



Variability in aerosol optical properties over an urban site, Kanpur, in the Indo-Gangetic Plain: A case study of haze and dust events



Kirpa Ram ^{a,*}, Sunita Singh ^a, M.M. Sarin ^b, A.K. Srivastava ^c, S.N. Tripathi ^d

^a Institute of Environment & Sustainable Development, Banaras Hindu University, Varanasi, India

^b Geosciences Division, Physical Research Laboratory, Ahmedabad, India

^c Indian Institute of Tropical Meteorology (Branch), New Delhi, India

^d Department of Civil Engineering, Indian Institute of Technology, Kanpur, India

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ABSTRACT

In this study, we report on three important optical parameters, viz. absorption and scattering coefficients (b_{abs} , b_{scat}) and single scattering albedo (SSA) based on one-year chemical-composition data collected from an urban site (Kanpur) in the Indo-Gangetic-Plain (IGP) of northern India. In addition, absorption Ångström exponent (AAE) was also estimated in order to understand the wavelength dependence of absorption and to decipher emission sources of carbonaceous aerosols, in particular of black carbon. The absorption and scattering coefficients ranged between 8.3 to 95.2 Mm^{-1} ($1 \text{ Mm}^{-1} = 10^{-6} \text{ m}^{-1}$) and 58 to 564 Mm^{-1} , respectively during the study period (for $n = 66$; from January 2007 to March 2008) and exhibit large seasonal variability with higher values occurring in winter and lower in the summer. Single scattering albedo varied from 0.65 to 0.92 whereas AAE ranged from 0.79 to 1.40 during pre-monsoon and winter seasons, respectively. The strong seasonal variability in aerosol optical properties is attributed to varying contribution from different emission sources of carbonaceous aerosols in the IGP. A case study of haze and dust events further provide information on extreme variability in aerosol optical parameters, particularly SSA, a crucial parameter in atmospheric radiative forcing estimates.

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1. Introduction

The ubiquitous presence of aerosols in the atmosphere affects radiation budget of the Earth, directly by scattering and absorbing solar radiation, or indirectly through modification of microphysical properties and thus, influences climate on a local as well as global scale (IPCC, 2013). The impacts of aerosols on radiation budget, hydrological cycle and climate are significant, but remain highly uncertain (Gadhavi and Jayaraman, 2010; IPCC, 2013; Menon et al., 2002a; Menon et al., 2002b; Ramanathan et al., 2001; Satheesh and Moorthy, 2005). The abundance of atmospheric aerosol and its constituents, their physico-chemical and optical properties are highly variable with respect to time and space (Ram and Sarin, 2010; Ram et al., 2012b; Ramanathan and Carmichael, 2008; Satheesh and Moorthy, 2005). Carbonaceous aerosols, an ubiquitous component of atmospheric aerosols, can contribute about 20–70% of particulate matter over urban environment and are mainly associated with fine-fraction of aerosols (i.e. aerodynamic diameter $\leq 2.5 \mu\text{m}$, defined as $\text{PM}_{2.5}$ aerosols) (Fuzzi et al., 2006; Jimenez et al., 2009; Ram and Sarin, 2011; Ram and Sarin, 2015). Carbonaceous aerosol play a significant role in regional atmospheric chemistry, visibility and air-quality (Ram et al., 2012a; Rengarajan et al.,

2011; Srivastava et al., 2014) and also contribute to the formation of haze and fog events (Huang et al., 2014; Kang et al., 2013; Ram and Sarin, 2011; Ram et al., 2012a; Tan et al., 2016; Tiwari et al., 2014).

Carbonaceous aerosols mainly include organic aerosols (OA) which account for the majority of aerosol mass whereas contribution of elemental carbon (EC), or black carbon (BC), is relatively low ($\leq 10\%$) (Ram and Sarin, 2010; Ram et al., 2010b; Ram et al., 2014; Tripathi et al., 2005). Although, both organic carbon (OC) and EC are emitted from the same emission sources, they have different optical, chemical and physical properties (Lan et al., 2013; Ram and Sarin, 2009; Schwarz et al., 2008). Black carbon (BC) is also called as elemental carbon or graphitic carbon and is a principal light absorbing species in atmospheric aerosols. However, light absorption due to BC varies temporally and geographically depending on emission sources and their emission strength (Andreae and Gelencser, 2006; Bond and Bergstrom, 2006; Bond et al., 2013; Bond et al., 2006; Ram and Sarin, 2009).

Asia is one of the most polluted regions in the world and is often characterized by atmospheric brown cloud formation in the Indo-Gangetic plain (IGP), the Himalayas and south-east Asia (Ramanathan et al., 2005). Aerosol optical properties exhibit a large spatial and temporal variation depending on aerosol composition, type of aerosols sources, prevailing meteorological conditions, ageing and transport of aerosols (Dey and Tripathi, 2007; Ram and Sarin, 2010; Ram and

* Corresponding author. Tel.: +91 542 6703555.

E-mail addresses: ram.iesd@bhu.ac.in, kirpa81@gmail.com (K. Ram).

Sarin, 2012; Ram and Sarin, 2015). Light absorbing aerosol species enhances global warming by supplementing to the greenhouse effect, however the assessment of their climatic impact still poses a greater challenge with relatively large uncertainty (Gadhavi and Jayaraman, 2010). It is, thus, important to assess aerosol optical properties and their climatic effect on a regional scale with a combination of real-time measurements as well as satellite retrievals. The present study aims to evaluate a few crucial optical properties of aerosols, namely absorption and scattering coefficients (b_{abs} and b_{scat} , respectively) and single scattering albedo (SSA) using a one-year chemical composition data of PM₁₀ samples ($n = 66$; January 2007 to March 2008) collected from an urban location, Kanpur, in the Gangetic plain of northern India. In addition, we also studied the wavelength dependence of aerosol absorption coefficient and estimated absorption Ångström exponent (α ; AAE) for different seasons to decipher emission sources of aerosols. Finally, aerosol optical parameters during haze and dust events, as a case study, have been examined with the help of real-time measurements as well network of ground-based sun photometers AERONET (Aerosol Robotic Network) at Kanpur.

2. Methodology

2.1. Ambient aerosol sampling

PM₁₀ samples (particulate matter with aerodynamic diameter $\leq 10 \mu\text{m}$) were collected, during January 2007–March 2008, from Kanpur using a high-volume sampler (APM 450, Environtech Pvt. Ltd., New Delhi, India), operated at a flow rate of $1.0 \pm 0.1 \text{ m}^3 \text{ min}^{-1}$. A total of 66 PM₁₀ samples were collected during the study period and details of aerosol sampling are described in Ram et al. (2010b). Of these, 22 samples were collected during wintertime (December–February), 9 samples during March, 25 samples during summer (April–June) and 10 samples during post-monsoon (October–November). July–September months represent the wet-season when south-west monsoon rain causes efficient washout of the atmosphere and hence, no aerosol samples were collected during July–September period (Ram et al., 2010b).

2.2. Site description and meteorological details

The sampling site, Kanpur (26.5°N , 80.3°E , 142 m above mean sea level), is located in the central part of the IGP. The site is mostly influenced by large-scale anthropogenic emission sources (biomass burning, fossil-fuel combustion and industrial activities) within the IGP and mineral dust transported from western India which directly influence optical properties of aerosols at Kanpur (Ram et al., 2010b). Due to shallow boundary layer in the wintertime and the Himalayan mountains parallel to the IGP, aerosols are confined to lower atmosphere resulting in a high concentration of anthropogenic aerosols (Ram and Sarin, 2011; Ram and Sarin, 2012; Ram and Sarin, 2015; Ram et al., 2010b). The average fractional contribution of carbonaceous aerosols in the IGP is about 30–35% of PM₁₀; however, it can be as high as 70% of PM_{2.5} during the wintertime (Ram and Sarin, 2011; Ram et al., 2010b). It is noteworthy to mention here that variability in aerosol composition, absorption and scattering properties over IGP is important to decipher climate-relevant optical properties and radiative forcing (Ram et al., 2012b).

2.3. Chemical composition of aerosol (EC, OC, Ionic species)

An aliquot of filter paper (area: 1.5 cm^2) was used for the measurement of EC, OC concentration on a thermo-optical EC-OC analyzer (Sunset laboratory Inc., USA) using NIOSH-5040 (National Institute of Occupational Safety and Health) based on thermal-optical transmittance (TOT) protocol (Birch and Cary, 1996; Ram et al., 2008). The analytical procedure of EC, OC consists of two stages of heating in an inert and an oxidizing medium. The first stage includes volatilization of OC

in a non-oxidizing medium through stepwise heating. In the second stage, the oven is subjected to a stepwise temperature increase (up to 900°C) in an oxidizing condition where all the fractions of pyrolyzed carbon and EC are converted to CO_2 . The evolved CO_2 in each volatilization step was converted to methane and were measured using a flame ionization detector. The thermograph obtained in the first and second stages, when corrected for pyrolyzed carbon, gives OC and EC fractions, respectively. The initial absorbance at 678 nm was used to define the split-point between OC and EC. All the reported OC concentrations are corrected for pyrolyzed and carbonate carbon (CC). The detection limits for EC and OC was reported to be 0.01 and $0.3 \mu\text{g m}^{-3}$, respectively whereas the average blank concentration for OC is $1.8 \pm 0.2 \mu\text{g m}^{-3}$ ($n = 8$). Replicate analysis of samples indicated good analytical precision, with relative percent deviations $< 3.0\%$ for OC and better than 5.0% for EC analysis ($n = 16$) (Ram et al., 2010b).

For water-soluble ionic species, one-fourth of filter area ($\sim 105 \text{ cm}^2$) is soaked in 50 ml milli-Q water (resistivity $18.2 \text{ M}\Omega \text{ cm}$) for approximately 6–8 h. An intermittent ultrasonic treatment was performed to disintegrate aerosol particles from the filter. An aliquot of water-extract was used to determine concentration of water-soluble ionic species using an ion-chromatograph. For separating anions, 1.8 mM $\text{Na}_2\text{CO}_3/1.7 \text{ mM NaHCO}_3$ eluent was used and for cations 20 mM methanesulfonic acid was used as an eluent. The replicate analysis provides a precision of better than 5% for ionic species and the concentration of blank filter was subtracted to assess the concentration of cations and anions. The analytical details of water-soluble ionic species have been described elsewhere (Ram et al., 2010b; Rengarajan et al., 2007).

2.4. Assessment of aerosol absorption and scattering coefficient

Aerosol absorption coefficient (b_{abs}) was estimated at 678 nm with the help of measured optical-attenuation (ATN) as per the methodology provided in an earlier publication (Ram and Sarin, 2009). The total uncertainty in the estimated b_{abs} values is of the order of $\sim 23\%$ arising from the ATN, area, volume of air sampled, multiple scattering and shadowing corrections (Ram and Sarin, 2009).

Scattering coefficient (b_{scat}) is calculated using the following Eq. (1):

$$b_{\text{scat}} = \sum M_i * MSC_i \quad (1)$$

where M_i represents mass concentrations ($\mu\text{g m}^{-3}$) of $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , organic matter (OM) and BC, and MSC_i is the mass scattering cross-section (in m^2g^{-1}) of the chemical species (Bond and Bergstrom, 2006; Magi, 2009). Eq. (1) can be rewritten as

$$b_{\text{scat}} = M_{\text{ionic}} * MSC_{\text{ionic}} + M_{\text{carbon}} * MSC_{\text{carbon}} \quad (2)$$

In Eq. (2), M_{ionic} represents sum of the mass concentrations of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 whereas M_{carbon} represents sum of mass concentrations of OM and BC (Magi, 2009, 2011). The mass concentration of SO_4^{2-} and NO_3^- are converted to $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 for calculating the scattering coefficient (Magi, 2009). A factor of 1.6 is used to convert OC to OM (Ram et al., 2010b; Rengarajan et al., 2007). Scattering coefficient was initially estimated at 550 nm in the present study as mass scattering cross-sections are reported at 550 nm and latter extrapolated to 678 nm in order to match the wavelength of absorption coefficient (Levin et al., 2010). The values of MSCs for different chemical species, used in the present study, are listed in Table 1.

2.5. Assessment of single scattering albedo (SSA; Ω)

Single scattering albedo is an important parameter which explains absorbing/scattering nature of aerosols and is used in the estimation of direct aerosol radiative forcing. The scattering and absorption characteristics of aerosols in combination with surface reflectance determine

Table 1
Mass Scattering Cross-Section (MSC) data at 550 nm is used in the present study#.

Species	MSC (m^2g^{-1})
SO_4^{2-}	6.1 ± 2.1
NO_3^-	5.5 ± 2.1
OC	3.8 ± 0.5
BC	2.9 ± 0.8

as per the values reported by Magi (2011).

whether aerosol contribute toward cooling or heating of the atmosphere (Lewis et al., 2008). It is defined as the ratio of scattering to extinction coefficients due to aerosols and is a measure of the fraction of radiation absorbed by atmospheric aerosols. It can be represented by the following equation:

$$\text{SSA} = \frac{b_{\text{scat}}}{b_{\text{scat}} + b_{\text{abs}}} = \frac{b_{\text{scat}}}{b_{\text{ext}}} \quad (3)$$

where b_{scat} is scattering coefficient and b_{ext} is extinction coefficient which is sum of scattering and absorption coefficients (i.e. $b_{\text{ext}} = b_{\text{scat}} + b_{\text{abs}}$). The value of SSA ranges from zero (for purely absorbing) to 1 for purely scattering type aerosols. It is pertinent to mention that all the values of scattering and absorption coefficients and SSA are reported at 678 nm in order to maintain the consistency.

2.6. Assessment of Ångström coefficient (α)

The absorption Ångström exponent (AAE, α) was estimated from the wavelength dependence of light absorption by particle was assessed by using the power law relationship as shown in Eq. (4):

$$\text{norm-}b_{\text{abs}} = K\lambda^{-\alpha} \quad (4)$$

where, α is absorption Ångström coefficient which gives spectral dependence of aerosol absorption and $\text{norm-}b_{\text{abs}}$ is normalized absorption with respect to that at the wavelength at 950 nm. The value of α is approximately 2 for biomass burning, and a value of α close to 1 is taken for fossil-fuel burning whereas intermediate values of α indicate mixed contribution from biomass, bio-fuel burning or mineral dust (Aruna et al., 2013; Bergstrom et al., 2007; Fialho et al., 2005; Gadhavi and Jayaraman, 2010; Ganguly et al., 2005; Kirchstetter et al., 2004; Sandradewi et al., 2008; Weingartner et al., 2003).

3. Results and discussion

Aerosol optical properties such as aerosol optical depth (AOD) and single scattering albedo (SSA) are important parameters to assess direct aerosol radiative forcing and climate at local as well as global scale (Menon et al., 2002a; Srivastava et al., 2012a). The temporal variability in these optical parameters at Kanpur on daily, monthly and seasonal scales are described in the following sections.

3.1. Day-to-day variability in aerosol optical properties

Absorption coefficient values ranged from 8.3 to 95.2 Mm^{-1} (average: $46.5 \pm 19.9 \text{ Mm}^{-1}$; $n = 66$, $1 \text{ Mm}^{-1} = 10^{-6} \text{ m}^{-1}$) whereas scattering coefficient values varied between 58 and 564 Mm^{-1} (average: $192 \pm 118 \text{ Mm}^{-1}$, $n = 66$). Similarly, single scattering albedo varied between 0.65 and 0.92 (average: 0.78 ± 0.06 , $n = 66$) indicating a large temporal change in absorbing/scattering nature of aerosols. Absorption Ångström Exponent varied from 0.79 (12 May 2007) to 1.40 (28 January 2008) suggesting that carbonaceous aerosols over Kanpur are derived from mixed emission sources during the study period. Fig. 1 represents daily variations in the estimated absorption

coefficient (Fig. 1a), scattering coefficient (Fig. 1b) and single scattering albedo (Fig. 1c) values at 678 nm wavelength during the measurement period (January 2007–March 2008) at Kanpur.

3.2. Monthly variability in aerosol optical properties

Monthly average values of estimated aerosol optical parameters are shown in Fig. 2 and also depicted in Table 2. Monthly average absorption coefficient values range from 22.5 to 88.8 Mm^{-1} during the study period. Generally, higher values of b_{abs} are associated with winter and post-monsoon months while lower values were observed during pre-monsoon or summer season. The seasonal average values of b_{abs} during pre-monsoon, post-monsoon and winter of year 2007 are 31.3 ± 8.5 ($n = 25$), 66.8 ± 5.6 ($n = 10$) and 53.7 ± 5.5 ($n = 22$) Mm^{-1} , respectively (Table 3). It is clear that b_{abs} values during winter and post-monsoon is about two-times higher than that in summer season. Aerosol sampling could not be performed during monsoon season and hence, optical properties data for monsoon season is not available. Absorption coefficient is found to be significantly correlated with mass concentration of EC, indicating EC as a principal absorber in ambient aerosol at Kanpur with a non-zero intercept (Ram and Sarin, 2009; Ram et al., 2012b). The non-zero intercept value of absorption were also obtained at urban and high-altitude sites of India (Ram and Sarin, 2009; Ram et al., 2010c) and were attributed to the absorption from brown carbon and/or dust (Bergin et al., 2014). It is also reported that more than 90% of total absorption was due to BC alone in a study conducted in China (Andreae et al., 2008) though, a small fraction of absorption (~20%) was attributed to less-absorbing aerosols such as humic-like substances (HULIS) and brown carbon (Cheng et al., 2008; Ma et al., 2011). Thus, higher b_{abs} values during post-monsoon and winter seasons generally resulted due to high EC mass concentration mainly originating from fossil-fuel and increased biomass burning activities (Ram et al., 2010b). Similar results were also observed recently at Delhi, an urban megacity, situated at the western part of the IGP (Srivastava et al., 2014) (Table 3).

A large spatio-temporal variability in b_{abs} values has been reported over sampling sites in India and the variability has been attributed to different emission sources for BC aerosols having different physico-optical properties, boundary layer dynamics and changes in aerosol composition and mixing state (Ram and Sarin, 2009; Ram et al., 2010c; Ram et al., 2012b). The variability in b_{abs} values observed, in this study, is similar to those reported at other sites in India. For example Ram et al. (2012b) reported that b_{abs} ranged from 20 Mm^{-1} (in April 2009) to 90 Mm^{-1} (in December 2009) at Kanpur whereas in a recent study, Srivastava et al. (2014) reported that absorption coefficient at another urban site, Delhi, varies between 7.2 and 183.8 Mm^{-1} during 2011–2012 (mean value $\sim 52 \pm 36 \text{ Mm}^{-1}$). The average absorption coefficient values of 69.7 ± 19.6 , 39.9 ± 9.1 , 66.1 ± 17.2 , $12.9 \pm 4.6 \text{ Mm}^{-1}$ were reported at Jaduguda (rural), Hisar (urban), Allahabad (urban) and Manora Peak (a high-altitude in the Himalaya) (Ram and Sarin, 2009). Furthermore, the average value of b_{abs} observed in the present study ($46.5 \pm 19.9 \text{ Mm}^{-1}$) is comparatively lower than that reported by Wu et al. (2009) at urban location, Guangzhou, China ($82 \pm 23 \text{ Mm}^{-1}$) during 2004–2007. In addition, a similar trend, as reported in the present study, was also observed at a high-altitude site, Manora Peak, in the Central Himalaya (Ram et al., 2010a).

Monthly mean scattering coefficient (Fig. 2b) varied between 82.0 Mm^{-1} and 409.3 Mm^{-1} during entire sampling period. Seasonal average values of b_{scat} during pre-monsoon, post-monsoon and winter seasons are 104.3 ± 25.1 , 228.9 ± 27.4 and $272.0 \pm 116.6 \text{ Mm}^{-1}$, respectively. The values of b_{scat} during winter and post-monsoon seasons are approximately two-three times higher compared to those during summer season. The enhancement in scattering coefficient is even higher than that in the case of absorption coefficient. This significant increase in scattering coefficient during winter is mainly due to high concentrations of scattering aerosols with high mass scattering cross-

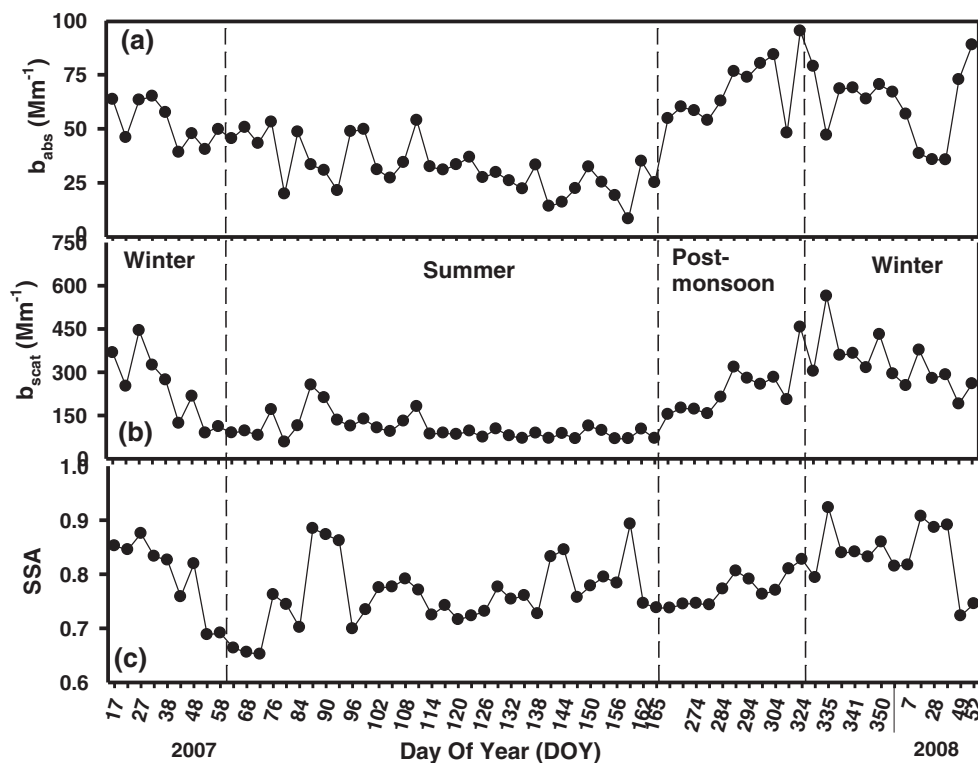


Fig. 1. Daily variations of absorption and scattering coefficients and SSA at 678 nm at the study site (Kanpur).

section (MSC; Table 1), and further enhancement in scattering due to an increase in their mass and growth under acidic condition (Ram et al., 2010b). For example, the average concentration of organic carbon (OC) during post-monsoon and winter are two to three times higher compared to those in summer (Ram et al., 2010b). Organic carbon (MSC = $3.8 \pm 0.5 \text{ m}^2\text{g}^{-1}$; Table 1), SO_4^{2-} and NO_3^- (MSC = $6.1 \pm 2.1 \text{ m}^2\text{g}^{-1}$; and $5.5 \pm 2.1 \text{ m}^2\text{g}^{-1}$, respectively; Table 1) have high mass scattering cross-section (Magi, 2009; Magi, 2011) and thus, contribute maximum to the total scattering coefficient. There was an overall factor of two increase in the mass concentration of water-soluble inorganic species (WSIS; sum of mass concentrations of inorganic ions), especially concentrations of SO_4^{2-} , NO_3^- and NH_4^+ ions which have high MSC values (Ram et al., 2010b).

Fig. 2c present monthly variations of SSA during the measurement period at Kanpur. The higher SSA values (Fig. 2c) during winter season (0.81 ± 0.06 , $n = 22$) resulted due to relatively higher enhancement in scattering coefficient compared to that of absorption coefficient. The lowest value of SSA (0.65) was observed in the month of March representing the presence of high absorbing aerosols. The month represents transition period between winter and summer seasons, and a temporal shift in the emission characteristics and/or changes in type of biomass burning (wood fuel and agricultural waste). It should be noted that an average oc/ec ratio of 2.9 ± 0.5 was found during March which is much lower than those in the winter and post-monsoon. In addition, the concentrations of SO_4^{2-} , NO_3^- and NH_4^+ were lower during March. These data suggest relative dominance of absorbing EC over scattering aerosols which probably resulted in lower SSA values in March. Relatively lower OC/EC ratios were observed in March, coincidence with lower SSA values, further indicate higher contribution from fossil-fuel emission sources (Ram et al., 2010b).

3.3. Seasonal mean variability in aerosol optical properties

Seasonal average values of estimated aerosol optical properties are shown in Fig. 3. The average SSA (0.79 ± 0.04 , $n = 66$) during January 2007–March 2008 is comparable to 0.81 ± 0.04 reported in the Pearl

River Delta region of China (Wu et al., 2009). The SSA values during pre- and post-monsoon seasons are comparable (0.76 ± 0.02 vs 0.77 ± 0.01) but lower than those obtained during winter season. These observations suggest that aerosol samples collected during pre- and post-monsoon seasons are relatively more absorbing in nature. However, SSA values in the present study are lower than those reported over other stations in India (Jethva et al., 2005; Srivastava et al., 2014). The values of SSA reported by Ram et al. (2012a) during post-monsoon (0.83) were relatively higher than the value obtained in the present study for the same month (0.77). In contrast, SSA value for winter season (0.81), observed in the present study, is comparable with the value reported by (Ram et al., 2012b). However, ground-based measured/estimated values of SSA are lower than the columnar average SSA estimated using sun/skyradiometer measurements. For example, Gadhavi and Jayaraman (2010) reported significantly higher value of SSA at 500 nm (range: 0.92–0.99) at a rural site in Gadanki using Skyradiometer data.

Table 4 shows a comparison of optical properties of aerosols estimated at Kanpur, in the present study, with earlier studies reported in the IGP and different parts of the world. In general, relatively higher value of b_{abs} and b_{scat} were found during wintertime which are in close agreement with earlier studies conducted over urban atmosphere of India and China (Gadhavi and Jayaraman, 2010; Man and Shih, 2001; Ram et al., 2012b; Wu et al., 2009; Yan et al., 2008). For example, Srivastava et al. (2011a) reported SSA values of 0.92 during pre-monsoon 2009 at Kanpur in the IGP, which is relatively higher than that observed at Gandhi College (0.89) of the eastern IGP region. These values are relatively higher than the average SSA of 0.70 ± 0.06 (range: 0.63 to 0.79) at 500 nm at New Delhi, in the western IGP region during 2007 (Srivastava et al., 2012b). In another study, Singh et al. (2010) reported annual mean SSA value of $\sim 0.79 \pm 0.05$ at $0.5 \mu\text{m}$ ranging from 0.89 to 0.70 over Delhi for the year 2006. In other studies, Pandithurai et al. (2008) found that SSA (at 500 nm) ranged between 0.74 and 0.84 during pre-monsoon at New Delhi. However, in a study conducted in northern China at rural site Shangdianzi (in Miyun County of Beijing), Yan et al. (2008) reported different trend compared to

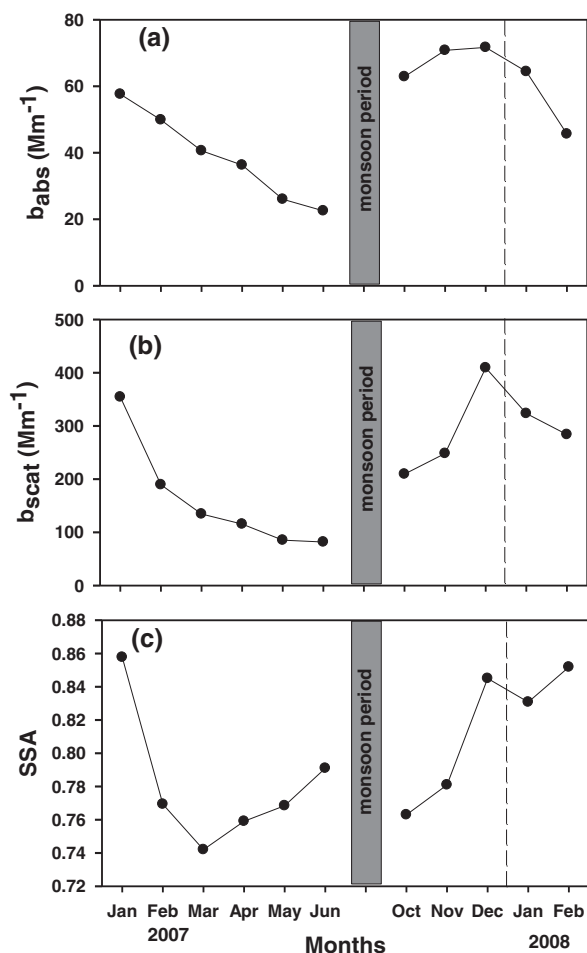


Fig. 2. Monthly variations of absorption, scattering coefficients and SSA at 678 nm during the study period at Kanpur.

the present study. The authors found the higher value of b_{abs} in summer and lower in winter while b_{scat} was higher in fall (September–November) and lower in winter. However, the higher value for SSA

Table 2

Monthly averages of aerosol absorption and scattering coefficients, single scattering albedo at 678 nm and Ångstrom exponent (α) reported in the present study.

Sampling period	n^*	$b_{abs} \pm S.D.^{\#}$ (Mm^{-1}) ^s	$b_{scat} \pm S.D.$ (Mm^{-1})	$SSA \pm S.D.$	$\alpha \pm S.D.$
January 2007	3	57.7 ± 10.1	354.4 ± 97.3	0.86 ± 0.01	1.05 ± 0.11
February 2007	6	49.9 ± 9.9	189.5 ± 96.6	0.77 ± 0.06	1.21
March 2007	8	40.6 ± 11.5	134.5 ± 70.2	0.74 ± 0.09	1.10 ± 0.06
April 2007	10	36.3 ± 10.7	115.5 ± 30.6	0.76 ± 0.04	1.05 ± 0.04
May 2007	10	26.0 ± 7.4	85.3 ± 15.2	0.77 ± 0.04	1.0 ± 0.13
June 2007	5	22.5 ± 9.8	81.9 ± 16.8	0.79 ± 0.06	1.10 ± 0.06
October 2007	7	62.9 ± 8.9	209.5 ± 64.8	0.76 ± 0.02	1.18 ± 0.22
November 2007	3	70.8 ± 19.8	248.2 ± 39.5	0.78 ± 0.02	1.04
December 2007	5	71.7 ± 17.5	409.3 ± 102.2	0.84 ± 0.04	1.05 ± 0.12
January 2008	4	64.4 ± 5.8	323.4 ± 76.0	0.83 ± 0.02	1.22 ± 0.16
February 2008	4	45.6 ± 18.1	283.9 ± 76.4	0.85 ± 0.08	1.14 ± 0.10
March 2008	1	88.8	259.8	0.74	1.11
Annual average		53.1 ± 19.8	224.6 ± 107.6	0.79 ± 0.04	1.10 ± 0.07
Maximum		88.8	409.3	0.86	1.22
Minimum		22.5	82.0	0.74	1.00

[#] S.D. = Standard Deviation.

^s $1 Mm^{-1} = 10^{-6} m^{-1}$.

* n = Number of samples collected in respective months.

was in accordance with the present study (highest in winter). The probable reason for different trend of measured optical properties at Shangdianzi was attributed to change in emission sources depending on season, transport mechanism, chemical conversion/transformation of aerosols present in the atmosphere and wind direction shifting (Yan et al., 2008). The present study revealed that the aerosols in winter and post-monsoon seasons are relatively more scattering in nature, although mass concentration and fractional contribution of BC is higher during these seasons (Ram et al., 2010b).

4. Spectral dependence of light absorption

Wavelength dependence of light absorption by aerosols is an important parameter which needs to be taken into account in the estimation of radiative forcing (Kirchstetter et al., 2004). Aerosol absorption due to BC aerosols exhibits spectral dependence and shows an inverse relationship with wavelength (Aruna et al., 2013; Bergstrom et al., 2004; Bergstrom et al., 2002). To study the spectral dependence of BC, measured absorption coefficient was normalized with respect to the absorption coefficient at 950 nm ($norm-b_{abs}$) and absorption Ångstrom exponent (α ; AAE) values were estimated for different seasons. The value of AAE ranged from 0.79 (for 12 May 2007 sample) to 1.40 (for 28 January 2008 sample) at Kanpur during the sampling period. The seasonal variation of AAE is presented in Fig. 4. It is evident from the figure that AAE exhibit slightly different seasonal variation to that of SSA (Fig. 3c) with the highest value in winter and lowest in pre-monsoon season, similar to studies conducted at other locations (Favez et al., 2009; Sandradewi et al., 2008).

The variability in AAE value represents BC aerosols derived from different emission sources (Aruna et al., 2013; Kompalli et al., 2014). This is also supported by the study of Ram et al. (2010b) wherein authors reported a change in emission sources as well as their strength on the basis of OC/EC and K^+/OC ratios at Kanpur. In another study, the values of α was found to be in the range of 0.96 to 1.13 suggesting the presence of more UV-absorbing aerosols in winter than summer season (Sandradewi et al., 2008). Sandradewi et al. (2008) also reported a significant positive correlation between AAE and CO/NO_x ratio under the influence of primary traffic emissions. However, Favez et al. (2009) suggested that the change in atmospheric chemical composition and the presence of absorbing species other than BC led to the seasonal trend of AAE values. In addition, a significant correlation was obtained between AAE values and organic aerosols indicating the presence of Humic Like Substances (HULIS) and brown carbon (Favez et al., 2009).

The unique topography of the IGP and variety of emission sources led to a large spatial and temporal heterogeneity in distribution and characterization of physico-chemical and optical properties (Srivastava et al., 2011a; Tiwari et al., 2013). The value of AAE was found to be 1.0 for samples collected on 18th May 2007 (representative of summer; Fig. 5a) and 1.40 for 28th January 2008 (representative of winter season, Fig. 5b). Furthermore, the value of AAE is higher during winter months and lower during summer months, which indicate that the emission sources and their strength are different during winter and summer seasons. Generally, biomass burning activities increases during winter months (Ram et al., 2010b) which lead to an increase in the value of AAE in wintertime. Srivastava et al. (2011a) have estimated the AAE values at Kanpur and Gandhi College (a rural station in the eastern IGP region) during pre-monsoon period using ground-based AERONET sun/sky radiometer measurements. The AAE values were >1 at both the stations and relatively higher AAE values (1.62 at Kanpur vs 1.46 at Gandhi College) suggest the dominance of biomass burning aerosols at both locations. In a recent study, Tiwari et al. (2013) showed a negative gradient in the magnitude of AAE from western to the eastern IGP region during the pre-monsoon period.

Table 3
Optical properties of aerosol at 678 nm and their comparison with other studies.

Sampling sites	Sampling period	b_{abs} (Mm^{-1})	b_{scat} (Mm^{-1})	SSA	References
Kanpur	Jan 07–Mar. 08	46.5 ± 19.9	191.5 ± 118.3	0.78 ± 0.06	Present study
	Jan–Feb. 07	53.7 ± 5.4	272.0 ± 116.6	0.81 ± 0.06	Present study
	Mar–Jun. 07	31.4 ± 8.5	104.3 ± 25.1	0.76 ± 0.02	Present study
	Oct–Nov. 07	66.8 ± 5.6	228.9 ± 27.4	0.77 ± 0.01	Present study
	Dec 07–Feb. 08	60.6 ± 13.4	338.9 ± 64.1	0.84 ± 0.01	Present study
	Mar 08	88.8	259.8	0.74	Present study
	Oct–Nov. 08	79.0 ± 8.9	401.0 ± 57.0	0.83 ± 0.02	Ram et al. (2012a)
	Dec 08–Jan. 09	62.1 ± 22.8	295.0 ± 236.3	0.80 ± 0.06	Ram et al. (2012a)
	Mar–Apr. 09	24.1 ± 3.6	418.1 ± 273.8	0.93 ± 0.02	Ram et al., (2012a)
	Apr–Jun	62.5 ± 22.3 ^d	110.5 ± 36.1 ^d	0.63 ± 0.06 ^d	Soni et al. (2010)
	Jan–Mar	189 ± 86.0 ^d	565.6 ± 274.6 ^d	0.74 ± 0.03 ^d	Soni et al. (2010)
	Delhi	2009			0.85 ± 0.052 ^a
Wuqing, China	Jul–Aug. 09	43 ± 27 ^e	379 ± 251 ^d	0.87 ± 0.5	Ma et al. (2011)
Wuqing, China	Mar–Apr. 09	47 ± 38 ^e	280 ± 253 ^d	0.83 ± 0.05	Ma et al. (2011)
Jaduguda	Dec 04	69.7 ± 19.6			Ram and Sarin (2009)
Hisar	Dec 04	39.9 ± 9.1			Ram and Sarin (2009)
Allahabad	Dec 04	66.1 ± 17.2			Ram and Sarin (2009)
Manora peak	Dec 04	12.9 ± 4.6			Ram and Sarin (2009)
Manora peak	Feb 05–Jun. 07	12.2 ± 6.6			Ram and Sarin (2009)
Mt Abu	Dec 05–Feb. 06	8.0 ± 5.5			Ram and Sarin (2009)
Mt Abu	May 05–Feb. 06	5.8 ± 4.3			Ram and Sarin (2009)
Delhi	Dec 11–Mar. 12	71.7 ± 54.6 ^h			Tiwari et al. (2014)
Delhi	2008–2009			0.70 ± 0.07 ^d	Soni et al. (2010)
Guangzhou, China	Oct–Nov. 04	91 ± 60 ^f	418 ± 159 ^f	0.83 ^f	Andreae et al. (2008)
Guangzhou, China	2004–2007	82 ± 23 ^c	358 ± 107 ^b	0.81 ± 0.04 ^b	Wu et al. (2009)
Beijing, China	Aug–Sept. 06	57.8 ± 31.1 ^g	364.0 ± 324.3 ^b	0.80 ± 0.11 ^d	Han et al. (2014)
Shangdianzi, China	Apr 03–Jan. 05	17.5 ± 13.4 ^h	174.6 ± 189.1 ^b	0.88 ± 0.05 ^b	Yan et al. (2008)

^a 440 nm.^b 525 nm.^c 532 nm.^d 550 nm.^e 637 nm.^f 540 nm.^g 670 nm.^h 880 nm.

5. Influence of haze and dust event on chemical composition and optical properties of aerosols in IGP: a case study

The hazy and foggy weather conditions over urban areas around the globe are becoming very common, mainly due to higher emission of anthropogenic pollutants from industries and transportation along with secondary aerosol formation (Huang et al., 2014; Ram et al., 2012a). The IGP is a densely populated area in northern India and is influenced by emissions from various anthropogenic activities which led to high concentration of pollutants (Nair et al., 2007). The haze (during winter) and dust (during pre-monsoon) events are common phenomena in the IGP due to its unique topography and favorable meteorological conditions (Chinnam et al., 2006; Dey and Tripathi, 2007; Dey et al., 2004; Ram et al., 2012a). In the following sections, we have investigated a couple of dust and haze events to examine the differences in chemical and optical properties of aerosols at Kanpur.

PM₁₀ samples collected during wintertime (December 2007–February 2008) show persistently high aerosol loading at Kanpur. PM₁₀ mass concentration exhibits a continuous build up from 161.4 $\mu\text{g m}^{-3}$ (on 30th November 2007) up to 252 $\mu\text{g m}^{-3}$ on 1st December 2007 and reached to a maximum value of 312.5 $\mu\text{g m}^{-3}$ on 16 December 2007. A significant change in chemical composition (especially OC, EC, NH₄⁺, NO₃⁻ and SO₄²⁻) and optical properties of aerosol samples

collected during haze and fog weather conditions was observed during the study period. In this study, we have included only those haze and fog events, which were captured by Moderate Resolution Imaging Spectro-radiometer (MODIS) on NASA's Aqua satellite. The two haze events were captured on 10 December 2007 (Fig. 6a) and 16 December 2007 (Fig. 6b) whereas dust storm events were captured on 18th May and on 10th June 2007 (Fig. 6c & d). A comparison of chemical composition and optical properties of aerosols on these haze and dust events are presented in Table 4 and a detailed discussion is provided below.

5.1. Haze event

The MODIS Aqua satellite captured a thick haze on 10 December 2007 (Fig. 6a) which persisted for several days in India, especially south of the Himalaya (Fig. 6b). The concentrations of PM₁₀ and its chemical constituents, mainly carbonaceous and inorganic aerosols, were significantly higher during haze events at Kanpur (Table 4). Relatively high OC/EC ratio and elevated K⁺ concentration observed on hazy days indicate enhanced biomass burning activities during these days (Ram et al., 2012a). A recent study has suggested enhancement in the yield of WSOC in presence of transition metals due to oxidation of carbonyl derivatives into respective organic acids, esters and

Table 4
A comparison of chemical composition and optical properties of aerosols on haze and dust events.

Parameters	Measured chemical species ($\mu\text{g m}^{-3}$)										Optical properties		
	PM ₁₀	OC	EC	NH ₄ ⁺	K ⁺	Ca ²⁺	Cl	NO ₃ ⁻	SO ₄ ²⁻	OC/EC ratio	b_{abs}	b_{scat}	SSA
Dust event (12 May 07)	104.3	11.3	2.2	1.4	0.34	1	0.11	1.68	4.2	5.3	25.9	79.4	0.75
Haze event (10 & 16 Dec. 07)	273.6	49.6	4.8	9.7	2.3	2.3	4.2	22.6	11.5	10.4	68.7	361.7	0.84

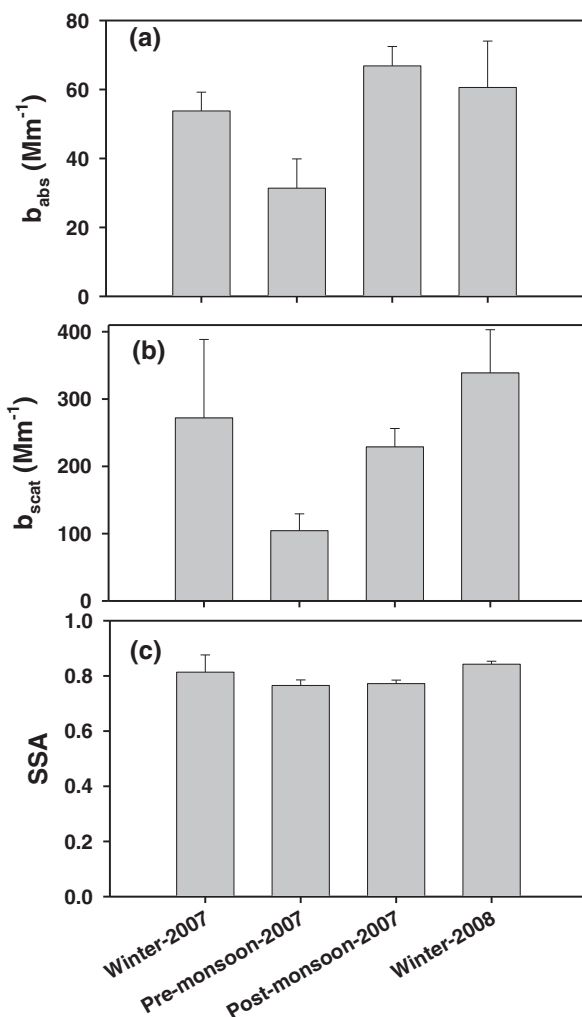


Fig. 3. Seasonal variation of absorption and scattering coefficients and SSA at Kanpur during the study period. Winter 2008 includes December 2007, January & February 2008.

other derivatives during haze days in the Gangtic plain (Singh and Gupta, 2016). This is also evident from WSOC/OC ratio in aerosol samples collected during haze events (Ram et al., 2012a). The enhancement in scattering type of aerosols is reflected from aerosol optical parameters such as scattering coefficient and SSA which are relatively higher

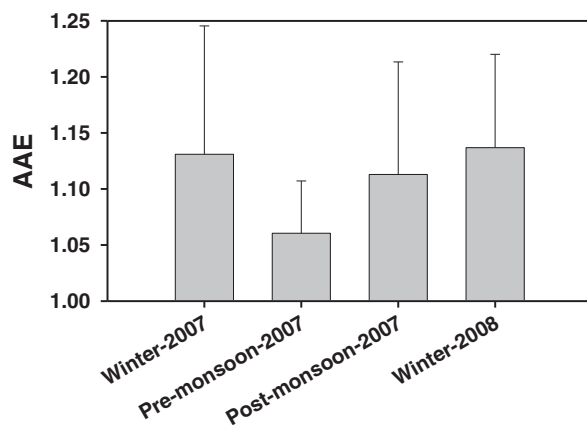


Fig. 4. Seasonal variation of Absorption Ångström exponent (AAE) at Kanpur during Sampling Period. Winter 08 includes December 2007, January 2008 & February 2008.

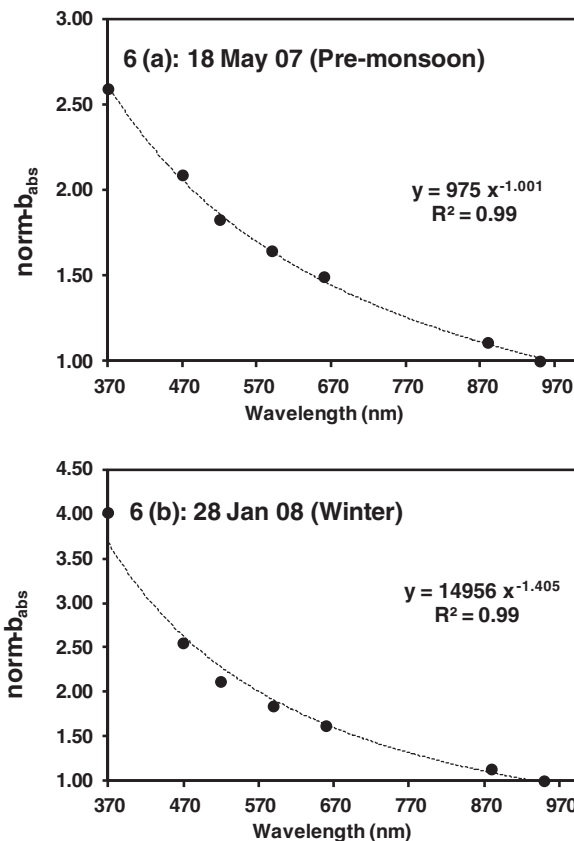


Fig. 5. Spectral dependence of absorption during (a) summer and (b) winter seasons. The absorption coefficient (i.e. $norm-b_{abs}$) was normalized with respect to 950 nm.

during hazy days (Table 4). These observations are similar to an earlier observation taken at Allahabad and Hisar in the IGP (Ram et al., 2012a). Similar findings were also observed in Guangzhou, China where local emission sources and SOAs contributed to the formation of haze events (Huang et al., 2014; Tan et al., 2009; Yu et al., 2013). The higher concentration of NO_3^- and SO_4^{2-} may result from secondary formation from their precursor gases in the presence of high relative humidity during haze event (Kaul et al., 2011; Li and Han, 2016; Ram et al., 2012a; Ram et al., 2014). It is noteworthy to mention here that concentration of K^+ , Ca^{2+} , Cl^- , and HCO_3^- was also higher on the 16th December 2007. Higher concentrations of Ca^{2+} indicate an enhanced contribution from mineral dust whereas that of Cl^- may be attributed to contribution of local industrial sources and biomass burning emissions. Thus, some of the haze events indicate that mineral dust, probably locally emitted in the vicinity of sampling site, were mixed with biomass burning emission and anthropogenic aerosols.

5.2. Dust storm

Ground-based measurement of PM_{10} mass concentration showed significantly high PM_{10} concentration of $201.2 \mu g m^{-3}$ on 21 May 2007. The chemical composition of aerosol samples collected on the dust event indicates that concentrations of carbonaceous and inorganic species were a factor of 3–4 lower on dust events compared to those on haze events (Table 4). In contrast, concentration of Ca^{2+} ($4.6 \mu g m^{-3}$), a tracer for mineral dust, was highest on 21 May 2007 indicating an increased contribution from mineral dust. In addition, AOD and SSA values were also higher on 21 May 2007 possible due to a relative increase in scattering from mineral dust. Relatively high concentration of crustal species in aerosol samples has been reported in Beijing, China during dust storms (Wang et al., 2005; Yuan et al., 2008) and at Kanpur

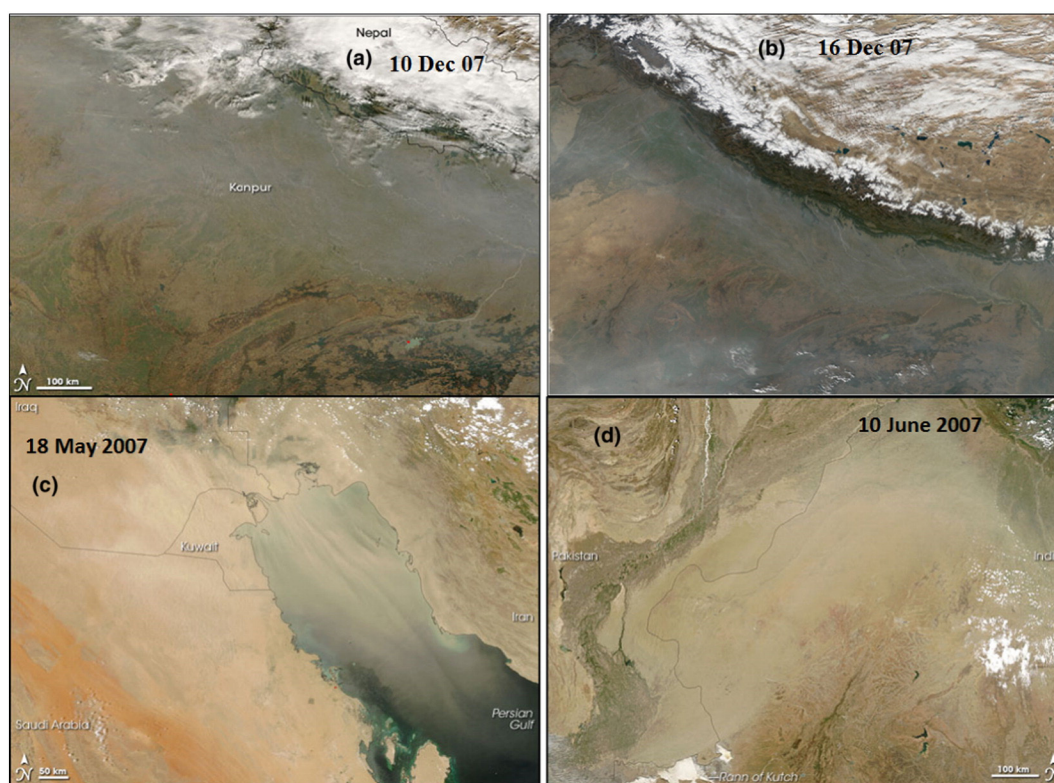


Fig. 6. The Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Aqua satellite images (a): dark, dirty and gray haze blanketing the area over and around Kanpur (captured on 10 Dec. 2007); (b): Haze lingered in India south of the Himalaya for several days in December 2007 (captured on 16 Dec. 2007); (c): Dust over the Persian Gulf on May 18, 2007; (d): Dust plume captured on 10 June 2007.

(Chinnam et al., 2006). The Himalaya acts as a barrier to the pollutants, including dust carried by westerly winds, thus increasing dust concentration in the foothills of the Himalayan region (Srivastava et al., 2015). Dust storms can have significant influence on direct aerosol radiative forcing due to changes in aerosol optical properties and thus, can influence climate on a local and regional level (Gómez-Amo et al., 2011; Srivastava et al., 2011b). In addition, mixing of dust particles with carbonaceous and acidic species could significantly influence physico-chemical and optical properties of aerosols (Sullivan et al., 2010).

More recent study has suggested that local meteorology, emission sources of aerosols and their long range transport influence the formation of haze event in China (Li and Han, 2016). It is estimated that ~44% of surface $PM_{2.5}$ in Beijing was contributed by local emissions with the remaining 56% coming from surrounding sources through regional trans-boundary transport. Higher concentration of carbonaceous and inorganic aerosols was observed during haze events (Yuan et al., 2015) whereas Ca^{2+} (tracer of mineral dust), AOD and SSA was higher during dust event (Ram et al., 2012b). Hygroscopic aerosols produced by secondary processes may directly influence scattering properties of aerosols (Yang et al., 2015). In addition, the formation of carbonyls, formaldehyde, acetaldehyde and acetone by photochemical reactions have been higher during haze days in Beijing, China (Duan et al., 2012).

6. Conclusions and implications

One-year chemical composition data of ambient aerosols have been used to assess their optical properties (absorption and scattering coefficients and single scattering albedo) from an urban site, Kanpur, in northern India. Absorption Ångström exponent (AAE) is also estimated in order to understand the wavelength dependence of absorption and to

decipher sources of carbonaceous aerosols (especially BC). Our study helps to make following important conclusions:

1. Absorption and scattering coefficients ranged between 31.4 ± 8.5 to $66.8 \pm 5.7 \text{ Mm}^{-1}$ and 104 ± 25 to $339 \pm 64 \text{ Mm}^{-1}$, respectively, during the study period. The absorption and scattering coefficients exhibit a strong seasonal variability with higher values in the winter and lower in summer season. The single scattering albedo varied from 0.65 (pre-monsoon) to 0.92 (winter), suggesting significant change in the sources, composition and optical properties of aerosols at Kanpur.
2. Wavelength dependence of absorption coefficient shows conspicuous seasonal pattern with higher values in winter and lower in summer/post-monsoon seasons. The absorption Ångström exponent shows variability in the mixing proportion of carbonaceous aerosols and dust particles during the winter and summer over Kanpur.
3. Aerosol chemical composition and optical properties are distinctly different during haze and dust events at Kanpur in the IGP region.

Overall, this study documents strong seasonal variability in optical properties of aerosols at an urban site in the Gangetic Plain attributable to variable emission sources, aerosol chemical composition, long-range transport and meteorological conditions in the IGP. The changes in aerosol properties during haze and dust events can significantly affect direct aerosol radiative estimation on a regional scale and needs further detailed investigation. Relatively higher WSOC/OC ratio, NO_3^- and SO_4^{2-} concentrations during haze event are attributed to enhanced secondary formation influencing mixing state of BC which in turn can affect the absorption, scattering and light extinction properties of aerosols. The current understanding on the mixing state of BC particles and their size-distribution is rather limited which further add to the uncertainties in the estimation of optical properties and atmospheric radiative forcing. In addition, optical properties of light absorbing brown

carbon and mineral dust have gained significant importance in recent years (Bergin et al., 2014; Srinivas et al., 2016) for their role in modifying optical properties that need further assessment in an urban environment dominated by pollution sources.

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