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High aerosol loading over mega city Delhi in the western Indo-Gangetic plain : Optical characteristics

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सार – शहरी महानगर दिल्ली, जो उत्तर भारत के भारत-गांगेय मैदानी क्षेत्र (आई जी पी) में स्थित है, में वायुविलय प्रकाशीय लक्षणों का वर्ष 2006 से 2008 के दौरान स्वचालित सूर्य/व्योम विकरण मापी का उपयोग करके मापन किया गया। इस अध्ययन से यह पता चला कि इस केन्द्र में उच्च वायु विलय है जो विभिन्न प्राकृतिक मानव-जनित उत्सर्जन स्रोतों द्वारा घिरी हुई उसकी स्थ्लाकृति के कारण हो सकते है तथा जिनका स्वास्थ्य, वायु गुणवत्ता और जलवायु प्रणाली पर गंभीर प्रभाव पड़ सकता है। परिणामों से अध्ययन की अवधि के दौरान ए ओ डी में बहुत परिवर्तिता दिखी जिसमें शीत (0.67 ± 0.06) और ग्रीष्म (0.71 ± 0.11) ऋतु में लगभग समान मान देखे गए। एंगस्टार्म घातांक मान शीत (1.19 ± 0.07, जिससे फाइन मोड़ वायु-विलय के प्रभाव का पता चलता है) में अधिक और ग्रीष्म (0.74±0.06, जिससे कोर्स मोड़ वायु-विलय का पता चलता है) में कम पाए गए। अध्ययन की अवधि के दौरान एकल प्रकीर्णन एलबीडो (एस एस ए) में थोड़ी कमी देखी गई, जिसका माध्य मान ~0.9 रहा। एस एस ए मॉनसूनोत्तर ऋतु के दौरान लगभग 0.93 और शीत ऋतु में 0.96 पाया गया जबकि ग्रीष्म और मानसून ऋतु में एस एस ए लगभग 0.95 पाया गया। इस स्टेशन में अनुमानित मासिक और अब शोषण एंगस्टार्म घातांक (ए ए ई) मान 0.11 से 1.87 तक रहे, जो 1.0 से ~55 प्रतिशत बार कम (मुख्यत: शीत और मानसून के दौरान), तथा 1.0 से ~45 प्रतिशत बार ज्यादा (मुख्यत: ग्रीष्म और मानसूनोत्तर के दौरान) पाया गया।

ABSTRACT. Measurements of aerosol optical properties were carried out at an urban mega city Delhi, which is situated in the western Indo-Gangetic Plain (IGP) region in north India using an automatic sun/sky radiometer during 2006-2008. The present study revealed high aerosol loading over the station, which could be due to its topography surrounded by different natural and anthropogenic emission sources, and may have major implications towards health, air quality and climate system. Results show a large variability in AOD during the study period, with nearly equal values during winter (0.67 ± 0.06) and summer (0.71 ± 0.11). The Ångström exponent (AE) values were found to be relatively higher during winter (1.19 ± 0.07, suggests dominance of fine-mode aerosols) and lower during summer (0.74±0.06, suggests dominance of coarse-mode aerosols). A slight decrease in single scattering albedo (SSA) was observed during the study period, with a mean value of ~0.9. SSA was found to be about 0.93 during post-monsoon and 0.96 during the winter period whereas during summer and monsoon, SSA was about 0.95. The estimated monthly absorption Ångström exponent (AAE) values over the station varied from 0.11 to 1.87, which were found to be less than 1.0 by ~55% time (mostly during winter and monsoon), and greater than 1.0 by ~45% time (mostly during summer and post-monsoon).

Key words – Indo-Gangetic plain, Optical properties, Single scattering albedo, Absorption Ångström exponent.

1. Introduction

Atmospheric aerosols are tiny liquid or solid particles, suspended in to the atmosphere with negligible terminal fall speed and injected from different natural as well as anthropogenic sources (Kaskoutis *et al*., 2007). Natural aerosols share nearly ~80% and play an important role in the global climate change while rest 20% cover by the anthropogenic sources, which cause the regional scale of climate feature (Kaskoutis *et al*., 2009). As a result, net effect of natural as well as anthropogenic aerosols is crucial for the climate change (Charlson *et al*., 1992; Charlson and Wigley, 1994; Anderson *et al*., 2003; Bellouin *et al*., 2005; IPCC, 2007). Aerosols play a crucial role in Earth's energy budget and affect the climate, human health and enviourment due to their direct and indirect effect (Sateesh and Ramanathan, 2000; Bellouin *et al*., 2005). For better understanding of mechanism which defines the optical state of the atmosphere, the sound knowledge of aerosol characteristics, their temporal and spatial variability and their interaction with other atmospheric parameters and processes are very

important (Smirnov *et al*., 2000). Recent research reveals that assessing aerosol effects on climate require knowledge of not only the regional and global distribution of aerosol amount, but also their various properties (Myhre, 2009 and references therein). This creates a large uncertainty in the assessment of global climate as well as climate change and reduces the level of scientific understanding of aerosols (IPCC, 2007).

The Indo-Gangetic Plain (IGP) traversed by the Ganga river and its tributaries is one of the largest, densely populated, industrialised and developing region of the world where aerosols not only affect the Indian monsoon but also the global climate system (Satheesh *et al*., 2006). 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. The topography of IGP from west to east and variable meteorological condition controls the dynamical behaviour of atmospheric aerosols over the region in both space and time. The region is dominated by the urban/industrial aerosols (Guttikunda *et al*., 2003; Sharma *et al*., 2003; Monkkonen *et al*., 2004; Tiwari *et al*., 2009), which has been demonstrated significant seasonal variability based on the complex combination of anthropogenic factors mixed with the contribution from the natural sources (transported dust), particularly during the pre-monsoon period (Srivastava *et al*., 2012a). The large increase in anthropogenic aerosols over IGP is hypothesized to cause considerable changes to regional monsoonal climate (Ramanathan and Ramana, 2005; Lau *et al*., 2006; Gautam *et al*., 2010; Moorthy *et al*., 2013). In contrast, the IGP experiences an enhanced convective and turbulent boundary layer and witnesses a large influx of westerly wind driven dust loaded air masses during the pre-monsoon/summer season (Gautam *et al*., 2011; Srivastava *et al*., 2011). This period also marks the development and evolution of the monsoon type circulation over the Indian subcontinent. Numerous studies using satellite and ground-based measurements have revealed high aerosol loading over India, especially in IGP region (Massie *et al*., 2004; Prasad and Singh, 2007; Lawrence and Lelieveld, 2010; Dey and Di Girolamo, 2010; Kaskaoutis *et al*., 2011; Ramachandran *et al*., 2012; Tiwari *et al*., 2013a). Thus, it is important to improve aerosol characterization over the IGP with high spatio-temporal resolution measurements.

In the present study, crucial aerosol optical parameters were retrieved using ground-based automatic sun/sky radiometer at New Delhi - An important region over the western part of IGP and studied extensively during three years period from 2006 to 2008.

2. Site description

The measurements were carried out in the campus of the India Meteorological Department, New Delhi (28.63° N, 77.22° E and ~250 m above msl) using a sun/sky radiometer. New Delhi is the capital city of India and one of the most polluted mega cities in Asia. The station climate is semi-arid and has maximum temperature of ~45-48 °C during summer and minimum of ~1-2 °C during winter (Tiwari *et al*., 2009). The high concentrations of fine aerosol particles (from the variety of anthropogenic sources) during the winter period constitute formation of early morning fog/haze, which turns into the production of smog (polluted fog), causing severe reduction in visibility (Tiwari *et al*., 2011) and leading to road accidents, health hazards, delay in air traffic. On the other hand, during summer, the region receives frequent dust storms from its neighboring Desert regions, which largely affects the air quality of the region due to large quantity of sediments coming from arid and semi-arid regions (Pandithurai *et al*., 2008; Srivastava *et al*., 2011). Further, it was also observed to influence of emissions from biomass/crop residue burning over the station from various agricultural activities over the north-west part of India during pre-monsoon and post-monsoon periods (Badarinath *et al*., 2009).

3. Instrumentation and data analysis

The sun/sky radiometer (Model: POM-01 of Prede, Japan) is capable for measuring direct solar and diffuse sky radiance at five spectral channels from visible to near-infrared spectral regions (340, 400, 500, 670 and 1020 nm), with a half band width of 3 nm for 340 nm wavelength and 10 nm for other wavelengths. The crucial columnar aerosol parameters such as aerosol optical depth (AOD), single scattering albedo (SSA) etc are derived at the above five discrete wavelengths using Skyrad. Pack (version 4.2) radiative transfer code (Nakajima *et al*., 1996). The algorithm takes care of cloud contamination using in-built threshold checks. In addition, manual cloud screening for questionable data was performed using weather observations obtained from India Meteorological Department. Sun/sky radiometer measures diffuse radiation at various scattering angles from the Sun at different wavelengths in narrow band. The measured sky spectral radiances can be used to obtain different optical and size related properties of aerosols in the total atmospheric column (Tonna *et al*., 1995; Kaufman *et al*., 1994; Nakajima *et al*., 1996). There are certain errors, which could occur during the measurements; these are sun or sky channel calibration, inaccurate azimuth angle pointing during sky radiance measurements and inaccuracy in accounting for surface reflectance measurements. However, detailed calibration and data

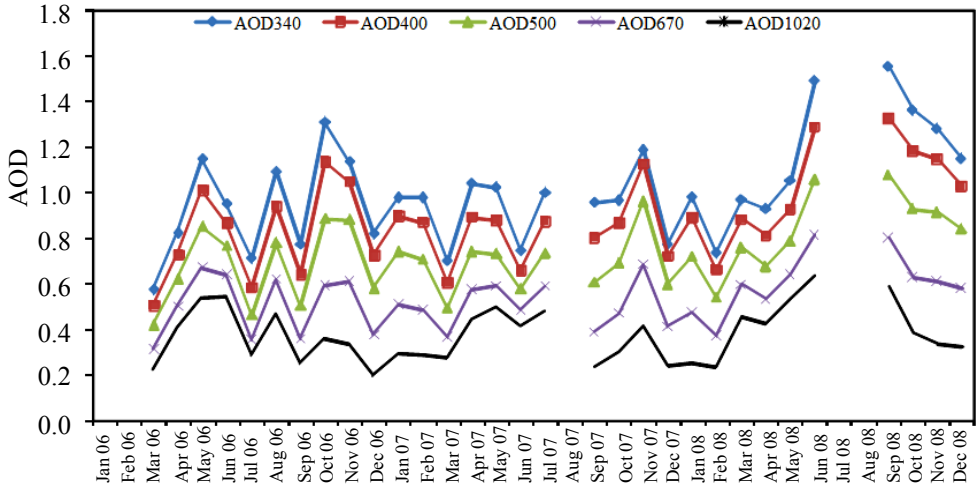

Fig. 1. Monthly variation of AOD at five different wavelengths during the period from 2006-2008

reduction procedures for this instrument have been described elsewhere (Nakajima *et al*., 1996; Pandithurai *et al*., 2007). Measurements were carried out during clear sky days and in the present study, we have utilized the processed data for about three years period from March 2006 to December 2008.

The spectral dependence of AOD, derived from sun/sky radiometer, is typically approximated by using Ångström's empirical formula (Ångström, 1964) as

$$\tau_a(\lambda) = \beta\lambda^{-\alpha} \quad (1)$$

where, λ is the wavelength in micrometre, τ_a is AOD, α is Ångström exponent (AE) and β is turbidity coefficient, which equals to τ_a at λ = 1 μm. Since α depends on wavelength (Tiwari *et al*., 2013a) but in above equation (1), α is assumed to be independent to wavelength. Therefore this equation is a special case of a more complicated equation for a limited range of particle diameters and a limited interval of wavelength (Kaskaoutis *et al*., 2009). Taking the logarithms at both sides, equation (1) becomes

$$\ln \tau_a(\lambda) = -\alpha \ln \lambda + \ln \beta \quad (2)$$

Parameter τ_a gives an indication of the amount of the aerosols present in the atmosphere, and α is a good indicator of the fraction of accumulation-mode particles (r < 1 μm) to coarse-mode particles (r > 1 μm), which describes the aerosol size distribution. For two different wavelengths (λ_1 and λ_2), α can be given as

$$\alpha = - \ln (\tau_{\lambda 1} / \tau_{\lambda 2}) / \ln (\lambda_2 / \lambda_1) \quad (3)$$

The above equation implied that α is invariant in the spectral range λ_1 to λ_2. This is valid only when the size distribution of aerosols in vertical column follows at least inverse power law (Beegum *et al*., 2009). But the real size distributions show slight variation from the power law distribution mainly when aerosols are injected from different sources and the spectrum has a very wide range (Schuster *et al*., 2006; Beegum *et al*., 2009; Kaskaoutis *et al*., 2009). Under such condition, a second order polynomial fit to AOD spectra can be used to quantify the curvature in spectral distribution of AODs (Eck *et al*., 1999) as

$$\ln\tau_\lambda = \alpha_2 (\ln\lambda)^2 + \alpha_1 \ln\lambda + \alpha_0 \quad (4)$$

where, the coefficient α_2 represents the curvature observed in the spectral distribution of AOD during the measurements. It is also a good indicator for aerosol particle size, having negative value (α_2<0) indicates aerosol size distributions dominated by fine-mode and positive value (α_2>0) indicates the size distribution dominated by coarse-mode aerosol particles (Eck *et al*., 1999).

4. Results and discussion

4.1. *AOD and Ångström exponent*

Fig. 1 shows monthly spectral variation of AOD during the entire study period (March 2006 to December 2008) which comes along with seasonal variations. From the figure we observed that the AOD has large spectral variability throughout the study period with lower AODs at higher wavelength and *vice-versa*. A significant increase in spectral AODs was observed during summer/

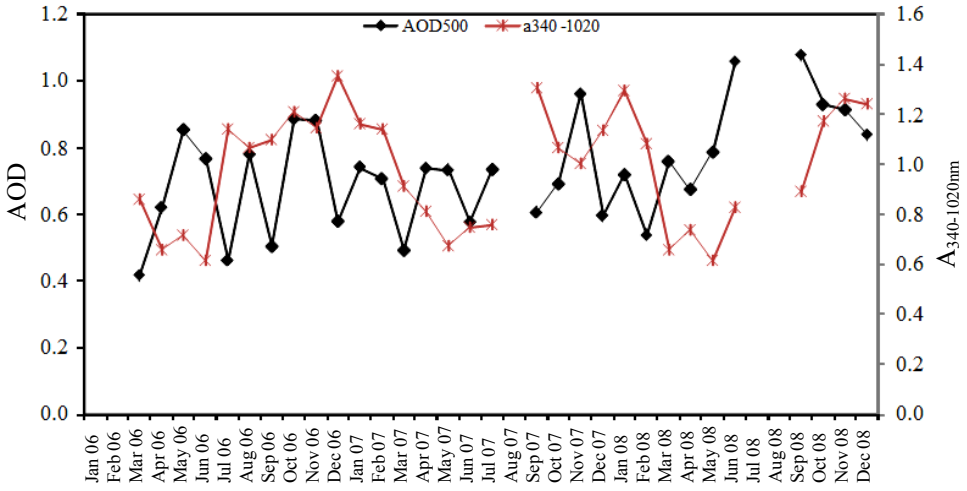

Fig. 2. Monthly variation of AOD_{500} and $\alpha_{340\text{-}1020}$

monsoon and post-monsoon months in each year. From the figure, we also observed that the variation at shorter wavelengths (340 and 400 nm) and mid-visible wavelength (500 nm) is quite different from the observed AODs at longer wavelength (1020 nm). AOD at shorter and mid-visible wavelengths have highest values during post-monsoon seasons (October and November), except during 2008 when a maximum was observed in September month while thereafter it decrease throughout the whole year. Apart from this, AOD have maximum values at longer wavelength during monsoon season (July, August). The AOD values observed over Delhi is little higher than rest part of India, like Ahmedabad (Ramchandran *et al*., 2012), Pune (Pandithurai *et al*., 2007), Vishakhapatnam (Niranjan *et al*., 2004), Bangalore (Sreekanth, 2013). However, it is quite comparable with the values observed at other locations in IGP, *e.g*., at Kanpur (Singh *et al*., 2004; Kaskaoutis *et al*., 2012), Varanasi (Tiwari *et al*., 2013b) and Kharagpur (Niranjan *et al*., 2006). Higher concentration of aerosols loading over IGP is mainly due to the unique topography, large population density and various anthropogenic activities such as industrial, agricultural and vehicular activities.

Fig. 2 shows monthly variation of AOD at 500 nm and AE (α) for the pair of wavelength 340-1020 nm calculated by using equation (3). Being at the vicinity of desert regions, an increase in AOD is observed from March to May, except during 2008 when a slight decrease was observed in April month, which is further observed to be decrease in the month of monsoon, indicating the washout processe of aerosol loading due to rain activities. Further, it increases in post-monsoon months (relatively higher in October) and finally shows a decraease in winter period. In contrast to this, AE have nearly opposite monthly variations to AOD throughout the study period. Similar types of results are also observed by Tiwari *et al*. (2013a) over the entire IGP region. A large monthly variation was observed in AOD (at 500 nm), varied between 0.42 (March 2006) and 1.1 (September 2008). On the other hand, AE has minimum value in June 2006 (~0.5) while maximum was observed in the month of December 2006 (~1.3), suggesting different types (coarse- and fine-modes) of aerosol loading over Delhi. Interestingly, higher AODs was also observed with higher AE values, mostly during winter period, which suggest well admixing types of aerosol over the station. Recently, Tiwari *et al*. (2013a) also reported similar types of aerosol loading over IGP region. In earlier study, Badrinath *et al.* (2009) have reported the transport of biomass burning aerosols from north-west India and other industrial emissions over the IGP region.

Monthly mean values of AOD (500 nm) and AE (340-1020 nm) during the entire study period are given in Table 1. It is noticed from the table that the AOD varied from 0.56 ± 0.18 in March to 0.92 ± 0.04 in November. Relatively large AODs were observed during winter, post-monsoon and summer months with interestingly different magnitude of AE. The AE values were found to be highest during winter and post-monsoon months. Result suggests high loading of fine-mode aerosols over the station, which largely influenced from various anthropogenic emission sources. However, during summer months, the AE values were lowest, suggesting that the aerosol loading is largely influenced with the coarse-mode particles. The AE values were found to be decrease rapidally from January (high value 1.23 ± 0.09) to May (low value 0.67 ± 0.05) and thenafter a concurrent increase was observed from June onwards, which reached to its maximum value in December (1.24 ± 0.11). A decrease in AE from March to May is associated with the dominance of coarse-mode

TABLE 1

Monthly mean values of AOD, AE, SSA and AAE during the entire study period from 2006-2008

Month (2006 - 2008)	AOD (500 nm)	AE (340 - 1020 nm)	SSA (500 nm)	AAE (340 - 1020 nm)
January	0.73 ± 0.02	1.23 ± 0.09	0.98 ± 0.01	0.11 ± 0.01
February	0.62 ± 0.12	1.11 ± 0.04	0.97 ± 0.01	0.37 ± 0.08
March	0.56 ± 0.18	0.81 ± 0.13	0.95 ± 0.02	0.19 ± 0.08
April	0.68 ± 0.06	0.74 ± 0.08	0.95 ± 0.02	1.07 ± 0.33
May	0.79 ± 0.06	0.67 ± 0.05	0.94 ± 0.01	1.87 ± 0.70
June	0.80 ± 0.24	0.73 ± 0.11	0.96 ± 0.02	0.55 ± 0.22
July	0.60 ± 0.19	0.95 ± 0.27	0.96 ± 0.01	0.24 ± 0.27
August	0.78 ± 0.00	1.07 ± 0.00	0.90 ± 0.01	0.21 ± 0.00
September	0.73 ± 0.31	1.10 ± 0.21	0.98 ± 0.01	0.78 ± 0.47
October	0.84 ± 0.13	1.15 ± 0.08	0.95 ± 0.03	1.27 ± 0.13
November	0.92 ± 0.04	1.14 ± 0.13	0.92 ± 0.06	1.65 ± 0.41
December	0.67 ± 0.15	1.24 ± 0.11	0.93 ± 0.08	0.79 ± 0.59

dust particles from the long-range transportation in the western side, mainly from Middle East and Thar Desert regions (Srivastava *et al*., 2011; Tiwari *et al*., 2013a). On the other hand, the higher value of AE represents the dominance of fine mode particles over the station, which could be mainly attributed to the emissions coming from industries, vehicles and biomass burning from north-west India (Srivastava *et al*., 2012b). Similar variations in AE was also reported to be observed at various other locations (Bhuyan *et al*., 2005; Gogoi *et al*., 2011; Lodhi *et al*., 2013).

For the better understanding of aerosol climatology over Delhi, the entire study period is divided in to four different seasons, *viz*., summer (March, April, May, June), monsoon (July, August, September), post-monsoon (October, November) and winter (December, January, February). The seasonal mean variation of spectral AOD was plotted and depicted in Fig. 3 during 2006 - 2008. Large spectral variability in AOD was observed during post-monsoon period, with relatively high AOD values at lower wavelengths and low AODs at higher wavelength; however, *vice-versa* was observed during summer with relatively less spectral variability in AOD. Similar spectral variability in AOD was also reported in previous studies over the IGP using measurements (Srivastava *et al*., 2011; Kaskaoutis *et al*., 2012; Tiwari *et al*., 2013a) and modeling tools (Srivastava *et al*., 2012a). The seasonal mean AOD (at 500 nm) and the corresponding AE (340-1020 nm) values are depicted in Table 2. The AOD values are found to vary from 0.67 ± 0.06 in winter to 0.88 ± 0.06 in post-monsoon whereas nearly similar values were observed during summer (0.71 ± 0.11) and

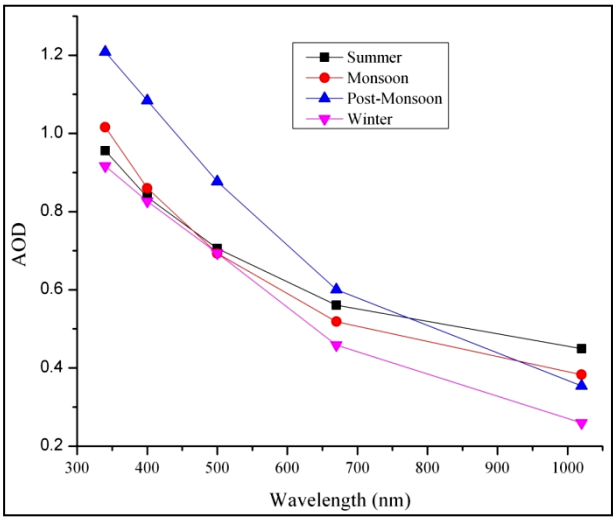

Fig. 3. Seasonal variation of spectral AOD

monsoon (0.70 ± 0.09) seasons. Further, the corresponding AE values observed over the station show relatively lower magnitude during summer (0.74 ± 0.06), suggesting enhanced coarse-mode particles. However, higher AE values were observed during the winter (1.19 ± 0.07) and post-monsoon (1.15 ± 0.01) seasons suggest the enhanced fine-mode aerosols over the station.

For the study of wavelength dependence of AE, it has been calculated for three different wavelength bands, *e.g*., shorter (340-500 nm), longer (500-1020 nm) and broad/total wavelengths (340-1020 nm). Fig. 4 represents the scatter plot of AE between shorter and longer

TABLE 2

Seasonal mean values of AOD AE, SSA and AAE during the study period from 2006-2008

Season (2006 - 2008)	AOD (500 nm)	AE (340 - 1020 nm)	SSA (500 nm)	AAE (340 - 1020 nm)
Summer	0.71 ± 0.11	0.74 ± 0.06	0.95 ± 0.02	0.42 ± 0.34
Monsoon	0.70 ± 0.09	1.04 ± 0.08	0.96 ± 0.03	0.92 ± 0.73
Post-monsoon	0.88 ± 0.06	1.15 ± 0.01	0.93 ± 0.04	0.41 ± 0.32
Winter	0.67 ± 0.06	1.19 ± 0.07	0.96 ± 0.05	1.46 ± 0.27

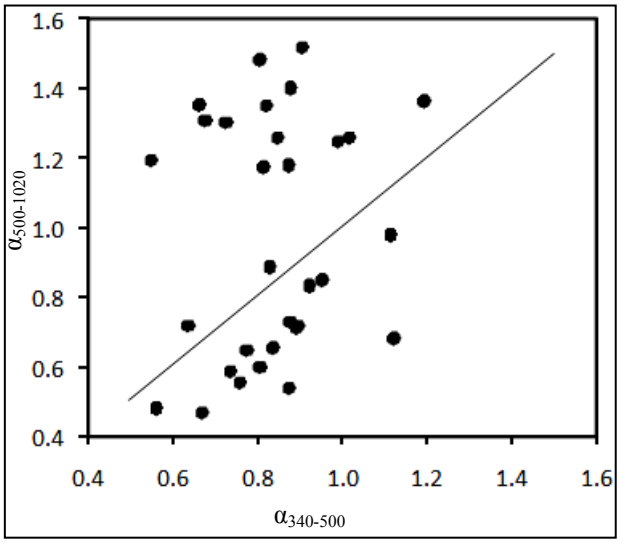

Fig. 4. Scatter plot of Alpha (500-1020 nm) *vs.* Alpha (340-500 nm) during the 2006-2008

wavelengths for different seasons. The straight line represents the slope at unity, on which the AE values for shorter and longer wavelengths would have the same. This straight line also represents the zero curvature line, *i.e.*, $\alpha_2 = 0$ as represented by the equation. The values $\alpha_2 < 0$ lie above the straight line, represents the positive curvature and indicating the dominance of fine-mode aerosol particles (mostly during winter and post-monsoon seasons). On the other hand, $\alpha_2 > 0$ lie below the straight line, represents the negative curvature and indicating the dominance of coarse-mode aerosol particles (mostly during summer and monsoon seasons). Further, ~55% aerosol loading is found to be dominated by the positive curvature and rest 45% is dominated by the negative curvature. A well admixing of aerosol loading was observed, which may be due to the transported natural dust aerosols from desert regions and anthropogenic aerosols from fossil fuel combustion and various industrial activities. In the case of positive curvature, the rate of change of α is more significant at shorter wavelengths whereas in the case of negative curvature, the rate of change is more significant at longer wavelengths.

4.2. *Single scattering albedo*

Single scattering albedo (SSA) is very important parameter for the measurement of fraction of radiation absorbed by the atmospheric aerosols (expressed through 1-SSA) and to study of radiative properties of atmospheric aerosols. It represents the combined effect of scattering and absorption properties of aerosol particles which in combination with surface reflectance determine whether the aerosols are contributing in cooling or heating effects (Satheesh, 2002). SSA is calculated from the scattering optical thickness, which is obtained from the normalized aerosol phase function using diffuse radiance measured at different angles. The detailed methodology for determining the SSA is given by Dubovik *et al*. (1998). SSA varies between 0 and 1, having value close to 1 for highly scattering aerosols (*e.g*., sulphate) and around 0.76 for low scattering aerosols (*e.g*., black carbon) (Saeed *et al*., 2013). Fig. 5 shows monthly spectral variation of SSA during the study period. The spectral behavior of SSA largely depends on the nature of aerosol particles. It is noticed from the figure that relatively large variability in SSA at 1020 nm as compared to rest of the wavelengths, with higher magnitude during May 2006 (0.98) and lower during September 2008 (0.74). The variations in SSA at larger wavelength may be due to presence of near water vapour band (Singh *et al*., 2003). The maximum value of SSA at 500 nm is ~0.99 in the month of November 2006 and minimum value is observed in the month of December 2008, with a value of ~0.84. Apart from this, maximum value of SSA at 400 nm is ~0.98 in November 2006 and January 2008 and minimum value is observed in December 2008 (0.83). We further observed that except at 1020 nm, the magnitude of SSA is <0.95 at other wavelengths during summer season. On the other hand, SSA>0.95 was observed in winter season throughout the study period, suggesting dominance of less absorbing aerosols over Delhi. Based on chemical analysis of sampled aerosols, Tare *et al*. (2006) at Kanpur and Tiwari *et al.* (2009) at New Delhi have observed an enhancement in water-soluble aerosols, mostly scattering type SO_4 aerosols, during winter and suggested to

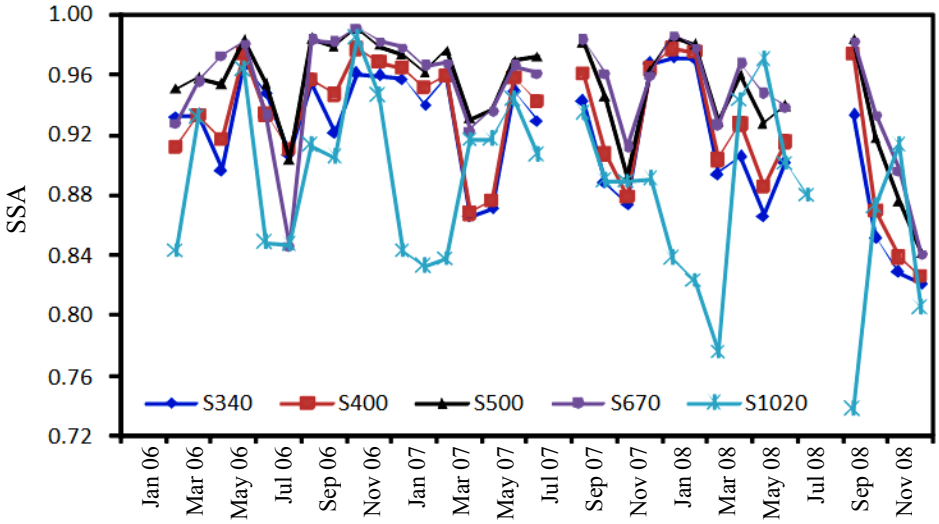

Fig. 5. Monthly variation of SSA at five different wavelengths during the period from 2006-2008

associate with the anthropogenic emission sources, primarily from the burning of fossil fuel and biomass. The monthly mean SSA (at 500 nm) is given in Table 1. It is noticed from the table that the mean SSA over the station was greater than 0.90. A maximum SSA value is observed in the months of January and September (0.98) while minimum is in August (0.90).

Seasonal spectral variation in SSA is shown in Fig. 6. Interestingly different variations in SSA were observed during all the seasons, showing an increase from lower wavelength (340 nm) up to the mid-visible (500 nm) from where it decreases up to the higher wavelength (1020 nm). In a recent study, Giles *et al.* (2012) have reported an increase in SSA with wavelength from UV to visible and suggested the presence of brown or organic carbon. However, in another study, Giles *et al.* (2011) have reported a decrease in SSA with increasing wavelength, and suggested due to the dominance of black carbon. Thus spectral dependence of SSA observed in the present study may suggest the presence of mixture of black carbon, brown and/or organic carbon along with its coating over mineral dusts, transported from the Middle East and Thar Desert regions over the station. In few earlier studies, Singh *et al.* (2004); Pandithurai *et al.* (2008) and Srivastava *et al.* (2011) have shown an increase in SSA with wavelength (measured with sun/sky radiometers) during pre-monsoon or summer seasons, suggesting an influence of transported mineral dusts from the Desert regions. Further, similar spectral variability in SSA was reported over Delhi (model derived) by Singh *et al.* (2010) and Srivastava *et al.* (2012b). The seasonal mean SSA at 500 nm is given in Table 2, and was found to be 0.93 ± 0.02 during post-monsoon and 0.96 ± 0.03 during winter. On the other hand, SSA was observed to be similar during summer and monsoon seasons (~0.95).

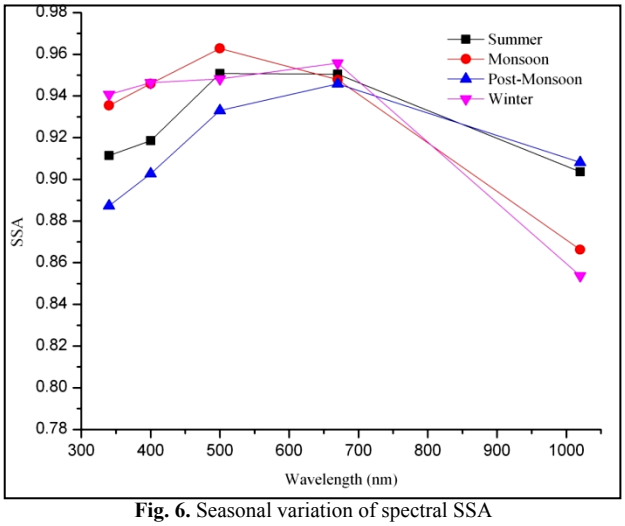

Fig. 6. Seasonal variation of spectral SSA

4.3. *Absorption Ångström exponent*

The Absorption Angström Exponent (AAE), which gives the change in light absorption as a function of wavelength (λ), is computed from the negative slope of the fitted line of the natural logarithm of spectral information about the absorption aerosol optical depth (*i.e.*, AAOD), which is analogues to the Ångström relationship and can be obtained as by Russell *et al.* (2010) as

$$AAOD(\lambda) = [1-SSA(\lambda)] \times AOD(\lambda) \quad (5)$$

This is a well known theoretical equation and the magnitude of AAE is obtained from the negative of the slope of the fitted line of the natural logarithm of spectral variations in AAOD. Theoretically, the AAE value near to unit suggests the existence of freshly emitted black

carbon; however, the AAE value between 1.0 and 2.0 suggests biomass burning aerosols and a value greater than 2.0 suggests the contribution from mineral dust aerosols (Russell *et al*., 2010). The present study location is surrounded by the variety of anthropogenic emission sources and situated at the proximity of Desert regions, which may consequently have slightly different AAE values due to complex mixture of natural desert dusts and anthropogenic aerosols. The estimated monthly and seasonal mean values of AAE (340-1020 nm) over the station during the entire study period are shown in Table 1 and 2, respectively. Significant variability in the magnitude of AAE can be seen from Table 1, which is found to vary between 0.11 ± 0.01 (January) and 1.87 ± 0.70 (May). From the large variability of AAE, the present study suggests a possible mixing of variety of aerosols from different emission sources over the station. In another study, Kirchstetter *et al*. (2004) have suggested that the deviations in AAE from 1.0 is likely due to 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.

On seasonal basis (Table 2), AAE values are found to about 0.42 ± 0.34, 0.92 ± 0.73, 0.41 ± 0.32 and 1.46 ± 0.27 during winter, summer, monsoon and post-monsoon seasons, respectively. Relatively large AAE value during the post-monsoon period (1.46 ± 0.27) is mainly due to the influence of biomass burning aerosols (Russell *et al*., 2010) from the agricultural activities, which is one of the major sources over the station during the period mainly from north-west part of India. Further, fire crackers burning during Diwali (celebrated during the post-monsoon period) may be the other possible source over the station. On the other hand, the magnitude of AAE less than 1.0 (by ~55% time, mostly during winter and monsoon) suggests possible coating of absorbing or non-absorbing substance over absorbing aerosols (particularly BC) as suggested by Gyawali *et al*. (2009). The mean AAE value during the entire study period is found to be about 0.84 ± 0.66. In a previous study over Delhi, Soni *et al*. (2010) have reported the AAE value close to unity and suggested due to the abundance of BC aerosols from fossil fuel combustion sources whereas large AAE value (>1.0) indicates absorbing aerosols from biomass/biofuel burning sources, as suggested by Russell *et al*. (2010).

5. Conclusions

(*i*) Relatively large enhancement in AOD from automatic sun/sky radiometer during 2008 as compared to 2006 and 2007, with nearly equal values during winter (0.67 ± 0.06) and summer (0.71 ± 0.11) were observed over Delhi during 2006-2008.

(*ii*) AE value was found to be higher during winter (1.19 ± 0.07) indicating dominance of fine-mode aerosols whereas a lower AE during summer (0.74 ± 0.06) suggests dominance of coarse-mode aerosols.

(*iii*) A slight decrease in SSA was observed over the station during the entire study period (with a mean value of ~0.9), suggesting absorbing nature of aerosols. Seasonally, the mean SSA was about 0.93 during post-monsoon and about 0.96 during the winter period whereas during summer and monsoon, SSA was ~0.95.

(*iv*) The estimated AAE value was found to vary between 0.11 (January) and 1.87 (May), suggesting mixed type aerosols from the variety of emission sources. AAE was found to be less than 1.0 by ~55% time (mostly during winter and monsoon), suggesting the possible coating of absorbing or non-absorbing substance over absorbing aerosols (particularly BC). On the other hand, AAE greater than 1.0 was observed by ~45 time (mostly during summer and post-monsoon) suggesting the dominance of mixing of aerosols from desert dust and biomass burning sources.

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