $\alpha_1$  and  $\alpha_2$  are the coefficients of the polynomial fit, which can be utilized to get information on the dominant type of aerosols (whether fine or coarse) in the size distribution. Schuster et al. (2006) showed that though different aerosol size distributions can produce the same value of  $\alpha$ , the coefficients of polynomial fit can be distinctly different. AODs measured in the wavelength region of  $0.44\text{--}0.87 \mu\text{m}$  during 2006–2010 over Kanpur and Gandhi College are used for the calculation of  $\alpha$ ,  $\alpha_0$ ,  $\alpha_1$  and  $\alpha_2$  by applying a polynomial fit to  $\ln \tau$  versus  $\ln \lambda$  data, and monthly/seasonal means are obtained. AODs in the  $0.44\text{--}0.87 \mu\text{m}$  range have been chosen because AERONET channels in the  $0.44\text{--}0.87 \mu\text{m}$  wavelength range are more accurate (Holben et al., 1998; Eck et al., 1999).

## 4. Results and discussion

### 4.1. Aerosol optical depths, fine mode fraction and Ångström coefficients

The gradual increase in AOD from March to May over both the locations in IGP (Fig. 3a) can be ascribed to the increase in dust particle concentration as the southwesterly winds (Fig. 2) are from the arid, and semiarid regions. AODs decrease during monsoon over both the locations which is consistent with the earlier results obtained over Kanpur only as in Eck et al. (2010). This decrease is attributed to wet deposition and change in the origin of the air parcels (originating from the Arabian Sea (Fig. 2)). The decrease in AOD, however, is not significant as the air parcels travel across western India and the Thar desert before reaching the study locations (Figs. 2 and 3a). During winter and postmonsoon, air masses are from west and the wind speeds are lower (Table 1); the trajectories and winds are more local during postmonsoon than in winter. Therefore, in these seasons fine mode aerosols emitted by the local anthropogenic activities such as fossil fuel and biomass combustion will dominate. Aerosols advected from agriculture field waste burning emissions in the fine mode (Fig. 2) will also contribute to the aerosol content over the IGP during these seasons. These results show that during the year the IGP is influenced by aerosol emissions in the fine mode from coal fire plants, vehicles, industries, biomass and domestic biofuel burning, and transport of mineral dust and sea salt in coarse mode (Fig. 3). Therefore, it is important to examine and quantify whether fine and coarse mode aerosol population remain the same throughout the year, or aerosols in a particular mode decrease (increase) to compensate for the

increase (decrease) in the other mode. This is an important information as anthropogenic aerosols, which contribute about 20–40% to the global AOD, are smaller in size and more absorbing than natural aerosols (Myhre et al., 2009).

The FMF value can vary from 0 (single coarse mode aerosol) to 1 (single fine mode aerosol), and provides quantitative information on the nature of the aerosol size distribution. Seasonal mean FMF for Kanpur (Gandhi College) are:  $0.84 \pm 0.12$  ( $0.89 \pm 0.08$ ) – winter,  $0.42 \pm 0.07$  ( $0.55 \pm 0.07$ ) – premonsoon,  $0.49 \pm 0.14$  ( $0.57 \pm 0.12$ ) – monsoon and  $0.77 \pm 0.15$  ( $0.84 \pm 0.08$ ) – postmonsoon. Over both the locations the highest value of FMF is observed during November–January when fine mode aerosols dominate the IGP (Fig. 3b). This is due to (i) the influence of high anthropogenic emissions over the entire IGP during winter (Singh et al., 2004; Di Girolamo et al., 2004; Prasad et al., 2006; Eck et al., 2010; Dey and Di Girolamo, 2010; Giles et al., 2011), (ii) the trapping of aerosols which get transported from west to east due to northwesterly winds because of a lower boundary layer height (Dey and Di Girolamo, 2010) and (iii) the intense crop residue burning (Badarinath et al., 2009). FMF is minimum during April–June when coarse mode aerosols are abundant in the atmosphere (mean FMF values are 0.36 in Kanpur and 0.48 in Gandhi College (Fig. 3b) due to increase in the concentration of dust particles (Fig. 2) (Eck et al., 2010; Giles et al., 2011).

During the entire study period and for all the months  $\alpha$  is found to be higher over Gandhi College (varies between 0.64 and 1.46, annual mean  $1.13 \pm 0.27$ ) than Kanpur (in the range of 0.41–1.37, annual mean  $0.98 \pm 0.32$ ). This is in good agreement with features seen in FMF. An increase in the value of  $\alpha$  occurs either due to increase in fine mode aerosol concentration or decrease in concentration of coarse mode aerosols in the atmosphere. However, comparatively higher FMF over Gandhi College than Kanpur confirm that fine mode aerosol concentration remains higher over Gandhi College. This could be attributed to the transport of fine mode pollutants downwind, and higher amount of local biomass/biofuel burning (Badarinath et al., 2009; Dey and Di Girolamo, 2010), as it is a rural location. Similar monthly variation in  $\alpha$  with a maximum during October–February and a minimum during April–June has been earlier reported over Kanpur (Eck et al., 2010).

$\beta$  is found to exhibit significant variation (Fig. 3d), similar to the variation in AOD (Fig. 3a), indicating changing columnar aerosol loading during different months. Over both the locations  $\alpha$  is higher while  $\beta$  is lower during winter and postmonsoon indicating higher fine mode aerosol concentration which is a characteristic feature of the locations in IGP (Singh et al., 2004; Dey et al., 2004; Tripathi et al., 2005; Eck et al., 2010). The dense population in this region is associated with large sources of aerosols (due to fossil fuel consumption and biofuels used for domestic purposes) which produce higher fine mode aerosols (Fig. 6, Dey and Di Girolamo, 2010). During monsoon  $\alpha$  increases while  $\beta$  decreases because of relatively lower AOD values (Fig. 3a).

In summary, AODs are more or less similar in both the locations, however FMF and  $\alpha$  are higher over Gandhi College confirming the dominance of fine mode aerosols throughout the year in Gandhi College. It should be noted that Kanpur is also affected by pollutants that are transported from Delhi and western Uttar Pradesh (Fig. 1), yet it has lower FMF when compared to Gandhi College. It is, therefore, worth investigating whether it is the local dust emissions or the biomass burning emissions that are more at the rural Gandhi College that give rise to higher FMF than the urban Kanpur. This classification becomes important over the IGP where the biomass and biofuel sources co-exist in close vicinity of fossil fuel burning sources. This aspect is examined by utilizing the MODIS cloud corrected fire count version 5 monthly data available at  $1^\circ \times 1^\circ$  latitude resolution corresponding to Kanpur and Gandhi College. It is

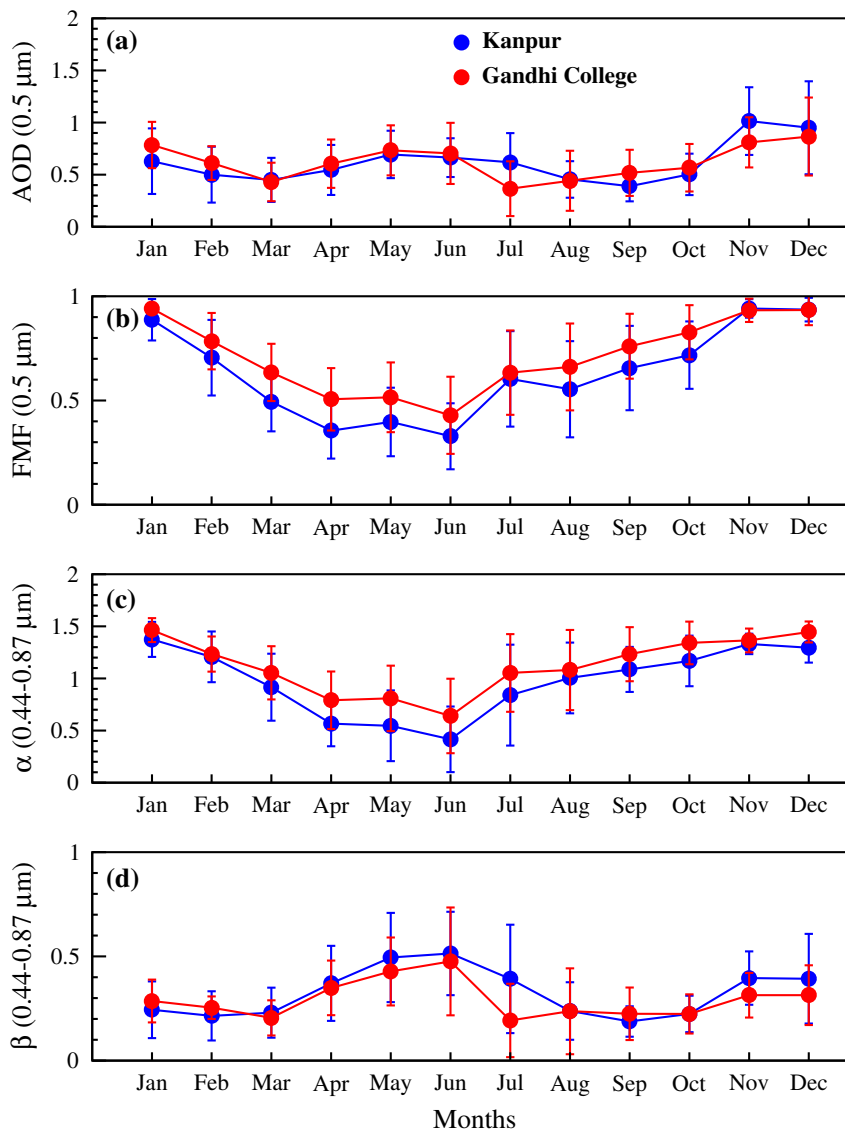


Fig. 3. Monthly mean and  $\pm 1\sigma$  standard deviation in (a) aerosol optical depth (AOD), (b) fine mode fraction (FMF), and Ångström coefficients (c)  $\alpha$  and (d)  $\beta$  during 2006–2010 over Kanpur and Gandhi College.

clear that the fire count is higher over Gandhi College than Kanpur during the year (Fig. 4). Fire counts exhibit large inter-annual variations over both the locations, however, the fire count is higher during premonsoon and peaks in April (Fig. 4, Table 3). The higher MODIS fire count is consistent with the spring time agricultural waste/residue burning over the IGP; the fire counts are significantly higher over Gandhi College confirming the dominance of local biomass burning activities. Seasonal mean fire count over Kanpur (Gandhi College) is:  $1 \pm 1$  ( $2 \pm 2$ ) – winter,  $7 \pm 6$  ( $19 \pm 22$ ) – premonsoon,  $2 \pm 3$  ( $5 \pm 4$ ) – monsoon and  $2 \pm 2$  ( $0 \pm 0$ ) – postmonsoon. During premonsoon the fire count over Gandhi College is a factor of three higher than Kanpur, and the standard deviation is higher than the mean value corroborating the large inter-annual variability in fire count (Fig. 4). Fire counts are also higher over the IGP during postmonsoon (October–November) due to the burning of rice crop residues (Badarinath et al., 2009) and the resultant transport of the sub-micron size aerosols. However, there are fire counts only over Kanpur and not over Gandhi College (Table 4). The absence of fire counts over Gandhi College during

postmonsoon indicates the spatial limit of long range transport of biomass burning emission produced aerosols over the IGP; this occurs because the winds are more local and wind speeds are lower during postmonsoon. This aspect is further highlighted in the AOD,

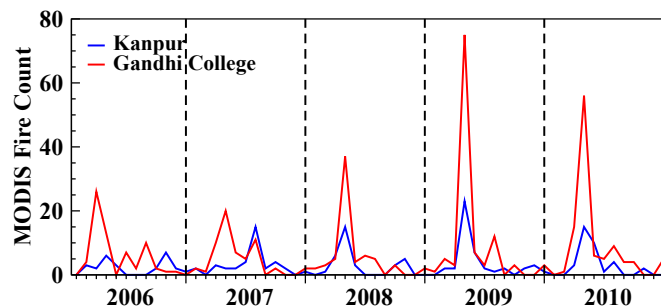


Fig. 4. Monthly variation of cloud corrected MODIS fire count over Kanpur and Gandhi College during 2006–2010.

**Table 3**

Monthly mean MODIS cloud corrected fire count over Kanpur and Gandhi College during 2006–2010 along with  $\pm 1\sigma$  variation from the mean.

Month	Kanpur	Gandhi College
Jan	0 $\pm$ 0	1 $\pm$ 1
Feb	1 $\pm$ 1	3 $\pm$ 2
Mar	3 $\pm$ 2	12 $\pm$ 9
Apr	12 $\pm$ 8	40 $\pm$ 26
May	5 $\pm$ 3	5 $\pm$ 3
Jun	1 $\pm$ 2	5 $\pm$ 1
Jul	4 $\pm$ 6	8 $\pm$ 4
Aug	1 $\pm$ 1	3 $\pm$ 4
Sep	2 $\pm$ 2	3 $\pm$ 1
Oct	4 $\pm$ 2	0 $\pm$ 0
Nov	1 $\pm$ 1	0 $\pm$ 0
Dec	1 $\pm$ 1	2 $\pm$ 2

$\beta$  and FMF variations during March–May (Fig. 3); AODs and  $\beta$  are similar and increase from March to May over Kanpur and Gandhi College, however, FMF and  $\alpha$  values are higher for Gandhi College. Thus, it is clear that, coarse mode dust particles are more in Kanpur when compared to Gandhi College even though the source regions look similar during premonsoon (Fig. 2). The lower FMF over Kanpur during premonsoon indicates that there exists a gradient in the amount of dust that is transported over the IGP because dust is more in Kanpur than Gandhi College.

Aerosol optical properties varied over a wide range – 0.5  $\mu\text{m}$  AOD varied from 0.1 to 2, FMF in the range of <0.1 to 1, and  $\alpha$  in the 0.44–0.87  $\mu\text{m}$  region varied from 0.1 to 2, over both Kanpur and Gandhi College during the 5-year study period (2006–2010). In order to accommodate the wide range of aerosol characteristics encountered over the IGP, AOD and  $\alpha$  values are segregated into 4 bins in the range of 0–0.5, 0.5–1.0, 1.0–1.5 and 1.5–2.0, and FMF is classified in four bins as 0–0.25, 0.25–0.5, 0.5–0.75 and 0.75–1.0. A broader segregation bin is chosen to study the frequency occurrence owing to the large variability in aerosol characteristics. Over both the study locations, AOD in the range of 0.5–1 is found to be the most frequent (>50%) for all the seasons except during winter when AODs <0.5 are more frequent over Kanpur. More than 70% of FMF values are in the range of 0.75–1.0 during winter and postmonsoon due to the dominance of fine mode aerosols over both the study locations. The frequency of occurrence of FMF values that are greater than 0.75 is higher throughout the year in Gandhi College when compared to Kanpur (Fig. 5a). During premonsoon 70% of FMF values in Kanpur are less than 0.5, confirming the presence of coarse mode (dust) particles. In contrast over Gandhi College about 50% of FMF values are greater than 0.5 during premonsoon. This comparison in a robust manner brings to the fore the limit in terms of spatial distance dust gets transported during premonsoon over the IGP despite the back trajectories look seemingly similar for both the locations (Fig. 2). It is however worth noting that back trajectories below 1000 m reach Gandhi College sweeping through south and central India. Dust emissions showed a gradient over the Indo-Gangetic Plain (Fig. 3, Habib et al., 2006).

The  $\alpha$  exhibits a skewed distribution with peaks in the range of 1.0–1.5 during winter and postmonsoon. The  $\alpha$  is >1 for  $\geq 90\%$  of spectra during postmonsoon over Kanpur and Gandhi College indicating the dominance of fine mode aerosols which is consistent with the FMF distribution. During winter >90% of  $\alpha$  values are >1 over Gandhi college indicating fine mode dominance. Over Kanpur ~30% of AOD spectra with  $\alpha < 1$  is observed, indicating the presence of coarse mode aerosols in spite of higher fine mode aerosol concentration (Fig. 5). During premonsoon and monsoon  $\alpha$  is  $\leq 1$  for ~60% of spectra and the frequency distribution in  $\alpha$  is

broad which indicates the presence of mixed type aerosols with a bimodal size distribution. The percentage of AOD spectra with  $\alpha \leq 1$  is higher over Kanpur than Gandhi College during all the seasons indicating higher coarse mode aerosol concentration over Kanpur throughout the year. Similar features in the frequency distribution curve of AODs and  $\alpha$  were reported earlier over Kanpur during 2001–2003 which suggested the dominance of fine mode aerosols during winter/postmonsoon and coarse mode aerosols during premonsoon/monsoon (Singh et al., 2004). Srivastava et al. (2012) also observed that the fine mode aerosol concentration remains higher while coarse mode concentration remains lower over Gandhi College when compared to Kanpur during premonsoon. Seasonal differences in AOD, FMF and  $\alpha$  observed in the present study confirm the existence of strong variability in aerosol size distribution. Nevertheless, further analysis is necessary to determine the type of aerosols responsible for producing the variability in aerosol size distribution which is attempted next.

#### 4.2. Spectral distribution of aerosol optical depths

In the ambient atmosphere aerosol size distribution is multimodal, therefore  $\alpha$  need not be single valued. In such cases the difference in  $\alpha$  value calculated in the short (0.44–0.50  $\mu\text{m}$ ) and the long (0.675–0.87  $\mu\text{m}$ ) wavelength spectral regions helps in determining whether fine or coarse mode aerosols are dominant in an aerosol size distribution (Gobbi et al., 2007). A positive value of  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  indicates higher coarse mode aerosol concentration, while negative value suggests higher concentration of fine mode aerosols. Near zero value of  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  represents a constant value of  $\alpha$  and unimodal aerosol size distribution. The  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  values exhibit large spread with non-zero values over both the study locations confirming multimodal aerosol size distribution and non-linear behavior of  $\ln \tau$  versus  $\ln \lambda$  (Fig. 6). During winter and postmonsoon  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  is negative for >60% of AOD spectra over Kanpur and Gandhi College (Table 4) confirming the dominance of fine mode aerosols corroborating the findings of Fig. 3. During premonsoon  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  is positive for ~80% of AOD spectra over Kanpur and Gandhi College (Fig. 6) indicating higher coarse mode abundance over IGP which is in league with results shown earlier (Fig. 3). During monsoon 77% of AOD spectra is found to have positive  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  values over Kanpur, while over Gandhi College  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$  is positive for 69% of AOD spectra. The results indicate that although both Kanpur and Gandhi College are situated in IGP region and influenced by more or less similar meteorology (Fig. 2, Table 1), size distributions of aerosols and the type of aerosols governing the aerosol optical properties can be different on seasonal time scales.

To get further information about the nature of aerosol size distribution and to strengthen the findings, coefficients of polynomial fit are calculated to examine the aerosol distribution. It should be noted that the average rms error, which indicates the goodness of fit, between the measured AOD spectra and estimated spectra using polynomial fit is found to be very less over Kanpur (0.006  $\pm$  0.004) and Gandhi College (0.004  $\pm$  0.003). As the error in the coefficient of polynomial fits is not significant they are not considered in the ensuing discussion. Schuster et al. (2006) showed that the relation  $\alpha_2 - \alpha_1 = \alpha$  can be considered valid for a bimodal aerosol size distribution. Under such conditions  $\alpha_2 - \alpha_1$  is  $\geq 2$  represents AOD spectra dominated by fine mode aerosols, while  $\alpha_2 - \alpha_1$  is  $\leq 1$  represents AOD spectra dominated by coarse mode aerosols. AOD spectra with  $\alpha_2 - \alpha_1$  value between 1 and 2 represents wide range of fine mode fractions or mixture of modes. Both Kanpur and Gandhi College (Table 5) are marked by the dominance of wide

**Table 4**

Percentage of AOD spectra classified on the basis of  $\alpha_{0.44-0.50} - \alpha_{0.675-0.87} < 0$  (fine mode) or  $\geq 0$  (coarse mode) over Kanpur and Gandhi College.

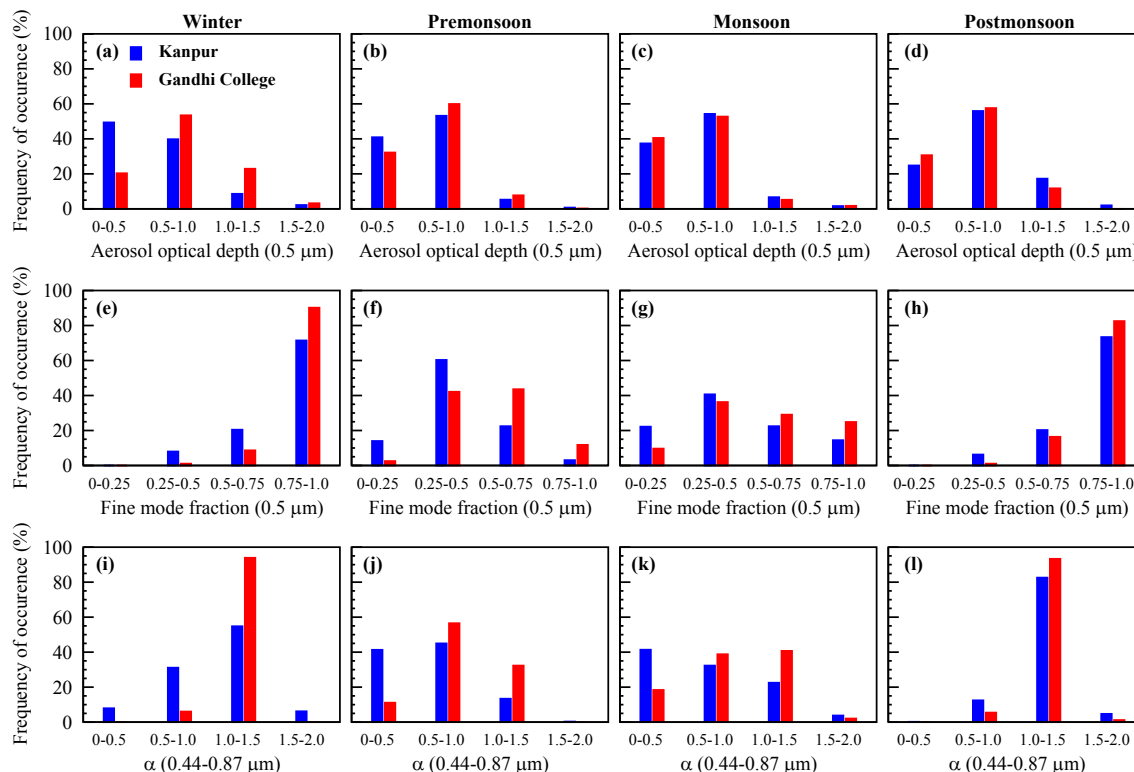
Location	$\alpha_{0.44-0.50} - \alpha_{0.675-0.87}$	Winter	Premonsoon	Monsoon	Postmonsoon
Kanpur	< 0	62	19	23	76
	$\geq 0$	38	81	77	24
Gandhi College	< 0	86	21	31	86
	$\geq 0$	14	79	69	14

range of fine mode fractions or mixture of modes during winter as  $\alpha_2 - \alpha_1$  values lie between 1 and 2 for 61% of the AOD spectra over Kanpur and for 95% of spectra over Gandhi College (Fig. 6). It is notable that 39% of AOD spectra over Kanpur have  $\alpha_2 - \alpha_1 \leq 1$  indicating the presence of coarse mode aerosols during winter, while over Gandhi College only 4% of AOD spectra showed coarse mode aerosols (Table 5) which has been observed earlier in the frequency distribution of FMF (Fig. 5e). During premonsoon and monsoon coarse mode aerosols (mostly dust) become dominant over the IGP with a significantly higher concentration over Kanpur (86%) as compared to Gandhi College (67%), confirming the earlier result. During postmonsoon >85% of AOD spectra are dominated by fine mode aerosols over both the locations. These findings strongly correlate with results shown in Table 4 proving the robustness of the methodology. The results indicate that the coarse mode aerosol concentration is higher while fine mode concentration is lower over Kanpur than Gandhi College throughout the year. This analysis of Ångström exponents at different spectral ranges and second order polynomial fits to the spectral distribution of AODs reveal in a

robust and quantitative manner that fine mode particles are always higher over Gandhi College in IGP when compared to Kanpur.

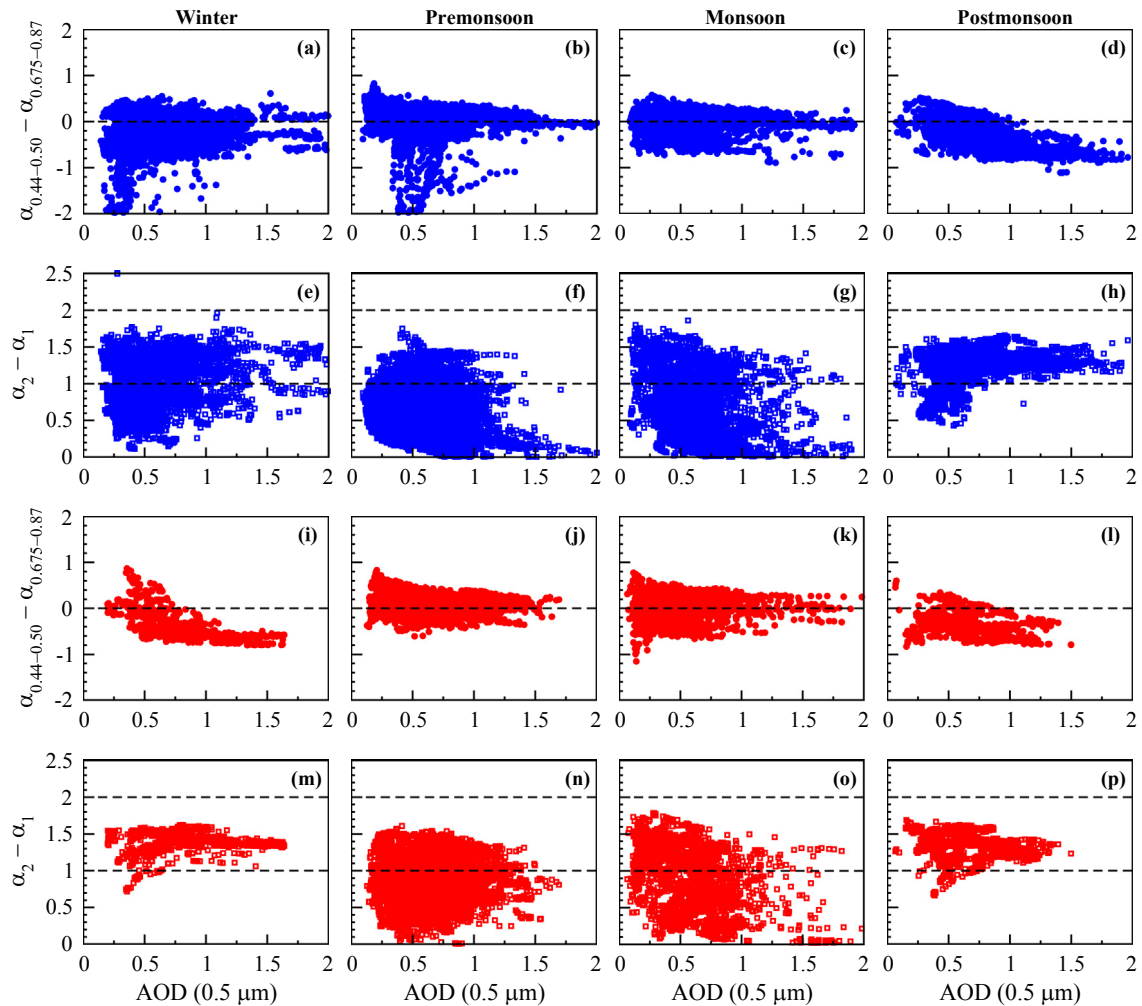
### 4.3. Single scattering albedo

Single scattering albedo (SSA), the ratio of scattering to total extinction of aerosols, can be used as an index to determine the relative dominance of scattering versus absorbing aerosols. SSA can range from 0 (purely absorbing) to 1 (purely scattering). Spectral variation in SSA gives information on the abundance of absorbing and scattering aerosols (e.g. black carbon, organic carbon, dust, sulfate) in different size ranges due to spectral absorption characteristics of aerosol mixture. Spectral dependence of SSA shows an increasing trend with wavelength for dust aerosols while SSA decreases with wavelength when BC is the sole absorber; spectral dependence of SSA can exhibit different wavelength dependency in the presence of aerosol mixture (Dubovik et al., 2002; Russell et al., 2010; Giles et al., 2011). SSA is found to be >0.85 and exhibit large seasonal and wavelength dependence over the study locations during 2006–2010 (Fig. 7). SSA decreases with wavelength during winter and is lower (Fig. 7) which can be attributed to the dominance of absorbing urban aerosols such as BC (Singh et al., 2004; Tripathi et al., 2005). During premonsoon and monsoon SSA is found to increase with increasing wavelength over both the locations due to the dominance of coarse mode dust particles in the atmosphere. As dust particles scatter the radiation more efficiently, their dominance in the aerosol size distribution during premonsoon and monsoon result in increase of SSA at wavelengths >0.5  $\mu\text{m}$ . Increase in SSA with wavelength is more sharper in Kanpur than over Gandhi College, which can be attributed to a higher coarse mode dust concentration over Kanpur as seen earlier (Table 5). A higher dust concentration over Kanpur than Gandhi College is consistent with the back trajectories; winds originate



**Fig. 5.** Frequency of occurrences of AOD (a, b, c, d), FMF (e, f, g, h) and Ångström exponent ( $\alpha$ ) (i, j, k, l) in Kanpur and Gandhi College during winter, premonsoon, monsoon and postmonsoon seasons of 2006–2010.





**Fig. 6.** Differences in  $\alpha$  values obtained between short (0.44–0.50  $\mu\text{m}$ ) and long (0.675–0.87  $\mu\text{m}$ ) wavelengths versus 0.5  $\mu\text{m}$  aerosol optical depth (AOD) over Kanpur (a, b, c, d) and Gandhi College (i, j, k, l). Difference in coefficient of polynomial fits versus 0.5  $\mu\text{m}$  aerosol optical depth (AOD) over Kanpur (e, f, g, h) and Gandhi College (m, n, o, p) during winter, premonsoon, monsoon and postmonsoon of 2006–2010.

from and pass through arid and semi-arid regions for all the seasons at all the altitudes over Kanpur (Fig. 2). The spectral dependence in SSA is weaker during postmonsoon over both the locations, nevertheless SSA is lower in Gandhi College as compared to Kanpur (more scattering type). This is due to the higher concentration of fine mode absorbing aerosols over Gandhi College, a rural location, where biomass/biofuel burning emissions dominate as evidenced earlier (Fig. 4).

Over the course of the year, these two sites in IGP exhibit aerosol optical characteristics of a range of different environments. For example, during winter fine mode aerosols (urban, industrial and biomass burning emissions) dominate the IGP (Fig. 5e) and SSA decreases as a function of wavelength – a characteristic feature of urban-industrial and mixed type environment as seen in Dubovik et al. (2002) and Russell et al. (2010) respectively. During premonsoon and monsoon fine mode aerosol contribution decreases (Fig. 5f, g) because of the increase in coarse mode dust and sea salt aerosol respectively, giving rise to SSA that increases with wavelength – a typical feature over desert dust and oceanic type of environments (Dubovik et al., 2002; Russell et al., 2010). This is because the IGP is one of few regions that experience strongly seasonally varying air masses and therefore, aerosol sources, which result in aerosol characteristics that vary widely across space and

time. The results from this study can serve as inputs in satellite retrieval of aerosols and in regional models to assess the radiative impacts as this region exhibits aerosol characteristics that represent a variety of aerosol environments in different seasons.

#### 4.4. Type of absorbing aerosols

Black carbon and dust are the two major absorbing aerosol types in the atmosphere. It is important to characterize the type of absorbing aerosol particles present in the atmosphere as they strongly influence Earth's climate through direct (radiative forcing) and indirect/semi-direct (cloud optical properties and lifetime) effects. Information on aerosol size distribution, and absorption and extinction Ångström exponents have been used to differentiate the percentage of black carbon and dust aerosols. Giles et al. (2011) showed that dominant absorbing aerosol types can be determined using a combination of optical and physical characteristics of aerosols, namely, aerosol absorption (absorption Ångström exponent ( $\alpha_{abs}$ )), size (extinction Ångström exponent ( $\alpha_{ext}$ ), fine mode fraction at 0.675  $\mu\text{m}$  (FMF<sub>0.675</sub>)) and sphericity fraction. Giles et al. (2011) characterized absorbing aerosols in three major categories such as Mostly Dust (aerosol mixture where iron oxide in dust is dominant absorber), Mostly BC (mixture of biomass burning,

**Table 5**

Percentage of AOD spectra classified on the basis of  $\alpha_2 - \alpha_1 \leq 1$  (coarse mode aerosol dominance) and between 1 and 2 (wide range of fine mode fractions or mixture of modes) over Kanpur and Gandhi College.

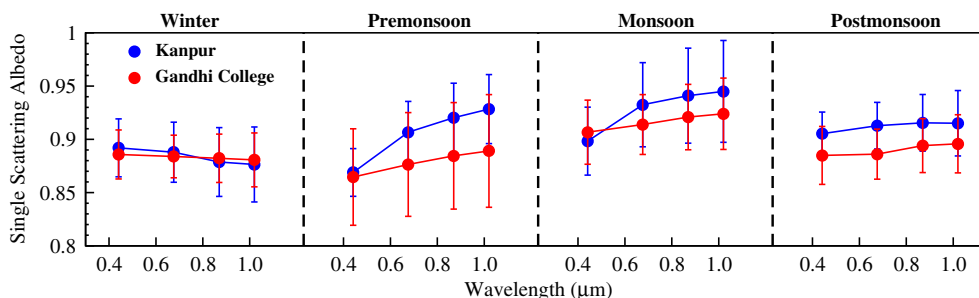
Location	$\alpha_2 - \alpha_1$	Winter	Premonsoon	Monsoon	Postmonsoon
Kanpur	$\leq 1$	39	86	73	13
	1–2	61	14	27	87
Gandhi College	$\leq 1$	4	67	56	5
	1–2	96	33	44	95

urban/industrial emissions with BC as the dominant absorber), and Mixed BC and Dust (optical mixture of fine mode BC and coarse mode dust as dominant absorber). Following Giles et al. the absorbing aerosol type over Kanpur and Gandhi College have been classified based on the above parameters into: *Mostly Dust* ( $\alpha_{abs} > 2.0$ ,  $\alpha_{ext} \leq 0.5$ ,  $FMF_{0.675} \leq 0.33$ , sphericity fraction  $< 0.2$ ), *Mostly BC* ( $1.0 < \alpha_{abs} \leq 2.0$ ,  $\alpha_{ext} > 0.8$ ,  $FMF_{0.675} > 0.66$ , sphericity fraction  $\geq 0.2$ ), and *Mixed BC and Dust* ( $\alpha_{abs} \sim 1.5$ ,  $\alpha_{ext} \sim 0.5$ ,  $FMF_{0.675} \sim 0.33$ ). The absorbing aerosols present over both the study locations during winter and postmonsoon are Mostly BC followed by Mixture of BC and Dust (Fig. 8, Table 6), however, their percentage contributions vary. The sphericity fraction is typically low for coarse mode dust particles as compared to fine mode BC aerosols (Fig. 8). During premonsoon and monsoon, atmosphere over both the locations is characterized by presence of mixed BC and Dust absorbing aerosols ( $\geq 80\%$ , Table 6); however, the concentration of BC remains higher over Gandhi College than Kanpur throughout the year (Table 6), and significantly so during winter and postmonsoon. It is observed that Mostly dust aerosols are nearly absent in Gandhi College throughout the year while their presence has been observed over Kanpur during premonsoon and monsoon. The findings are consistent with Giles et al. (2011) who reported the presence of Mixed BC and Dust over Kanpur during premonsoon seasons of 2002–2008.

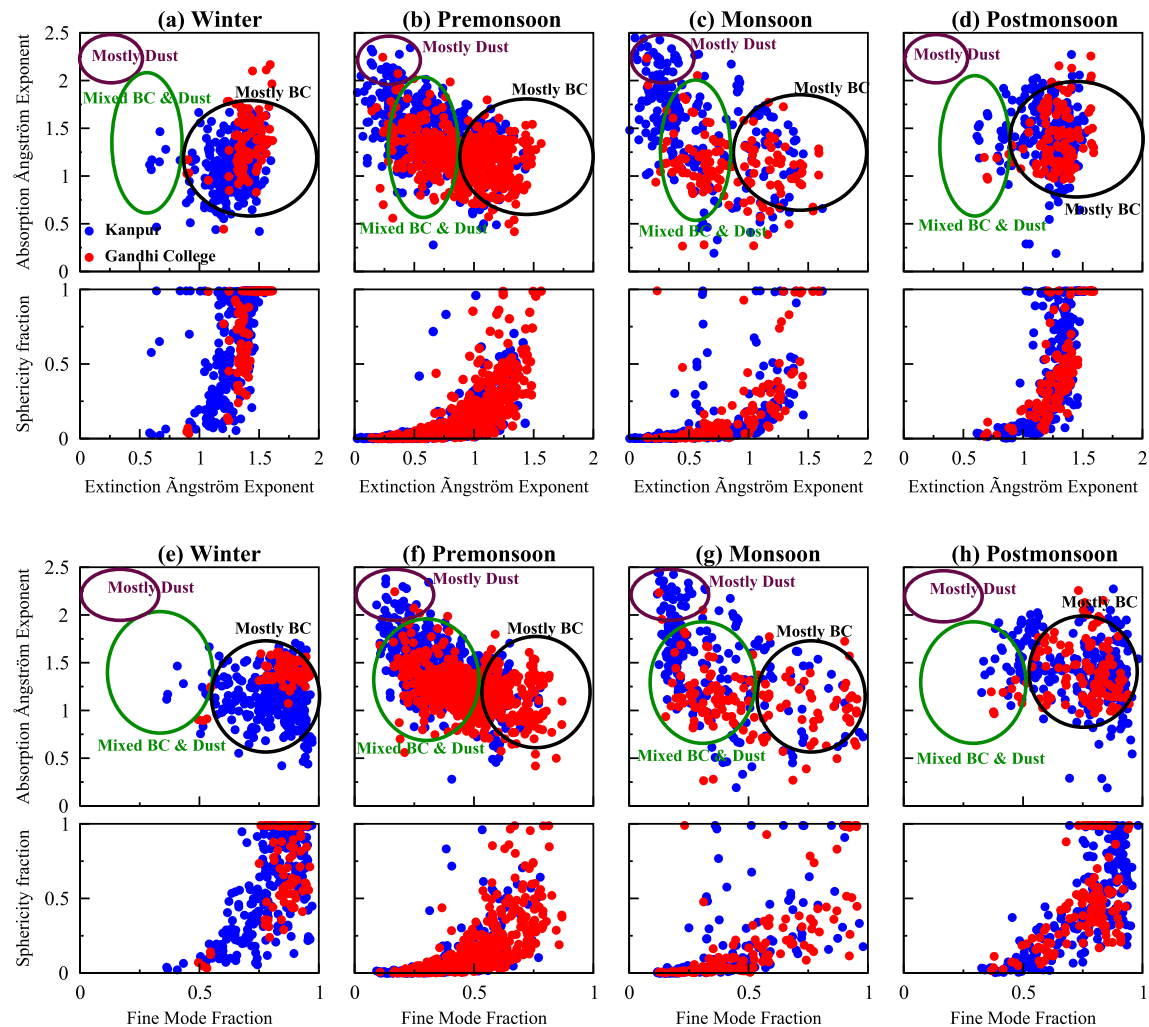
Organic carbon (OC) is produced from fossil fuel, biofuel and biomass burning, and natural biogenic emissions. Organic material is found to be the dominant component of biomass burning aerosols (Artaxo et al., 1998). OC is mainly a scattering type aerosol. However, OC particles that are freshly emitted from biomass burning emissions can contribute to absorption in the ultraviolet and blue wavelength regions of the spectrum (Kirchstetter et al., 2004). Arola et al. (2011) attributed the enhanced absorption at 0.44  $\mu\text{m}$  in several AERONET sites to organic carbon. They also pointed out that some amount of this enhanced absorption could be associated with dust contamination. Arola et al. cautioned that there exists significant uncertainty in their OC retrievals mainly due to assumptions made on the refractive index and density of OC. Higher OC concentrations over the IGP are also likely to be affected by urban pollution (Arola et al., 2011). SSA for biomass burning

aerosols decreased rapidly with increasing wavelength in the spectral region of 0.30–1.5  $\mu\text{m}$  (Russell et al., 2010). SSA for biomass burning aerosols at 0.5  $\mu\text{m}$  was about 0.87 and decreased to  $\sim 0.77$ . However, over Gandhi College SSA increases as a function of wavelength (Fig. 7) even during premonsoon when the fire counts from biomass burning emissions are the highest (Table 3), and therefore the abundance of OC is expected to be higher. SSA values are comparable over Kanpur and Gandhi College during winter, while in the other seasons SSA in Gandhi College is lower than that of Kanpur in the 0.44–1.02  $\mu\text{m}$  spectral region (Fig. 7), which confirms that the aerosols over Gandhi College are more absorbing. However, the near similar values of SSA at 0.44  $\mu\text{m}$  (Fig. 7) in Kanpur (urban) and Gandhi College (rural), and pattern of spectral dependence of SSA in Gandhi College suggests that the enhanced absorption due to OC in Gandhi College may not be significant, and that the enhanced absorption resulting in lower SSA in Gandhi College occurs due to the dominance of Mostly BC, followed by Mixed BC and dust (Table 6).

These results on the quantification of absorbing aerosol type have both radiative and climatic implications. The amount of absorbing aerosols over a regional aerosol hotspot and the gradient will be important in quantifying the anthropogenic aerosols as there exists no concurrence on the anthropogenic absorption AOD (IPCC, 2013 and references therein), and the amount of dust, its mixture with BC and its gradient is crucial for monsoon rainfall and hydrological cycle. The absorbing aerosols can heat the lower atmosphere (from near the surface to up to 4 km) during premonsoon and monsoon (Ramachandran and Kedia, 2010). The aerosols can re-emit this heat as thermal radiation, thereby heat the air mass and may cause evaporation of cloud droplets (*semi-direct effect*) (IPCC, 2013), and can influence the hydrological cycle and monsoon rainfall. It has been postulated that the absorbing aerosols (dust mixed with BC) over the Indo-Gangetic Plain in the premonsoon season can heat the lower atmosphere; this aerosol heating accompanied with the anti-cyclone circulation over the Tibetan plateau can result in advancement of the monsoon rainy season over north India and can subsequently lead to a higher rainfall over India (Lau and Kim, 2006). Further Lau et al. (2008) pointed out that the above is a working hypothesis and relies on specific assumptions regarding aerosol types, their horizontal and vertical distribution, and radiative properties; all of these need to be validated by more detailed observations on aerosol type, mixing and further numerical experiments to test the sensitivity of mixing to aerosol forcing. The present results are important to perform the sensitivity studies and to determine the spatial and temporal extent of aerosol impact on hydrological cycle and monsoon rainfall as it is shown clearly that the amount of dust, and dust mixed with BC do exhibit spatiotemporal variations between Kanpur and Gandhi College which are in IGP and separated only by about 500 km.



**Fig. 7.** Seasonal mean spectral variation in single scattering albedo (SSA) and  $\pm 1\sigma$  standard deviation in the wavelength range of 0.44–1.02  $\mu\text{m}$  over Kanpur and Gandhi College during winter, premonsoon, monsoon and postmonsoon of 2006–2010.



**Fig. 8.** Absorption Ångström exponent and sphericity fraction as a function of extinction Ångström exponent and fine mode fraction of AOD at  $0.675 \mu\text{m}$  over Kanpur (a, b, c, d) and Gandhi College (e, f, g, h) respectively. The boxes in different colors represent categories of absorbing aerosols (Mostly Dust, Mostly BC, and Mixed BC and Dust) over IGP. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

These results confirm that black carbon aerosols contribute dominantly to absorption over the entire IGP during winter and postmonsoon, while during premonsoon and monsoon the contribution of BC to the total aerosol decreases significantly. Kanpur and Gandhi College located in IGP though influenced by similar meteorological patterns, are governed by different aerosol types during different seasons. Both fine and coarse mode aerosols remain abundant throughout the year in varied proportion resulting in a bimodal aerosol distribution and non-linear AOD spectra over both the locations. During the entire study period and for all the seasons, aerosols seem to be more anthropogenic and more absorbing in nature over Gandhi College when compared to Kanpur, even though it is classified as a rural site and Kanpur is an urban site. This is due to the higher concentration of fine mode absorbing aerosols over Gandhi College, a rural site, which are emitted locally because of significant biomass/biofuel burning emissions, and from long range transport of fine mode anthropogenic aerosols as Gandhi College is situated downwind of major urban cities. Dust is more abundant over the entire IGP during premonsoon due to its proximity to and transport from the Thar desert (Fig. 2), however, amount of dust shows a gradient – it is higher over Kanpur when compared to Gandhi College. The dust

concentration remains higher over Kanpur during all the seasons than that of Gandhi College. This detailed analysis of spatiotemporal variations in aerosol characteristics over IGP region confirms the existence of large heterogeneity in aerosol properties over these two locations which must be taken into account in aerosol radiative forcing estimates and climate impact assessment.

## 5. Summary and conclusions

Optical and physical characteristics of aerosols are examined over two contrasting locations, Kanpur (urban, industrialized location) and Gandhi College (rural site) in Indo-Gangetic plain during 2006–2010 using AERONET data. Spectral distribution of aerosol optical depths (AODs) are examined by deriving the Ångström exponent ( $\alpha$ ) in the  $0.44\text{--}0.87 \mu\text{m}$  spectral range, in narrow spectral ranges ( $0.44\text{--}0.50 \mu\text{m}$  and  $0.675\text{--}0.87 \mu\text{m}$ ), and by using the coefficients of second order polynomial fit. Types of absorbing aerosols present over IGP and the dominant aerosol mode that governs the aerosol size distribution and spatiotemporal variability of optical and physical properties over the two locations are quantified.

The major findings of the study are:

**Table 6**

Percentage contribution of different aerosol types to absorption based on the analysis of absorption and extinction Ångström exponents, fine mode fraction at 0.675  $\mu\text{m}$  and sphericity fraction over Kanpur and Gandhi College.

Location	Aerosol type	Winter	Premonsoon	Monsoon	Postmonsoon
Kanpur	Mostly Dust	0	1	5	0
	Mostly BC	60	2	6	59
	Mixed BC and Dust	40	97	89	41
Gandhi College	Mostly BC	82	8	13	68
	Mixed BC and Dust	18	92	87	32

- (1) Monthly mean AOD at 0.5  $\mu\text{m}$  ranged between 0.39 and 1.01 (annual mean  $0.62 \pm 0.19$ ), and between 0.37 and 0.87 (annual mean  $0.62 \pm 0.16$ ) over Kanpur and Gandhi College respectively, and exhibit prominent seasonal variation.
- (2) Both FMF and  $\alpha$  are higher over Gandhi College than Kanpur indicating higher fine mode aerosol concentration over Gandhi College throughout the year. FMF values are higher over Gandhi College (annual mean  $0.71 \pm 0.18$ , median 0.71) than Kanpur (annual mean  $0.63 \pm 0.22$ , median 0.63). Ångström parameters  $\alpha$  and  $\beta$  varied in the range of 0.41–1.37 and 0.19–0.51 over Kanpur, and 0.64–1.46 and 0.19–0.48 over Gandhi College respectively. Frequency distribution of AOD,  $\alpha$  and FMF corroborate the dominance of fine mode aerosols during winter and postmonsoon, and mixed (both fine and coarse mode) aerosols during premonsoon and monsoon.
- (3) An analysis of MODIS fire counts reveals that it is the local biomass burning emissions over rural Gandhi College and not the local dust emissions that give rise to higher FMF than the urban Kanpur. Fire counts are higher over Gandhi College than Kanpur, and peak during premonsoon which is consistent with the spring time agricultural waste/residue burning over the IGP. This classification becomes important over the IGP where the biomass and biofuel sources co-exist in close vicinity of fossil fuel burning sources.
- (4) Spread in the differences of  $\alpha$  values determined in narrow wavelength bands confirmed that the aerosol size distribution is multimodal. The differences between the coefficients of polynomial fit,  $\alpha_2$  and  $\alpha_1$ , revealed the presence of wide range of fine mode fractions or mixture of modes during winter and postmonsoon over both the locations. The coarse mode aerosol concentration ( $\alpha_2 - \alpha_1 \leq 1 > 50\%$ ) increases during premonsoon and monsoon over both the locations. The coarse mode aerosol concentration is found to be higher over Kanpur than Gandhi College throughout the year as confirmed from a higher percentage of  $\alpha_2 - \alpha_1 \leq 1$  over Kanpur than Gandhi College throughout the year.
- (5) SSA showed strong seasonal and wavelength dependence over both the locations. During winter SSA decreases with wavelength consistent with the presence of absorbing aerosols, while during postmonsoon SSA is almost linear with wavelength in the 0.4–1.02  $\mu\text{m}$  wavelength region, however, SSA is lower in Gandhi College. This is due to the higher concentration of fine mode absorbing aerosols over Gandhi College than Kanpur due to admixture of both urban and rural emissions. An increase in SSA with wavelength over Kanpur and Gandhi College during premonsoon and monsoon is observed which is attributed to the presence of coarse mode dust which is scattering in nature, however, SSA values are lower in Gandhi College confirming the abundance of absorbing aerosols when compared to Kanpur. Near similar values of SSA in the blue wavelength region over

Kanpur and Gandhi College, and the spectral pattern of SSA indicates that the enhanced absorption due OC aerosols over Gandhi College (a rural site where biomass burning emissions are abundant) is negligible.

- (6) An analysis of aerosol absorption characteristics (absorption Ångström exponent) and size information (extinction Ångström exponent, sphericity fraction, and fine mode fraction) reveals that Mostly BC or Mixed BC and Dust during winter and postmonsoon make up the absorbing aerosols, while Mixed BC and Dust are dominant absorbing aerosols during other seasons over both the locations.
- (7) The analysis of aerosol optical properties reveal that there exists a gradient in the amount of dust that gets transported across the IGP. The amount of coarse mode (dust particles) encountered over Kanpur throughout the year is higher when compared to Gandhi College which is dominated by fine mode (biomass/biofuel burning emissions, and transport of fossil fuel emissions from major urban centers).

The study in a robust manner brings out quantitatively the variation in aerosol optical and physical characteristics due to change in the contribution of natural and anthropogenic aerosols produced from different sources, formation mechanisms and long range transport. The uniqueness of the results lies in the fact that the two locations in the IGP exhibit characteristics that vary from urban-industrial and mixed, to desert type environments in different seasons. The results have radiative and climatic consequences, and will be useful in validating/tuning the aerosol composition obtained from satellites/models, and can serve as crucial inputs in modeling the radiative effects of aerosols.

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