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Inferring aerosol types over the Indo-Gangetic Basin from ground based sunphotometer measurements

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ABSTRACT

A discrimination of aerosol types over the Indo-Gangetic Basin (IGB) region during pre-monsoon period was made using multi-year ground based sun/sky radiometer measured aerosol products associated with the size of aerosols and radiation absorptivity. High dust enriched aerosols (i.e. polluted dust, PD) were found to contribute more over the central IGB station at Kanpur (KNP, 62%) as compared to the eastern IGB station at Gandhi College (GC, 31%) whereas vice-versa was observed for polluted continental (PC) aerosols, which contain high anthropogenic and less dust aerosols. Contributions of carbonaceous particles having high absorbing (mostly black carbon, MBC) and low absorbing (mostly organic carbon, MOC) aerosols were found to be 11% and 10%, respectively at GC, which was ~46% and 62% higher than the observed contributions at KNP; however, very less contribution of non-absorbing (NA) aerosols was observed only at GC (2%). Variability in aerosol types together with single scattering albedo (SSA) at both the stations were also studied during the forenoon (FN) and afternoon (AN) hour, which suggests their strong association with emission sources. Results were well substantiated with the air mass back-trajectories and the fire products. Spectral information of SSA for each aerosol type discriminates the dominance of natural dust (SSA increases with increasing wavelength) with anthropogenic aerosols (SSA decreases with increasing wavelength) at both the locations. The estimated absorption Ångström exponent (AAE) values suggest relative dominance of absorbing type aerosols over the central part of IGB (due to dominant dust absorption) as compared to the eastern part during pre-monsoon period.

1. Introduction

The climatic and environmental effects of atmospheric aerosols are the critical issues in global science community because aerosols, derived from natural and anthropogenic emission sources, are well known to affect the air quality, human health and radiation budget (Pöschl, 2005; IPCC, 2007). Uncertainty in quantifying the climatic impacts of aerosols continues to be greater than that of greenhouse gases (IPCC, 2007) due to variety of their sources, varying trends in aerosol loading and extreme heterogeneity in the spatial and temporal variability of their optical and microphysical properties (Morgan et al., 2006; Kaskaoutis et al., 2007). Accurate assessment of the aerosol impacts on radiative forcing is a complex task, since various aerosol types cause different effects on the solar radiation (Kaskaoutis and Kambezidis, 2008) and also have different effects on the sign and magnitude of the aerosol radiative forcing (Heintzenberg et al., 1997; Satheesh and Moorthy, 2005). Thus, detailed knowledge of the optical properties of the key aerosol types is essential.

The Indian subcontinent is one of the densely populated, industrialized and developing regions where aerosols not only affect the Indian monsoon but also the global climate

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system (Satheesh et al., 2006). It is one such region, where heterogeneity in aerosol optical and microphysical properties over a wide range of spatial and temporal scales continues to hinder in improving the estimates of aerosol-induced climate forcing. Thus, it is important to improve aerosol characterization with high spatio-temporal resolutions; particularly over the Indo-Gangetic Basin (IGB) region, which supports nearly 70% of the country's population and is one of the highly polluted regions of the world. The problem is also critical due to lack of adequate long-term measurements of aerosol properties and large uncertainty in emission factors, leading to poor representation of aerosol distribution by General Circulation Model (GCM) (Ganguly et al., 2009). Although, aerosol properties have been measured at many sites in India in continuous and campaign modes (Dey and Girolamo (2010) have summarized the in-situ observations) in the last two decades, only few of them have fairly long-term data of aerosol microphysical properties (Moorthy et al., 2007; Devara et al., 2008; Dey and Tripathi, 2008). Satellite data can provide the required space-time coverage of aerosol optical depth (AOD) but the retrieval of aerosol absorption (in form of single scattering albedo, SSA) is not reliable compared to the ground based measurements (Dey and Girolamo, 2010), which calls for better representative aerosol composition/types for this region.

In this paper, we focused on the Ganga basin region where AOD is highest within the subcontinent. The region is of great research interest due to its unique topography surrounded by the Himalayas to the north, moderate hills to the south, Thar Desert and Arabian Sea in the west, and Bay of Bengal in the east. It is dominated by the urban/industrial aerosols (Guttikunda et al., 2003; Sharma et al., 2003; Monkkonen et al., 2004; Tiwari et al., 2009), which demonstrate significant seasonal variability based on the complex combination of anthropogenic factors mixed with the contribution from the natural sources (mostly dust), particularly during the pre-monsoon period. Recent studies over the region, have documented the seasonality in aerosol characteristics and direct radiative forcing (Dey and Tripathi, 2008; Srivastava et al., in press) based on in-situ measurements. A recent study over the IGB region with the Aerosol Robotic Network (AERONET) data revealed significant gradient in various aerosol characteristics from central to the eastern sectors of the IGB during pre-monsoon period, which suggests diverse nature of aerosols and their effects over the region (Srivastava et al., 2011a). Mixing of natural dust with anthropogenic particles during this period, has been hypothesized (Dey et al., 2008) in the IGB and corroborated with the AERONET data (Giles et al., 2011), further complicating the satellite retrieval of aerosol characteristics and quantifying the climatic effects (Mishra et al., 2010). Hence, it is important to understand the diurnal and spatial variability of aerosol types in the IGB to make realistic assessments of aerosol–hydroclimate interplay.

2. Site description and experimental set-up

The present study involves aerosol products from the CIMEL sun/sky radiometer measurements at two stations over the IGB region: (i) Kanpur, hereinafter referred as KNP (26.4°N, 80.4°E) and (ii) Gandhi College, hereinafter referred as GC (25.8°N, 84.2°E). The sun/sky radiometers deployed at these stations are as a part of AERONET/TIGERZ Program of NASA, USA. KNP is one of the highly polluted cities in Asia situated in the central part of the IGB. The AERONET site is located in the campus of Indian Institute of Technology Kanpur (IITK), which is northwest to the main industrial region of KNP. On the contrary to KNP, GC AERONET site is located in Ballia at the eastern part of the IGB, which is a rural–urban fringe site. Aerosol emissions over the IGB result from biomass burning in open field (e.g. agricultural waste), domestic fuel in rural settings (e.g. wood and dung cakes) and emissions from fossil fuel sources such as diesel and kerosene mixed-fuel vehicles and various industrial processes (Habib et al., 2006; Prasad et al., 2006; Tiwari et al., 2009; Eck et al., 2010; Ram and Sarin, 2010; Ram et al., 2010; Srivastava et al., 2011a, in press). Previous studies over the IGB showed distinct seasonal patterns of aerosol properties controlled by the northeast and summer monsoon (Singh et al., 2004; Jethva et al., 2005; Dey et al., 2005; Eck et al., 2010).

The CIMEL sun/sky radiometer measures the direct Sun radiances at eight spectral channels (0.34, 0.38, 0.44, 0.5, 0.67, 0.87, 0.94 and 1.02 μm), where 0.94 μm channel is used to estimate the columnar water vapor content, but the remaining channels are used to retrieve spectral AODs. On the other hand, almucantar sky radiance measurements at four spectral channels (0.44, 0.67, 0.87 and 1.02 μm) are used to retrieve size distribution, single scattering albedo (SSA) and refractive indices of the aerosols (Holben et al., 1998). The processed aerosol related data are available on-line at the AERONET site (http://aeronet.gsfc.nasa.gov/) in three categories: cloud contaminated (level 1.0), cloud screened (level 1.5) and quality assured (level 2.0) (Smirnov et al., 2000). Level 2.0 data are the final AERONET product, which have been utilized for the present study during the pre-monsoon period (April–June) in 4 years (2006–2009), where 103 and 168 days data were available at KNP and GC, respectively. The pre-monsoon period is of particular interest because this is the key period when locally generated and regionally transported aerosol loading peaks over the IGB region and spread up to the foothills of Himalayas (Venkataraman et al., 2005; Eck et al., 2010; Arola et al., 2011; Devi et al., 2011; Gautam et al., 2011; Srivastava et al., 2012a), which has been linked to influence the monsoon circulation in India (Lau et al., 2006, 2010). Due to high level of anthropogenic emissions, aerosol distribution in terms of type and loading undergo strong variability associated with the episodic yet strong influence of dust transport and biomass burning during the pre-monsoon period (Gautam et al., 2011). Dust was found to be one of the major components of aerosol composition (apart from other species) over the region (Chinnam et al., 2006), which significantly affects the region during pre-monsoon period due to enhanced surface convection activities (Dey et al., 2004; Pandithurai et al., 2008; Gautam et al., 2009; Eck et al., 2010; Giles et al., 2011; Srivastava et al., 2011a, 2011b, 2012b), and thus essential to quantify its contribution over the region. The total uncertainty in the retrieved parameters is discussed elsewhere (Dubovik et al., 2000; Holben et al., 2001; Ogunjobi et al., 2008; El-Metwally et al., 2011).

3. Synoptic conditions over IGB

Synoptic meteorology (wind pattern, air temperature and specific humidity) over the IGB region (measurement

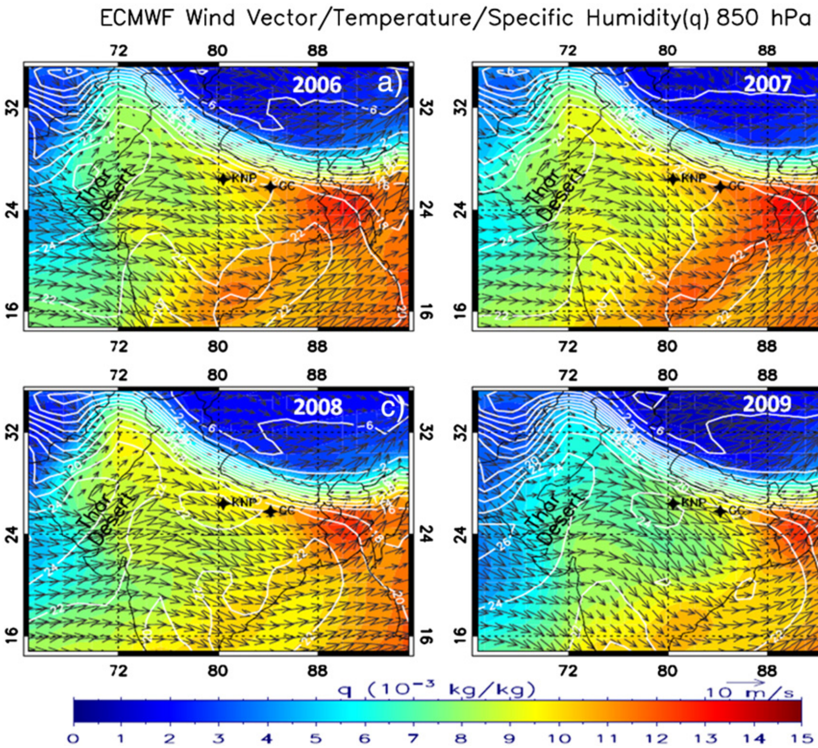

Fig. 1. Synoptic meteorological conditions derived from ECMWF at 850 hPa pressure level over the entire IGB region during the pre-monsoon period of the year (a) 2006, (b) 2007, (c) 2008 and (d) 2009. The location of measurement sites KNP and GC are marked by the star.

locations are shown by stars) along with its surroundings during the per-monsoon period from 2006 to 2009 are shown in Fig. 1 (a–d), respectively. The European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis monthly data of weather parameters such as wind, air temperature, and specific humidity at 850 hPa pressure level were used to study the synoptic meteorological conditions over the stations. In all the figures, winds are shown with arrows pointing toward the wind direction, where length of arrows defines the magnitude of wind speed (in ms^{-1}), line contour represents air temperature (in °C) and shaded color contour represents specific humidity (in kg kg^{-1}) (shown in blue color for low and red for high magnitude). A west to east temperature gradient is revealed from the figures in each year, which is more pronounced during 2008 and 2009, and may influence the increase of pressure gradient. This pressure gradient caused winds to be intensified from west to eastward. Further, results reveal that the study region over IGB is generally characterized by northwesterly and westerly to southwesterly winds during the pre-monsoon period, intensifying from 2006 to 2009. The study region over IGB is found to be relatively drier during 2008–2009 than during 2006–2007.

4. Methodology

Aerosol composition was measured with the chemical analysis method over IGB during different periods of time (Tare et al., 2006; Tiwari et al., 2009; Ram and Sarin, 2010). Retrieval of columnar black carbon and organic carbon has been carried out over IGB using the AERONET data (Dey et al., 2006; Arola et al., 2011). Moreover, Ganguly et al. (2009) have integrated AERONET and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) data into atmospheric GCM to infer aerosol types at two AERONET sites in the IGB. However, in the present study, we adopted an approach to infer aerosol types (in terms of major air mass types) over

Table 1
Major aerosol categories along with the mean (±1σ) of AOD, AE and AAE at KNP and GC.

Major aerosol composition[a]	$AOD_{(0.5)}$ (at KNP)	$AOD_{(0.5)}$ (at GC)	AE (at KNP)	AE (at GC)	AAE (at KNP)	AAE (at GC)
PD	0.66 ± 0.18	0.63 ± 0.21	0.43 ± 0.26	0.45 ± 0.12	1.70 ± 0.31	1.30 ± 0.26
PC	0.68 ± 0.18	0.72 ± 0.22	0.79 ± 0.14	0.78 ± 0.12	1.43 ± 0.30	1.18 ± 0.29
MBC	0.68 ± 0.12	0.70 ± 0.24	1.09 ± 0.06	1.12 ± 0.14	1.28 ± 0.14	1.09 ± 0.32
MOC	0.88 ± 0.31	0.80 ± 0.23	1.14 ± 0.06	1.24 ± 0.13	1.31 ± 0.21	0.99 ± 0.32
NA	–	1.22 ± 0.30	–	1.15 ± 0.12	–	0.51 ± 0.12

[a] PD—polluted dust, PC—polluted continental, MBC—mostly black carbon, MOC—mostly organic carbon and NA—non-absorbing.

the IGB during pre-monsoon period based on various parameters retrieved by AERONET.

Our approach to infer major aerosol types is based on the combination of particle size and absorption/scattering information, following Lee et al. (2010). CIMEL sun/sky radiometer derived fine mode fraction (FMF) at 0.5 μm has been used to represent the dominant aerosol size mode, because FMF provides quantitative information for both fine- and coarse-mode aerosols. SSA has been used to quantify the aerosol absorption/scattering at 0.44 μm, which is the shortest wavelength of AERONET channels and generally used to distinguish absorbing from non-absorbing aerosols (Lee et al., 2010). On contrary to the present method, other methods were also established to characterize the aerosol types at various locations (Eck et al., 1999, 2010; Kaskaoutis et al., 2009 and references therein), which are mainly based on the combination of AOD and Ångström exponent (AE). It generally provides qualitative information due to AE, which is a qualitative indicator of particle size. However, FMF used in the present method, provides quantitative information for both fine- and coarse-mode aerosols (Lee et al., 2010).

Based on the seasonality of aerosol properties studied previously (Dey and Tripathi, 2008; Srivastava et al., in press-a) and emission factors (Streets et al., 2003), the following major categories of aerosols (Table 1) are expected at both the stations over IGB: polluted dust (PD, dominantly dust mixed with anthropogenic particles), polluted continental (PC, dominantly anthropogenic mixed with dust), mostly black carbon (MBC, highly absorbing carbonaceous particles), mostly organic carbon (MOC, low absorbing carbonaceous particles) and non-absorbing (NA) fine-mode aerosols (e.g. sulfate, nitrate etc.).

Further, the absorption aerosol optical depth (AOD_{abs}) at different wavelengths (λ) for both the stations during study period can be obtained as suggested by Russell et al. (2010)

$$AOD_{abs}(\lambda) = [1-SSA(\lambda)] \times AOD(\lambda) \quad (1)$$

The absorption Ångström exponent (AAE) has been computed as the negative of the slope of the fitted line of the natural logarithm of AOD_{abs} vs. the natural logarithm of four retrieval wavelengths (0.44, 0.67, 0.87 and 1.02 μm) and used to substantiate the inferred aerosol types over IGB. In general, the mean AOD for PD, PC and MBC type aerosols are by-and-large similar at both the stations, and are lower relative to MOC and NA aerosols (only at GC) as depicted in Table 1. Results thus suggest an important role of meteorology in dispersing the pollutants across the IGB during this period. However, the variability in aerosol loading is much higher at GC compared to KNP.

5. Results and discussion

5.1. Discrimination of aerosol types

Fig. 2a shows density plot of SSA versus FMF at KNP and GC for different aerosol types. A wide range of SSA was observed at GC as compared to KNP for each aerosol type. However, wide range of FMF at GC was observed only for MBC, MOC and NA. Results suggest relatively large variations in the emission sources of aerosols reaching at GC as compared to KNP. The mean ($\pm 1\sigma$) SSA and FMF based on cluster analysis of daily-averaged data at KNP and GC, associated with the above five aerosol categories are also shown in Fig. 2b. The horizontal and vertical lines indicate the standard deviations of SSA and FMF, respectively from their respective means, indicating the variability of these parameters for different aerosol types. Although similar magnitude of SSA was observed for PD, PC and MBC type aerosols, they are further distinguished based on FMF thresholds following Lee et al. (2010), i.e. FMF<0.4 indicates dominantly coarse-mode and hence is assigned to PD aerosols, FMF>0.6 indicates dominantly fine-mode and hence is assigned to MBC aerosols, and PC aerosols are considered for 0.4≤FMF≤0.6. MOC and NA type aerosols have similar FMF, but higher scattering relative to the other aerosol types. The contributions of PD, PC, MBC and MOC type aerosols were found to be ~62%, 28%, 6% and 4%, respectively at KNP and ~31%, 46%, 11% and 10%, respectively at GC. Moreover, very less contribution of NA aerosols was found only at GC (~2%). Results depict that the contribution of PD is ~50% higher at KNP as compared to their contribution at GC whereas the contributions of PC, MBC and MOC are found to be higher at GC by ~39%, 46% and 62%, respectively than those observed at KNP. Recently, Kumar et al. (2012) have carried out a

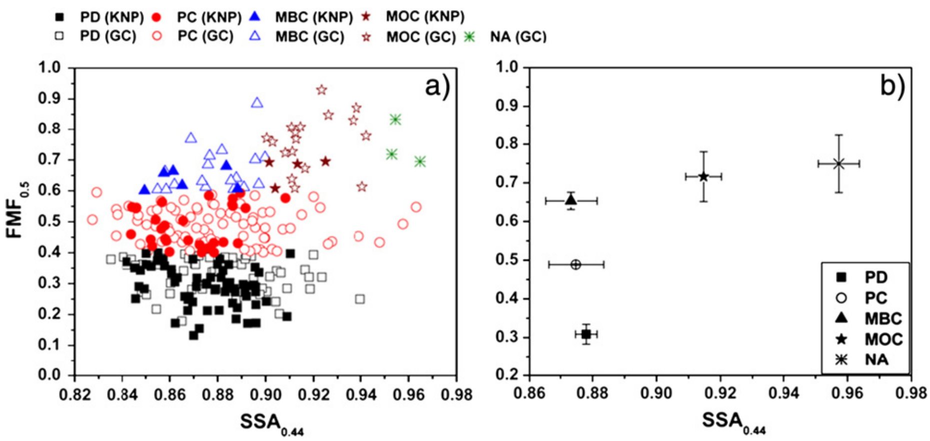

Fig. 2. (a) Density plot and corresponding (b) cluster plot of AERONET-derived SSA vs. FMF for two stations over the IGB region (showing different aerosol types) during pre-monsoon period.

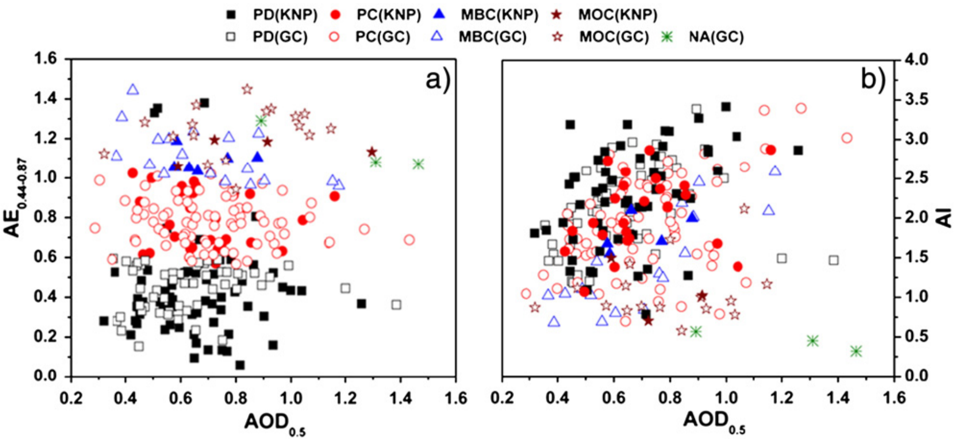

Fig. 3. (a) Same as Fig. 2a for AOD vs. AE, (b) same as Fig. 2a for AOD vs. AI for different aerosol types.

detailed analysis of the various aerosol properties from AERONET at multi- environmental sites in India, having dominance of biomass burning, urban/industrial pollution, marine origin and Desert dust aerosols and demonstrated their impacts on the regional aerosol radiative forcing and heating rates.

A density plot of AOD versus AE was also done for KNP and GC, associated with the five aerosol categories mentioned above, as shown in Fig. 3a. Figure showed a clear discrimination of AE for PD (contains mostly coarse dust particles), PC (contains mostly fine particles) and the remaining aerosol types (such as MBC, MOC and NA) at both the stations. The magnitude of AOD (corresponding AE) was found to be about 0.66 ± 0.18 (0.43 ± 0.26), 0.68 ± 0.18 (0.79 ± 0.14), 0.68 ± 0.12 (1.09 ± 0.06) and 0.88 ± 0.31 (1.14 ± 0.06) at KNP and 0.63 ± 0.21 (0.45 ± 0.12), 0.72 ± 0.22 (0.78 ± 0.12), 0.70 ± 0.24 (1.12 ± 0.14), 0.80 ± 0.23 (1.24 ± 0.13) and 1.22 ± 0.30 (1.15 ± 0.12) at GC, respectively for PD, PC, MBC, MOC and NA aerosol types. In another study, Kaskaoutis et al. (2009) have studied seasonal characteristics of aerosols and their types over a tropical urban station at Hyderabad using Sun photometer measurements. They have classified the aerosols, based on threshold values of AOD and AE, as marine influenced (MI), urban/industrial aerosols under high AOD (HUI), Desert dust aerosols under high AOD (HDD) and mixed type (MT). Although these aerosol classes are not exactly the same as mentioned in the present study, they are close enough for a rough comparison. The overall behavior of aerosol types such as PD, PC and MBC, MOC, NA together in the present study was found to be comparable with the HDD, MT and HUI aerosol types, respectively of Kaskaoutis et al. (2009) during pre-monsoon period. However, some discrepancies can be expected due to different geographic locations, synoptic meteorological conditions and the emission sources at these locations. Further, a density plot of AOD versus AI, associated with the five aerosol categories was done for both the stations and shown in Fig. 3b. It is to be noted that the magnitude of AI increases with increasing AOD at both the stations for PD, PC and MBC aerosol types; however, not much change in AI was observed for other aerosols.

5.2. Diurnal variation of aerosol types

The diurnal variability of aerosol types is critical to understand the climatic effects. There may be some sampling biases in the forenoon (FN) or afternoon (AN) measurements due to different sky conditions and also possibly due to ambiguity in the solar zenith angle (SZA). The SZAs at KNP and GC during

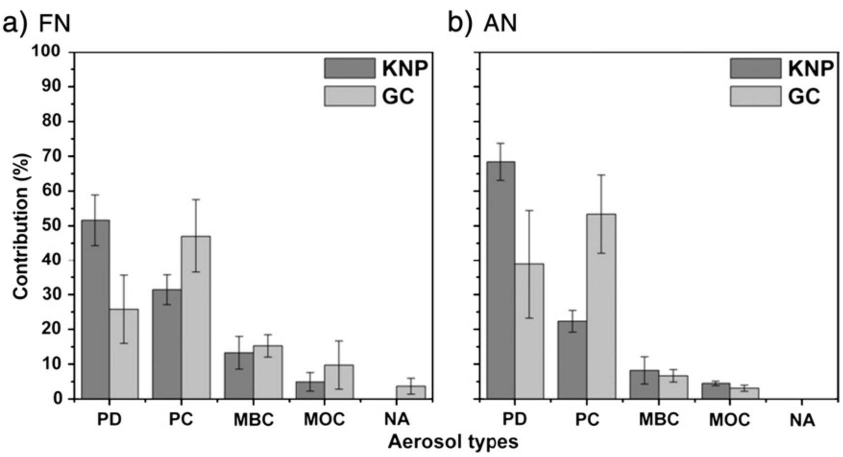

Fig. 4. Percentage contribution of different aerosol types at KNP and GC during (a) forenoon (FN) and (b) afternoon (AN) hours.

the FN and AN sampling periods are in the range 50–60°, suggesting that the SZA may not be a major factor. Further, sampling biases due to different sky conditions in the FN and AN periods may also be reduced in the present analysis as large sample density was found for the respective sampling periods at both the stations. To reduce the above uncertainty, we have done cluster analysis of the relative contribution of inferred aerosol types at KNP and GC for the entire FN and AN periods instead of individual time period and shown in Fig. 4a and b, respectively. The FN samples were clustered between the time period from 06:30 to 08:30 IST, which were found to be 162 and 224 respectively for KNP and GC. The AN samples were clustered between the time period from 15:30 to 18:00 IST, and which were found to be 207 and 215 respectively for KNP and GC.

PD was found to be relatively higher (52 ± 7% and 68 ± 5% during FN and AN, respectively) at KNP than at GC (26 ± 10% and 39 ± 16% during FN and AN, respectively) whereas vice-versa was observed for PC. This may be attributed to the proximity of KNP to the dust source regions (e.g. Thar Desert and Middle-East Asia) as compared to GC. A gradient in dust particles from western (high) to eastern (low) parts of IGB has also been observed by satellite data (Dey and Girolamo, 2010) and ground based observations (Srivastava et al., 2011a). The expansion of boundary layer in the AN facilitates the transport of dust from source regions in the west, leading to an increase in relative occurrence of PD from FN at both these sites. While this has resulted in a decrease in relative occurrence of PC in the AN at KNP, no noticeable change is

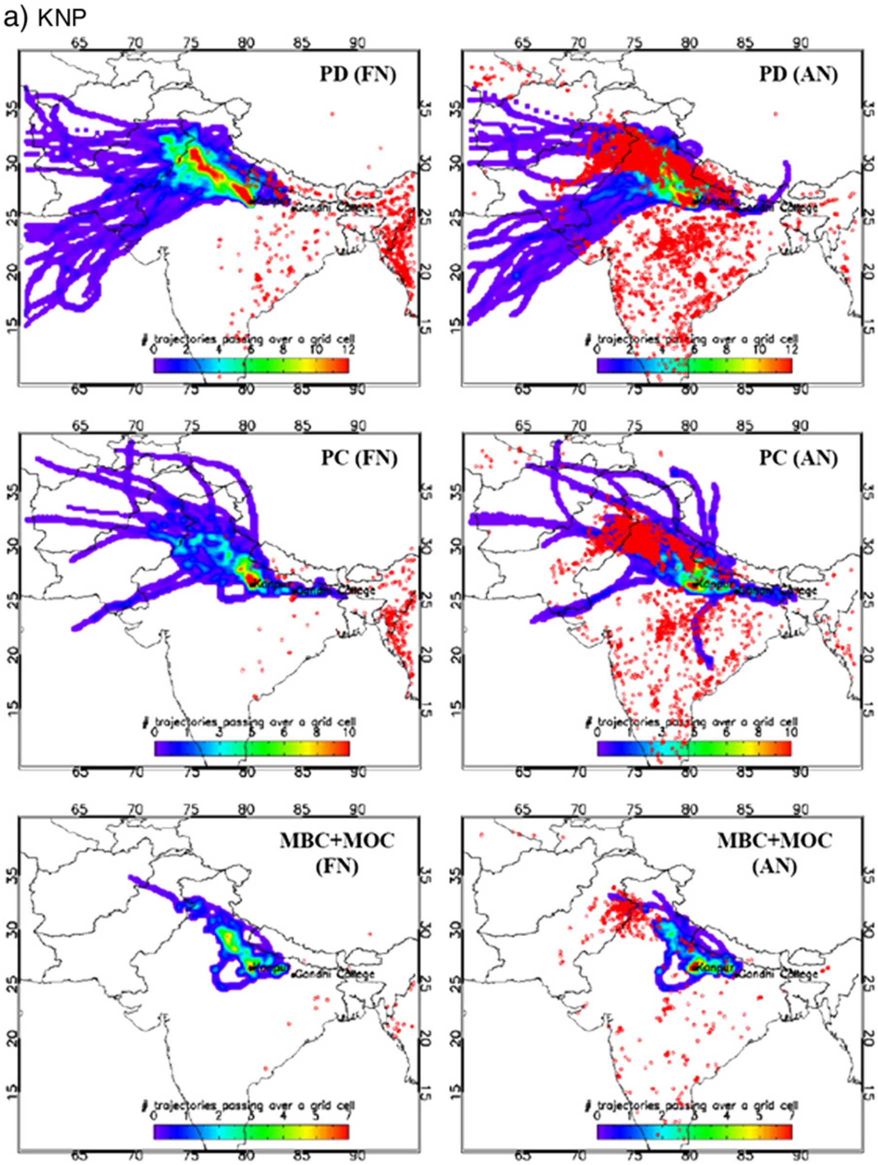

Fig. 5. a. Air mass back-trajectories and fire counts for the observed aerosol types during FN and AN for KNP. b. Same as Fig. 5a for GC.

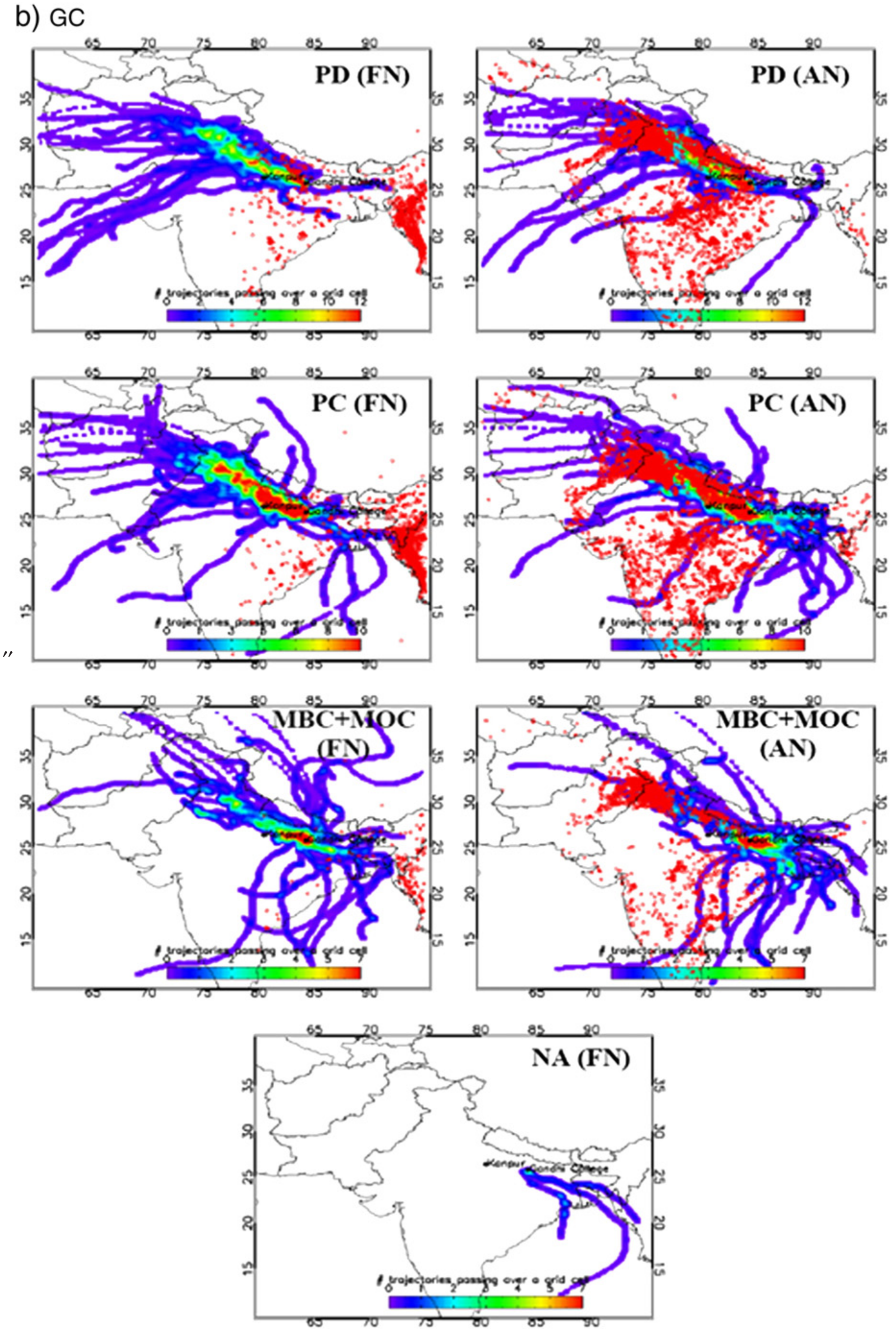

Fig.5 (*continued*).

observed at GC. Since GC site is at rural area, dust particles raised from local soils (Dey and Girolamo, 2010) may compensate for the loss of dust particles transported from long range. The contributions of MBC and MOC were found to be as compared higher during FN relative to AN at both the stations. NA type aerosols were observed only at GC in the FN hour. The corresponding mean value of SSA (at 0.44 μm) was also examined during FN and AN hour at both the stations. It is noted that no significant change in SSA was observed for PC and MOC during FN and AN whereas a slight decrease in the magnitude of SSA was observed for PD from FN (0.88 ± 0.01) to AN (0.87 ± 0.01) at KNP, which was found to be opposite at GC. Similar feature with slightly less magnitude of SSA was observed for MBC at both the stations during FN and AN hour.

However, relatively scattering particles having large magnitude of SSA (0.96 ± 0.01) was found for NA, which was the case observed only at GC in the FN hour.

5.3. Source and transport analysis

To understand the possible sources of these aerosols at both the stations during FN and AF hour, 5-day air mass back-trajectories at 1000 m were studied respectively, at 02 GMT (for FN) and 11 GMT (for AN) using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration (Draxler and Rolph, 2003) and displayed corresponding to the inferred aerosol types (Fig. 5a for KNP and Fig. 5b for GC). The corresponding fire counts data product from the Moderate Resolution Imaging Spectroradiometer (MODIS), launched on Terra platform was also analyzed (Kaufman et al., 1998). It is superimposed on air mass back-trajectory plots to understand the possible transport pathways of biomass/forest fire burning aerosol sources from the surrounding regions to both the stations during the observed aerosol types during FN and AN hour. More details of the MODIS fire product and fire detection algorithm is given elsewhere (Justice et al., 2002). As aerosol measurements were done during the daytime from sunrise to sunset, fire data was classified as a mean between sunrise to 12 noon for FN and 12 noon to sunset for AN.

Harvesting of wheat system is usually done in the north-west Indian region during the pre-monsoon period, mainly in the months of May and June, which leaves behind large quantities of straw in the field as crop residues (Badarinath et al., 2009). These crop residues are subjected to open burning to clear the wastes and to prepare the field for sowing the rice system. The burning activities are generally done in the afternoon time, and the intensity of burning gets weakened as time progresses, which substantiate the observed difference in the number of fires, as shown in the figures during FN and AN hour at both the stations. Slightly different characteristics in air mass trajectories were observed for each aerosol type during FN and AN hour at both the stations, which are found to be well associated with the observed fire counts and the synoptic meteorological conditions prevailed in and around the stations.

During the period of observed PD aerosols at KNP, air masses are transported mostly from the Thar Desert and Middle East regions, which are effective dust sources for the Indian subcontinent because of the prevailing westerlies (Bollasina and Nigam, 2009; Dey and Girolamo, 2010). Relatively large spreading in air mass was observed during the AN, which results an increase contribution of dusts in PD aerosols. However, at GC, air masses are mostly confined over Thar Desert region during the AN hour. During the period when PC aerosols were observed, air masses at KNP are mostly from the north-west region, which has relatively large transport pathway during the FN. On the other hand, air masses at GC cover north-west to the central parts predominantly during the AN. Results are expected to be caused due to open field burning of harvested wheat straw, which is usually done in the northern India during the pre-monsoon period, and their long-rang transport at both the stations (Badarinath et al., 2009; Sharma et al., 2010). The influence of aerosols from forest-fires, occurs mostly over the central Indian region during pre-monsoon period (Ramachandran and Cherian, 2008), can also be expected at both the stations, which is predominantly at GC during AN.

Though there is a considerable difference in the absorption properties (i.e. SSA) of MBC and MOC type aerosols (as can be seen in Fig. 2b), it is very difficult to discriminate the sources of these aerosols based on air mass back-trajectories as they have mostly the common sources (Ito and Akimoto, 2007), and hence we have considered MBC and MOC type aerosols together for generating back-trajectories. Note that the air masses during the period of observed MBC and MOC aerosols at KNP are localized (high during AN) and transported from further north-west region, and can be expected from the combination of biomass burning and urban/industrial sources. On the other hand at GC, air masses cover large continental regions from north-west to the central part of India, and results a larger contribution (about two times higher) of MBC and MOC aerosols at GC than at KNP during the FN. Air mass trajectories show marine source of NA aerosols at GC during FN, which probably mix with the other continental aerosols during transportation.

The above results are highly associated with the synoptic meteorology over the region, discussed in the previous section, which revealed that the stations are generally characterized by westerly to southwesterly winds during the pre-monsoon period. These winds are found to pass through arid regions of the western India (particularly from the Thar Desert) and southwest Asia to bring dry air masses over the stations (Pandithurai et al., 2008; Srivastava et al., 2011a) and one of the significant sources for PD aerosol type at both the stations apart from the dust raised from local soils due to convection activities. Further, it is noticed from the figures that the stations are also characterized by north-westerly winds (Fig. 1) and are responsible for the transport of biomass/fossil fuel burning aerosols from the north-west region (major open field burning region of harvested wheat straw during pre-monsoon period), which highly influence the contribution of other aerosol types, e. g. PC, MBC and MOC at both the stations. KNP, being proximity to the Thar Desert region, is found to be affected by the relatively large amount of transported dust aerosols during the pre-monsoon period, as already discussed in the previous section, the probability of its mixing with black carbon aerosols can be expected to be relatively larger as compared to GC, which makes the investigation a real challenge (Das and Jayaraman, 2011) and one of the future perspectives of the present study.

5.4. Absorption characteristics of aerosols

Analysis of the aerosol spectral absorption (i.e. spectral SSA) was further carried out for the inferred aerosol types at KNP and GC (Fig. 6a and b, respectively). As expected, PD and PC aerosols show spectrally increasing SSA, suggesting the relative importance of dust. PD has higher spectral trend relative to PC due to larger fraction of dust, which was found to be dominated at KNP as compared to GC. On the contrary to PD and PC type aerosols at GC, NA aerosols show spectrally decreasing SSA with relatively larger magnitude at all the wavelengths. However, relatively less spectral dependence in SSA can be seen for MBC and MOC aerosols,

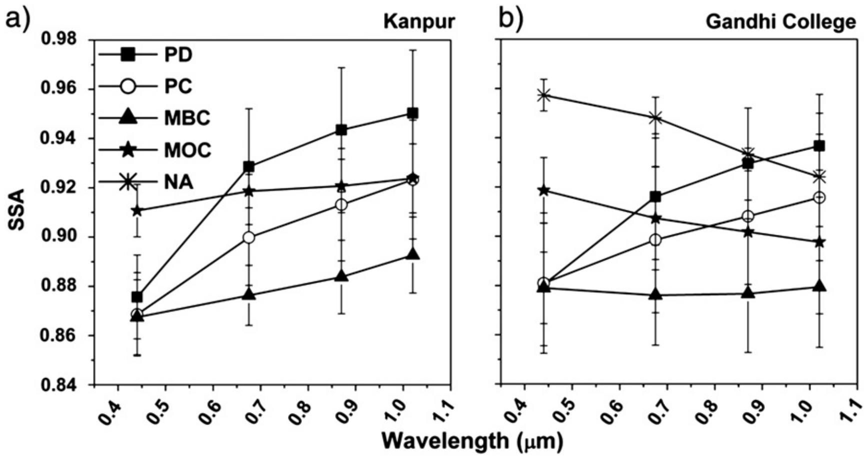

Fig. 6. Spectral SSA for different aerosol types at (a) KNP and (b) GC.

which shows slight decrease in spectral SSA at GC and opposite at KNP.

The estimated mean ($\pm 1\sigma$) AAE for each inferred aerosol class at both the stations are shown in Fig. 7 and also summarized in Table 1. The magnitude of AAE near to 1.0 (marked by dotted line in Fig. 7) represents a theoretical AAE value for black carbon (Bergstrom et al., 2007). AAE values for PD and PC aerosol types were found to be 1.70 and 1.43, respectively at KNP and 1.30 and 1.18, respectively at GC. However, for MBC and MOC type aerosols, AAE values were relatively higher at KNP (~20%) than at GC, where values were found to be closer to the theoretical AAE value for black carbon (i.e. AAE ≈ 1.0), thus indicating the presence of fresh BC at GC, which can be expected from the potential source of combustion of fossil fuel and biomass burning used for domestic purposes. On the other hand, aged BC or mixed BC can be expected at KNP (mostly from biomass burning and urban/industrial sources), which is favorable scenario during the summer months (Dey et al., 2008; Giles et al., 2011). Results obtained in the present study over the IGB are by and large consistent with those reported by Bergstrom et al. (2007) and Russell et al. (2010) near to the major emission source regions of these aerosols.

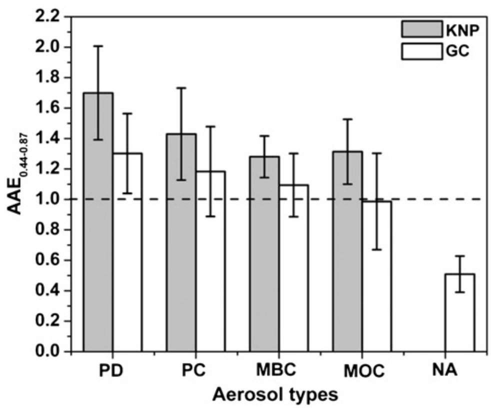

Fig. 7. Mean AAE values for each aerosol type at KNP and GC.

In a recent study over Delhi, Soni et al. (2010) have reported that the AAE value close to unity is due to the abundance of black carbon from fossil fuel burning whereas large AAE value (>1.0) indicates absorbing aerosols from biomass/biofuel burning. In the previous studies, Bergstrom et al. (2002) suggested that the small size BC aerosols ($r<0.01$ μm) with constant refractive index would have AAE of 1.0 (as observed over GC); however, optically effective and relatively large size BC aerosols ($r>0.01$ μm) with constant refractive index can have an AAE of 1.3 (as observed over KNP). The deviations in the magnitude of AAE from 1.0 is likely due to the spectral changes in the imaginary part of the refractive index as a result of the spectral absorption response of the aerosol particle related to its composition (Kirchstetter et al., 2004). Recently, Russell et al. (2010) have reported that the AAE value for organic species (sometimes called brown carbon) are larger than that for BC, which suggests for the present study that the sufficient amount of organic species are present at the central IGB region [consistent with Arola et al. (2011)] whereas sufficient amount of BC are at the eastern IGB region. The AAE value for NA type aerosols observed at GC is found to be less than 1.0, which suggests possible coating of non-absorbing substance over fine-particles, particularly over BC (Gyawali et al., 2009).

6. Summary and conclusions

Aerosol types have been characterized using ground-based measurements of aerosol properties carried out by sun/sky radiometer at KNP and GC- typical representative stations in the central and eastern IGB region, during pre-monsoon period of 2006–2009. Five major categories of aerosols were identified over the IGB based on the size and absorption characteristics. Relatively large contributions of dust dominated aerosols (PD) were observed at KNP (~62%) as compared to GC (~31%) due to proximity of KNP to the Desert regions, which was found to be higher during AN at both the stations. However, vice-versa was observed for PC aerosols. Carbonaceous aerosols, i.e. MBC and MOC were found to be 11% and 10%, respectively at GC, which was ~46% and 62% higher than their contributions at KNP. Further, they were found to be relatively higher during FN at GC whereas opposite was observed during AN. For both the reference locations over IGB where different aerosol types were observed, spectral mean SSA was found to be different for each aerosol type, which is due to the influence of different emission sources. The estimated AAE values at both the stations suggest relative dominance of absorbing type aerosols over the central part of IGB (due to dominant dust absorption) as compared to the eastern part during pre-monsoon period. Results show a significant spatial difference in the characteristics of aerosol types from central to the eastern IGB region, which may lead to a considerable asymmetry in aerosol radiative forcing between the regions.

Because of the advantages of the global coverage of ground based AERONET stations, the present study can be used to compare and validate aerosol types derived from different model and satellite studies, as well as to analyze the characteristics of aerosol types on regional and global scales over the land where satellite observations need to be improved (Kleidman et al., 2005; Lee et al., 2010; Andrews et al., 2011; Badarinath et al., 2011; Mielonen et al., 2009). Due to large uncertainty in satellite derived aerosol products over the IGB during pre-monsoon dust periods, long-term measurements during different seasons can indeed provide useful information of the characteristics of aerosol types over the region on seasonal and inter-annual basis, which are meager and crucial for the regional climate models. Furthermore, the proper assessment of mixing and/or coating of various aerosol species and their impacts on various aerosol characteristics have not been well quantified (Xue et al., 2011), which makes the investigation a real challenge (Das and Jayaraman, 2011). IGB, being in proximity to the Thar Desert region, is found to be affected predominantly by the enhanced dust aerosols during pre-monsoon period. As a result, the probability of this interaction (i.e. mixing) was suggested to be more over the region during this period (Dey et al., 2008; Giles et al., 2011) and is one of the future perspectives of the present study.

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