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Variability and Trends of Aerosol Properties over Kanpur, Northern India Using AERONET Data (2001–10)

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Variability and trends of aerosol properties over Kanpur, northern India using AERONET data (2001–10)

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Abstract

Natural and anthropogenic aerosols over northern India play an important role in influencing the regional radiation budget, causing climate implications to the overall hydrological cycle of South Asia. In the context of regional climate change and air quality, we discuss aerosol loading variability and trends at Kanpur AERONET station located in the central part of the Indo-Gangetic plains (IGP), during the last decade (2001–10). Ground-based radiometric measurements show an overall increase in column-integrated aerosol optical depth (AOD) on a yearly basis. This upward trend is mainly due to a sustained increase in the seasonal/monthly averaged AOD during the winter (Dec–Feb) and post-monsoon (Oct–Nov) seasons (dominated by anthropogenic emissions). In contrast, a neutral to weak declining trend is observed during late pre-monsoon (Mar–May) and monsoon (Jun–Sep) months, mainly influenced by inter-annual variations of dust outbreaks. A general decrease in coarse-mode aerosols associated with variable dust activity is observed, whereas the statistically significant increasing post-monsoon/winter AOD is reflected in a shift of the columnar size distribution towards relatively larger particles in the accumulation mode. Overall, the present study provides an insight into the pronounced seasonal behavior in aerosol loading trends and, in general, is in agreement with that associating the findings with those recently reported by satellite observations (MODIS and MISR) over northern India. Our results further suggest that anthropogenic emissions (due mainly to fossil-fuel and biomass combustion) over the IGP have continued to increase in the last decade.

Keywords: aerosol optical depth, trend, AERONET, Kanpur, Indo-Gangetic plains

S Online supplementary data available from stacks.iop.org/ERL/7/024003/mmedia

1. Introduction

Aerosols over India exhibit strong seasonal and inter-annual variability mainly driven by the regional monsoon system and seasonally changed airmass patterns. The Indo-Gangetic plains (IGP), in the northern part of India, are among the most densely populated as well as the most heavily aerosol-laden regions of the world. With the increase in

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population density and energy demands, aerosol emissions have been gradually increasing, mainly through fossil-fuel and bio-fuel combustions (Lawrence and Lelieveld 2010). The large increase in anthropogenic aerosols over IGP is hypothesized to cause considerable changes to regional monsoonal climate (Ramanathan *et al* 2005, Lau *et al* 2006, Dey and Tripathi 2008, Gautam *et al* 2010). The two main contrasting seasons over northern India (winter and pre-monsoon/summer), in terms of boundary-layer dynamics and wind patterns, dictate variations in aerosol type and their spatial, temporal and vertical distribution. During the relatively stable winter season, the area is often covered by a low lying thick fog/hazy layer (Gautam *et al* 2007). The aerosols are mainly of anthropogenic origin due to large carbonaceous and sulfate emissions from fossil-fuel and bio-fuel combustions (Prasad *et al* 2006). In contrast, the IGP experiences an enhanced convective and turbulent boundary layer and witnesses a large influx of westerly wind driven dust-laden air masses during the pre-monsoon/summer season (April–June) (Gautam *et al* 2011). This period also marks the development and evolution of the monsoon type circulation over the Indian subcontinent. The onset of the monsoon season typically sees a significant reduction in the dust-laden aerosols associated with heavy and continuous rainfall (Singh *et al* 2004) and by the end of the monsoon season, northwestern IGP is influenced by aerosols due to extensive crop-residue burning (Sharma *et al* 2010).

Numerous studies using satellite observations and ground-based measurements have revealed an overall increase in aerosol optical depth (AOD) over India especially in IGP (Satheesh *et al* 2002, Massie *et al* 2004, Sarkar *et al* 2006, Porch *et al* 2007, Prasad and Singh 2007a, Lawrence and Lelieveld 2010, Dey and Di Girolamo 2011, Kaskaoutis *et al* 2011, Kharol *et al* 2011, Ramachandran *et al* 2012). Systematic aerosol observations via the well-calibrated AERONET instruments have played a vital role in the determination of the increase of anthropogenic aerosols, especially in developing countries (Yoon *et al* 2011, Xia 2011, Wang *et al* 2011).

Kanpur AERONET station, in the central part of the IGP, has been operational since January 2001 (Singh *et al* 2004) and, for the first time, we present monthly long-term (2001–10) trend analyses of spectral AOD measurements. We have analyzed the 10 year period and also two 5 year sub-periods, (i) 2001–5 and (ii) recent, 2006–10, independently in view of the statistically changing aerosol properties. The Ångström exponents at different spectral bands are also analyzed in order to understand the trends associated with anthropogenic or natural processes and are discussed with respect to the modification of the columnar aerosol size distribution. In addition to the analysis of the aerosol loading trend over Kanpur for a 10 year period, the present work also supplements the satellite (MODIS, MISR) derived trends of AOD over IGP during the last decade.

2. Data and methodology

The intense haze and smog conditions over the IGP during the winter season, firstly observed by the ADEOS Polder satellite (Goloub *et al* 2001) as a blanket of heavy aerosol layer over the area, motivated the Indian Institute of Technology (IIT) Kanpur and NASA/GSFC to establish the first long-term AERONET station in India at Kanpur (26.5°N, 80.2°E) in 2001 (Singh *et al* 2004). During the last decade, several studies have dealt with aerosols over Kanpur focusing on aerosol type classification (Gobbi *et al* 2007, Eck *et al* 2010, Giles *et al* 2011), model simulations (Chin *et al* 2009) and satellite validation (Tripathi *et al* 2005, Jethva *et al* 2007, Prasad and Singh 2007a). Direct-beam and sun/sky almucantar radiance measurements using a CIMEL sunphotometer provide column-integrated spectral AODs at eight wavelengths, from 340 to 1640 nm, and water vapor content at 940 nm (Holben *et al* 1998). Furthermore, via the almucantar measurements and the spectral deconvolution algorithm (SDA) retrievals, aerosol columnar size distribution (CSD), single scattering albedo (SSA), asymmetry parameter, refractive index, fine and coarse-mode AODs are also available for large solar zenith angles ($>50°$) and high aerosol loading conditions ($\mathrm{AOD}_{440} > 0.4$) (Dubovik *et al* 2000). The Level 2 (cloud screened and quality assured) AERONET data were used in the present work, following the uncertainties in the retrievals described elsewhere (Giles *et al* 2011). The aerosol properties were daily averaged and analyzed on a monthly and seasonal basis during the period January 2001 to December 2010.

Since aerosols over India are composed of both natural and anthropogenic components during different seasons, we also analyze the respective trends of fine and coarse particles by examining the variability in Ångström exponents (α) defined at shorter (380–500 nm) and longer (675–870 nm) wavelengths. According to Reid *et al* (1999), Schuster *et al* (2006), the former is indicative of the aerosol fine-mode size, while the latter provides useful information about the coarse-to-fine mode ratio. Thus, supplementary to AOD_{500} and $\alpha_{440-870}$, direct-sun retrievals of $\alpha_{380-500}$ and $\alpha_{675-870}$ are used for the trend analysis over Kanpur during 2001–10. AERONET almucantar retrievals are not used for analyzing trends in this study due to limited observations available, especially during the monsoon season, and to avoid potential bias in linear regression type analysis.

For all the available data series (AOD_{500}, $\alpha_{440-870}$, $\alpha_{380-500}$, $\alpha_{675-870}$), linear regression analysis was applied during the period 2001–10 using the monthly mean values. In addition to the monthly means, median-based analysis is a potential way to avoid outliers/skewed data, especially for those months with limited sample size. Therefore, linear regressions were also calculated using the monthly median values. If there is not much sampling/outlier bias then trends from both the median and mean methodologies would be close and not deviate significantly. With the usage of the median-based analysis, sampling and outlier biases were minimized, which help us to make the analysis more robust. Moreover, the trends in aerosol properties are analyzed on a monthly basis during the period 2001–10 using the daily mean AERONET retrievals. The percentage (%) variation in aerosol properties is calculated via the formula: $x(\%) = aN/\bar{x}100$, where x is the variable, a is the slope value from the linear

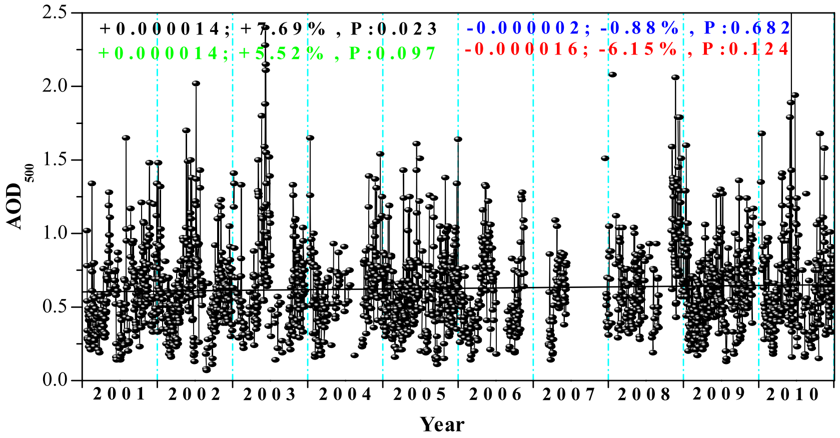

Figure 1. Inter-annual variability and trend of the AOD_{500} daily values over Kanpur for 2001–10. The slope of the regression analysis along with the % difference and the P value are given (black for AOD_{500}, green for α (440–870), blue for α (380–500) and red for α (675–870).)

regression analysis and N the whole number of days, months or years during the studied period, i.e. with and without data. For each case the statistical significance of the slope was checked by applying the P value, i.e. $P < 0.05$ for statistically significant variations at the 95% confidence level.

3. Results and discussion

3.1. Trend analysis from 2001 to 2010

Figure 1 shows the AOD_{500} trends using the daily mean values for the period 2001–10. Due to the presence of clouds and the calibration protocol, aerosol observations are not equally distributed within the months and years, and a large gap in data from December 2006 to February 2007 and from June 2007 to November 2007 exists. Furthermore, during the monsoon rainy season (June–September) there are limited observations that present large year-to-year fluctuations due to monsoon onset, intensity and duration (Gautam *et al* 2009). In order to perform a robust statistical analysis, all available measurements during the examined period are considered (including gaps in observations). In figure 1, the slope values from the regression analysis are provided corresponding to variation per day along with % difference and statistical significance tests indicated by p values, as described in section 2. The data series in figure 1 exhibit large day-to-day and seasonal variability in AOD_{500}, which is strongly influenced by the local and regional meteorological and atmospheric conditions, i.e. rainfall, air mass trajectories, aerosol emission rates, etc (Singh *et al* 2004). Especially over India, the role of rainfall in aerosol properties and variations is crucial during the monsoon season (Gautam *et al* 2009, Manoj *et al* 2011). On the other hand, the increase in aerosol loading over northern India may also affect precipitation and the hydrological cycle (Lau *et al* 2006). However, it is difficult to quantify the influence of rainfall in AOD trends over Kanpur, since anthropogenic emissions and dust transport play a significant role in influencing aerosol loading and properties, especially during the extended dry period, i.e. from October to June over northern India. For example, the deficit of rainfall during monsoon of 2002 and late pre-monsoon of 2003 caused an increase in dust activity and atmospheric aerosol lifetime over northern India (Kaskaoutis *et al* 2011) and high AOD_{500} values (figure 1).

The results show overall increasing trends in AOD_{500} (7.69%) and $\alpha_{440-870}$ (5.52%), which are found to be higher (13.8% and 18.1%, respectively) during the second half-period (2006–10), that are consistent with satellite observations (MODIS, MISR) of increasing AOD and enhanced anthropogenic emissions over IGP (Dey and Di Girolamo 2011, Kaskaoutis *et al* 2011). The increase of 7.69% in AOD_{500} during the period 2001–10 is considered as statistically significant at the 95% confidence level ($p < 0.05$), in contrast to the change in Ångström exponents. However, the observed trends and % variations are sensitive on the basis of daily, monthly or yearly data. Averaging the aerosol properties over a monthly period has the disadvantage of limited observations during some specific months that may influence the monthly mean and, in turn, introduce biases in trends. On the other hand, the monthly median values are a good way to avoid outliers/skewed data, especially for months with insufficient datasets. Thus, the trend analysis is also attempted on a monthly and yearly basis using the mean and median values. The results are summarized in table 1, excluding the year 2007, due to lack of data for the majority of the months (figure 1). On a monthly and yearly basis, the AOD_{500} increase is found to be slightly higher (~9–10%) compared to daily values, while similarly to the daily trends, the monthly and yearly ones are positive for $\alpha_{400-870}$ and negative for α at shorter and longer wavelengths (except for $\alpha_{380-500}$ for monthly and yearly median values). Overall, the results (figure 1, table 1) show a concurrent increase in both AOD_{500} and $\alpha_{440-870}$, suggesting an increase in anthropogenic aerosols over Kanpur, although the trends in all Ångström exponents are not statistically significant at the 95% confidence level. The consistency of the trends obtained using the mean or median values (table 1), with differences in the variations in the order of 1–5%, suggests that the datasets used for analyzing the aerosol trends provide reliable results despite the lack of data during specific months. The similar trends from both the median- and mean-based approaches suggest that the sampling/outlier bias in the monthly values

Table 1. Trend values and (%) variation of aerosol properties over Kanpur during the period 2001–10 using the yearly mean/median and the monthly mean/median values.

	Yearly mean trends		Yearly median trends		Monthly mean trends		Monthly median trends	
	Trend/year	(%)	Trend/year	(%)	Trend/month	(%)	Trend/month	(%)
AOD	0.0062	10.12	0.005 5	9.24	0.000 53	10.29	0.000 46	9.35
$\alpha_{440-870}$	0.0036	3.89	0.005 7	5.98	0.000 52	6.93	0.000 62	7.97
$\alpha_{380-500}$	−0.0045	−5.32	0.000 09	0.11	−0.000 06	−0.91	0.000 06	0.86
$\alpha_{675-870}$	−0.0091	−9.43	−0.006 5	−6.81	−0.000 42	−5.07	−0.000 26	−3.35

Table 2. Statistical parameters for the regression analysis in the monthly time series of the examined aerosol properties. The statistically significant trends at the 95% confidence level are presented in bold italic. (*N*: number of available days for each month)

	Trend/day (%)	*P*	*N*	Trend/day (%)	*P*	*N*	Trend/day (%)	*P*	*N*
	January			February			March		
AOD	0.000 04 (1.81)	0.876	155	0.000 19 (11.28)	0.536	196	***0.000 34 (23.79)***	***0.002***	206
$\alpha_{440-870}$	−0.000 11 (−2.64)	0.595		−0.000 22 (−5.45)	0.356		−0.000 30 (−11.03)	0.189	
$\alpha_{380-500}$	−0.000 18 (−5.85)	0.319		−0.000 29 (−8.25)	0.052		***−0.000 42 (−14.83)***	***0.016***	
$\alpha_{675-870}$	−0.000 36 (−7.79)	0.127		−0.000 21 (−5.25)	0.507		−0.000 19 (−7.89)	0.449	
	April			May			June		
AOD	***0.000 48 (26.33)***	***0.005***	211	***−0.000 65(−26.9)***	***<0.001***	224	−0.000 21 (−8.08)	0.518	187
$\alpha_{440-870}$	−0.000 21 (−12.54)	0.279		***0.001 1 (72.67)***	***<0.0001***		0.000 31 (20.54)	0.264	
$\alpha_{380-500}$	−0.000 16 (−7.79)	0.457		***0.001 2 (65.35)***	***<0.0001***		0.000 33 (18.01)	0.206	
$\alpha_{675-870}$	***−0.000 36 (−25.14)***	***0.035***		***0.000 66 (50.68)***	***0.001***		−0.000 46 (−32.03)	0.138	
	July			August			September		
AOD	−0.000 55 (−27.59)	0.099	99	0.000 055 (2.83)	0.852	101	−0.000 006 (−0.36)	0.978	129
$\alpha_{440-870}$	***0.001 7 (67.86)***	***<0.0001***		0.000 03 (1.03)	0.943		***0.000 78 (22.61)***	***0.003***	
$\alpha_{380-500}$	***0.001 2 (50.25)***	***<0.001***		−0.000 29 (−10.9)	0.284		0.000 016 (0.49)	0.928	
$\alpha_{675-870}$	***0.001 07 (44.51)***	***0.015***		−0.001 05 (−35.8)	0.071		0.000 11 (3.07)	0.791	
	October			November			December		
AOD	0.000 026 (1.29)	0.889	208	***0.000 63 (24.60)***	***0.005***	211	***0.000 49 (21.06)***	***0.039***	168
$\alpha_{440-870}$	0.000 14 (3.62)	0.435		***0.000 26 (5.98)***	***0.001***		0.000 18 (4.26)	0.146	
$\alpha_{380-500}$	0.000 089 (2.70)	0.429		***−0.000 45 (−14.1)***	***<0.0001***		***−0.000 32 (110.35)***	***0.023***	
$\alpha_{675-870}$	***−0.000 65 (−15.61)***	***0.014***		0.000 22 (4.51)	0.061		0.000 15 (3.13)	0.324	

is not significant and, therefore, the trend analysis is more robust.

The trends in the AOD and α values for each month during the period 2001–10 are further examined using the daily AERONET observations and the results are summarized in table 2. The period from May to October exhibits mostly neutral to negative trends in AOD_{500}; there is a statistically significant decrease (−26.9%) in AOD_{500} during May associated with a pronounced positive trends in all α values. Furthermore, significant increases in α values ($p < 0.05$) are observed in July associated with a considerable (−27.6%) decrease in AOD_{500}, although they are not statistically significant due to large temporal variation and limited number of observations due to cloudiness. The main cause for these downward trends during May–July can be attributed to the extreme values of high AOD_{500} and low α associated with frequent and intense dust activity during 2002 (monsoon) and 2003 (late pre-monsoon) (see the peaks in figure 1), in turn influencing the aerosol trends during the whole decade (Kaskaoutis *et al* 2012). On the other hand, a statistically significant increase in AOD_{500} is observed in March (23.8%), April (26.3%), November (24.6%) and December (21.1%) associated with a decrease in $\alpha_{440-870}$ during the pre-monsoon months and an increase for late post-monsoon and winter, respectively. The negative trends in α suggest an increase in natural coarse-mode aerosols, whereas an increasing trend is associated with the increase in fine anthropogenic emissions (a statistically significant trend in $\alpha_{440-870}$ is observed only for November). It should also be noted that a statistically significant decrease in $\alpha_{380-500}$ is found for March, November and December, thus suggesting a shift of the CSD to a larger fine-mode radius indicative of gas-to-particle conversion and coagulation processes. In contrast, and opposite to the satellite observations of large increase in anthropogenic AODs over IGP during winter (Dey and Di Girolamo 2011), January and February exhibit rather neutral AOD_{500} trends over Kanpur and slight negative trends in α values.

The AOD trends from MODIS and MISR data also exhibited strong monthly variability over northern India (Dey and Di Girolamo 2011, Kaskaoutis *et al* 2011). Regarding the consistency of the monthly trends between AERONET, MODIS and MISR only a qualitative comparison can be provided since the non-overlapping periods in the

Table 3. Statistical parameters from the comparison of the aerosol optical properties over Kanpur AERONET between the sub-periods 2001–5 (group 1) and 2006–10 (group 2). The mean values of AOD_{500}, $\alpha_{440-870}$, $\alpha_{380-500}$ and $\alpha_{675-870}$ are given for each month and group. N is the number of daily observations for each group, while t and P values correspond to the statistical variables of the two-pair t-test. The statistically significant differences (95% confidence level) between the two groups are highlighted in bold italic, while the differences that are in the limits of the statistical significance at the same level are presented in italic.

	Jan 1	Jan 2	t	P	Feb 1	Feb 2	t	P	Mar 1	Mar 2	t	P
N	82	73			113	83			107	99		
AOD	0.697	0.673	−0.46	0.64	0.463	0.491	0.89	0.38	0.424	0.463	1.77	0.08
$\alpha_{440-870}$	1.306	1.275	−0.81	0.42	1.149	1.121	−0.67	0.50	0.854	0.832	−0.49	0.62
$\alpha_{380-500}$	0.963	0.942	−0.66	0.51	***1.017***	***0.956***	***−2.22***	***0.03***	0.901	0.852	−1.41	0.16
$\alpha_{675-870}$	***1.479***	***1.381***	***−2.30***	***0.02***	1.149	1.101	−0.85	0.39	0.759	0.732	−0.56	0.57
	Apr 1	Apr 2	t	P	May 1	May 2	T	P	Jun 1	Jun 2	t	P
N	107	104			99	125			97	90		
AOD	*0.518*	*0.577*	*1.92*	*0.056*	0.777	0.723	−1.57	0.12	0.828	0.727	−1.63	0.10
$\alpha_{440-870}$	0.487	0.518	0.91	0.36	***0.351***	***0.563***	***5.56***	***<0.001***	0.428	0.479	0.98	0.33
$\alpha_{380-500}$	0.593	0.640	1.27	0.21	***0.603***	***0.711***	***3.07***	***0.002***	0.519	0.582	1.27	0.21
$\alpha_{675-870}$	0.432	0.428	−0.13	0.89	***0.326***	***0.465***	***3.85***	***<0.001***	0.470	0.388	−1.40	0.16
	Jul 1	Jul 2	t	P	Aug 1	Aug 2	T	P	Sep 1	Sep 2	t	P
N	61	38			54	47			74	55		
AOD	0.658	0.553	−1.46	0.15	0.536	0.561	0.47	0.64	0.489	0.509	0.46	0.64
$\alpha_{440-870}$	***0.641***	***0.995***	***3.92***	***<0.001***	0.861	0.944	1.00	0.32	0.993	1.091	1.78	0.07
$\alpha_{380-500}$	***0.651***	***0.884***	***3.60***	***<0.001***	0.834	0.815	−0.34	0.73	0.964	0.968	0.11	0.91
$\alpha_{675-870}$	***0.659***	***0.883***	***2.39***	***0.02***	0.966	0.845	−1.02	0.31	1.105	1.035	−0.84	0.40
	Oct 1	Oct 2	t	P	Nov 1	Nov 2	T	P	Dec 1	Dec 2	t	P
N	133	50			129	82			112	56		
AOD	0.646	0.588	−1.61	0.11	***0.691***	***0.890***	***5.09***	***<0.001***	*0.689*	*0.784*	*1.96*	*0.051*
$\alpha_{440-870}$	1.205	1.189	−0.47	0.64	***1.287***	***1.332***	***3.03***	***0.002***	1.298	1.332	1.35	0.18
$\alpha_{380-500}$	1.007	1.047	1.78	0.08	***1.009***	***0.884***	***−7.09***	***<0.001***	***0.978***	***0.917***	***−2.11***	***0.04***
$\alpha_{675-870}$	***1.359***	***1.169***	***−3.76***	***<0.001***	***1.434***	***1.514***	***3.75***	***<0.001***	1.474	1.507	1.14	0.26

two data series, the coarse (1° × 1° for MODIS and 0.5° × 0.5° for MISR) Level 3 spatial resolution over Kanpur and the unequal distribution of the observations during a month cause difficulties in deriving a quantitative comparison. Despite these difficulties, the AERONET data (using monthly mean/median values) show positive trends in late post-monsoon and winter months and neutral or even negative AOD_{500} trends during the period May–October that are found to be consistent with MODIS and MISR retrievals. A higher spatial resolution of Level 2 (10 km × 10 km) satellite data over Kanpur may be more valid for examining the trends over the site, but such a quantitative comparison is beyond the scope of the present work.

3.2. Comparison of sub-periods (2001–5 and 2006–10)

Since there are gaps in daily data, especially during monsoon and winter seasons, statistical significance tests (t-test) of the changes in the monthly mean values of aerosol properties between the two sub-periods (2001–5 and 2006–10) are applied and the results are summarized in table 3. In this approach, the variations and trends in aerosol properties are grouped into mean values and are therefore smoothed. Thus, statistically significant increase of AOD_{500} during 2006–10 is observed only for November, while the increase in AOD_{500} in December and April is at the limits of the statistical significance at the 95% confidence level (p values close to 0.05). Consistent with the results of table 2, there is a significant increase from the first period to the second in all α values in May and July and a decrease or small change of AOD_{500} during May–October. The results of tables 2 and 3 are, in general, similar and statistically significant changes can be emphasized for AOD_{500} and α values during November, which indicate an increase in anthropogenic component, whereas those observed in May and July suggest a decrease in natural aerosols from 2001–5 to 2006–10.

The same approach of grouping aerosol observations in the two sub-periods is applied for the four dominant seasons and the results are summarized in figure 2 (view of box charts, details are given in the figure caption). The boxes show the range of the 25% and 75% values of the observations, the mean value (square) and the median value (line), as well as the 1% and 99% and minimum and maximum values. Using the t-test for independent groups at the 95% confidence level, the statistically significant changes in the mean values for AOD_{500} and alpha are examined for two contrasting periods: 2001–5 and 2006–10.

In winter, the second period exhibits slightly higher AOD_{500} (0.631 against 0.608) and slightly lower $\alpha_{440-870}$ and $\alpha_{675-870}$ values without statistically significant differences. In contrast, a statistically significant difference is revealed for $\alpha_{380-500}$ with lower values during 2006–10, suggesting an increase in particle size of fine aerosols (Reid *et al* 1999, Schuster *et al* 2006). A pronounced shift towards larger values for the fine-mode radius and an increase in

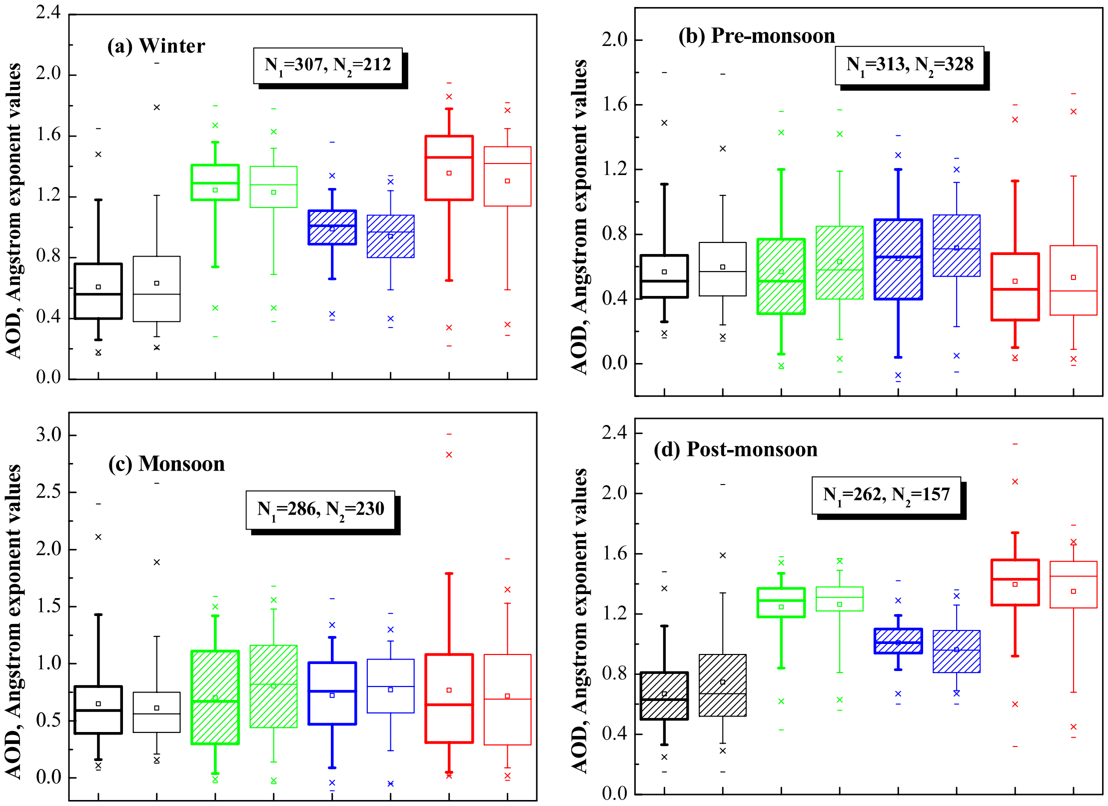

Figure 2. Box charts for AOD_{500} (black), α (440–870) (green), α (380–500) (blue) and α (675–870) (red) for Kanpur AERONET data (2001–5) (bold boxes) and 2006–10. The statistically significant differences between the means of the two periods are defined with the filled patterns. The number of observations for each group (N_1 for 2001–5 and N_2 for 2006–10) is also provided.

the accumulation-mode fraction is observed (figure 3(a)), which is consistent with the decrease in $\alpha_{380-500}$. A decrease in the $dV/d\ln R$ is also noted for the coarse mode around 4 μm during 2006–10, which can partly offset the increase in the coarse mode for the radius range 0.8–2.0 μm. The combination of these two results in the small difference in $\alpha_{675-870}$ values between the two periods. These modifications in the CSD curves have negligible effect in $\alpha_{440-870}$ as also shown by Eck *et al* (2005).

In the pre-monsoon season (figure 2(b)) the contrasting results of March, April and May (tables 2 and 3) lead to a small and not statistically significant increase in AOD_{500}. During the pre-monsoon as well as the monsoon season, much larger variation in all α values is found compared to the homogeneous anthropogenic-aerosol-laden atmosphere during post-monsoon and winter. Regarding changes in $\alpha_{440-870}$ and $\alpha_{380-500}$, statistically significant increases are observed in the second period strongly influenced by the considerable differences observed in May (tables 2 and 3). In contrast, the opposite trends in $\alpha_{675-870}$ between the pre-monsoon months (tables 2 and 3) result in smoothing the differences in this parameter. During pre-monsoon a pronounced decrease in the fine-mode aerosols (figure 3(b)) is observed during 2001–5, whereas an increase of the $dV/d\ln R$ is observed for the coarse mode during 2006–10. The concurrent increase in $dV/d\ln R$ for both fine and coarse modes during 2006–10 indicates a slight increase in AOD_{500} (figure 2(b)). According to Reid *et al* (1999) a similar increase in fine and coarse modes causes much larger variation in α at shorter and mid-wavelengths than at the longer wavelengths and so the statistically significant increase in $\alpha_{380-500}$ and $\alpha_{440-870}$ (figure 2(b)). The contrasting AOD and alpha trends in the pre-monsoon months (tables 2 and 3) are clearly detected by the CSDs in these months during the two sub-periods (see supplementary figure 1 available at stacks.iop.org/ERL/7/024003/mmedia). More specifically, the CSDs in March and April reveal a larger coarse-mode fraction and dominance of coarser aerosols during 2006–10 leading to lower α values. In contrast, the coarse-mode fraction dominates the CSD in May during the first sub-period (2001–5), thus affecting the α values in all wavelength ranges (statistically significant increase in α during 2001–10) (see table 2). This is attributed to the enhanced presence of dust aerosols and frequent dust outbreaks over Kanpur during May 2003 (Prasad and Singh 2007b, Kaskaoutis *et al* 2012).

During the monsoon period (figure 2(c)), the only statistically significant difference is that for $\alpha_{440-870}$ it increased during 2006–10. The seasonal differences during monsoon are mainly influenced by the July values (tables 2 and 3), while the decrease in AOD_{500} in June and July is partly recovered by the increase in August and September (table 3). The CSD curves in the two sub-periods are similar except for the larger coarse-mode fraction in the first period that leads to the lower $\alpha_{440-870}$ (figure 3(c)). Finally, the variations in aerosol properties during post-monsoon (figure 2(d)) are similar to those in winter with statistically significant increase in AOD_{500} influenced by the large increasing November trend. The strong decrease in $\alpha_{380-500}$ in November modulates the

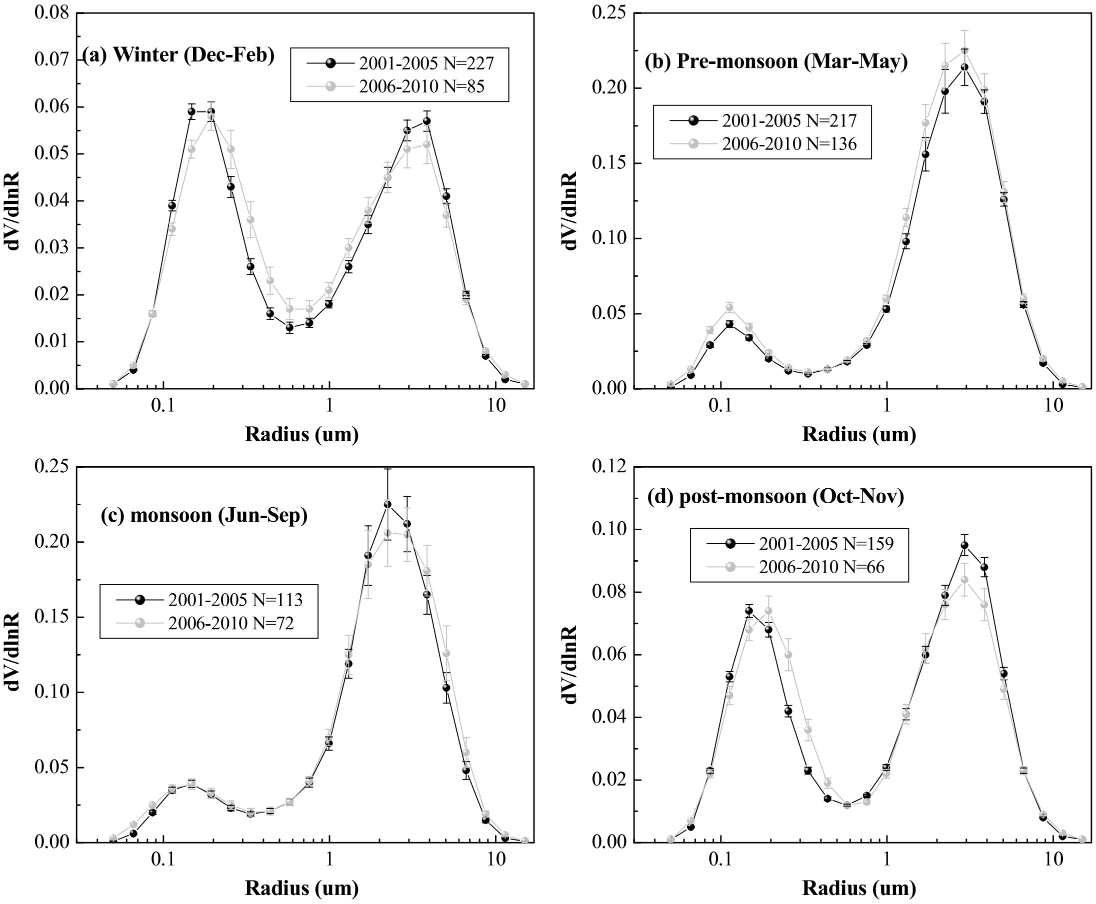

Figure 3. Mean seasonal CSDs at Kanpur AERONET station during the periods 2001–5 and 2006–10. The number of CSD retrievals for each period is given on each graph. The vertical bars correspond to the standard error from the seasonal mean.

seasonal variation, while the contradictory results in $\alpha_{675-870}$ for October and November smooth the differences between the two periods. The changes in CSD during post-monsoon (figure 3(d)) are similar to those observed in winter, thus introducing similar results regarding the changes in the aerosol parameters. The increase in fine-mode radius and the associated shift towards accumulation mode are both highlighted, leading to a decrease in $\alpha_{380-500}$. The analysis showed that the differences in the size distribution curves in all seasons are not statistically significant at the 95% confidence level. It is to be noted that the apparent difference between the changes in the three Ångström exponent values is attributed to the curvature in the spectral AOD in the log–log plot (Eck *et al* 1999) suggesting different values of α and, hence, different trends for each spectral band.

4. Conclusions

The present work examined the variation and trends in aerosol optical properties at Kanpur AERONET site in northern India during the last decade (2001–10). Linear regression analysis along with statistical significance tests were applied for examining robustness of the trends in the daily and monthly averaged aerosol data record. Significant variations in the AOD and Ångström exponent trends were observed depending on the month and season. In general, the AOD_{500} increased significantly during the November–December period as well as in the months of March and April. In contrast, a neutral or slight decreasing trend during May–October was found, which is attributed to large inter-annual variations of AOD, particularly associated with dust loading during May–July for the years 2002 and 2003; this is in agreement with satellite (MODIS, MISR) observations and model (GOCART) simulations over the region (Dey and Di Girolamo 2011, Kaskaoutis *et al* 2011). Large changes in $\alpha_{440-870}$ were not observed, except from an overall slight increase, indicating a larger contribution of anthropogenic aerosols. Recently, Lu *et al* (2011) also showed significant increases in sulfate and carbonaceous aerosol emissions from India associated with the remarkable energy consumption growth. The respective trends in α defined at shorter and longer wavelengths were associated with changes in columnar aerosol size distribution, mostly exhibiting a shift towards a larger fine radius and accumulation-mode fraction during post-monsoon and winter, thus corroborating the increase in fine-mode aerosol concentrations during the extended dry period. It is to be noted that despite the fact that the presented CSDs can satisfactorily explain the aerosol differences between the two sub-periods, there are serious difficulties for a direct comparison between figures 2 and 3, mainly due to larger uncertainties (Dubovik *et al* 2000) and the much smaller number of almucantar retrievals for each period, which may not be equally distributed on all days of each season. Overall, this study confirms the seasonal dependency in the aerosol loading trends, recently reported by satellite observations (MODIS and MISR) over northern India, and further provides an in depth assessment based on columnar particle size variations from monthly decomposition of the observed trends.

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