Seasonal variability in aerosol optical and physical characteristics estimated using the application of the Ångström formula over Mohal in the northwestern Himalaya, India

RAJ PAUL GULERIA¹, JAGDISH CHANDRA KUNIYAL^{1,*},
NAND LAL SHARMA¹ and PITAMBER PRASAD DHYANI²

¹*G.B. Pant Institute of Himalayan Environment and Development, Himachal Unit, Mohal-Kullu 175 126, India.*

²*G.B. Pant Institute of Himalayan Environment and Development, Kosi-Katarmal, Almora 263 643, India.*

**Corresponding author. e-mail: jckuniyal@gmail.com jckuniyal@rediffmail.com*

Investigations of aerosol optical and physical characteristics using the application of Ångström formula and second order polynomial fit were carried out from April 2006 to March 2009 at Mohal in the Kullu valley. The measurements of spectral aerosol optical depths (AODs) were conducted using multi-wavelength radiometer (MWR). The AOD at 0.5 μm wavelength on daily basis (mean $\pm$ standard deviation) for the entire three-year study period is obtained as 0.24 ± 0.08 . Seasonal variations show the highest AOD at 0.5 μm wavelength with $\sim 0.34 \pm 0.08$ during pre-monsoon (April–July), followed by $\sim 0.26 \pm 0.08$ during monsoon (August–September), $\sim 0.21 \pm 0.05$ during post-monsoon (October–November) and $\sim 0.20 \pm 0.07$ during winter (December–March). The seasonal values indicate that the AOD at 0.5 μm wavelength is decreasing from pre-monsoon to winter with a notable reduction around 41%. The Ångström parameters using least square method is not found appropriate for size distribution particularly when coarse mode aerosols dominate. The coefficients of second order polynomial fit are more appropriate for the discrimination of aerosol size or irrespective to the dominance of either of the aerosols size. The difference in coefficient of polynomial fit is used to get confirmation on the dominant mode during different seasons. Study reveals that about 93%, 72% and 59% of AOD spectra are dominated by a wide range of fine mode fractions or mixture of modes during post-monsoon, winter and monsoon, respectively. On the other hand, during pre-monsoon, 72% of AOD spectra are found to be dominated by coarse mode aerosols.

1. Introduction

Aerosols are produced by a variety of natural as well as anthropogenic activities, and get distributed in the atmosphere through turbulent mixing and transport which result in a large variability in their size distribution (Meszaros 1981; Reist 1984; Eck *et al* 1999; Kedia and Ramachandran

2009). At global level, the natural sources (e.g., volcanic eruption, transport of desert dust, etc.) of aerosols are dominant over the anthropogenic sources (e.g., biomass burning, vehicular emission, etc.), but at regional level the contribution of anthropogenic sources of aerosol is significant (Ramanathan *et al* 2001). The long-range transport of desert dust also contributes to the aerosol

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size distribution (ASD) (Moorthy and Satheesh 2000; Gogoi *et al* 2008; Kedia and Ramachandran 2009, 2010). The investigation on aerosol transport is carried out by Kuniyal *et al* (2009) and Guleria *et al* (2011a, 2011b, 2012) at Mohal in the northwestern part of the Indian Himalaya. Their investigations suggest that Mohal gets affected by dust aerosol due to its transport from the western deserts. The understanding of the variability in the ASD is the prime concern to quantify the radiative effect of aerosol, but it is a challenging task due to inadequate information (IPCC 2007).

Spectral aerosol optical depth (AOD, $\tau_{p\lambda}$) contains information pertaining to ASD and is used to compute Ångström exponent (α) and turbidity coefficient (β) using Ångström Power Law (Schuster *et al* 2006; Kaskaoutis *et al* 2007, 2010; Gogoi *et al* 2008; Kedia and Ramachandran 2009; Sharma *et al* 2010, 2011). Ångström exponent is a quantitative indicator of the ASD and the turbidity coefficient is a measure of total aerosol loading in a vertical column (Schuster *et al* 2006; Gogoi *et al* 2008; Kedia and Ramachandran 2009; Kaskaoutis *et al* 2010; Sharma *et al* 2011). In general, Ångström Power Law is applicable to the entire wavelength spectrum, only if the ASD fits to the Junge Power Law (Kedia and Ramachandran 2009). But, when the spectral range is extended and the ASD is generally multi-modal, it can introduce significant errors (Kaskaoutis *et al* 2010). Therefore, the use of Ångström Power Law is not applicable to all the environments and regions (Eck *et al* 1999, 2001; Reid *et al* 1999; Kaskaoutis and Kambezidis 2006; Kaskaoutis *et al* 2007; Kedia and Ramachandran 2009; Kalapureddy and Devara 2010). When the spectral range is extended, Ångström Power Law departs from its linear behaviour of $\ln \tau_{p\lambda}$ versus $\ln \lambda$ and this results in different values of α for different wavelength ranges (Kedia and Ramachandran 2009). This leads to a curvature in $\tau_{p\lambda}$ which contains useful information about the ASD (Eck *et al* 1999). The value of $\alpha \leq 1$ indicates that the size distributions are dominated by coarse mode aerosols such as dust or sea salt. The value of $\alpha \geq 2$ indicates that the size distributions are dominated by fine mode aerosols that are usually attributed to anthropogenic activities (Eck *et al* 1999; Schuster *et al* 2006).

Numerous measurements to investigate the curvature in AOD spectra and impact assessments are reported from the Indian region in recent years (Kedia and Ramachandran 2009; Kalapureddy and Devara 2010; Kaskaoutis *et al* 2010, 2011; Kedia *et al* 2010; Sharma *et al* 2010; Kumar *et al* 2011). However, these measurements are focused either on urban landmass or oceans adjacent to densely populated coastal regions. Investigations from remote

and sparsely inhabited regions of the mountain ecosystems are very rare. This paper deals with the study of characterization of atmospheric aerosols, investigation of the errors and uncertainties computed from the application of the Ångström Power Law (linear fit) as well as from the second order polynomial fit in the $\ln \tau_{p\lambda}$ versus $\ln \lambda$ relationship. This study also highlights the seasonal variability in contribution of fine mode as well as coarse mode aerosols from different source origins. Thus, the reported results on seasonal variations in aerosol characteristics from the Kullu valley located in the northwestern Indian Himalaya will be useful in modelling the radiative effect of aerosols at a regional level.

2. Experimental site description and surface meteorology

The present experimental site – Mohal (31.9°N, 77.12°E, 1154 m amsl) is located in the Kullu valley of the northwestern part of Indian Himalaya (figure 1). This part of the Indian Himalaya is mostly affected due to dust aerosol transport from

Figure 1. View of the Kullu valley (source: Google Earth), showing installation location of MWR.

the desert prone regions such as the Sahara Desert (Kuniyal *et al* 2009; Guleria *et al* 2011a, 2011b, 2012), the Taklimakan Desert (Guleria *et al* 2011b, 2012), and the Thar Desert (Kuniyal *et al* 2009; Guleria *et al* 2011a, 2012) during pre-monsoon and the smoke aerosol transport from the plains of India such as Punjab (Guleria *et al* 2011b, 2012) during post-monsoon. The Kullu valley is surrounded by mountains ranging from 3000–5000 m. The River Beas flows in the middle part of this valley. The Kullu valley is about 80-km long and 2-km wide. It begins from Larji (957 m amsl) in the lower Beas basin in the south and extends up to the Rohtang crest (4038 m amsl) in the upper Beas basin in the north (Kuniyal *et al* 2009). Mohal is a semi-urban experimental site under the programme of Aerosols Radiative Forcing over India (ARFI), a national project under the Indian Space Research Organization Geosphere Biosphere Programme (ISRO-GBP) (Kuniyal *et al* 2009; Guleria *et al* 2011b).

2.1 Surface meteorology

It is a well-known fact that meteorology and atmospheric dynamics play an important role in affecting and modulating the spatio-temporal variabilities in aerosol characteristics over any region (Singh *et al* 2004; Ganguly *et al* 2006; Kaskaoutis *et al* 2009). Therefore, following Kuniyal *et al* (2009) and Guleria *et al* (2011a), the entire observation period is classified into four local seasons: pre-monsoon (April–July), monsoon (August–September), post-monsoon (October–November) and winter (December–March) depending on the prevailing local meteorological parameters and climatic characteristics. The monthly variations of meteorological parameters are shown in figure 2. The surface wind is noticed as high as 1.4 ms^{-1} during August 2006. In general, the surface wind speed remains low ($<1.0 \text{ ms}^{-1}$) with less significant monthly/seasonal variations. However, the average value of wind speed is found to be slightly lower during post-monsoon period. The predominant surface wind direction over Mohal is generally south–southwest. During the study period, relative humidity is observed highest in monsoon ($\sim 70\%$) and lowest in post-monsoon and winter ($\sim 61\%$). The monthly temperature is found to vary between 7°C and 26°C . In general, the temperature is found to increase from January to August. Figure 2(e) indicates that the maximum rainfall occurs in winter months (38% of total rainfall) and minimum in post-monsoon months (2% of the total rainfall). However, this rainfall pattern is in contrast to the other parts of India. The lower part of the Kullu valley starting from Aut in the south to Katrain

Figure 2. Monthly variation in (a) surface wind speed, (b) surface wind direction, (c) relative humidity, (d) temperature, and (e) monthly total rainfall from April 2006 to March 2009.

in the north lies in the rain shadow zone (Kuniyal *et al* 2009; Guleria *et al* 2011b) and hence receives less amount of rainfall during monsoon period as compared to winter period.

Figure 3 shows the average seasonal pattern of the prevailing winds at 700 hPa, averaged over the period 2006–2009. The map of prevailing synoptic wind vectors is obtained from the National Centers for Environmental Prediction (NCEP) reanalysis website (<http://www.cdc.noaa.gov>). Figure 3 shows that the wind pattern at 700 hPa over Mohal is calm and is of mixed origins. During pre-monsoon, the north-westerly wind flows towards Mohal with a velocity of about 3 ms^{-1} . In monsoon, the prevailing winds flow towards the present

Figure 3. NCEP re-analysis mean vector wind patterns at 700 hPa level over the Indian region for pre-monsoon (April–July), monsoon (August–September), post-monsoon (October–November) and winter (December–March) seasons during the measurement period (April 2006–March 2009). The shades represent the magnitudes of wind in ms^{-1} , the arrows show direction and the star denotes the location of Mohal.

experimental site from the northwestern part and the Bay of Bengal. Whereas in post-monsoon and winter, the westerly winds seem to be dominant with a velocity of about 3 ms^{-1} .

3. Instrumentation, data and analysis

3.1 AOD measurement

The measurements of spectral AOD were carried out at Mohal for the period ‘April 2006 to March 2009’ using multi-wavelength radiometer (MWR). The MWR instrument is designed and developed at Space Physics Laboratory, Vikram Sarabhai Space Centre, Thiruvananthapuram (Moorthy *et al* 1999). The MWR having a field of view $\sim 2^\circ$,

takes measurements of the spectral extinction of ground reaching solar flux as a function of solar zenith angle at 10 wavelengths, centred at 0.38, 0.4, 0.45, 0.5, 0.6, 0.65, 0.75, 0.85, 0.935 and $1.025 \mu\text{m}$ (Moorthy *et al* 1999). The MWR works on the principle of filter wheel radiometer as described by Shaw *et al* (1973) and was operated on all cloud-free days. During the measurement period, the prevailing weather conditions in majority of the days were cloudy. This limits the availability of MWR data. There were only 181 data points over the entire study period as the measurements were restricted to clear sky days. However, Mohal region received maximum amount of rainfall in winter months of 2008, but the MWR measurements could become possible due to less number of cloudy days as compared to June–July 2008. The

raw data obtained through MWR is further analysed to obtain AOD following Langley techniques (Shaw *et al* 1973). In analysing the MWR data, the data collected during the forenoon and afternoon parts of the day is considered as a single dataset and the average spectral AOD is retrieved following Langley technique (Gogoi *et al* 2008, 2009). A detailed description of the MWR instrument and data analysis techniques have been published elsewhere (Moorthy *et al* 1999, 2001; Sagar *et al* 2004; Bhuyan *et al* 2005; Gogoi *et al* 2008).

Long-term temporal stability of the Langley intercept, corrected for the daily variation of the Sun–Earth distance is used as an indirect calibration of the instrument. Since Langley intercept remains invariant over years and any change in zero air mass intercept should arise from variations in the MWR parameters. Errors in the AOD measurements can arise from contribution of forward scattering to the measured flux; air mass calculation, deviation in the calibration coefficient, and in estimation of the optical depth by molecular scattering and absorption. Estimates have shown that if these errors are added, there may have the maximum uncertainty of ~ 0.03 (Gogoi *et al* 2008). For more details about the errors involved in MWR-measured AOD, see earlier papers, e.g., Moorthy *et al* (1999, 2001), Sagar *et al* (2004), Bhuyan *et al* (2005) and Gogoi *et al* (2008).

3.2 Ångström exponent (α) and turbidity coefficient (β)

Ångström proposed an empirical formula to approximate the spectral dependence of atmospheric extinction caused by aerosols. Spectral AOD contains information pertaining to size distribution used to compute α and β using empirical formula which is famously known as the Ångström Power Law (Ångström 1961).

$$\tau_{p\lambda} = \beta \lambda^{-\alpha}, \quad (1)$$

where wavelength (λ) is expressed in μm . The logarithmic scale of equation (1) yields a straight line as:

$$\ln \tau_{p\lambda} = \ln \beta - \alpha \ln \lambda. \quad (2)$$

The plots of $\ln \tau_{p\lambda}$ versus $\ln \lambda$, yield a straight line of slope $-\alpha$ and intercept $\ln \beta$. The validity of the Ångström Power Law is stated after Junge Power Law (Junge 1955) for a limited range of particles, where significant extinction takes place and the spectral variation of the refractive index does not impose any significant variations on the Mie extinction factor (Kaskaoutis *et al* 2006). For more details of α and β and its computation techniques,

see Bhuyan *et al* (2005); Gogoi *et al* (2008) and Sharma *et al* (2011).

3.3 Second order polynomial fit

Due to varied sources and their formation mechanism, the aerosol size distribution in the atmosphere is rarely uni-modal. When it is multi-modal, the wavelength dependence of AOD does not follow Ångström Power Law (Eck *et al* 1999). Therefore, departure from the linear behaviour of $\ln \tau_{p\lambda}$ versus $\ln \lambda$ data is expected. The second order polynomial fit to examine the curvature in the AOD spectra can be followed to estimate the error for each spectral AOD dataset. The second order polynomial fit to examine the curvature in the AOD spectra is expressed as:

$$\ln \tau_{p\lambda} = a_2 (\ln \lambda)^2 + a_1 \ln \lambda + a_o, \quad (3)$$

where a_1 , a_2 and a_o are constants. The coefficient a_2 represents the curvature observed in the spectral distribution of AOD when $\ln \tau_{p\lambda}$ is plotted against $\ln \lambda$. The curvature is an indicator of the aerosol particle size. The positive curvature indicates that the aerosol size distribution is dominated by coarse mode aerosols, while negative curvatures with fine mode aerosols (Eck *et al* 1999; Schuster *et al* 2006). Near to zero values are indicative of bi-modal aerosol distributions with similar contribution of coarse mode and fine mode aerosols (Eck *et al* 1999; Schuster *et al* 2006).

4. Results and discussion

4.1 Seasonal variations in AOD and Ångström parameters

The seasonal variations in AOD averaged over the period April 2006 to March 2009 obtained at 0.38, 0.5 and 1.025 μm wavelengths are shown in figure 4(a). The vertical bars represent the standard deviation from the mean of the observations. We chose these wavelengths for the analysis because these are representative of the ultra violet, visible and near-infrared wavelength spectrum, where the effect of scattering due to aerosols of different size can be differentiated (Schuster *et al* 2006). It is also made clear that the AOD in the short wavelength region are more influenced by fine mode aerosols, while in the longer wavelengths by coarse mode aerosols (Kedia and Ramachandran 2010). The variation in AOD in all seasons is found to be wavelength-dependent. The standard deviations of AODs are large during the pre-monsoon and monsoon, which indicate higher day-to-day variability in the characteristics of aerosols

Figure 4. Seasonal variations in (a) AOD obtained at 0.38, 0.5 and 1.025 μm wavelengths and (b) α and β estimated from the linear fit of spectral AOD in the spectral band of 0.38–1.025 μm . The vertical bar represents the standard deviations from the mean of the observations.

(Holben *et al* 2001). The daily average (mean $\pm$ standard deviation) AOD at 0.38, 0.5 and 1.025 μm wavelengths for the entire study period is obtained as 0.33 ± 0.10 , 0.24 ± 0.08 and 0.14 ± 0.07 , respectively. The seasonal highest AOD at 0.38, 0.5 and 1.025 μm wavelengths in pre-monsoon are obtained as 0.43 ± 0.09 , 0.34 ± 0.08 , and 0.24 ± 0.09 , respectively. This is due to enhanced loading of dust aerosols, which are transported from the desert regions (e.g., Sahara Desert, Thar Desert, etc.) (Kuniyal *et al* 2009; Guleria *et al* 2011a, 2011b, 2012). The seasonal lowest AOD at 0.38 and 0.5 μm wavelengths are obtained as 0.29 ± 0.08 and 0.20 ± 0.07 at 0.5 μm in winter, whereas AOD at 1.025 μm wavelength is obtained as 0.09 ± 0.03 in post-monsoon. The AOD at 0.38, 0.5 and 1.025 μm wavelengths decreased from pre-monsoon to winter with notable reduction of around 33%, 41% and 58%, respectively. The sharp change in AOD at longer wavelengths indicates that the coarse mode aerosols decreased significantly from pre-monsoon to winter.

The seasonal variations in α and β averaged over the period April 2006 to March 2009 is estimated from the linear fit of spectral AOD in the spectral band 0.38–1.025 μm (figure 4b). From figure 4(a, b), it is known that during pre-monsoon,

large AOD values are associated with small α values. This indicates that the relative concentration of coarse mode aerosols is higher than fine mode aerosols. Kuniyal *et al* (2009) and Guleria *et al* (2011a, 2011b, 2012) conducted the study over Mohal on aerosol transport using the back trajectories in conjunction with Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) analysis. Their studies report that during pre-monsoon, the AOD at Mohal is mostly affected due to the transport of dust aerosols from the desert regions such as Sahara Desert, Thar Desert, etc. This could be the cause of a large concentration of coarse mode aerosols over Mohal. During post-monsoon, small AOD values are associated with large α values. This indicates that during post-monsoon, the contribution of fine mode aerosols in the atmosphere is relatively more than coarse mode aerosols. Guleria *et al* (2011b, 2012) report that during post-monsoon, AOD at Mohal is affected by the transport of smoke aerosols from the plains of India. This could be one of the major causes for the accumulation of more fine aerosols over Mohal. The seasonal mean values show that the β decreased from pre-monsoon to post-monsoon with notable reduction of 58%. Whereas the α increased from pre-monsoon to post-monsoon with a notable increase around 67%. The daily average values of α and β for the entire study period are obtained as 1.20 ± 0.41 and 0.12 ± 0.06 , respectively.

4.2 Errors and uncertainties in the retrieval of Ångström parameters

Kaskaoutis and Kambezidis (2006) reported that the computation of Ångström parameters (α , β) using least square method (linear regression fit) is not applicable for all atmospheric conditions and locations, because this method is least wavelength-dependent. Mohal experimental site is characterized by natural and anthropogenic aerosols which show distinct seasonal characteristics over the region (Guleria *et al* 2011b). The pre-monsoon season is a key period when dust aerosol loading peaks over Mohal (Kuniyal *et al* 2009; Guleria *et al* 2011a, 2011b). The prevailing westerly air-mass transports dust aerosols originating from the western desert regions (Kuniyal *et al* 2009; Guleria *et al* 2011a, 2011b). During post-monsoon season, the transport of anthropogenic aerosols from the plains of India and polluted continent of Pakistan and Afghanistan cause increase in aerosol loading over the Mohal region (Guleria *et al* 2011b). Local sources such as fuel wood burning and seasonal forest fires are also the main contributors to the building up of aerosol layer over Mohal. Thus, the

Figure 5. Seasonal variations in spectral AOD in spectral band of 0.38–1.025 μm . The vertical bar represents the standard deviations from the mean of the observations. The linear and the second order polynomial fit equations for each season are shown.

Table 1. Seasonal average (mean $\pm$ error) linear fit parameters and second order polynomial fit coefficients computed in the spectral band of 0.38–1.025 μm during April 2006 to March 2009.

Season	Linear fit parameters			Second order polynomial fits coefficients			
	$\alpha \pm \text{error}$	Intercept $\pm \text{error}$	R^2	$a_2 \pm \text{error}$	$a_1 \pm \text{error}$	$a_0 \pm \text{error}$	R^2
Pre-monsoon	0.90 ± 0.13	1.71 ± 0.08	0.84	$+0.55 \pm 0.48$	-0.39 ± 0.49	-1.65 ± 0.09	0.89
Monsoon	0.98 ± 0.11	2.02 ± 0.06	0.91	$+0.43 \pm 0.39$	-0.57 ± 0.39	-1.97 ± 0.08	0.93
Post-monsoon	1.42 ± 0.07	2.51 ± 0.04	0.98	$+0.29 \pm 0.28$	-1.12 ± 0.28	-2.48 ± 0.05	0.98
Winter	1.13 ± 0.08	2.32 ± 0.05	0.96	$+0.05 \pm 0.32$	-1.08 ± 0.32	-2.31 ± 0.06	0.96

present study is conducted using the application of second order polynomial fit in order to understand the applicability of Ångström Power Law. The seasonal variations in spectral AOD in the spectral band of 0.38–1.025 μm are shown in figure 5. The seasonal average (mean $\pm$ error) values of linear fit parameters and second order polynomial fit coefficients computed in the spectral band of 0.38–1.025 μm are shown in table 1. From this table, it is clear that during pre-monsoon, the linear and second order polynomial fits have exhibited large uncertainties. However, the linear regression fit method ($R^2 = 0.84$) is found to be least wavelength-dependent compared to second order polynomial fit ($R^2 = 0.89$). The coefficient of determinant (R^2) is a measure to know how well the regression line approximates the real data points

and provides information about the effectiveness of the linear and second order polynomial fits (Kedia and Ramachandran 2009). During monsoon, the linear and second order polynomial fits have also exhibited large uncertainties, but they are relatively low compared to pre-monsoon. The linear regression fit method ($R^2 = 0.91$) is found to be least wavelength-dependent compared to second order polynomial fit ($R^2 = 0.93$). The large values of positive a_2 and small values of α indicate that during pre-monsoon and monsoon, the coarse mode aerosols are dominant. During post-monsoon and winter, both the fits have exhibited low uncertainties with strong dependency on wavelength (see table 1). The small values of positive a_2 and large values of α indicate that during post-monsoon and winter, the fine mode aerosols are dominant. From

the results of this study, it is found that the linear and second order polynomial fits have exhibited higher accuracy during post-monsoon and winter in comparison to pre-monsoon and monsoon, when significant uncertainties and errors in the retrieved parameters, i.e., α , a_1 and a_2 occurred in close association with coarse mode aerosols. But, the use of least square method is found least wavelength-dependent compared to second order polynomial fits.

Earlier studies reported that computation of α using least square method is least wavelength-dependent and is not appropriate for all seasons or the days where the size distribution is dominated by significant presence of coarse mode aerosols (Eck *et al* 1999; Reid *et al* 1999; Schuster *et al* 2006; Kaskaoutis *et al* 2007). High value of R^2 means a good agreement between the linear and second order polynomial fits indicating that aerosol size distribution is dominated by significant presence of fine mode aerosols (Kedia and Ramachandran 2009). So, an attempt is made to draw a plot between R^2 and α computed in the spectral band of 0.38–1.025 μm (figure 6). From correlation plot, it is noticed that at $\alpha < 1.0$, second order polynomial fit in 61% cases has shown wavelength dependent behaviour with $R^2 > 90$, whereas in case of linear fit it is 40%. At $\alpha > 1.0$, second order polynomial fit in 98% cases has shown wavelength dependent behaviour with $R^2 > 90$, whereas in case of linear fit it is 84%. Therefore, this study signifies that the computation of α and β using least square method is least wavelength-dependent. So, it is not appropriate for all the seasons or days where the aerosol size distribution is dominated by significant concentrations of coarse mode aerosol.

A detailed analysis is carried out to estimate the errors and uncertainties of the second order polynomial fit to the spectral AOD. The reason of the errors arising in the estimation of a_1 and a_2 is worked out. The errors in a_1 and a_2 show the

Figure 6. Correlation between the coefficients of determinant (R^2) and α for spectral band of 0.38–1.025 μm .

relative skill of the second order polynomial fit in simulating the AOD spectral distribution and express the scatter of the AOD around the polynomial curve (Schuster *et al* 2006; Kaskaoutis *et al* 2007). Figure 7(a, b) shows the correlation of the errors in a_1 and a_2 evaluated in spectral bands of 0.38–1.025 μm and 0.38–0.75 μm with AOD at 0.5 μm . From the plots, it is found that error a_1 and error a_2 decreased with AOD. However, despite the fact that error a_1 and error a_2 indicate an exponential decrease in spectral band of 0.38–0.75 μm and in spectral band of 0.38–1.025 μm seems to be rather neutral. A large number of errors in the computed coefficients are observed for low AOD ≤ 0.25 which themselves exhibit large variations. Nevertheless, as the AOD increases (AOD > 0.35), the error in the estimate decreases dramatically. Schuster *et al* (2006) reported that $\alpha = a_2 - a_1$, therefore large errors in a_1 and a_2 lead to a larger uncertainty in the linear fit. AOD values at 0.5 μm in 63% cases are ≤ 0.25 . This reveals that a larger uncertainty occurred in retrieval of α in close association with low AOD values. Kaskaoutis *et al* (2006) reported that this feature is common in sunphotometric measurements and is attributed to the higher variability of the spectral AOD.

4.3 Curvatures in AOD spectra

To verify the validity of relation $\alpha = a_2 - a_1$, the correlation between α and $a_2 - a_1$ in different spectral bands is shown in figure 8(a, b). The data points lying on 1:1 line or closely around it are found to confirm the validity of the relation $\alpha = a_2 - a_1$. The linear fit and second order polynomial fit computed in the wavelength interval 0.38–1.025 μm are found in good agreement with root mean square deviation (RMSD), mean absolute bias deviation (MABD) and correlation coefficient of 0.08, 0.01 and 0.98 for the daily observations, respectively having intercepts close to zero. While, in the wavelength interval 0.38–0.75 μm , the RMSD, MABD and correlation coefficients for daily observations are obtained as 0.31, 0.14 and 0.74, respectively with the intercepts 0.39. There is almost no systematic overestimation of the $a_2 - a_1$ against α (figure 8a), while it seems to be present in figure 8(b) at $\alpha < 1.5$. These agreements provide the validation of the linear and polynomial fit equations computed in the spectral band of 0.38–1.025 μm . Whereas in the spectral band of 0.38–0.75 μm , the linear and polynomial fit-derived parameters are not found in good agreement and the retrieved parameters are found to discard the validity of the relation $\alpha = a_2 - a_1$. From the analysis, it is found that as the wavelength region shifts towards

Figure 7. Correlation of the errors in a_2 and a_1 obtained from the polynomial fit of the $\ln \tau_{p\lambda}$ versus $\ln \lambda$ in the two spectral bands with the AOD at $0.5 \mu\text{m}$.

Figure 8. Correlation between $a_2 - a_1$ and α for (a) spectral band of 0.38-1.025 μm and (b) spectral band of 0.38-0.75 μm . The dotted black lines represent the 1:1 line and the dark line, the linear equation.

shorter wavelengths, the use of least square is not appropriate for the retrieval of Ångström parameters. Therefore, this study indicates that there is a need to look into the curvature effect in retrieval of size parameter using Ångström Power Law at shorter wavelength spectrum. This feature is further investigated during some important cases. A typical example of 30 May 2008 is shown in figure 9(a, b) whereas the wavelength region used for α determination shifts towards shorter wavelengths,

higher α value is observed. The positive curvature in the $\ln \tau_{p\lambda}$ versus $\ln \lambda$ relationship indicates significant fraction of coarse mode aerosols. The linear and second order polynomial fit equations computed in different spectral bands are obtained as:

$$\ln \tau_{p\lambda} = -0.66 (\pm 0.15) \ln \lambda - 1.75 (\pm 0.09);$$

$$R^2 = 0.71; (0.38 - 1.025 \mu\text{m}), \quad (4)$$

Figure 9. Typical examples of spectral variation in AOD *versus* wavelength. The linear and the second order polynomial fits for each case are shown in the illustration.

$$\begin{aligned} \ln \tau_{p\lambda} &= 0.91 (\pm 0.49) \ln \lambda^2 \\ &\quad + 0.21 (\pm 0.49) \ln \lambda - 1.64 (\pm 0.10); \\ R^2 &= 0.81; (0.38-1.025 \mu\text{m}), \end{aligned} \quad (5)$$

$$\begin{aligned} \ln \tau_{p\lambda} &= -1.01 (\pm 0.17) \ln \lambda - 2.00 (\pm 0.12); \\ R^2 &= 0.88; (0.38-0.75 \mu\text{m}), \end{aligned} \quad (6)$$

$$\begin{aligned} \ln \tau_{p\lambda} &= -1.22 (\pm 0.84) \ln \lambda^2 \\ &\quad - 2.57 (\pm 1.08) \ln \lambda - 2.43 (\pm 0.30); \\ R^2 &= 0.92; (0.38-0.75 \mu\text{m}), \end{aligned} \quad (7)$$

The large value of positive a_2 and small value of α (see equations 4 and 5) indicate that on 30 May 2008, the AOD spectra is dominated by coarse mode aerosols. When the wavelength region shifts towards shorter spectral band of 0.38–0.75 μm (figure 9b), the large value of negative a_2 and large value of α (see equations 6 and 7) indicate that on 30 May 2008, the AOD spectra is dominated by fine mode aerosols. The α value is found to change from 0.66 to 1.01 when the wavelength region used for its determination shifts from longer wavelengths (0.38–1.025 μm) towards shorter wavelengths (0.38–0.75 μm). A typical example of 8 January 2009 is shown in figure 9(c, d), whereas the wavelength region used for α determination shifts towards shorter wavelengths, smaller α value is observed. The negative curvature in the

$\ln \tau_{p\lambda}$ *versus* $\ln \lambda$ relationship indicates significant fraction of fine mode aerosols. The linear and second order polynomial fit equations computed in different spectral bands are obtained as:

$$\begin{aligned} \ln \tau_{p\lambda} &= -1.47 (\pm 0.20) \ln \lambda - 2.10 (\pm 0.12); \\ R^2 &= 0.87; (0.38-1.025 \mu\text{m}), \end{aligned} \quad (8)$$

$$\begin{aligned} \ln \tau_{p\lambda} &= -1.87 (\pm 0.37) \ln \lambda^2 \\ &\quad - 3.27 (\pm 0.37) \ln \lambda - 2.32 (\pm 0.07); \\ R^2 &= 0.97; (0.38-1.025 \mu\text{m}), \end{aligned} \quad (9)$$

$$\begin{aligned} \ln \tau_{p\lambda} &= -0.72 (\pm 0.10) \ln \lambda - 1.54 (\pm 0.07); \\ R^2 &= 0.91; (0.38-0.75 \mu\text{m}), \end{aligned} \quad (10)$$

$$\begin{aligned} \ln \tau_{p\lambda} &= -0.81 (\pm 0.48) \ln \lambda^2 \\ &\quad - 1.76 (\pm 0.61) \ln \lambda - 1.82 (\pm 0.18); \\ R^2 &= 0.95; (0.38-0.75 \mu\text{m}). \end{aligned} \quad (11)$$

The very large values of negative a_2 and α (see equations 8 and 9) indicate that on 8 January 2009, the AOD spectra is dominated by fine mode aerosols. When the wavelength region shifts towards shorter spectral band of 0.38–0.75 μm (figure 9d), the small values of negative a_2 and α relative to equations (9) and (8), respectively indicate that on 8 January 2009, the AOD spectra

Figure 10. Scatter plot of coefficients a_2 against AOD at $0.5 \mu\text{m}$.

is dominated by coarse mode aerosols. The α value is found to change from 1.47 to 0.72, when the wavelength region used for its determination shifts from longer wavelengths ($0.38\text{--}1.025 \mu\text{m}$) towards shorter wavelengths ($0.38\text{--}0.75 \mu\text{m}$). From the above discussion, it is concluded that there is a considerable change in aerosol size distribution when the wavelength region shifts towards shorter spectral bands.

Figure 10(a, b) shows the scatter plot between polynomial coefficient a_2 and AOD at $0.5 \mu\text{m}$. The correlation between a_2 and AOD at $0.5 \mu\text{m}$ provides information on the atmospheric conditions under which α is independent from wavelengths (Sharma *et al* 2010). The data points lying on or around $a_2 = \text{zero}$ line in figure 10(a, b) indicate a bi-modal aerosol distribution without curvature (Schuster *et al* 2006; Kaskaoutis *et al* 2007). The negative values of a_2 suggest the presence of significant fraction of fine mode aerosols and positive values of a_2 provide the information about coarse mode aerosols. In 39% cases, the a_2 values over Mohal are obtained negative when they were computed in the spectral band $0.38\text{--}1.025 \mu\text{m}$. When the wavelength region shifts towards shorter spectral band of $0.38\text{--}0.75 \mu\text{m}$, the a_2 values in 70% cases are obtained negative. This study further signifies that as the wavelength region shifts towards shorter spectral band of $0.38\text{--}0.75 \mu\text{m}$, a considerable change in aerosol size distribution is observed. After evaluating the results, it is observed that there is need for great effort and attention on the accuracy of AOD at shorter wavelengths. So, the use of $0.38\text{--}1.025 \mu\text{m}$ spectral band for the calculation of α , a_1 and a_2 values can lead to the most precise results with the lowest curvature.

4.4 Seasonal variability in relative contribution of different mode aerosols

Eck *et al* (1999) and Schuster *et al* (2006) reported that for the multi-modal aerosol size distribution, the polynomial coefficients (a_2 and a_1) contain significant information about aerosol type. For bi-modal aerosol distribution, where distributions of fine mode and coarse mode aerosols are almost with equivalent contributions, the α is to a close approximation of $a_2 - a_1$ (i.e., $\alpha = a_2 - a_1$) (Schuster *et al* 2006). For values, where $a_2 - a_1 \geq 2$, the AOD spectra are dominated by fine mode ($<1 \mu\text{m}$)

Figure 11. Seasonal variations in correlation between the coefficients a_2 and a_1 computed in the spectral range of $0.38\text{--}1.025 \mu\text{m}$. Dotted lines correspond to $a_2 - a_1 = 1$ and $a_2 - a_1 = 2$.

Figure 12. Percentage of AOD spectra classified on the basis of seasons $a_2 - a_1 \leq 1$ (coarse mode aerosols are dominant), $1 < a_2 - a_1 < 2$ (a wide range of fine mode fractions or mixture of modes), ≥ 2 (fine mode aerosols are dominant).

aerosols, where $a_2 - a_1 \leq 1$, the AOD spectra are dominated by coarse mode ($>1 \mu\text{m}$) aerosols. For intermediate values of $a_2 - a_1$ between 1 and 2, the AOD spectra represent a wide range of fine mode fractions or mixture of modes. To identify different modes of aerosols prevailing over Mohal, coefficients a_2 and a_1 as a function of season are plotted in figure 11. The percentage of AOD spectra having $a_2 - a_1 \leq 1$ and $1 < a_2 - a_1 < 2$ is found to exhibit a strong seasonal variation (figures 11 and 12). Figure 11 shows that even at same value of a_2 , the value of a_1 may vary, but this feature depends on the contribution of coarse mode aerosols. Therefore, this study confirms that the use of single coefficient is not appropriate to discriminate fine mode as well as coarse mode aerosols. During pre-monsoon, due to dust aerosol transport from the desert prone region to Mohal (see Kuniyal *et al* 2009; Guleria *et al* 2011a, 2011b, 2012), coarse mode aerosol become dominant. This leads to a higher percentage of AOD spectra, i.e., 72% with $a_2 - a_1 \leq 1$ (figure 12). During monsoon, the coarse mode aerosols are dominant in 41% of AOD spectra, whereas the contribution of fine mode fractions or mixture of modes is 59% of AOD spectra. A wide range of fine mode fractions or mixture of modes is observed during post-monsoon as for 93% of the AOD spectra, $a_2 - a_1$ lies between 1 and 2. For 5% of AOD spectra, $a_2 - a_1 \geq 2$ indicates a strong dominance of fine mode aerosols. This large concentration of fine mode aerosol is attributed to anthropogenic aerosols, released into the atmosphere by bio-mass burning, forest fire at local levels and smoke aerosol transport from external sources (Guleria *et al* 2011a, 2011b, 2012). During winter, the fine mode aerosols are dominant in 72% of AOD spectra $a_2 - a_1$ lies between 1 and 2 (figure 12).

This study confirmed that during post-monsoon and winter, the size distribution over Mohal computed in the spectral band of $0.38\text{--}1.025 \mu\text{m}$ is found with equivalent contribution of fine mode

and coarse mode aerosols. But in pre-monsoon and monsoon, aerosol size distribution is found to discard the validity of the relation, i.e., $\alpha = a_2 - a_1$, which signifies the multi-modal aerosol size distribution. Therefore, this study highlights the seasonal variability in contribution of fine mode as well as coarse mode aerosols from different source origins. This study reveals that it is possible to depict the percentage contribution of different fine mode as well as coarse mode aerosols in the measured AOD spectra. Thus, the seasonal variations in aerosol characteristics from a mountain ecosystem will be useful in examining the radiative effect of aerosol at a regional level.

5. Conclusion

Three years measurements (April 2006 to March 2009) of seasonal variability in aerosol optical and physical characteristics were made at Mohal in the northwestern Indian Himalaya. Moreover, the appropriateness using the Ångström Power Law and second order polynomial fit for aerosols size distribution showed that the latter gives more accurate results compared to the former especially when the coarse mode aerosols dominate. In a nutshell, the major conclusions of the present study are as follows:

- The AODs on daily basis (mean $\pm$ standard deviation) stood to be 0.33 ± 0.10 , 0.24 ± 0.08 and 0.14 ± 0.08 at 0.38 , 0.5 and $1.025 \mu\text{m}$, respectively.
- The AOD at 0.38 , 0.5 and $1.025 \mu\text{m}$ have shown a reasonable decrease from pre-monsoon to winter with a significant reduction of 33%, 41% and 58%, respectively.
- The AOD values over Mohal have shown a seasonal as well as wavelength dependent behaviour indicating an influence of local and external sources of aerosols.

- The Ångström exponent varied significantly from pre-monsoon (April–July) to winter (December–March) indicating the dominance of anthropogenic impacts on the aerosol characteristics.
- Ångström parameters using least square method show least wavelength-dependence compared to second order polynomial fit. It is also not appropriate for the days when the size distribution is dominated by coarse mode aerosols.
- There is a considerable change in aerosol size distribution when the wavelength region shifts towards shorter spectral bands. This indicates that adequate attention is required to have greater accuracy in measuring AOD at shorter wavelengths.
- During pre-monsoon, about 72% of AOD spectra are found to be dominated by coarse mode aerosols. Whereas, during monsoon, post-monsoon and winter about 59%, 93% and 72% of AOD spectra are found to be dominated by a wide range of fine mode fractions or mixture of modes, respectively.
- This study highlights the seasonal variability in terms of fine as well as coarse mode aerosols from different origin sources. Thus, the data obtained on seasonal variability in aerosol characteristics ecologically from a sensitive mountain ecosystem will be useful in modelling the radiative effect of aerosols at a regional level.

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