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LONG TERM AEROSOL CHARACTERIZATION OVER KODAIKANAL, A HIGH ALTITUDE STATION IN SOUTH INDIA

1. Knowledge of aerosols' characteristics is vital, for understanding the impact of aerosols on climatic perturbations, chiefly due to the large heterogeneity in aerosols' features given the multiple sources, volatility and mobility of aerosols. Numerous measurements and impact assessments are being reported from Indian region in the recent years (Moorthy, 1999 & 2004; Satheesh and Ramanathan, 2000; Ramanathan *et al.*, 2001; Babu *et al.*, 2002; Seinfeld *et al.*, 2004; Pandithurai *et al.*, 2004; Tripathy *et al.*, 2005). But, most of these studies focused to urban, semi-urban, rural and densely populated coastal areas. Investigations from high altitude regions that provides information quite away from potential sources, representative of free troposphere conditions, greater spatial extent & background level against which the urban impacts can be compared are very few (Ramana *et al.*, 2004; Pant *et al.*, 2006a,b; Kumar *et al.*, 2012). Realizing the importance of aerosol study from a high altitude location, monthly, seasonal, pentad and diurnal variation of aerosol properties over Kodaikanal, a high altitude station in south India, using regular mode of observations obtained from hand held MICROTOS II sunphotometer for the period 2008-2015 is focused in this paper.

2. MICROTOS II sun photometer instrument measures the total columnar AOT from the direct sun radiation centered at 368, 500, 675, 778 and 1028 nm (FWHM: ± 2 -10 nm) and subtracts the Rayleigh scattering component. Ångström exponent (α) in the UV-VIS range is calculated from optical thickness values at wavelengths 368 and 675 nm. The accuracy of measurements for precision and consistency of the MICROTOS-II instruments are discussed in detail by Srivastava *et al.* (2006). MICROTOS-II instrument measured AOT values are susceptible to occasional spurious values due to filter and other components degradation, temperature effects and poor pointing at the sun. The pointing accuracy of the optical filters towards the sun, during each measurement, is very crucial for accurate measurement of atmospheric constituents (Morys *et al.*, 2001). In view of the above, extreme outliers are rejected through a simple statistical technique. Individual data points outside three times of standard deviation, on either side of long term mean for AOT₅₀₀ nm and α were excluded.

3. Kodaikanal (10°14' N, 77°28' E, 2343 m) air pollution observatory is situated in southern tip of India about 200 km from coast in south, west and east directions. Influx of marine air from the Arabian Sea and

the Bay of Bengal has a regional scale influence while biomass burning contributes at both a local and sub-regional scale. There are no heavily traveled roads nearby, nearest railways about 80 km away and no small or large scale industries, mining activity etc nearby. A small factory manufacturing thermometers is situated about 6 km away. The soil in this region is acidic in nature. Rainfall is lowest during February and highest during November with well distributed annual average rainfall as 1568 mm. Mean surface wind speeds are minimum at 6.3 kmph during September and maximum at 10.5 kmph during March. Predominant direction of surface winds at the station during post-monsoon season is north-northwesterly to north-northeasterly, in winter and pre-monsoon seasons it is northeast to southeast whereas during June to September winds are Westerly to Northwesterly. Temperature is pleasant with highest maximum, lowest minimum and annual mean maximum and minimum as 21.4 °C (May), 8.6 °C (January), 19.1 °C and 10.8 °C respectively. Afternoons are more humid than mornings especially during June to November with humidity more than 80%.

4. Despite being a pristine high altitude location, the daily mean AOT₅₀₀ for the present study period (April 2008-October 2015) was as high as 0.46 ± 0.09 with an average value of α as 1.6 ± 0.50 . Ranges of mean monthly, daily mean and individual AOT₅₀₀ for the study period were 0.77 to 0.22, 0.77 to 0.22 and 0.81 to 0.19 respectively. Corresponding values of Ångström exponent (α) were 2.23 to 0.16, 2.4 to 0.07 and 2.59 to -0.07. Ramana *et al.* (2004) have reported range of daily mean AOT₅₀₀ as 0.06 to 0.55 with mean as 0.2 to 0.34 for three high altitude sites in Nepal for winter 2003 with mean α as 1.3 to 1.4. Similarly Kumar *et al.* (2012) have reported mean AOT₄₄₀ as 0.24 with α as 1.11 for Sinhgad, a high altitude station in Maharashtra for winter season 2007. AOT₅₀₀ and α values reported in the present study are also in the same order. Above AOT and α values not only indicate significant contribution of coarse mode aerosols but also the broad range of individual α value (2.59 to -0.07) over Kodaikanal suggests that accumulation / fine mode aerosols either from local or distant sources through long range transportation are combined with coarse mode particles. Despite proximity to coast (~250 km, ~145 km and ~200 km from south, west and east coasts respectively), the high α value is attributable to alteration / transformation of pristine environment at high altitude Kodaikanal by air masses. This site is affected by north-northwest to northerly winds during post monsoon and winter seasons (Fig. 7) which are mainly from Indian landmass and were responsible for high value of α .

5. The monthly mean AOT₅₀₀ nm averaged for the period of April 2008 - October 2015 (Fig. 1) showed an

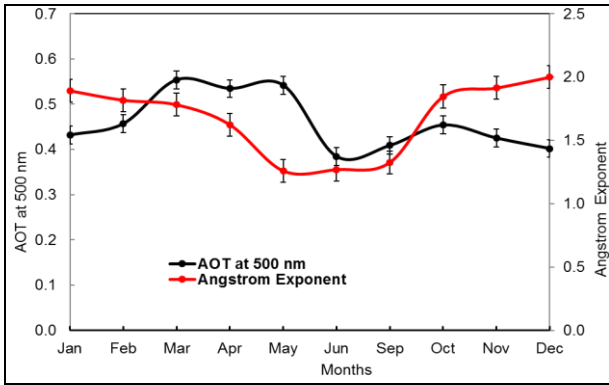
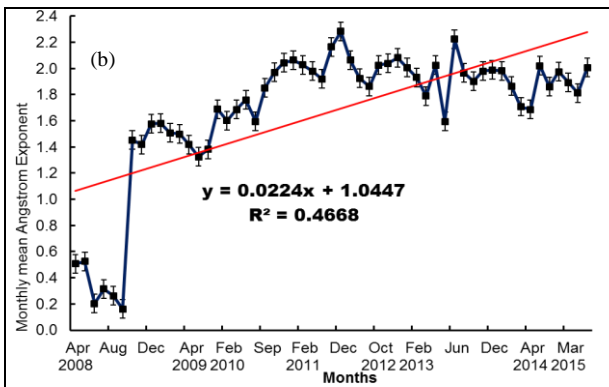
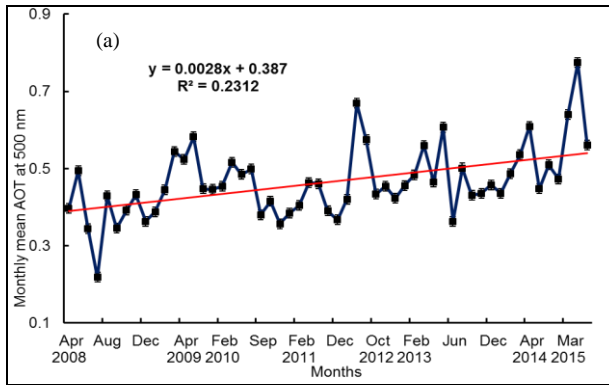
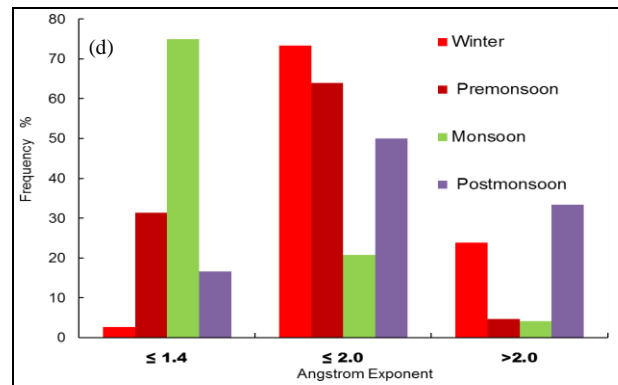
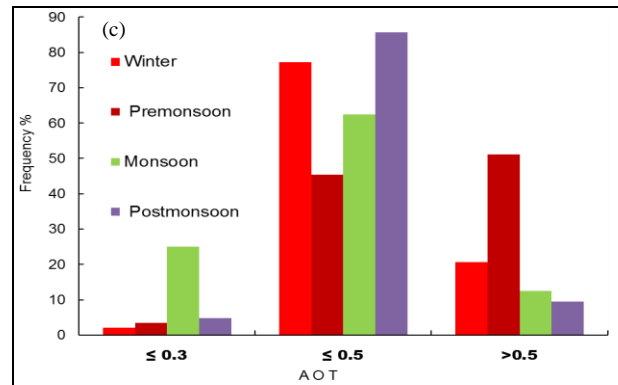
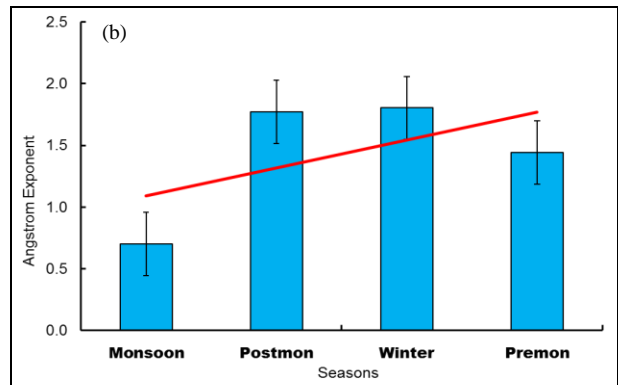
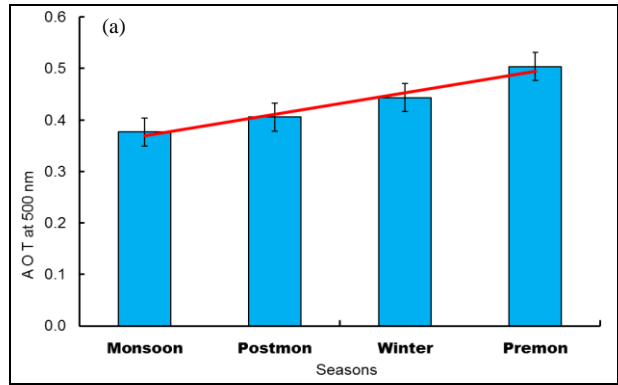


Fig. 1. Mean monthly variation of AOT₅₀₀ nm and Angstrom Exponent during 2008-15

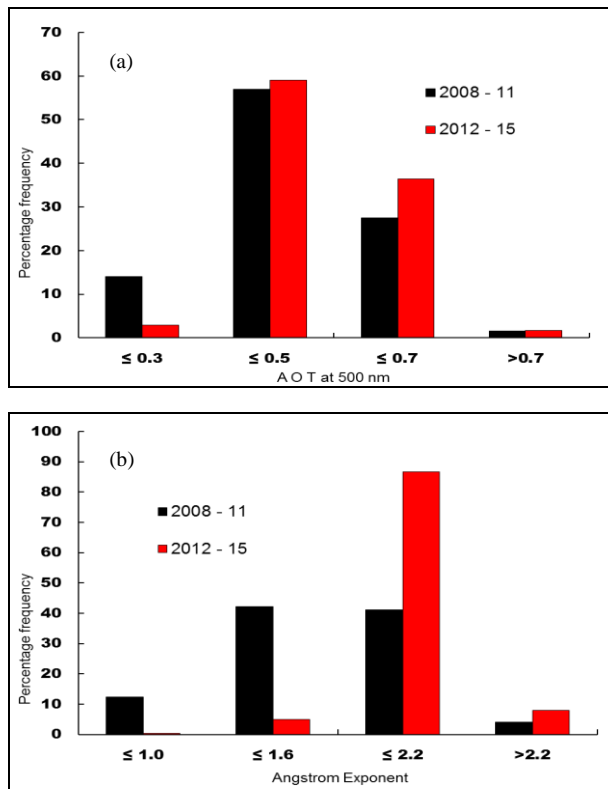


Figs.2(a&b). Monthly variation of (a) AOT₅₀₀ nm and (b) Angstrom Exponent, during 2008-15

increase from January (0.43) to March (0.55) and later on decreased till June (0.38) and again increased to reach a secondary maximum of 0.45 in the post monsoon season while the mean monthly α for the same period exhibited almost an inverse trend with decrease from January (1.89) to May (1.26) and an increase there onwards till December (2.00) in the post monsoon season. Inverse relation between AOT and α had been reported by Alam *et al.* (2012) over Lahore and Karachi; Pandithurai *et al.* (2008) over Delhi and Verma *et al.* (2013) over Jaipur.



Figs. 3(a-d). Seasonal mean of daily (a) AOT₅₀₀, (b) Angstrom Exponent (2008-2015), percentage frequency of different ranges of (c) AOT₅₀₀ and (d) Angstrom Exponent (2008-2015)



Figs. 4(a&b). Frequency of occurrence of (a) AOT and (b) Angstrom exponent in different periods

Removal of aerosols from the atmosphere due to rainout and washout and reduction in soil derived aerosols into atmosphere due to wet bare and extensive vegetative cover soils are responsible for low AOT during southwest (June-September) and northeast (November-December) monsoon seasons. The transition month, October leading to a secondary maximum. High / Low values of $AOT_{500 \text{ nm}}$ / α in March to May months can be attributed to the updraft of pollutants from foot hills regions below the observing site through the strong convective eddies, leading to increased vertical mixing that brings up the pollutants to higher levels. Occasional higher AOT that was observed may be due to the aerosol loading by the wind-blown dust particles that are originated at far off regions during intense dust storms and reaching to the elevated site through long range transport phenomenon. Low / high values of $AOT_{500 \text{ nm}}$ / α can be ascribed to prevalence of reverse meteorological conditions. Monthly mean $AOT_{500 \text{ nm}}$ and α value in the individual years are plotted in Figs. 2(a&b). Both $AOT_{500 \text{ nm}}$ and α displayed an increasing trend, indicating enhanced aerosol load over Kodaikanal with increase in anthropogenic fine mode particles in the atmosphere. This considerable variation in $AOT_{500 \text{ nm}}$ and α throughout the year is expected to show impact on the average aerosol radiation forcing during different seasons at Kodaikanal.

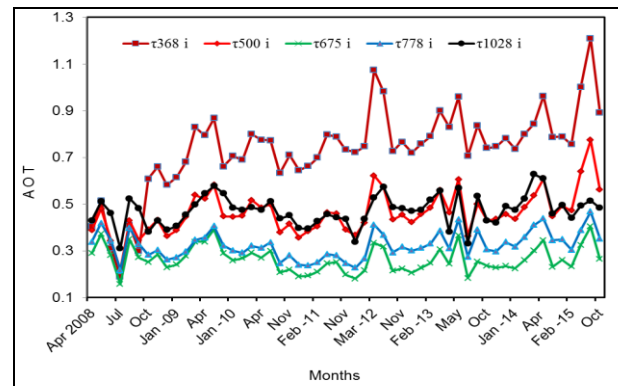
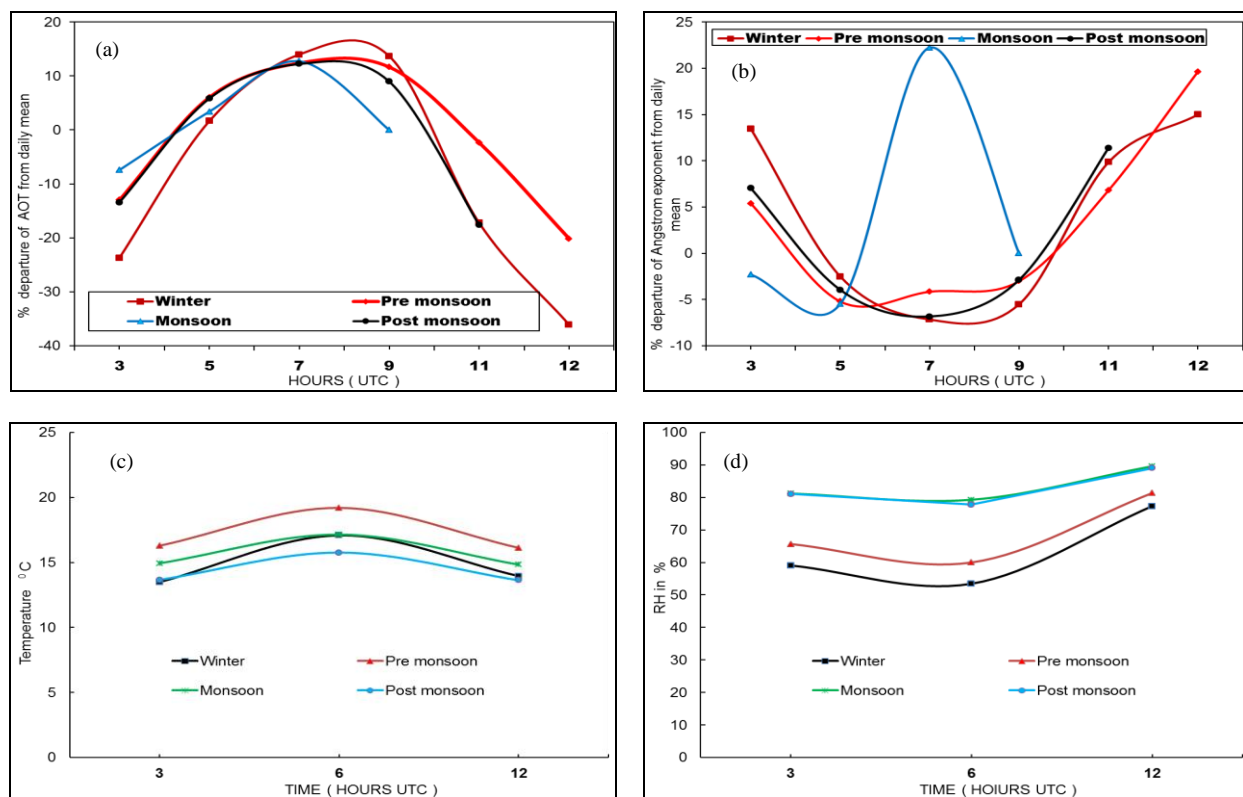


Fig. 5 Month wise spectral variation of AOT for the period 2008-2015

6. Seasonal variation of daily AOT_{500} and α is shown in Figs. 3(a-d). AOT_{500} showed increasing trend from monsoon (0.38) to pre-monsoon (0.50) season through post-monsoon (0.41) and winter (0.44) seasons [Figs. 3(a&b)] and Angstrom Exponent showed a reverse trend with a high values during post-monsoon and winter seasons and low value during pre-monsoon season [Figs. 3(c&d)]. Low Angstrom Exponent histogram in the monsoon season may due to lack of observations [Figs. 3(a&b)]. Analysis of seasonal variation of percentage frequency of AOT_{500} and α in different ranges revealed three characteristic features of aerosols over Kodaikanal. In the pre-monsoon season highest frequency of high AOT values (> 0.5) corresponded with lowest frequency of high α (2.0) indicating dominance of coarse mode aerosols. In the monsoon season, moderate frequency of moderate AOT (≤ 0.5) was associated with moderate frequency of moderate α (≤ 2.0) indicating mixed aerosols over Kodaikanal as the coarse particles get partially settled due to occasional rains. In post monsoon to winter seasons, lowest frequency of high AOT values (> 0.5) were associated with highest frequency of high values of α (2.0) [Figs. 3(c&d)] indicating dominant fine/accumulation aerosols in the atmosphere.

7. The percentage frequency of occurrence of AOT_{500} and Angstrom exponent in different class of intervals in the two half's of study period, *i.e.*, 2008-11 and 2012-15 is shown in Figs. 4(a&b). No significant difference in the aerosol loading between the two periods is noticed. A slight shift (increase in percentage frequency) of AOT_{500} from lower class (≤ 0.3) to higher class (0.5-0.7) AOT_{500} in the later half of study period was observed indicating increase trend in atmospheric aerosol loading. Whereas slight increase / decrease of percentage occasions of higher / lower class α (≥ 1.6) / (≤ 1.6) was witnessed as one moves from first to second half of study period indicating increase in fine / accumulation mode particle due to anthropogenic activity. Therefore, it can be inferred that the increasing trend in



Figs. 6(a-d). Season wise diurnal variation of (a) AOT₅₀₀, (b) Angstrom Exponent (c) Temperature and (d) Relative humidity, (2008-15)

atmospheric loading of aerosols is mainly due to anthropogenic activity.

8. Physical properties of aerosols can be ascertained through study of spectral dependence of AOT. Spectral variation of AOT at five wavelengths (368, 500, 675, 778 and 1028 nm) as obtained from MICROTOPS II observations during the study period is shown in Fig. 5 and it is seen from the figure that the spectral variation of AOT followed similar trends in all the months. AOT showed more values in all wavelengths during March to May and was less in all wavelengths during December to January. AOT values were found to be highest at 368 nm followed by 500 and 1028 nm and lowest at 675 nm.

9. Diurnal variability of aerosol optical properties has importance in atmospheric correction and validation of remote sensing data (Pawar *et al.*, 2012). It can also be used in the determination of aerosol radiative forcing and studying in the determination of interaction of aerosols with clouds and humidity (Smirnov *et al.*, 2002). Expressing the individual AOT₅₀₀ and α value in a day as a percentage difference of the daily mean and averaging the computed percentages season wise for each hour the diurnal variation of AOT₅₀₀ and α for all seasons is studied as suggested by many researchers (Smirnov *et al.*, 2002;

Wang *et al.*, 2004; Panditurai *et al.*, 2007; Pawar *et al.*, 2012). Diurnal departures of AOT₅₀₀ and α from the daily mean for four seasons is shown in Figs. 6(a&b). The AOT₅₀₀ departure was low (-7 to -24%) in the morning (0830 AM) and gradually increased to reach peak values of 12 to 14% from the daily mean by afternoon (1230 PM) and decreased thereafter in all seasons to reach lowest values of -20 to -36% [Fig. 6(a)]. Day time departures of AOT₅₀₀ are found to be positive during 1030 AM to 330 PM and negative there onwards for all seasons. The diurnal AOD discrepancy with higher AOD in the afternoon can be attributed to the vertical transport of pollutants from the near by polluted urban and foot hill regions, which were initially confined to lower heights in the night and early morning due to the low level inversions, but are released to greater heights (>2 km), as the boundary layer evolves upward. Apart from the above, increased aerosol input through lifting loose soil and other particulates and formation of clouds, caused by surface heating and vertical convective mixing over the hill top and increase in local anthropogenic activity including intermittent small-scale vehicular traffic in the day time are attributed to increase of AOT from morning to afternoon. Prevalence of reverse conditions and consequent less vertical mixing during afternoon to evening hours might have led to decrease in AOT.

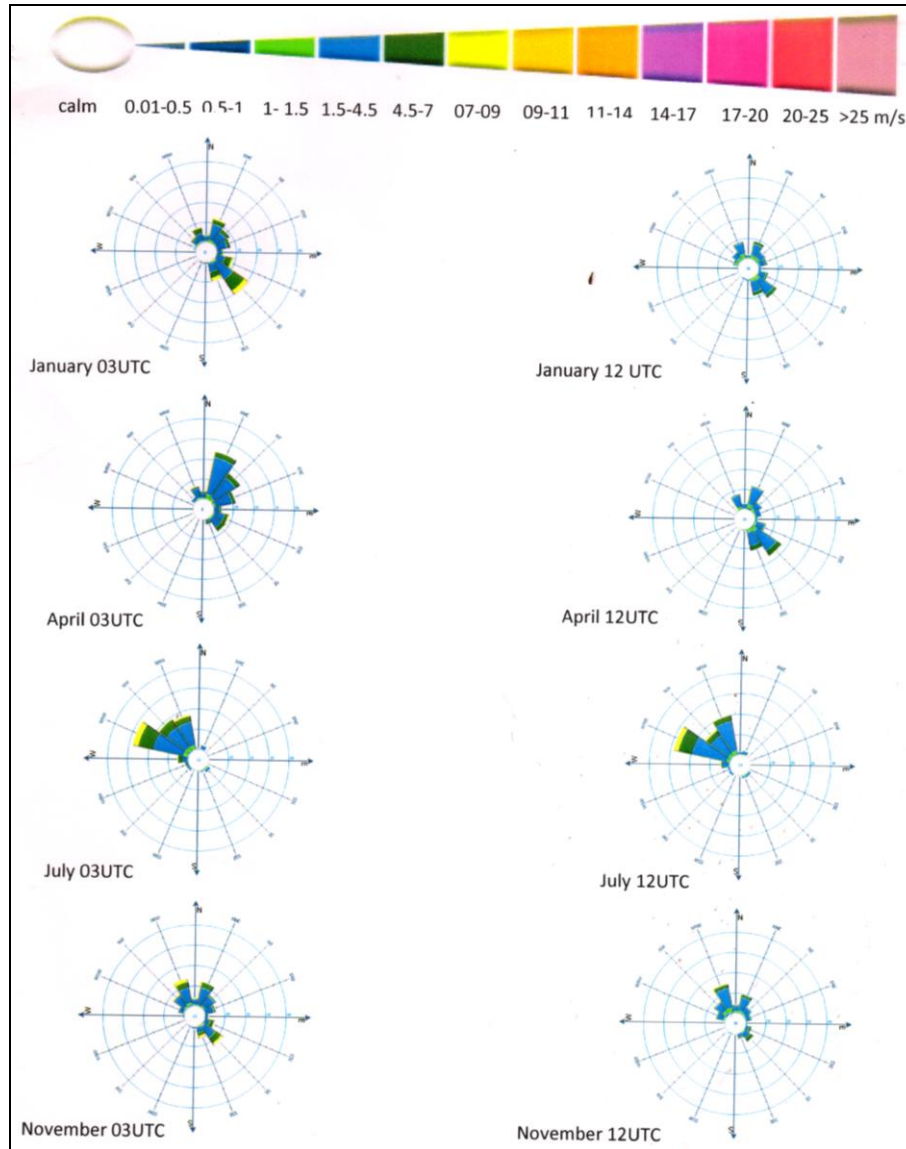


Fig. 7. Wind rose diagram (1971-2000) of Kodaikanal for representative months of winter (January), pre-monsoon (April), monsoon (July) and post-monsoon (November)

Percentage departure of α from daily mean indicated high values of +5 to 13% in the morning (0830 AM) which gradually decreased to reach lowest departures of -4 to -7% by 1230 PM and increased thereafter to attain 15 to 20% positive departure by evening during Post-monsoon, winter and pre-monsoon seasons a opposite behavior of AOT_{500} [Fig. 6(b)]. Percentage departures of α depicted a decrease from -2 to -5% during 0830 to 1030 AM. There onwards it sharply increased to reach highest value of 22% by 1230 PM and later decreased abruptly at 0230PM in the monsoon season [Fig. 6(b)]. The varying nature of diurnal departure of α between monsoon and other seasons

can be attributed to varying source regions and meteorological process between seasons (Verma *et al.*, 2013). Pawar *et al.* (2012) calculating the back trajectories of wind at various levels for winter and pre-monsoon months over Pune have shown arrival of different types of air masses from various source regions carrying continental aerosols.

10. The prevailing meteorological conditions and incursion of aerosols from different sources as depicted by the wind rose diagram (Fig. 7) together, in any season, play major role in diurnal variation of AOT in the

corresponding season. Seasonal mean air temperature, relative humidity, wind direction and speed over Kodaikanal are analyzed to study their influence on the diurnal variation of seasonal AOT. Seasonal mean air temperature and relative humidity at different hours of the day for the four seasons is shown in Figs. 6(c&d). Low air temperatures (13-16 °C) found in the morning hours rises rather rapidly to reach a peak value (16-19 °C) by 1130AM and falls gradually there onwards in all the seasons [Fig. 6(c) and a reverse trend of high relative humidity values (59-81%) observed in morning hours falling continuously to reach the lowest (53-78%) by about 1130 AM with rise (77-89%) there onwards till evening was witnessed in the case of relative humidity [Fig. 6(d)]. Wind rose diagrams constructed using the 10 minute mean wind speed for the period 1971-2000 is shown in Fig. 7. Wind is weak to moderate (3-12 knots) (20% calm) and mainly from northeast to southeast direction during winter and pre-monsoon seasons. Southeasterly winds can cause incursion of coarse mode marine aerosols from Bay of Bengal and Indian ocean. It is also weak to moderate (17% calm) and predominantly west-northwesterly during monsoon. Moderate speed winds (25% calm) chiefly from north-northwest to north-northeast occur during post-monsoon season (Fig. 7) which are mainly from land. Westerly wind during monsoon season can cause incursion of coarse mode aerosols from the desert and marine sources through long range transportation. It is conspicuous from the Fig. 6(a) and Fig. 6(c) that departures of AOT₅₀₀ tend to follow the temperature variations. Positive during hours of rise of temperature (1000 AM-1230 PM) and negative during low and fall of temperatures (before 0900 AM & after 3 PM). During nighttime, the shallow nocturnal boundary layer acts as a capping inversion (Stull, 1999), confining the emissions well below the hill top and as a result the concentration at hill top is mainly due to the residual layer (of the preceding day). After the sunrise, the heating of the land surface results in setting up of convective motions, which gradually raise the inversion to higher altitudes (Vladimir and Eischinger, 2004), leading to increased vertical mixing that brings up the air with high aerosol concentration to higher levels. This would result in the increase in the concentration of anthropogenic emissions at the peak as is observed in the present study. Even though this is the common feature in all seasons, the magnitude of departure is found to be complex leading to overlapping of seasonal departure curves. This may be due to the prevalence of meteorological factors that have contrasting influence on AOT in any season. Highest temperatures, moderate speed northeast to southeast wind (Fig. 7) conditions that can lead to higher AOT are associated with lower humidity level dry air conducive for record of lower AOT, during pre-monsoon season. Similarly highest humidity levels throughout the day are

combined with moderate to low temperatures, in different hours of the day, during monsoon and post-monsoon seasons caused complex nature of seasonal diurnal variations.

11. The daily mean AOT₅₀₀ for the period April 2008-October 2015 was 0.46 ± 0.09 and the mean Angstrom exponent was 1.6 ± 0.50 . Monthly AOT₅₀₀ and Angstrom Exponent indicated an increasing trend of both, revealing enhanced aerosol load with anthropogenic activity.

Seasonal variation of daily AOT₅₀₀ and Angstrom exponent indicated dominance of coarse, mixed and anthropogenic types of aerosols in the atmosphere during pre-monsoon, monsoon and post monsoon and winter seasons respectively over Kodaikanal. AOT₅₀₀ increased from monsoon to pre-monsoon season through post-monsoon and winter seasons and vice versa for Angstrom Exponent.

Percentage diurnal departure of AOT₅₀₀ from daily mean showed peak at 1230 PM and dip at 0530 PM in all the seasons and almost an opposite feature was observed in case of Angstrom Exponent in all except monsoon season. Angstrom Exponent decreased from morning to 1030 AM and then increased and decreased sharply between 1030 AM to 1230 PM and after 1230 PM respectively during monsoon season and this varying diurnal variations between monsoon and other seasons is attributed to change in meteorological processes and source regions.

Diurnal variation of AOT₅₀₀ tend to follow the temperature variations in all seasons but the magnitude of departure is found to vary in different seasons due to the contrasting influence of meteorological factors on AOT in any season.

Spectral variation of AOT at five wavelengths showed similar trend in all wavelengths and magnitude of AOT was more during March to May and less during December to January. AOT values were highest at 368 nm and lowest at 675 nm.

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