

Comparative assessment of aerosol optical properties over a mega city and an adjacent urban area in India

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सार – यह शोध पत्र महानगर दिल्ली तथा निकटवर्ती शहरी क्षेत्र रोहतक में वायुविलय के प्रकाशिक गुणों के तुलनात्मक विश्लेषण से संबंधित है। यहाँ यह उल्लेख करना प्रासंगिक है कि काफी नजदीक का क्षेत्र होने और सदृश मौसम विज्ञानिक स्थितियों के बावजूद दोनों स्थानों के वायुविलय के लक्षणों में काफी भिन्नता पाई गई है। इस अध्ययन में धरातल पर संस्थापित स्काई रेडियो मीटर से प्राप्त एक वर्ष की अवधि के आँकड़ों को लेकर कार्य किया गया है। इसमें दिल्ली और रोहतक में 500 नैनो मी. की ऊँचाई पर औसत वार्षिक वायुविलय प्रकाशिक गहराई (AOD) क्रमशः 1.01 और 0.73 प्रेक्षित की गई है, इसमें सहसंबंध गुणांक 0.67 तथा औसत निरपेक्ष अंतर 0.51 है। मॉनसूनोत्तर ऋतु को छोड़कर पूरे वर्ष में दिल्ली में वायुविलय प्रकाशिक गहराई (AOD) की मात्रा रोहतक से अधिक पाई गई है। इन स्टेशनों के बीच आंगस्ट्रॉम एक्सपोनेंट (अल्फा) में अन्तर बहुत ही कम है तथापि रोहतक में अल्फा के परिणाम कम हैं जो खुरदरे प्रकार के कणों की अधिक सघनता की मौजूदगी को दर्शाते हैं। पूरे वर्ष के दौरान दिल्ली में एकल प्रकीर्णन एल्बीडो (SSA) भी ऋतुनिष्ठ भिन्नताओं के मान काफी कम दर्शाते हैं जिससे अवशोषक एरोसोल्स (काला कार्बन जैसा) के योगदान का पता चलता है। दिल्ली में सूक्ष्म कणों की सघनता की मात्रा रोहतक से अधिक पाई गई है जिसमें धूल, वाहन-चालन, जैविक कूड़ा का जलना तथा औद्योगिक उत्सर्जनों का संयुक्त प्रभाव है। वायुविलय प्रकाशिक गहराई (AOD) और अल्फा के संबंध के आधार पर एरोसोल-वर्गीकरण से पता चलता है कि शहरी/जैविक कचरा के जलने से उत्पन्न वायुविलय दिल्ली में अधिक रहता और रोहतक में सर्दी तथा मॉनसून पूर्व में मिश्रित प्रकार (MT) को एरोसोल रहता है।

ABSTRACT. The present work revolves around the comparative analysis of aerosol optical properties in a mega city, Delhi and in a nearby urban area, Rohtak. It is pertinent to note that despite of the close proximity and similar meteorological conditions, the two study locations show significant differences in aerosol characteristics. The study is conducted using ground based Sky-radiometer measurements for a period of one year. The mean annual Aerosol Optical Depth (AOD) at 500 nm over Delhi and Rohtak is observed to be 1.01 and 0.73 respectively, with correlation coefficient of 0.67 and mean absolute difference of 0.51. The magnitude of AOD in Delhi is higher than in Rohtak throughout the year, except in post-monsoon season. The difference in Angstrom exponent (alpha) between the stations is minimal. However, lower magnitude of alpha is observed in Rohtak, indicating presence of more concentration of coarse-mode particles. Single Scattering Albedo (SSA) also shows seasonal variation with significantly lower values in Delhi throughout the year, indicating contribution of absorbing type of aerosols (like black carbon). The volume concentration in fine-size is found to be higher in Delhi than in Rohtak, indicating combined effect of dust, vehicular, biomass burning and industrial emissions. The aerosol classification via relationship between AOD and alpha shows that urban/biomass burning (U/B) aerosols are dominant in Delhi and mixed type (MT) aerosols in Rohtak during winter and pre-monsoon.

Key words – Sky radiometer, Aerosol optical depth, Single scattering albedo, Volume fraction.

1. Introduction

The rapid growth in population and urbanization have led to significant rise in land use change, industrialization and transportation sector in the last few decades, resulting in steep rise in pollution level. The

increasing atmospheric aerosol load has a direct impact on the local, regional and global climate. Atmospheric aerosols play an important role in determining Earth's radiation budget directly by scattering and absorbing incoming solar radiation (Clement *et al.*, 2009; Junwei *et al.*, 2012) and indirectly by modifying the cloud

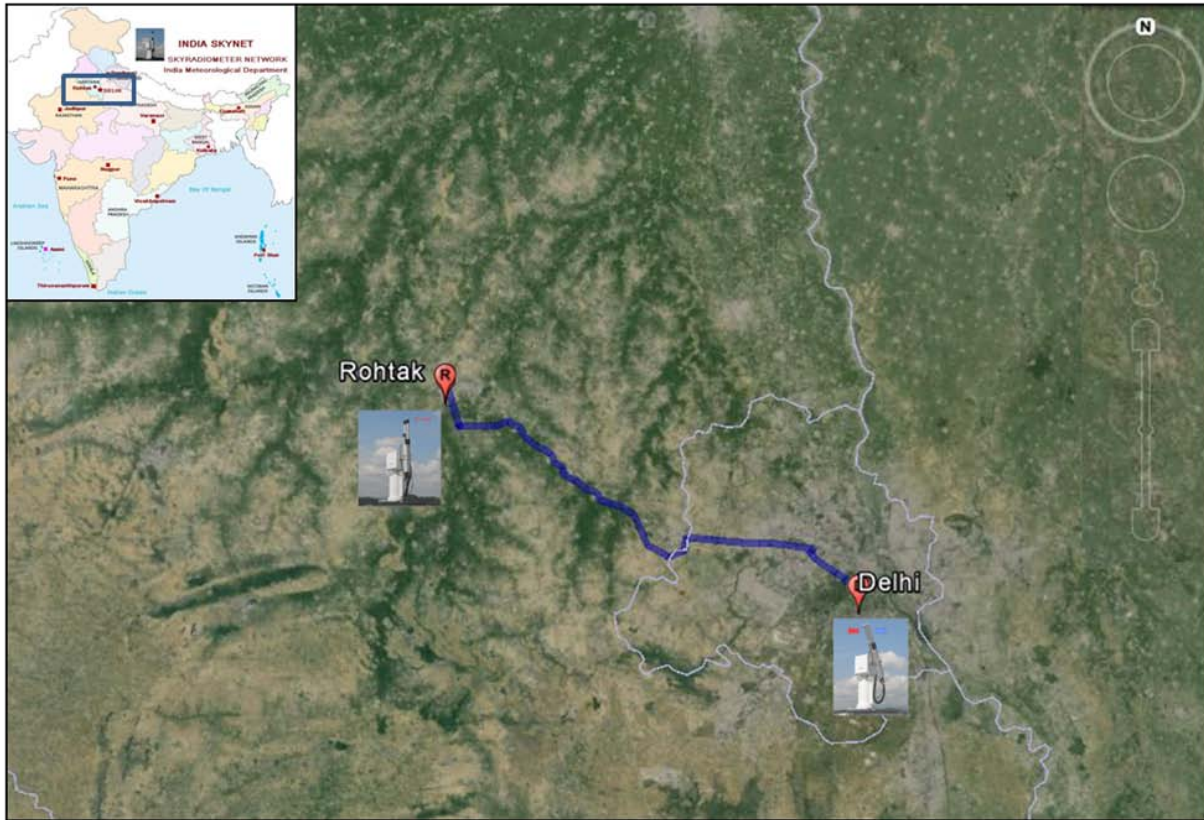


Fig. 1. Google earth map showing Delhi and Rohtak Skynet stations

microphysical processes (Haywood and Boucher 2000; Ramanathan *et al.*, 2001). The ability of aerosols to affect Earth's radiation budget, air quality and human health depends on their size and chemical characteristics (Che *et al.*, 2009; Dusek *et al.*, 2006; Dockery 2009). The chemical characteristics of aerosols depend on the nature of sources while size distribution depends on production mechanism and the meteorological conditions during their life time. Generally, anthropogenic aerosols are predominantly submicron in size while the naturally produced aerosols such as sea-salt and mineral dust tend to be super micron in size (coarse mode regime).

To understand the effect of aerosols on climate, it is essential to know their optical properties as well as spatial and temporal variability (Dubovik *et al.*, 2002). The aerosol optical properties are highly variable in space and time because of shorter atmospheric lifetime of aerosols (Kaufman *et al.*, 2002). Aerosol optical properties exhibit diurnal, seasonal and inter-annual variations even if a certain aerosol type is prevailing at one location, due to different meteorological conditions (Eck *et al.*, 2001; Pandithurai *et al.*, 2007). Several researchers have studied the spatial coverage of aerosol optical properties using satellite data (Dey and Girolamo, 2010; Guleria *et al.*,

2011; Ramachandran *et al.*, 2012; Taneja *et al.*, 2016). Ground-based instruments provide continuous and more reliable aerosol optical properties because of lower uncertainties as compared to satellite retrieved data. There are two well established ground-based aerosol monitoring networks called SKYNET (Nakajima *et al.*, 1996) and AERONET (Holben *et al.*, 2001), which use the PREDE skyradiometers and CIMEL CE-318 sunphotometers respectively. Che *et al.* (2008) reported that the difference between the two types of instruments was less than 1.3% for AOD and less than 4% for the single scattering albedo below the wavelength of 670 nm at Beijing. Single scattering albedo estimates retrieved by SKYRAD.PACK 4.2 inversion are larger relative to AERONET measurements.

The south Asia is one of the most densely populated regions of the world and recognized to be among hotspots of anthropogenic pollutants (Kaufman *et al.*, 2002; Srivastava *et al.*, 2012; Tripathi *et al.*, 2005). The atmospheric loading of aerosols over the Indian region is found to be increasing in the past decade (Dani *et al.*, 2010; Habib *et al.*, 2006; Lodhi *et al.*, 2013; Moorthy *et al.*, 1993; Moorthy *et al.*, 2005; Soni *et al.*, 2003; Soni *et al.*, 2012). Northern India along the Indo-Gangetic Plain

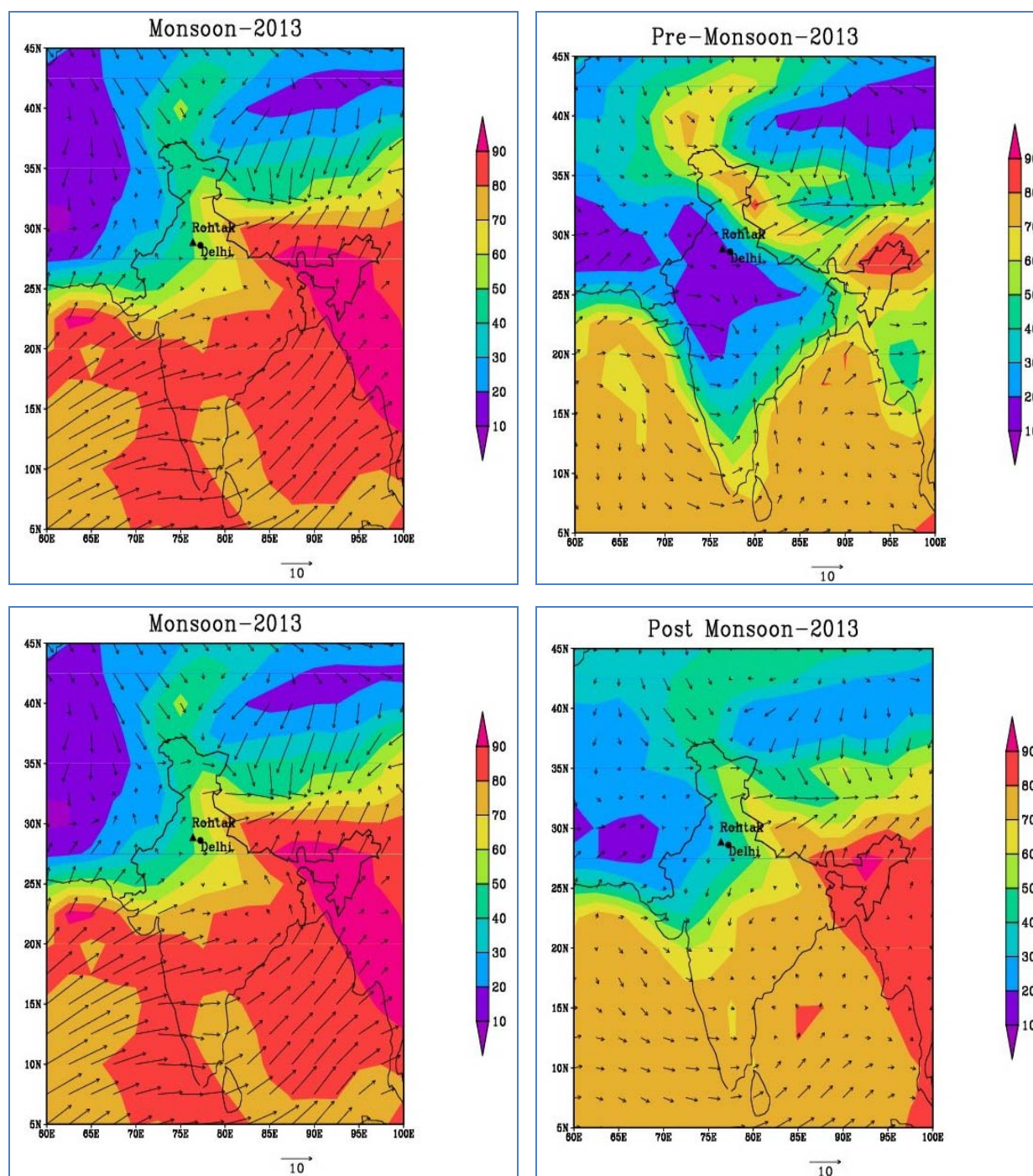


Fig. 2. Wind pattern and relative humidity (at 1000 hPa level) for four seasons over the Indian subcontinent and surrounding regions using NCEP/NCAR reanalysis dataset

exhibit more aerosol optical depth as compared to the southern part of India, mainly due to the influence of anthropogenic pollution and dust transported from the Thar Desert in northwestern India and from Arabian Peninsula (Prasad and Singh, 2007; Singh *et al.*, 2004;

Sarkar *et al.*, 2006; Dey and Girolamo, 2011; Dey *et al.*, 2004). A few studies have highlighted the variability of aerosols over sites with distinct weather and climate differences in India. Patel *et al.* (2015) demonstrated the variability of aerosol optical properties over a continental

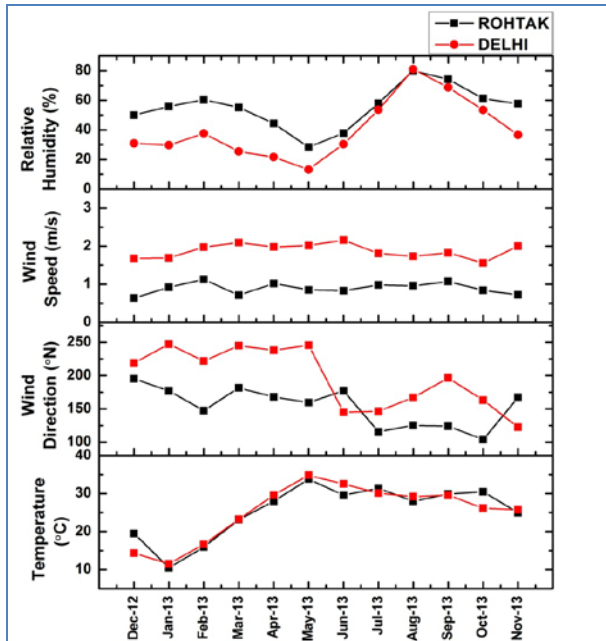


Fig. 3. Meteorological conditions during the study period at the experimental sites

site Dehradun and a marine site Kavaratti in India during pre-monsoon, highlighting the obvious differences in the aerosol characteristics, due to the different types and sources of aerosol and differences in local climate. Kant *et al.* (2015) analyzed the variation of optical and radiative properties of aerosols and black carbon over Dehradun and Patiala located in the northwest region of India, using synergy of ground based and satellite observations during pre-monsoon season. Despite of many research papers published in the recent years, studies on aerosols are far from being sufficient because of large differences in geographic regions in India.

In the recent years, the rapid increase in energy consumption and vehicular emission has caused severe aerosol pollution in Delhi. The aerosol concentration in Delhi is contributed by vehicular and industrial activities, dust transported from desert regions, soil originated particles and suspended dust generated by wind and excessive construction activities (Jain *et al.*, 2007; Dey *et al.*, 2004; Taneja *et al.*, 2014). Aerosol in Delhi region can be best described by a combination of urban and desert aerosols (Pandithurai *et al.*, 2008; Singh *et al.*, 2005).

Fan *et al.* (2015) studied the column-integrated aerosol optical and physical properties in an urban and suburban site on the North China Plain, revealing the comparative analysis of aerosol pollution at the two nearby sites. Very few similar studies have provided

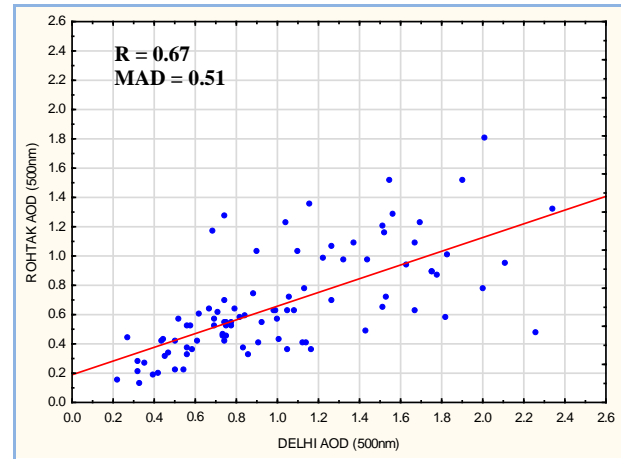


Fig. 4. Correlation between AOD (500 nm) at Delhi and Rohtak

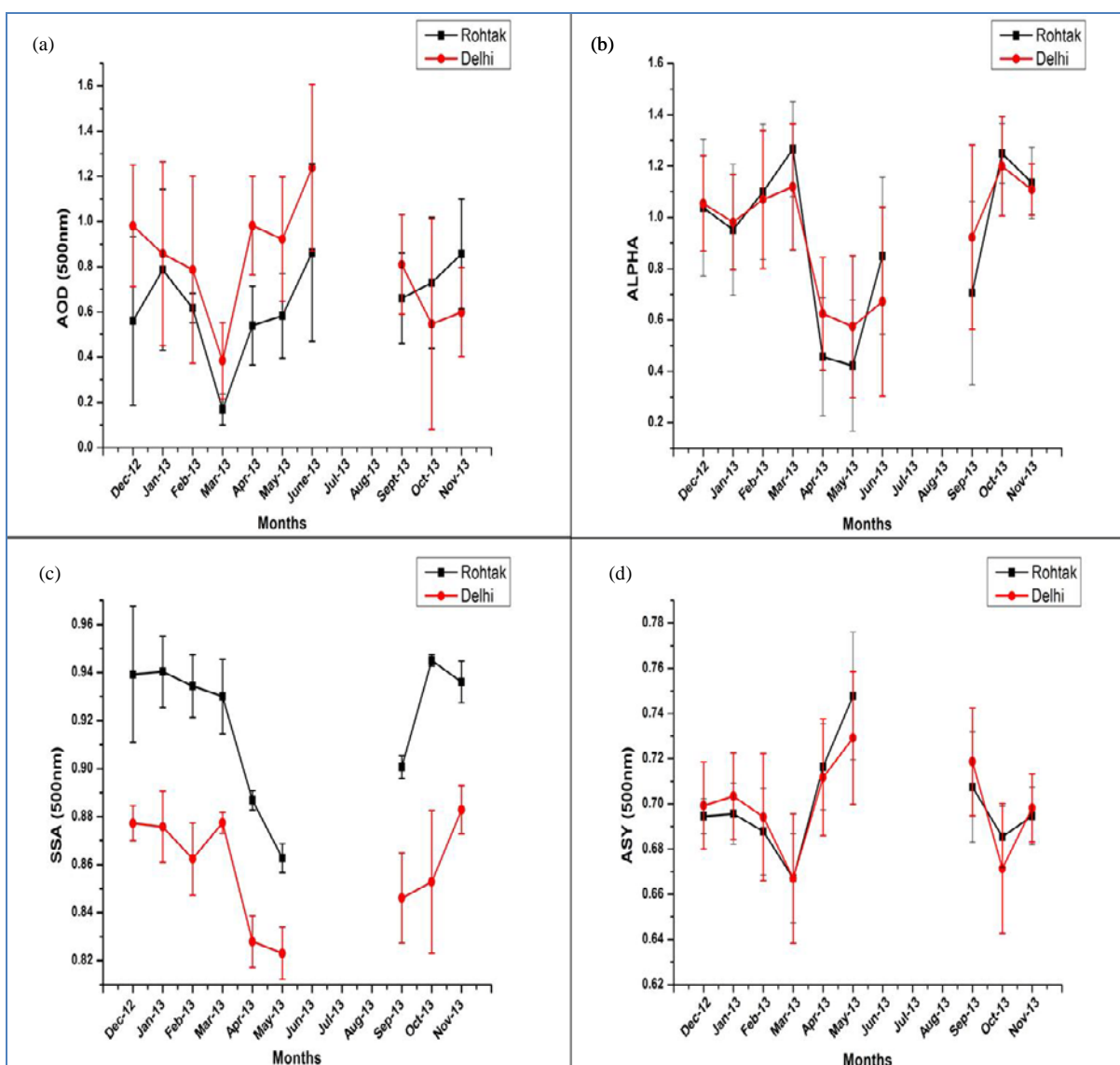
knowledge on aerosol variability over urban and suburban locations in India, using the ground based observations (Sharma *et al.*, 2014). A preliminary research on aerosol optical properties has been carried out by Mor *et al.* (2016). However, a comprehensive study on aerosol optical properties over Rohtak has not been published yet. In this paper, we present some results of aerosol optical properties and meteorological variables derived from the mega city Delhi and nearby urban area Rohtak from December 2012 to November 2013. Although both the regions are in close proximity to each other, there exists a significant difference in aerosol optical properties, which forms the basis of this study. The study examines and compares the aerosol characteristics of two stations which are having nearly the same meteorological conditions and topography but different aerosol optical properties. Diurnal and seasonal variations of aerosol optical properties and possible causes of their variation are discussed in the study.

2. Study regions and meteorology

2.1. Site description

Delhi (28.59° N, 77.22° E and 213 m amsl) is an urban megacity and national capital of India, with a population of over 16 million people as per Census of India, 2011. The total vehicle population was more than 7 million at the end of 2012 in Delhi (Economic survey of Delhi, 2012-13). Delhi is surrounded by the Indo-Gangetic plains in the North and East, the arid region Thar desert in the West and mountain range Aravalli in the South.

Rohtak city (28.89° N, 76.58° E, 214 m amsl), with a population of 0.37 million (as per the Census of



Figs. 5(a-d). Monthly variation of (a) Aerosol optical depth and (b) Angstrom Exponent (c) Single Scattering Albedo and (d) Asymmetry Parameter at 500 nm along with standard deviation at Rohtak and Delhi

India, 2011) is located 70 km northwest of Delhi (Fig. 1). The city is surrounded by agricultural land and a few small-scale industries. The city is connected to Delhi through National Highway. Due to proximity to the National capital, Rohtak has witnessed rapid industrialisation, urbanisation and diversification in agriculture.

2.2. Meteorological conditions

India Meteorological Department has categorized four main seasons for the country. Winter season comprises of January and February but December can also

be included in this season for northern parts of the country. March, April and May constitutes the summer season (pre-monsoon) in India. June, July, August and September form the core of south-west monsoon season in most parts of the country. The onset of monsoon season occurs in the first week of July and withdrawal starts during middle of September at Delhi and Rohtak. Post-monsoon season starts from October and continues till November (Attri and Tyagi, 2010).

Meteorological variables such as wind, relative humidity (RH), rainfall and temperature play a major role in distribution of aerosols throughout the atmosphere. The

wind and relative humidity patterns from NCEP/NCAR reanalysis (<http://nomads.ncep.noaa.gov>) from December, 2012 to November 2013 at 1000 hPa level over Indian region (0-40° N, 60°-100° E) for winter, pre-monsoon, monsoon and post-monsoon are depicted in Fig. 2. The graphical representation of monthly variations in meteorological conditions at the two stations is given in Fig. 3. The winter season is characterized by low temperature (10-20 °C) at both the locations; with RH around 30% in Delhi and 55-60% in Rohtak, accompanied with low wind speed in Rohtak (<1.0 m/s) and Delhi (< 2.0 m/s), mainly from south-southwesterly direction. The pre-monsoon months (March-May) over Delhi and Rohtak are characterized by strong winds, high temperature (30-40 °C), and low relative humidity (10-40%). The prevailing weather patterns during this season transport dust particles from western desert regions of the Thar Desert, Middle East, as well as the Arabian Peninsula. The monsoon season shows high RH (>70%) and low wind speeds (~1 m/s in Rohtak and 1.5 m/s in Delhi) with moderately high temperature (average 25-30 °C). During this season, the winds are generally south-southeasterly at the study locations. The post-monsoon season has temperature in the range of 25-30 °C, lower wind speed (~1 m/s in Rohtak and ~2 m/s in Delhi) and RH (55-65% in Rohtak and 40-60% in Delhi). The differences in meteorological parameters are minimal over both the locations. Slightly higher average RH, with lower wind speed and temperature are observed over Rohtak than Delhi during the study period. The aerosol optical properties are analyzed for seasons of winter, pre-monsoon and post-monsoon. The analysis during monsoon season is not presented in this study, as not enough data could be retrieved from sky radiometer due to lack of cloud free sky conditions during this season.

3. Data and methodology

Sky-radiometer (model POM-02) manufactured by Prede Co. Ltd, Japan, has been used in the Skynet-India network of India Meteorological Department (IMD). The data has been retrieved from Delhi and Rohtak stations for the period of one year from December 2012 to November 2013. The sky-radiometer measures direct solar and solar aureole radiance in narrow wavelength bands of the solar spectrum (340, 380, 400, 500, 675, 870, 1020 nm) at various scattering angles from the Sun (Nakajima *et al.*, 1996). These observations are used to retrieve optical parameters of aerosols such as Aerosol Optical Depth (AOD), Angstrom Exponent (Alpha), Single Scattering Albedo (SSA), Asymmetry parameter (ASY) and volume size distribution using the 'Skyrad.Pack v4.2' developed by (Nakajima *et al.*, 1996). One major advantage of using sky-radiometer POM-02 is the onsite calibration for solid view angle and for absolute sensitivity. In the normal

Langley method of calibration, it is assumed that AOD is constant during the calibration process which is not valid in most situations. An error of approximately 10% was estimated by Shaw (1976) while deducing calibration constant from the normal Langley method for a typical urban station. Thus, an improved/modified Langley method of calibration based on both direct and diffuse radiation data is used here (Tanaka *et al.*, 1986; Nakajima *et al.*, 1996). The uncertainties in retrievals of different optical properties of aerosols like AOD, SSA and volume size distributions have been analyzed for Delhi in 2006 by Pandithurai *et al.* (2008).

4. Results and Discussion

4.1. Seasonal variations of aerosol optical properties at Delhi and Rohtak

4.1.1. Aerosol Optical Depth (AOD)

AOD represents the aerosol load in the atmospheric column and an important parameter for the identification of source regions and evolution of aerosols. It is important to understand the diurnal and seasonal variability, as aerosols have very short life time and are influenced by meteorological parameters such as wind, temperature and relative humidity. The annual mean AOD at 500 nm over Delhi and Rohtak is observed to be 1.01 and 0.73 respectively, with correlation coefficient (R) calculated as 0.67 and mean absolute difference (MAD) as 0.51 (Fig. 4). There is approximately 37% variance in the AOD in Delhi and Rohtak. A moderately strong relationship between the AODs at the two locations indicates that the aerosol pollution in Delhi is regional in nature. However, there are significant seasonal differences in the aerosol optical properties at the two locations which are discussed in the following sections. Fig. 5(a) shows the comparison of monthly averaged AOD (at 500 nm) for Delhi and Rohtak with error bars showing standard deviations. Table 1 shows the monthly mean values along with $\pm 1\sigma$ standard deviation of aerosol optical properties at 500 nm for Delhi and Rohtak for the year 2013.

Seasonal variation in AOD occurs due to changes in atmospheric conditions and anthropogenic activities. The monthly variation in AOD is evident with moderate to high AOD found in winter months (0.875 ± 0.081 in Delhi and 0.575 ± 0.050 in Rohtak). During this period, fine mode particles dominate in the atmosphere. However, a sudden drop in AOD is noted in the month of March at both the stations, as it marks the onset of pre-monsoon season with change in wind regime. Similar decrease in AOD during March has been reported in different parts of IGP (Kedia *et al.*, 2014; Pandithurai *et al.*, 2008;

TABLE 1

Monthly mean values of aerosol optical properties (along with $\pm 1\sigma$ standard deviation) at 500 nm in Delhi and Rohtak for 2013

| Months | ALPHA | | AOD | | SSA | | ASY | |
|--------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| | Delhi | Rohtak | Delhi | Rohtak | Delhi | Rohtak | Delhi | Rohtak |
| Jan | 0.981 \pm 0.185 | 1.052 \pm 0.255 | 0.858 \pm 0.405 | 0.785 \pm 0.356 | 0.875 \pm 0.014 | 0.940 \pm 0.014 | 0.981 \pm 0.185 | 1.052 \pm 0.255 |
| Feb | 1.069 \pm 0.269 | 1.126 \pm 0.263 | 0.787 \pm 0.414 | 0.617 \pm 0.065 | 0.862 \pm 0.015 | 0.934 \pm 0.013 | 1.069 \pm 0.269 | 1.126 \pm 0.263 |
| Mar | 1.119 \pm 0.245 | 1.277 \pm 0.184 | 0.384 \pm 0.168 | 0.169 \pm 0.068 | 0.877 \pm 0.004 | 0.930 \pm 0.015 | 1.119 \pm 0.245 | 1.277 \pm 0.184 |
| Apr | 0.624 \pm 0.219 | 0.437 \pm 0.230 | 0.982 \pm 0.218 | 0.539 \pm 0.173 | 0.827 \pm 0.010 | 0.886 \pm 0.004 | 0.624 \pm 0.219 | 0.437 \pm 0.230 |
| May | 0.573 \pm 0.276 | 0.448 \pm 0.256 | 0.922 \pm 0.274 | 0.582 \pm 0.187 | 0.823 \pm 0.010 | 0.862 \pm 0.006 | 0.573 \pm 0.276 | 0.448 \pm 0.256 |
| Sep | 0.748 \pm 0.359 | 0.749 \pm 0.357 | 0.809 \pm 0.220 | 0.660 \pm 0.198 | 0.846 \pm 0.018 | 0.901 \pm 0.005 | 0.748 \pm 0.359 | 0.749 \pm 0.357 |
| Oct | 1.256 \pm 0.193 | 1.249 \pm 0.116 | 0.546 \pm 0.465 | 0.729 \pm 0.289 | 0.852 \pm 0.029 | 0.945 \pm 0.002 | 1.256 \pm 0.193 | 1.249 \pm 0.116 |
| Nov | 1.113 \pm 0.099 | 1.208 \pm 0.138 | 0.598 \pm 0.197 | 0.857 \pm 0.243 | 0.882 \pm 0.009 | 0.936 \pm 0.008 | 1.113 \pm 0.099 | 1.208 \pm 0.138 |
| Dec | 1.054 \pm 0.184 | 1.037 \pm 0.265 | 0.981 \pm 0.269 | 0.560 \pm 0.373 | 0.877 \pm 0.007 | 0.939 \pm 0.028 | 1.054 \pm 0.184 | 1.037 \pm 0.265 |

TABLE 2

Seasonal average values of volume size distribution parameters over Delhi and Rohtak

| Season | Location | R ^a _f | R _c | V ^b _f | V _c |
|---------------|----------|-----------------------------|----------------|-----------------------------|----------------|
| Winter | Delhi | 0.173 | 11.310 | 0.034 | 0.102 |
| | Rohtak | 0.173 | 16.540 | 0.025 | 0.128 |
| Pre- Monsoon | Delhi | 0.118 | 11.310 | 0.035 | 0.254 |
| | Rohtak | 0.791 | 7.734 | 0.028 | 0.133 |
| Post- Monsoon | Delhi | 0.791 | 5.289 | 0.033 | 0.160 |
| | Rohtak | 0.173 | 16.540 | 0.04 | 0.221 |

^aR: Volume median radius.^bV: Volume concentration (Subscripts 'f' and 'c' represent fine and coarse mode particles)

Singh *et al.*, 2010). As the season progresses, the dust raising convective activities also rise significantly in northern India. Significant increase in AOD has been observed from March to June, which can be ascribed to the increase in dust particle concentration as the westerly winds are from the arid and semiarid regions. The maximum AOD is observed in June with a value of 1.238 ± 0.369 at Delhi and 0.863 ± 0.393 at Rohtak, reflecting the influence of dust activities. A decline in AOD is observed for post-monsoon season, indicating the preferential removal of dust aerosols from atmosphere by rain and dominance of finer mode particles. The AOD value reduces to 0.546 ± 0.465 at Delhi and 0.729 ± 0.290 at Rohtak in October. It is pertinent to note that Delhi shows higher AOD as compared to Rohtak in all the seasons except in post-monsoon. This can be attributed to the fact that Rohtak is surrounded by agricultural land and

October month marks the harvesting period for Kharif crops. Due to short time available between harvesting of previous crop (late October and early November) and sowing of succeeding crop (November), farmers burn crop residues. This adds to the fine mode aerosols in the atmosphere during post-monsoon. Also, the biomass burning aerosols transported to the region from Punjab contributes to the rise in AOD during post-monsoon (Sharma *et al.*, 2010).

4.1.2. Angstrom Exponent (Alpha)

Alpha can be computed from the spectral values of the measured aerosol optical depth and is a useful quantity to assess aerosol size (Liu *et al.*, 2008). The value of Alpha is the most relevant qualitative indicator of aerosol size (Angstrom, 1929). Alpha is inversely

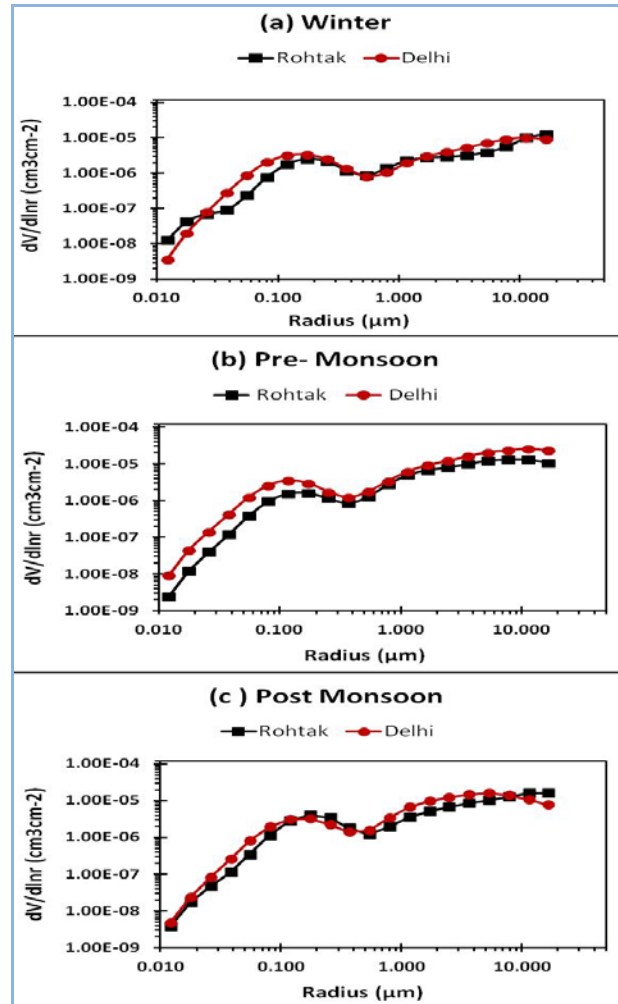
related to aerosol size, *i.e.*, smaller the aerosol larger the Alpha. Values of Alpha less than 1.0 indicate size distribution dominated by coarse mode aerosols that are typically associated with dust and sea salt. Whereas, values of Alpha generally range from more than 2.0 when size distribution is dominated by fine mode aerosols, usually associated with urban pollution and biomass burning (Eck *et al.*, 1999; Westphal and Toon, 1991).

Higher Alpha and AOD values were recorded during winter season at both the places indicating an increase in the contribution of fine mode aerosols [Fig. 5(b)]. A significant decrease in Alpha is observed during April and May with monthly average values less than 0.46 at Rohtak and 0.62 at Delhi. Dust raising convective activities is common during pre-monsoon season in northern India. Also the dust from Thar region and Arabian Peninsula gets transported during this season. Lower values of alpha during April and May at Rohtak indicate prominence of coarse mode particles, characterized by dust load. Higher alpha in Delhi is due to higher influence of fine mode particles, such as black carbon from vehicular, biomass burning and industrial emissions.

During post-monsoon, Alpha increased to 1.200 ± 1.93 at Delhi and 1.249 ± 0.117 at Rohtak, with corresponding decrease in AOD. The rain washes out the coarse mode particles in the atmosphere and hence results in lower alpha values.

4.1.3. Single Scattering Albedo (SSA)

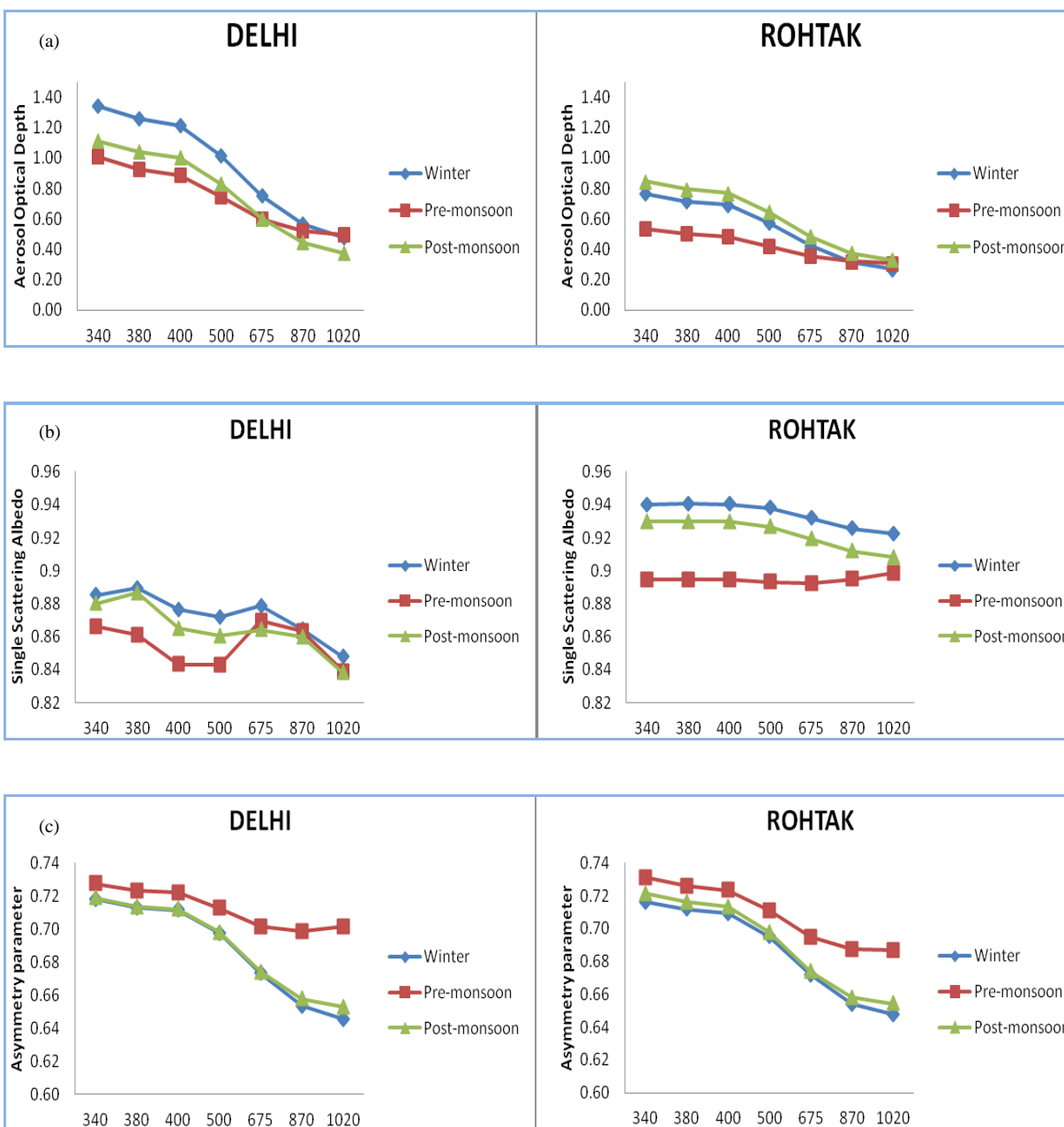
Single scattering albedo is the measure of reflectivity of aerosol particles and a key optical characteristic in assessing the radiative effects of aerosols. SSA is defined as the ratio of scattering efficiency to total extinction efficiency (a sum of scattering and absorption) of aerosol particles. SSA tends towards 1 if scattering dominates over absorption of radiation by atmospheric aerosols. Monthly mean SSA retrieved by sun/sky radiometer at 500 nm is shown in Fig. 5(c) for Delhi and Rohtak for the study period. During winter months, the average SSA is found to be 0.871 in Delhi and 0.938 in Rohtak. Whereas, a gradual decline in SSA is observed from March to May at both the stations, indicating the presence of more absorbing particles during the pre-monsoon months. This is in agreement with the earlier reported values over Indo-Gangetic Plain during pre-monsoon period by Pandithurai *et al.*, 2008. In a study by Moorthy *et al.* (2007), using the ground based and satellite data. It was found that dust from the Indian desert is more absorbing in nature as compared to the African dust. In the current study, a sudden rise in SSA is observed during post monsoon months, with average value of 0.927 in Rohtak and 0.860



FIGS. 6(a-c). Seasonal averaged volume size distributions at Delhi and Rohtak

in Delhi. This is due to the dominance of finer absorbing aerosols (Singh *et al.*, 2004).

Though, average SSA is observed to be more than 0.85 at both the stations, it is found to be relatively lower at Delhi. This can be attributed to the possible mixing of dust with absorbing aerosols (black carbon, sulphates and nitrates). Delhi is one of the most polluted urban regions where vehicular, thermal power plants and other industrial emissions are the major sources of pollution. Delhi SSA values are consistent with the values reported by Pandithurai *et al.* (2008), indicating stronger contribution from absorbing aerosols which are a mixture of both desert dust and anthropogenic (vehicular and industrial) aerosols. Moorthy *et al.* (2007) reported that the ability of dust particles to scatter and absorb light can be changed depending on which species that combine with dust aerosol. Higher SSA values over Rohtak are probably due to the fact that the soil and road dust of Rohtak have lesser



Figs. 7(a-c). Spectral variation of (a) AOD, (b) SSA and (c) ASY at different wavelengths over Rohtak and Delhi

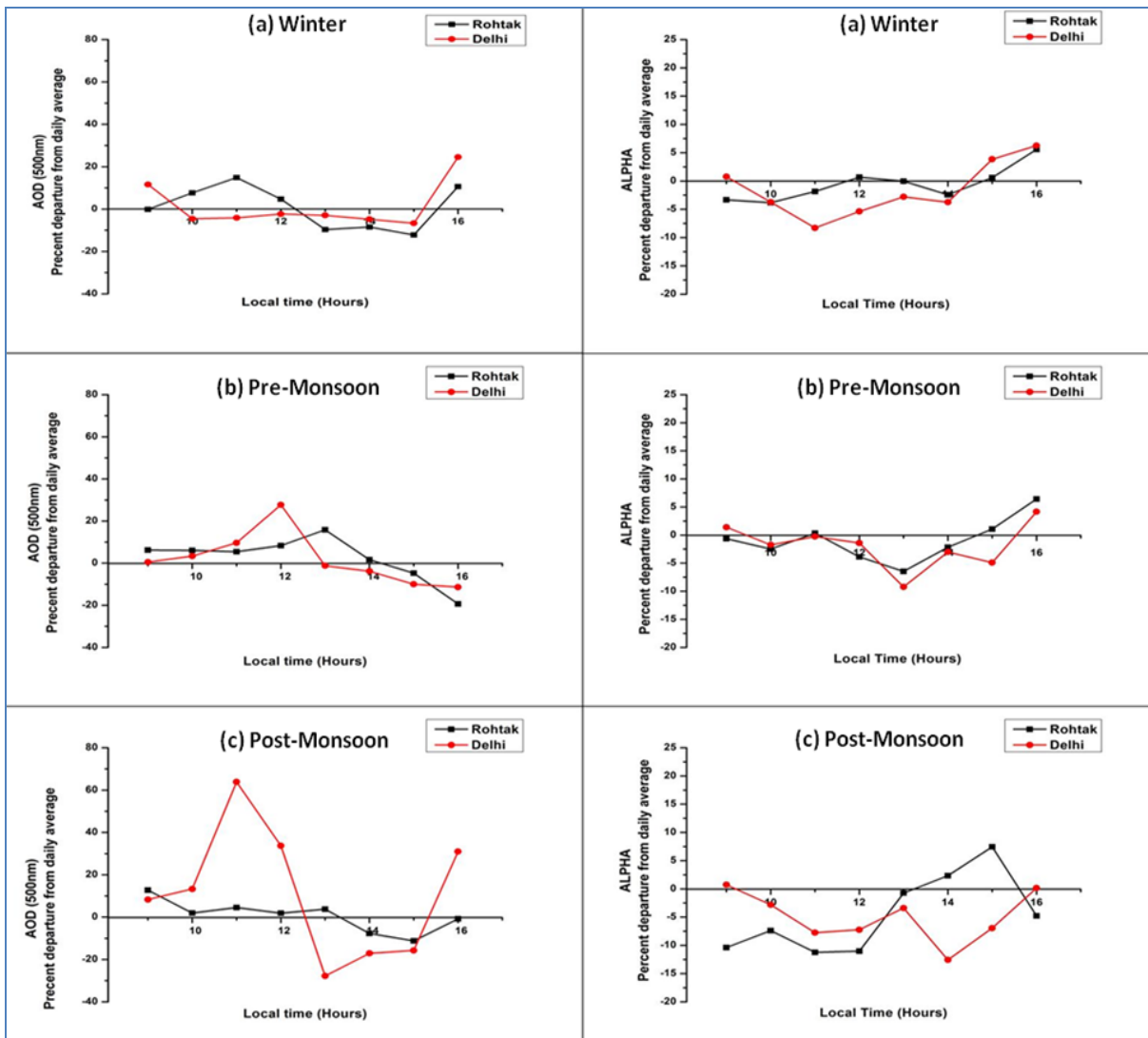
influence of black carbon, which are prominently found in the vehicular, biomass burning and industrial emissions in Delhi.

4.1.4. Asymmetry parameter (ASY)

The asymmetry parameter is a single-valued representation of the angular scattering and a key property controlling the aerosol contribution to radiative forcing (Pandithurai *et al.*, 2008). It depends on the size and

composition of the particles and is defined as the first moment of aerosol scattering phase function. The value of ASY ranges between -1 for entirely backscatterer to +1 for entirely forward scatterer. The monthly variation of ASY at 500 nm in Delhi and Rohtak is depicted in Fig. 5(d).

During winter season, a small negative gradient in the magnitude of ASY is observed at Delhi and Rohtak. However, ASY increases from April to May at



Figs. 8(a-c). Diurnal variability of AOD and Alpha as percent departure from daily mean over Rohtak and Delhi for (a) winter, (b) pre-monsoon and (c) post-monsoon seasons

Delhi (0.666-0.729) and Rohtak (0.667-0.747). Results suggest a gradient of coarse-to-fine mode particles over the study region. Post-monsoon months depict decreasing trend in ASY from 0.71 to 0.69 at both the locations. Results indicate that ASY is behaving opposite to the variations in Alpha, as a decrease in the magnitude of ASY is noticed with increasing Alpha values and *vice-versa*, at both the stations during the study period. It is observed that variations in asymmetry parameter do not reveal any significant difference at the two locations.

4.1.5. Aerosol size distribution

Particle size distribution is a key aerosol physical parameter for showing clearly the fine and coarse particle fractions in the atmosphere. The volume size distribution

is retrieved from the direct solar and diffuse sky radiance measurements as discussed by Nakajima *et al.* (1996). In the retrieval algorithm, it is assumed that the aerosols are composed of spherical and homogeneous particles. Scattering is simulated using Mie formulation, and multiple scattering effects are also taken into account. Whitby (1978) observed that most of the aerosol size distributions could be described with three volumetric modes: a nuclei mode with geometric mean radii of 0.0075-0.020 μm , an accumulation or fine mode with geometric mean radii of 0.075-0.25 μm and a coarse particle mode with geometric mean radii of 2.5-15 μm .

Seasonal average of aerosol volume particle size distribution ($dV(r)/d\ln r$ ($\text{cm}^3\text{cm}^{-2}$)) at Delhi and Rohtak is presented in Figs. 6(a-c). General patterns appear to be

bimodal but the volume mode depicts significant variations depending on the season and geographical location. In winter, Delhi and Rohtak have fine mode radius around 0.2 μm and coarse mode between 11-16 μm . However, the total volume of fine mode particles is found to be larger in Delhi than in Rohtak. A similar pattern of bimodal distribution is depicted at both stations in pre-monsoon (dust dominated case) with the mode radius of fine and coarse mode particles around 0.1 and 11 μm respectively at Delhi; 0.8 and 8 μm respectively at Rohtak. An overlapping pattern of volume size distribution is observed at the two stations during post monsoon with fine mode radius around 0.1 μm . The total volume of coarse mode particles is found to be higher in Delhi than in Rohtak, with coarse mode radius around 5 μm and 16 μm respectively. The seasonal averaged values of volume fraction and volume median radius for fine and coarse modes are depicted in Table 2.

Coarse mode volume fraction is found to be higher in Delhi by a factor of two during the pre-monsoon season when dust events are more prevalent over Delhi. The volume concentration in fine-size is higher in Delhi than that in Rohtak during pre-monsoon. This also correlates with the higher alpha value during dust period, indicating combined effect of dust and anthropogenic aerosols over Delhi. Post-monsoon season is marked with higher volume concentration of both fine and coarse mode particles over Rohtak. The volume concentration of coarse mode particle is almost double in Rohtak, as in comparison to Delhi during post-monsoon season.

4.2. Spectral variations

4.2.1. Aerosol optical depth

Fig. 7 (a) depicts seasonal variation of spectral AOD at seven different wavelengths (340, 380, 400, 500, 675, 870 and 1020 nm) for both the sites. The 340, 380, 400 and 500 nm represent the short wavelength region and are expected to be dominated by fine mode particles, while 675, 870 and 1020 nm represent the long wavelength region and are influenced by coarse mode particles. This is due to the fact that presence of high concentration of fine-mode particles selectively enhances the irradiance scattering at lower wavelength and therefore, the AOD values are high at the shorter wavelengths (Latha *et al.*, 2005). Likewise, the coarse-mode particles provide similar contributions to the AOD at relatively higher wavelengths (Schuster *et al.*, 2006). It is evident from the figure that AOD exhibits strong wavelength dependence during different seasons. Steeper AOD gradient is observed in winter months at both the locations. The winter AOD over Delhi depicts strong wavelength

dependence varying from 1.345 at 380 nm to 0.474 at 1020 nm, while it exhibits weak wavelength dependence over Rohtak ranging from 0.764 to 0.268 at 380 nm and 1020 nm respectively, showing insignificant decrease at longer wavelengths as compared to middle wavelengths.

The winter AOD is found to be higher than the post-monsoon AOD in Delhi and *vice-versa* in Rohtak. However, during pre-monsoon, the decreasing trend in AOD shows a gentler slope along the spectrum at both the locations. Also, increase in AOD at higher wavelengths (675-1020) during this period can be attributed to the presence of dust particles because coarse mode particles increase AOD only at longer wavelength. It is interesting to note that, at shorter wavelengths, the magnitude of AOD is higher over Delhi as compared to Rohtak. This indicates abundance of finer mode aerosols over Delhi as compared to Rohtak.

4.2.2. Single scattering albedo (SSA)

The spectral variation of seasonal average SSA (340-1020 nm) over study regions is shown in Fig. 7(b). Delhi shows an abrupt spectral variation in SSA, unlike the smooth decreasing pattern over Rohtak. In Delhi, the seasonal average SSA is observed to decrease from 340 to 500 nm, increase at 675 nm and again decrease at higher wavelengths. The lower magnitudes of SSA in Delhi can be attributed to the dominance of absorbing urban aerosols such as black carbon emitted from the industrial sources and diesel engines. While in Rohtak, a discernible decline is observed along the spectrum in winter and post-monsoon. However, during pre-monsoon months, a small increase in SSA is observed in Rohtak at $\lambda > 675$ nm, indicating abundant dust loading in the region. Due to the scattering state of the atmosphere with absorbing nature of dusts, SSA shows very small change. The different trend of spectral SSA over Delhi and Rohtak may be attributed to difference in aerosol sources and topography.

4.2.3. Asymmetry parameter (ASY)

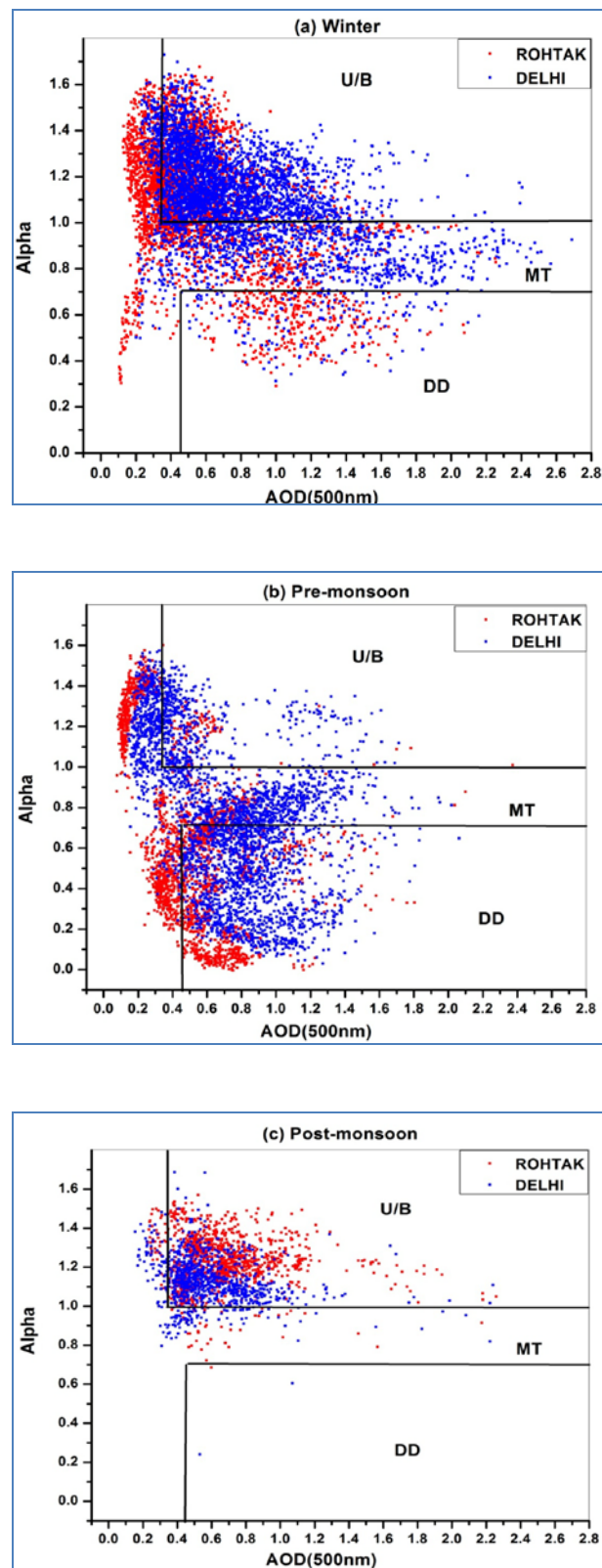
Fig. 7(c) shows the monthly mean spectral values of retrieved asymmetry parameter. The monthly averaged values of asymmetry parameter vary within 0.65-0.75 range for the whole period at both the sites. It is a notable observation that asymmetry parameter decreases with wavelength at 340-1020 nm range for all the seasons during the study period. However, the spectral dependence of asymmetry factor is almost constant at 670-1020 nm range during the pre-monsoon season at both the locations. Also the magnitude of ASY in pre-monsoon is found to be higher than in winter and post-monsoon. This

can be attributed to the coarse particles in the frequent dust weather. The asymmetry parameter for coarse particles is larger than those of fine particles at a given AOD. Similar decrease of ASY with wavelength in the visible spectral region and slight increase in the near infrared region during pre-monsoon months in New Delhi was reported by Pandithurai *et al.* (2008).

4.3. Diurnal variations in aerosol optical depth and Angstrom exponent

The aerosol optical properties over stations show a typical pattern due to complex synoptic processes and local aerosol emission sources. Knowledge of diurnal variability of aerosol optical properties is important for various applications such as validating satellite derived aerosol products, radiative forcing computation and aerosol interaction with clouds and other meteorological parameters (Kaufman *et al.*, 2002; Smirnov *et al.*, 2002). The diurnal variations in aerosol optical properties show seasonal dependence. Hourly statistics of AOD and Alpha is calculated as percentage departure from daily mean [Figs. 8(a-c)].

During winter, negative departure for Delhi and positive departure for Rohtak are depicted in the forenoon hours. This can be due to higher relative humidity and calmer winds (Fig. 3) in Rohtak as compared to Delhi during winter. Higher positive departure from daily mean is observed during evening hours at both the stations, which can be attributed to the low-level inversions leading to aerosol trapping in lower atmosphere. There is a corresponding negative departure for Alpha in forenoon at both the locations, followed by a positive departure in evening hours. In pre-monsoon, AOD departures are positive from 0900 hrs to 1300 hrs (IST), indicating that AOD is higher in the forenoon and reduces in the afternoon at both the locations. The difference is observed in the time of maximum AOD values which according to local time, is observed at 1200 hrs in Delhi and at 1300 hrs in Rohtak. Overall, diurnal departures fall within the range of $\pm 30\%$ for AOD and $\pm 10\%$ for Alpha, with larger AOD and smaller Alpha in the afternoon during pre-monsoon. During post-monsoon, AOD is higher in morning (till 1100 hrs) and evening (1600 hrs) compared to midday hours in Delhi. This can be attributed to the office traffic rush hours during morning and evening in Delhi (Mishra *et al.*, 2013). Whereas in Rohtak, the diurnal change in AOD is less than $\pm 10\%$ during the day. In Rohtak, the alpha departures are negative in morning and becomes positive (peak at 1500 hrs) in afternoon. On the contrary, the Alpha departure is negative for the whole day in Delhi. Enhancement in aerosol loading in Delhi particularly is subject to increased emissions from traffic during early morning and evening hours.



Figs. 9(a-c). The scatter plots of Alpha as a function of AOD at 500 nm over Delhi and Rohtak for winter, pre-monsoon and post-monsoon season

4.4 Aerosol classification: Scatter plot of Alpha-AOD

AOD represents columnar aerosol load in the atmosphere, while Alpha represents the aerosol size distribution. Therefore, Alpha-AOD scatter plots give qualitative indication to the aerosol load due to particles of different sizes. The particles of different sizes tend to cluster in different areas of the plot, which allows discerning of aerosols of different origin. Several earlier investigations have illustrated the discrimination of aerosol types by using this method of relating AOD and alpha (Eck *et al.*, 1999; Kaskaoutis *et al.*, 2007; Smirnov *et al.*, 2002; Toledano *et al.*, 2007). Following these studies, the aerosol types in the present work at the study locations have been classified into Urban/Biomass burning (U/B), Desert Dust (DD) and Mixed Type (MT). The U/B types of aerosols are of anthropogenic origin and those produced from biomass burning. The dust aerosols are the ones originated by the action of wind, particularly in western deserts, including the coarse-mode aerosols under high RH conditions. The remaining, undetermined cluster, which constitute combination of natural and anthropogenic factors (including RH, fuel types and emission characteristics) are termed as mixed type (MT). A mixed or indeterminate type of aerosols is formed due to aerosol-mixing processes in the atmosphere, such as coagulation, condensation, humidification, gas-to-particle conversion (Kaskaoutis *et al.*, 2009). According to Hess *et al.* (1998), for urban aerosol type, the magnitude of $AOD_{500\text{ nm}}$ is 0.643 for tropospheric aerosols. Also, the value for Alpha is 1.14 for spectral range 350-500 nm and 1.43 for 500-800 nm at a relative humidity (RH) of 80%. In the present study, threshold values for different aerosol types are taken as follows:

U/B: $AOD_{500\text{ nm}} > 0.35$ and $\text{Alpha} > 1$;

DD: $AOD_{500\text{ nm}} > 0.45$ and $\text{Alpha} < 0.7$ and remaining for MT.

Figs. 9(a-c) shows the scattergram of daily mean $AOD_{500\text{ nm}}$ against Alpha for Delhi and Rohtak during winter, pre-monsoon and post-monsoon. It depicts the sector wise cluster regions separated by solid lines, each corresponding to different aerosol types. During winter season, Alpha varies over a wide range at low $AOD_{500\text{ nm}}$ (< 0.3) for MT in Rohtak than in Delhi. On the contrary, Delhi shows more U/B fraction (approx. 65%) of aerosols during winter. It is evident from the graph that presence of DD aerosols during winter is insignificant. Although percentage of DD aerosols is found to be more in Rohtak (~9%) than in Delhi (~4%) during winter. However, significant number of points is accumulated

in the cluster region corresponding to aerosols of DD type in pre-monsoon season. The percentage contributions of dust aerosols over Delhi (~42%) and Rohtak (~41%) are almost the same, due to their close proximity to each other. The mixed type aerosols are equally contributing in this season over both the locations, with Rohtak having more MT (~53%) aerosols than Delhi (~44%). Post-monsoon season marks the wipe out of most of the coarse mode aerosols after rainfall. This is evident from the plot [Figs. 8(a-c)] that shows a higher density of U/B fine mode aerosols (80-90%) over both the study locations. It is seen that MT aerosols are more prominent over Delhi than Rohtak during this season. Overall, Delhi shows dominance of U/B aerosols and Rohtak of MT aerosols throughout the year except in post-monsoon.

5. Conclusions

Aerosol optical properties have been analyzed and compared over Rohtak with that of Delhi. Although both the regions are in close proximity to each other, there exists a significant variation of aerosol optical properties. The annual mean $AOD_{500\text{ nm}}$ over Delhi and Rohtak is 1.01 and 0.73 respectively, with correlation coefficient (R) calculated as 0.67. This gives a moderately strong relationship between AODs at the two locations, indicating that the aerosol pollution in Delhi is regional in nature. However, there are significant differences observed in this study on the seasonal aspect in the aerosol optical properties at the two locations. The salient conclusions of the present study are highlighted as follows:

(i) Delhi shows higher AOD in all the seasons except in post-monsoon. SSA over Delhi is less in comparison to Rohtak, suggesting more absorbing aerosols over Delhi. The total volume fraction of fine mode particles is higher in Delhi than in Rohtak, while that of coarse mode particles is almost the same. Spectral variation of AOD suggests that at shorter wavelengths, the magnitude of AOD is higher over Delhi as compared to Rohtak, indicating abundance of finer mode aerosols over Delhi. The different trend of spectral SSA over Delhi and Rohtak may be attributed to difference in aerosol sources and topography. The monthly averaged values of asymmetry parameter vary within 0.65-0.75 range for the whole period at both the sites.

(ii) Aerosol optical properties exhibit seasonal asymmetry in diurnal variation at both the study sites. The percentage departure from daily means show significant influence of meteorological conditions and anthropogenic activities during forenoon and afternoon hours in Delhi and Rohtak.

(iii) Dominance of U/B type of aerosols in winter season (approx. 65%) is evident in Delhi. During pre-monsoon season, DD type is equally prominent over both the locations. Post-monsoon season is characterized by dominance of MT aerosols over Delhi than Rohtak. Overall, Delhi shows dominance of U/B aerosols and Rohtak of MT aerosols throughout the year except in post-monsoon.

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