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Long term characteristics of aerosols over Pune, central India - Effect on radiative forcing

V. VIZAYA BHASKAR, V. K. SONI* and A. S. PANICKER**

India Meteorological Department, Pune – 411 005, India

**India Meteorological Department, New Delhi – 110 003, India*

***Indian Institute of Tropical Meteorology, Pune – 411 008, India*

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e mails : vvvbhaskar@yahoo.com

सार – पुणे और मध्य भारत के ऊपर वायुविलयों की प्रकाशीय और कालिक विशेषताओं और विकिरणित प्रणोदनों पर उनके प्रभाव का अध्ययन करने के लिए 2003-2013 तक की अवधि के सूर्य फोटोमीटर (MICROTOPS II) और सूर्य आकाश रेडियोमीटर (POM-02, Prede) आंकड़ो का उपयोग किया गया है। इस अध्ययन की अवधि में 500 nm (AOT_{500}) पर दैनिक माध्य वायु विलय की प्रकाशीय मोटाई और एंग्स्टार्म एक्सपोनेंट (α) क्रमश 0.77 ± 0.22 0.92 ± 0.24 रहा। सांता बारबरा डिस्क्रीट आर्डिनेट एटमॉस्फेयरिक रेडिएटिव ट्रांसफर (SBDART) मॉडल का उपयोग करते हुए वर्ष 2013 की लधुतरंग वायुविलय विकिरणित प्रणोदन (ARF) का आकलन किया गया। इससे यह पता चला है कि सतह (SUF) पर वायुमंडल के शीर्ष पर (TOA), ARF नकारात्मक रहा, जिससे वायुविलयों में बिखराव का पता चलता है। सूर्य आकाश से प्राप्त इकहरे रूप से फैलने वाला एल्बीडों (SSA) और एसेमिट्री पैरामीटर (ASP) अप्रैल/ मई से अगस्त तक AOT और ASP अधिक तथा SSA कम दिखाते हैं। इससे यह संकेत मिलता है कि इस अवधि में मिट्टी में धूल को सोखने की क्षमता थी। वायुमंडलीय उष्मण +14.3 से 20.5 W/m^2 तक भिन्न होती है और मानसून (+35.4 W/m^2) में और अधिक हो जाती है। SUF और ARF AOT_{500} ($R^2 = 0.90$) का क्रियान्वयन प्रबल रूप से पाया जाता है। SUF (-66.2 W/m^2/AOT) TOA (-0.2W/m^2/AOT) और वायुमंडल (ATM) (+66.0 W/m^2/AOT) पर विकिरणित प्रणोदन सक्षमता वायुविलय के फैलने की प्रकति पर निर्भर है। वायुमंडलीय उष्णन मानसूनोत्तर काल में 0.94 K/दिन से, मानसून के समय 2.78 K/दिन भिन्न होता है। AOT_{500} के मासिक मौसमी और दैनिक परिवर्तनशीलता और मौसम वैज्ञानिकघटकों पर α की भूमिका के साथ विवेचन किया गया है।

ABSTRACT. Sun photometer (MICROTOPS II) and Sun-sky radiometer (POM-02, Prede) data for 2003-2013 period are used to study the optical and temporal characteristics of aerosols and their effect on radiative forcing over Pune, central India. The daily mean aerosol optical thickness at 500 nm (AOT_{500}) and Angstrom exponent (α) for the study period were 0.77 ± 0.22 0.92 ± 0.24 respectively. Short wave aerosol radiative forcing (ARF) computed for the year 2013 using Santa Barbara Discrete-ordinate Atmospheric Radiative Transfer (SBDART) model, showed ARF at surface (SUF) and top of the atmosphere (TOA) was negative indicating dominance of scattering type aerosols. AOT, single scattering albedo (SSA) and asymmetry parameter (ASP) derived from Sun sky radiometer showed more AOT and ASP and less SSA during April / May to August indicating prevalence of absorbing soil dust in this period. Atmospheric heating varied from +14.3 to 20.5 W/m^2 and was more in monsoon (+ 35.4 W/m^2). ARF at SUF was found to be a strong function of AOT_{500} (R^2= 0.90). The radiative forcing efficiency inferred to scattering nature of aerosols at SUF (-66.2 W/m^2/ AOT), TOA (- 0.2 W/m^2 / AOT) and atmosphere (ATM) (+ 66.0 W/m^2 /AOT). The atmospheric heating rates varied from 0.94 K/day in post monsoon to 2.78 K/day in monsoon. Monthly, seasonal and diurnal variability of AOT_{500} and α with role of meteorological factors are also discussed.

Key words – Sun photometer (MICROTOPS II), AOT, AOT_{500}, Angstrom exponent, Heating rates.

1. Introduction

Aerosols albeit micron and submicron in size and constitute very small mass of the atmosphere, significantly influence the Earth's climate and radiation budget (Devara *et al*., 2005). Implications of aerosols on weather and climate are well documented. Nevertheless the real effect of aerosols on climate is one of the largest uncertainties in current global climate models and lack of extensive and reliable measurements in most regions, makes it difficult to quantify the global impact of aerosols on Earth's climate (IPCC, 2007). Aerosols modify incoming solar and outgoing longwave radiation directly through scattering and absorption and indirectly by changing the microphysical properties of clouds and their life span resulting in change in planetary albedo (Satheesh and Ramanathan, 2000; Houghton *et al*., 2001; Ramanathan *et al*., 2001). Differences in radiative fluxes at TOA (~100 km) and SUF due to aerosols are called TOA and SUF radiative forcings respectively and difference between these two is known as atmospheric radiative forcing. Magnitude and sign of ARF depend on the nature, size and chemical characteristics of the aerosols. However these effects show large variations due to short residence times of aerosols which lead to significant spatio-temporal variation in aerosol concentrations and their chemical and optical properties such as SSA, asymmetry factor etc. Therefore it emphasizes the need for widespread studies on aerosols and quantification of aerosol radiative properties and forcing because of changes in industrial practices, environmental regulation and biomass burning emissions. But reliable widespread studies with large and continuous data sets are meager over the globe.

Many studies have been undertaken to assess the impact of aerosols on the climate in the Indian region (Moorthy *et al*., 2005; Panicker *et al*., 2008, 2010; Pant *et al*., 2006; Ranjan *et al*., 2007; Srivastava *et al*., 2008; Dey and Tripathi, 2008; Pawar *et al*., 2012 ; Kumar *et al*., 2012a; Kumar *et al*., 2012b). Studies with large Sun photometer data sets (≥ 5 years) that computed ARF are few (Dey and Tripathi, 2008; Kumar *et al*., 2012b; Dani *et al*., 2012). Current investigation focused on the diurnal, monthly, seasonal and annual variation of AOT and its impact on climate through radiative forcing over Pune a tropical a station in Deccan traps of central India, for the consecutive ten year period during February, 2003 to March 2013. As such the present study assumes significance since this may serve as reference for measurement over the tropical region (Pune).

Pune is located in semiarid region close to Western Ghats in the west. Low temperatures are recorded during November to February with annual mean minimum temperature of 18 °C and December as the coldest month record mean minimum temperature of 12 °C in the night. Temperature rise rapidly after February till April and May which are usually hottest months that record mean maximum temperatures of 38 °C and 37 °C respectively with annual mean maximum temperature of 32 °C. The rainy season for Pune is during June to September when higher humidity levels (70-80%) exist. Annual precipitation is 717 mm and annual morning and evening relative humidity are 70% and 46% respectively. Summer (March- May) afternoons with 20-40% humidity are the driest. Thunderstorms occur during April-June and September-November. Winds are generally light during October-February, moderate in March-May season and increase in force (10-12 kmph) during June-September period. Station experiences marine influx through westerly to southwesterly winds from Arabian Sea during monsoon season. Aerosol loading diminishes with the onset of monsoon rains. The sources of aerosol are therefore fossil fuel burning (vehicles, incinerators), industries, biomass burning (domestic cooking and agricultural burning), oceanic source due to westerly to southwesterly winds from Arabian Sea during monsoon season and thus this region provides an excellent setting for studying the characteristics of anthropogenic and natural aerosols over the region.

2. Data and methodology

MICROTOPS II AOT data for 89 months in the ten year period of 2003-2013 and Sun-sky radiometer AOT, SSA and ASP data for the year 2013 at Pune (18° 32′ N, 74° 50′ E, 559 m) are used in the present study. Individual cloud free observations inferred from hand held MICROTOPS II Sun photometer (Solar Light Co., USA) at around 0300, 0500, 0600, 0700, 0900, 1100 & 1200 UTC are used to study the wavelength dependent optical characteristics of aerosols. The cloud free day implies that the sun was completely visible in the whole or intervals of the day and conducive for taking MICROTOPS II observations. This dataset is unique because of its relatively long-term continuous observations of AOT.

MICROTOPS 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 MICROTOPS II instruments are discussed in detail by Srivastava *et al*. (2006).

MICROTOPS 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 AOT_{500} nm and α were excluded.

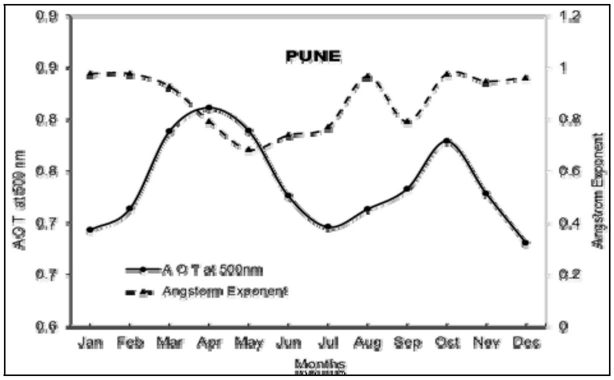


Fig. 1. Mean monthly variation of AOT_{500} nm and Angstrom exponent during 2003-13

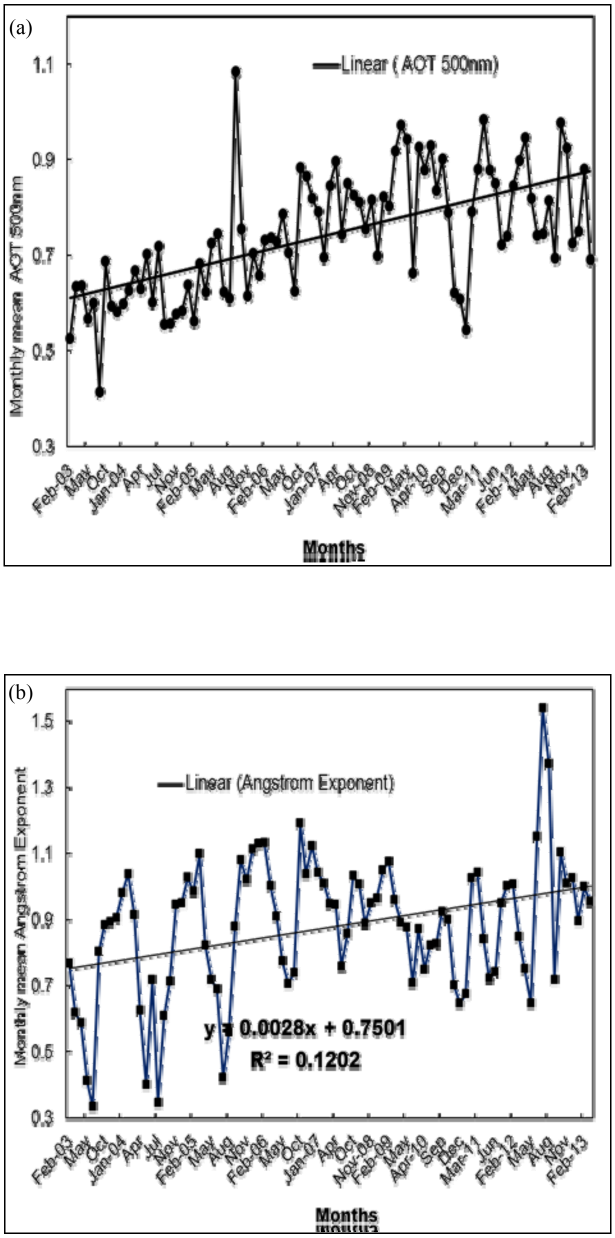
Using the individual AOT observations, daily and monthly mean AOT values have been computed. Monthly individual maximum and minimum AOT and monthly mean, maximum, minimum and standard deviation of daily AOT along with monthly mean and standard deviation of daily α for 89 months have been computed. Seasonal variation of daily AOT_{500} and α including percentage frequency in different ranges is studied. The four seasons are categorized as per India Meteorological Department convention. Monthly AOT_{500} and α were analyzed for linear trend. Spectral variation of AOT in the 368-1028 nm range and spectral variation of AOT with SSA & ASP are studied. Expressing the individual AOT_{500} 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 AOT_{500} and α for all seasons is studied. Influence of seasonal mean temperature and relative humidity at different hours and wind direction and speed on diurnal variation of seasonal AOT for four seasons is another aspect that is studied.

The station is part of Skynet-India network of India Meteorological Department. The Sun-sky radiometer (Model: POM-02 of Prede, Japan), installed at Pune is capable for measuring direct solar and diffuse sky radiance at various scattering angles from the sun at eleven spectral channels from visible to near-infrared spectral regions (315, 340, 380, 400, 500, 675, 870, 940, 1020, 1627 and 2200 nm), with a half band width of 3 nm for 340 nm wavelength and 10 nm for other wavelengths. The measured sky spectral radiances can be used to obtain different optical and size related properties of aerosols in the total atmospheric column. The aerosol optical parameters such as AOT, SSA and ASP are derived using Skyrad. Pack (version 4.2) radiative transfer code (Nakajima *et al.*, 1996). The operational details and errors associated with the instrument are already described elsewhere (Pandithurai *et al.*, 2008). ARF has been computed in the shortwave spectrum (0.2 - 4.0 μm) separately for SUF, ATM and TOA using the SBDART model and heating rates in the atmosphere are also calculated.

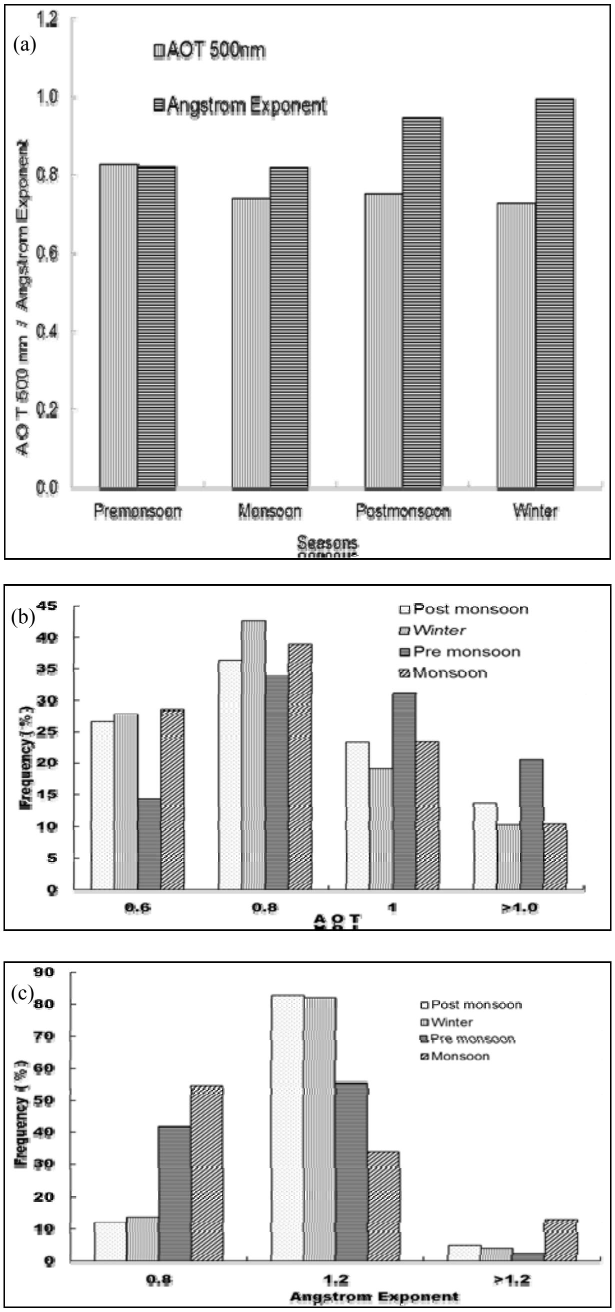
3. Results and discussion

3.1. *Monthly variation of AOT and angstrom exponent*

The daily mean AOT_{500} for the present study period (February 2003 - March 2013) was 0.77 ± 0.22 with an average value of α as 0.92 ± 0.24. Ranges of mean monthly, daily mean and individual AOT_{500} for the study period were 1.08 to 0.42, 1.72 to 0.25 and 2.17 to 0.13 respectively. Corresponding values of Angstrom exponent (α) were 1.54 to 0.33, 2.75 to 0.04 and 2.95 to -0.85. Above AOT and α values not only indicate significant contribution of coarse mode aerosols but also the broad range of individual α value (2.95 to -0.85) over Pune suggests that accumulation mode aerosols either from local or distant sources through long range transportation are combined with coarse mode particles. The monthly mean AOT_{500} nm averaged for the period of February 2003 - March 2013 (Fig. 1) showed an increase from January (0.69) to April (0.81) and later on decreased till July (0.7) and again increased to reach a secondary maximum of 0.78 in the post monsoon season while the mean monthly α for the same period exhibited almost an inverse trend with decrease from January (0.98) to May (0.68) and an increase their onwards till August (0.96) and again reduced to attain secondary minimum of 0.79 in the post monsoon season. 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 monsoon season (June-September) and the situation changes gradually leading to secondary maximum in post monsoon season. High / Low values of AOT_{500} nm / α in April and May months can be attributed to surface heating and consequent increased aerosol input because of lifting loose soil and other particulates, formation of clouds through vertical convection and occasional eastward transport of coarse mode dust aerosols from the Sahara and Thar deserts while low / high values of AOT_{500} nm / α can be ascribed to prevalence of reverse meteorological conditions. Inverse relation between AOT and α has been reported by Alam *et al.* (2012) over Lahore and Karachi; Pandithurai *et al.* (2008) over Delhi and Verma *et al.* (2013) over Jaipur. Monthly mean AOT_{500} nm and α value in the individual



Figs. 2(a&b). Monthly variation of (a) AOT_{500} nm during 2003-13 and (b) Angstrom exponent during 2003-13

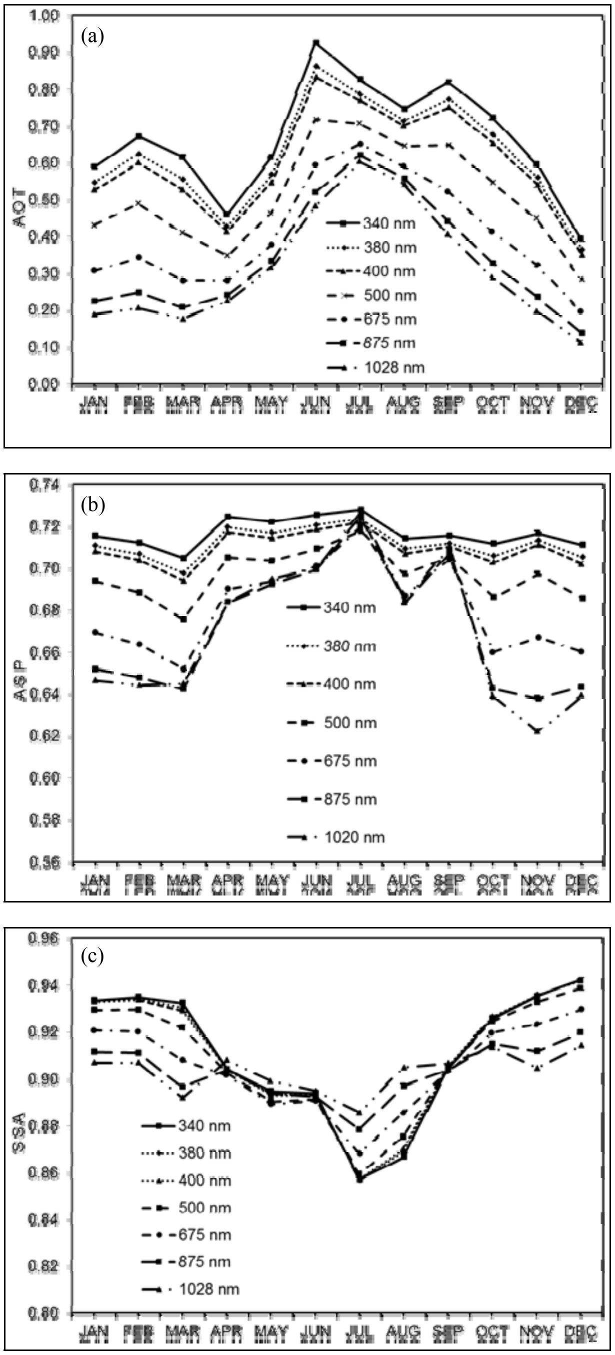


Figs. 3(a-c). (a) Seasonal mean of daily AOT_{500} and Angstrom exponent (2003-2013), (b) Percentage frequency of different ranges of AOT_{500} and (c) Angstrom exponent in different seasons

years are plotted in Figs. 2(a&b). Both AOT_{500} nm and α displayed an increasing trend, indicating enhanced aerosol load over Pune with increase in anthropogenic fine mode particles in the atmosphere. This considerable variation in AOT_{500} nm and α throughout the year is expected to show impact on the average aerosol radiation forcing during different seasons at Pune.

3.2. *Seasonal variation of AOT_{500} and Angstrom exponent*

Seasonal variation of daily AOT_{500} and α is shown in Figs. 3(a-c). AOT_{500} showed decreasing trend from pre-monsoon to winter season with exception of occurrence of secondary peak during post monsoon season and



Figs. 4(a-c). Month wise spectral variation of Sun sky radiometer derived (a) AOT for the year 2013, (b) ASP for the year 2013 and (c) SSA for the year 2013

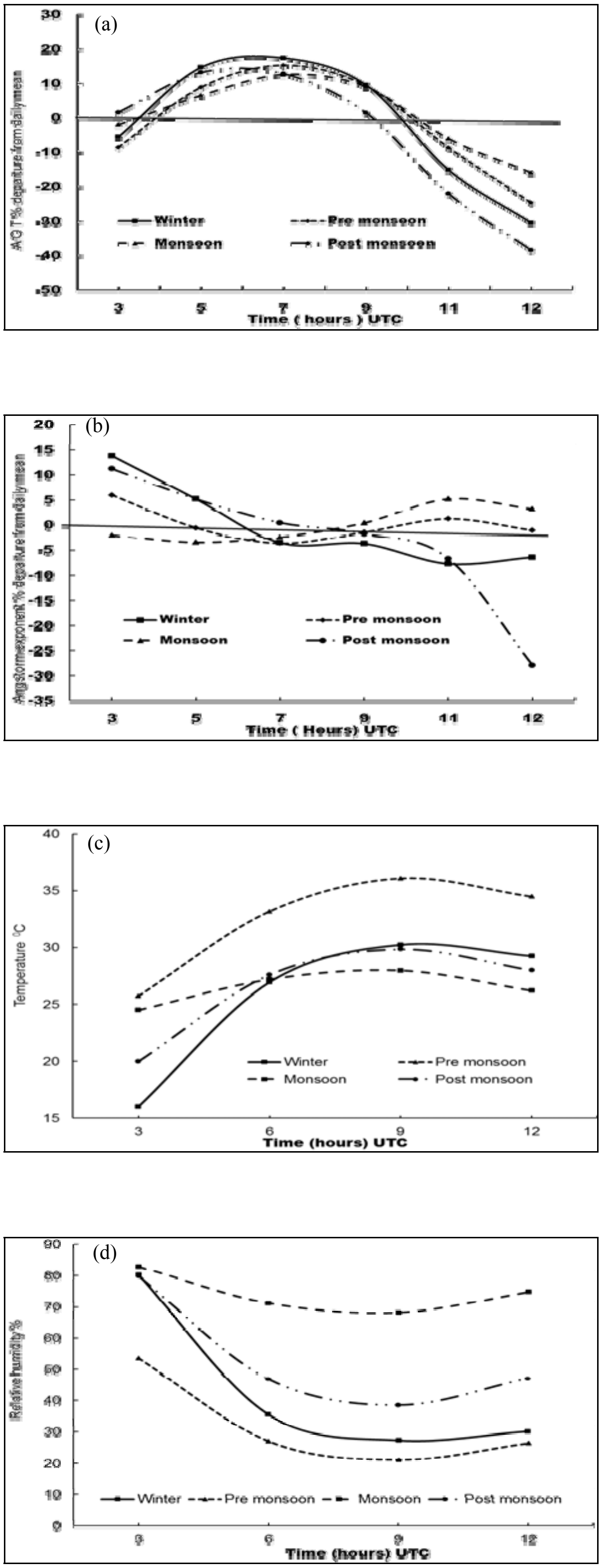
Angstrom exponent showed a reverse trend with a slight compromise during monsoon season [Fig. 3(a)]. Analysis of seasonal variation of percentage frequency of AOT$_{500}$ and α in different ranges revealed three characteristic features of aerosols over Pune. In post monsoon to winter seasons, highest frequency of low AOT values (0.6 to 0.8) were associated with highest frequency of high values of α (1.2) [Figs. 3(b&c)] indicating dominant anthropogenic aerosols in the atmosphere. In the pre-monsoon season highest frequency of high AOT values (1 & > 1.0) corresponded with lowest frequency of high α (1.2) indicating dominance of coarse mode aerosols. In the monsoon season, moderate frequency of high AOT was associated with moderate frequency of high α indicating mixed aerosols over Pune as the coarse particles get partially settled due to occasional rains. Ranjan *et al.* (2007) have also reported enhanced AOT values during pre-monsoon months over Rajkot which lies in the south of Thar desert.

3.3. *Spectral variation of AOT, SSA and ASP*

Physical properties of aerosols can be ascertained through study of spectral dependence of AOT. Spectral variation of AOT, single scattering albedo and asymmetry parameter at seven wavelengths (340, 380, 400, 500, 675, 870 and 1020 nm) as obtained from Sun-sky radiometer observations during the year 2013 is shown in Figs. 4(a-c) and it is seen from the figures that the respective spectral variation of all these optical parameters followed similar trends in all the months. AOT and ASP showed more values [Figs.4(a&b)] and SSA [Fig. 4(c)] was less in all wavelengths during April / May to August. At the same time, AOT and ASP were less whereas SSA was more in all wavelengths during September / October to March / April. A secondary maximum in AOT and ASP was observed during September. As an illustration, the average values of AOT$_{500}$, ASP and SSA were 0.633, 0.707 and 0.884 respectively during April / May to August; but these values were 0.422, 0.688 and 0.926 during September / October to March / April [Figs.4(a-c)]. Less AOT and ASP values along with high SSA indicates towards the more presence of scattering type aerosols like sulphate, non absorbing dust and sea salt, especially during post-monsoon through winter period (September / October to March / April). More AOT and ASP values and low SSA during pre-monsoon and early monsoon (April / May to August) signifies the presence of more absorbing type aerosols such as soil dust. Angstrom exponent showed higher values during September to March (1.041) than during April to August (0.523) which shows more prevalence of coarse size aerosols during pre monsoon to early monsoon seasons. Similar results have been reported by Ranjan *et al.* (2007) and kumar *et al.* (2012b).

3.4. *Percentage diurnal departure of AOT$_{500}$ and Angstrom exponent*

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



Figs. 5(a-d). Season wise diurnal variation of (a) AOT_{500}, (b) Angstrom exponent (2003-13), (c) Temperature and (d) Relative humidity (2003-13)

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 AOT_{500} 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 AOT_{500} 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 AOT_{500} and α from the daily mean for four seasons is shown in Figs. 5(a&b). The AOT_{500} departure was low (-9 to+2%) in the morning (0830am) and gradually increased to reach peak values of 13 to 17% from the daily mean by afternoon (1230pm) and decreased thereafter in all seasons to reach lowest values of -16 to -38% [Fig. 5(a)]. Day time departures of AOT_{500} are found to be positive during 0900am to 3pm and negative there onwards for all seasons. Increased aerosol input through lifting loose soil and other particulates and formation of clouds, caused by surface heating and vertical convective mixing and increase in local anthropogenic activity are attributed to increase of AOT from morning to afternoon. Reduction on surface heating and consequent less vertical mixing during afternoon to evening hours might have led to decrease in AOT. Percentage departure of α from daily mean indicated high values of -2 to +4% in the morning (0830am) which gradually decreased to reach lowest departures of -4 to 1% by noon and increased thereafter to attain 1 to 3% positive departure by evening during pre-monsoon and monsoon seasons a opposite behavior of AOT_{500} [Fig. 5(b)]. Percentage departures of α depicted a decrease throughout the day from high values of 11 to 14% in the morning to reach lowest values of -6 to -28% by evening during post monsoon and winter seasons [Fig. 5(b)]. The varying nature of diurnal departure of α between winter and pre monsoon months 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.

3.5. *Role of meteorological parameters on AOT variability*

The prevailing meteorological conditions and incursion of aerosols from different sources as depicted by the wind rose diagram (Fig. 6) 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 Pune are analyzed to study their influence on the diurnal variation of seasonal AOT. Seasonal mean air temperature and

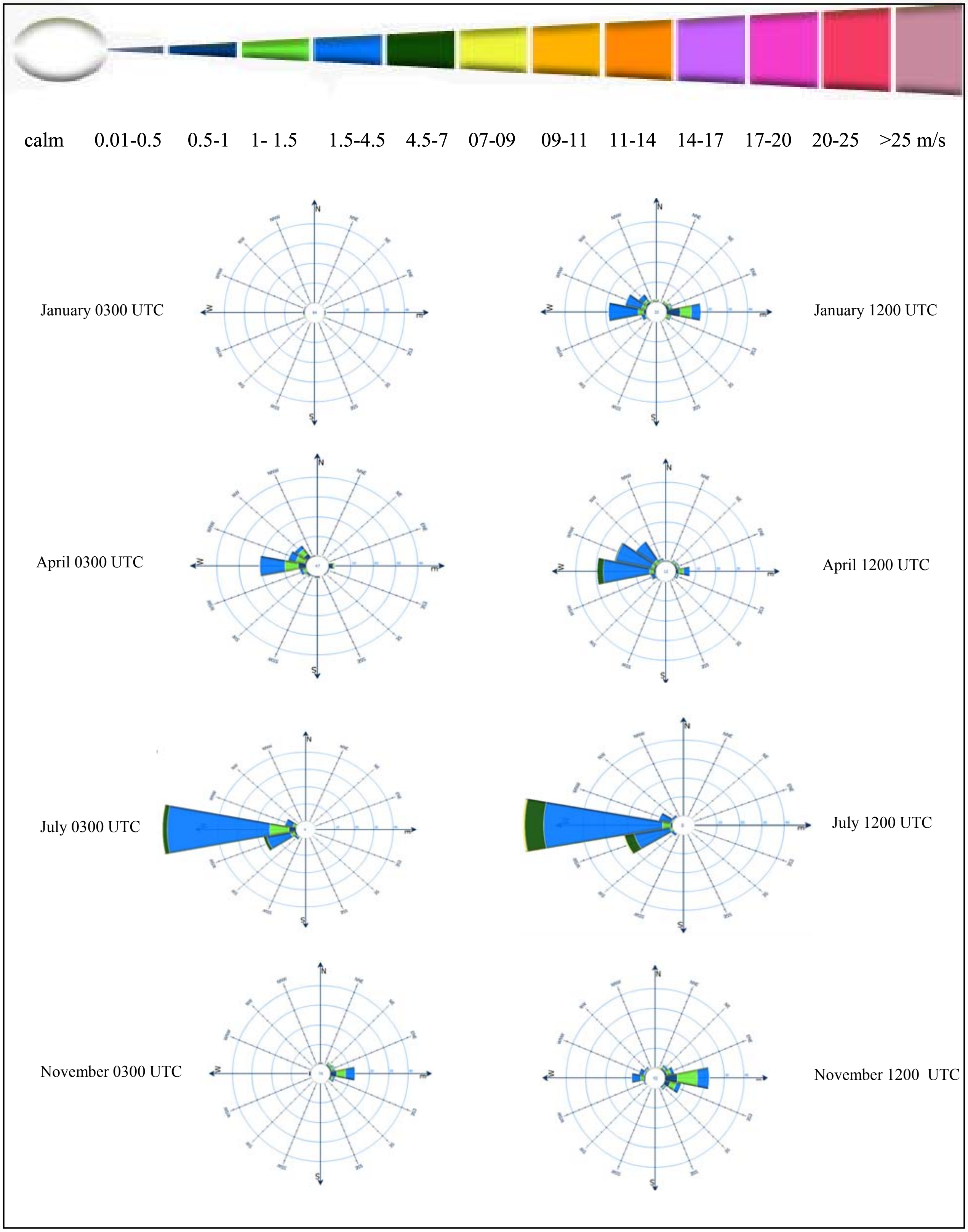


Fig. 6. Wind rose diagram (1970-2000) of Pune for representative months of winter (January), pre-monsoon (April), monsoon (July) and post-monsoon (November) seasons

relative humidity at different hours of the day for the four seasons is shown in Figs. 5(c-d). Low air temperatures (16-26 °C) found in the morning hours rises rather rapidly to reach a peak value (28-36 °C) by 0230pm and falls gradually there onwards in all the seasons [Fig. 5(c)] and a reverse trend of high relative humidity values (54-83%) observed in morning hours falling continuously to reach the lowest (21-68%) by about 0230pm with slight rise (26-75%) there onwards till evening was witnessed in the case of relative humidity [Fig. 5(d)]. Wind rose diagrams constructed using the 10 minute mean wind speed for the period 1970-2000 is shown in Fig. 6. Wind is calm (94%) during winter and light (74% calm) and mainly from east during post monsoon season. It is moderate (47% calm) and predominantly west to northwest during pre-monsoon and highest speed (only 4% calm) winds chiefly from west to southwest occur during monsoon season (Fig. 6). Westerly wind during monsoon and pre-monsoon seasons can cause incursion of coarse mode dust aerosols from the desert regions through long range transportation. It is conspicuous from the Fig. 5(a) and Fig. 5(c) that departures of AOT_{500} tend to follow the temperature variations. Positive during hours of rise of temperature (0900am - 0230pm) and negative during low and fall of temperatures (before 0900am and after 3pm). 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 particularly during 0830 to 0300pm. This may be due to the prevalence of meteorological factors that have contrasting influence on AOT in any season. Highest temperatures, moderate speed west to northwesterly wind (Fig. 6) conditions that can lead to higher AOT are associated with lowest humidity level driest 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 season and caused complex nature of seasonal diurnal variations.

3.6. *Comparison of MICROTOPS II and Sun-sky radiometer*

The AOT_{500} values obtained from the Sun-sky radiometer (Model POM-02, Prede Inc.) for the period from May 2011 to February 2013 have been compared with AOT_{500} values derived from MICROTOPS II sun photometer. In the 464 instantaneous AOT observations that are compared, the AOT values of MICROTOPS II were found to be higher than Sun Sky Radiometer values in all but only four cases and in 322 (70%) cases the difference was within 0.5. The maximum and minimum difference values were 1.36 and 0.011 respectively. The scatter plot shown in Fig. 7 indicates a correlation of 0.83 at 5% level of significance, suggesting that aerosol measurements with MICROTOP II and Sun-sky radiometer are in good agreement.

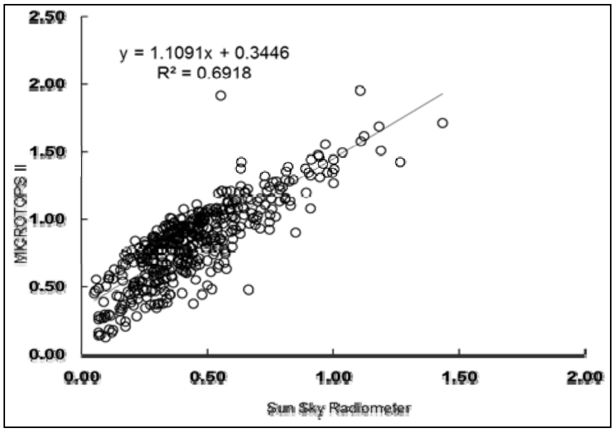
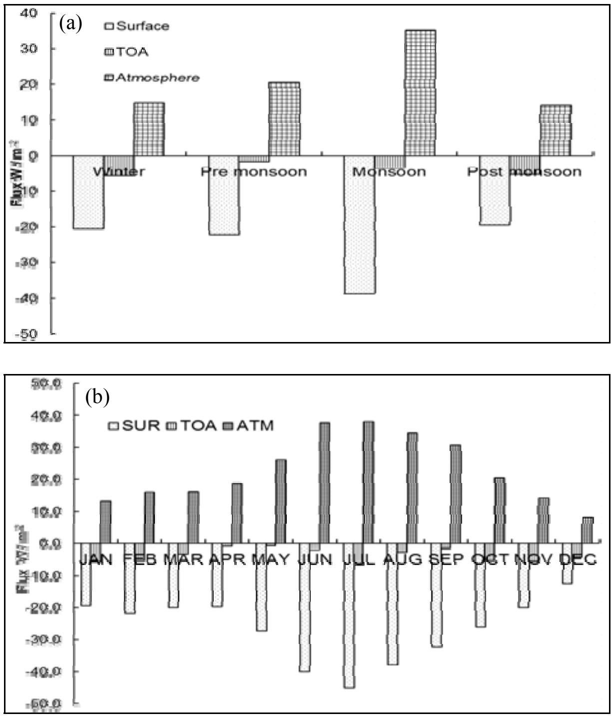


Fig. 7. Scatter plot of MICROTOPS & Sun Sky Radiometer AOT at 500 nm

3.7. *Short wave aerosol radiative forcing and heating rates*

Santa Barbara Discrete-Ordinate Atmospheric Radiative Transfer (SBDART) model developed at University of California (Ricchiazzi *et al.*, 1998) has been used to estimate the shortwave direct aerosol radiative forcing at top of the atmosphere, surface and atmosphere. SBDART model is a radiative transfer code based on Discrete- Ordinate method for a plane parallel and vertically inhomogeneous atmosphere for addressing a wide range of radiative transfer problems in remote sensing and atmospheric energy budget studies. The model computes plane-parallel radiative transfer calculations both in clear and cloudy sky conditions within the Earth's atmosphere and at the surface. ARF calculations are computed in the 0.2 - 4.0 μm shortwave spectrum range. Several studies have extensively used this model for ARF calculations over India (Pandithurai *et al.*, 2008; Singh *et al.*, 2010; Srivastava *et al.*, 2014). The ARF is strongly dependent upon spectral AOT, SSA, ASP as well as surface albedo and meteorological conditions. For the estimation of shortwave ARF, along with the observed spectral AOT, SSA and ASP at 500 nm; satellite derived O_3 and water vapor from OMI / AURA level 2 and MODIS level 3 respectively were used as inputs for the radiative transfer model. These input parameters were used to run the model for every change of 5° solar zenith angles initially from 0° to 89° once with aerosol condition and another without aerosol condition. Hence the output of the radiative transfer model is the monthly average of solar flux at TOA, SUF and ATM. Default tropical model



Figs. 8(a&b). Variation of SW ARF at TOA, SUF and ATM during year 2013 (a) Seasonal and (b) Monthly

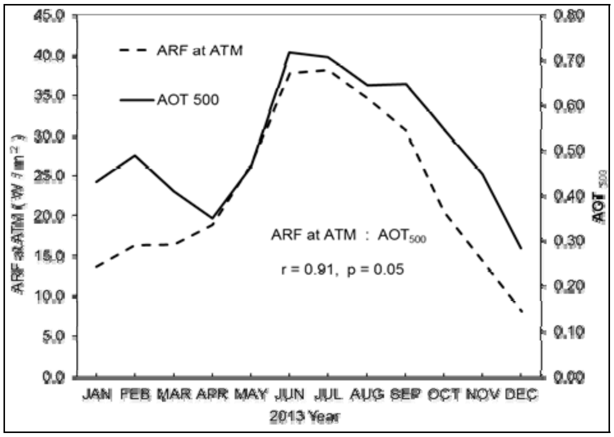


Fig. 9. Monthly variation of ARF at ATM and AOT at 500 nm during year 2013

and vegetative albedo also has been used in the model for estimating the forcing.

The uncertainties in shortwave ARF calculations could arise from various assumptions, such as model atmosphere as well as uncertainties in surface albedo, molecular scattering absorption, and errors in measured optical parameters. The overall uncertainty in ARF calculations does not exceed 20% (Pant *et al.*, 2006). The seasonally averaged values of ARF at TOA, SUF, and ATM were estimated for the year 2013.

As seen from Fig. 8(a), the SUF and TOA forcing was negative during all the seasons indicating dominance of scattering type aerosols. However, the negative forcing at TOA in pre-monsoon was comparatively less which denotes possible presence of absorbing dust particles as well as impact of combustion activities in the local surroundings. High wind speed and occasional long range transport of dust from the west during the pre-monsoon relative to other seasons cause higher loading of dust aerosols into the atmosphere during the pre-monsoon season. The dust is rich in carbonate and bicarbonate and these acts as absorption. The ATM forcing was positive in all the seasons indicating heating of the atmosphere and it is highest during monsoon. The SW ARF respectively for TOA, SUF and ATM over Pune was -5.57, -20.51 and 14.94 W/m^2 during winter; -1.72, -22.21 and 20.49 W/m^2 during pre-monsoon; -3.29, -38.65 and 35.35 W/m^2 during monsoon and -5.18, -19.50 and 14.32 W/m^2 during post-monsoon. Fig. 8(b) shows the monthly variation of SW ARF at TOA, SUF and ATM over Pune during the study period. It can be seen that ARF at ATM was more than double during June-September (mean 35.5 W/m^2) than that during rest of the year (mean 14.3 W/m^2).

Variation of monthly mean AOT$_{500}$ nm closely followed the variation of monthly mean ARF at ATM as seen from Fig. 9 and both showed significantly good correlation with each other (R^2 = 0.84, p = 0.05). Similarly, AOT$_{500}$ nm correlated well with ARF at SUF (R^2 = 0.90). A high degree of correlation between ARF and AOT indicates that ARF is a strong function of AOT. The slope of the linear regressions defines the aerosol forcing efficiency that is the rate of forcing per unit AOT and represents the effectiveness of the composite aerosols in perturbing the radiative balance (Santos *et al.*, 2008). In the present study, the radiative forcing efficiencies were found to be -0.2, -66.2 and +66 W/m^2 AOT^{-1} for TOA, SUF and ATM, respectively. The radiative forcing efficiency indicates scattering nature of aerosols at SUF and TOA which indicates cooling at surface and top of the atmosphere whereas, there is warming of the atmosphere in between.

Kumar *et al.* (2012b) in the long term study of aerosol modulation of atmospheric and surface solar heating over Pune reported SUF, TOA and ATM short wave direct radiative forcing (SWDRF) values in different years during 2004-2009 ranging between -28.68 to

-45.13, -8.88 to -2.67, 19.81 to 42.46 for post-monsoon; -31.02 to -41.46, -3.58 to -.8.88 and 24.95 to 36.92 for winter; -32.51 to -56.23, -4.83 to -10.56 and 24.83 to 51.24 W/m^2 for pre-monsoon seasons while the heating rates ranged from 0.56 to 1.19, 0.70 to 1.04 and 0.7 to 1.44 K/day for above order seasons. Pawar *et al.* (2012) while studying the SW ADRF for two locations over Pune during March-April, 2011 have reported SWDRF for different days ranging between -17.3 to -54.2 and -15.1 to -36.6 W/m^2. Kumar *et al.* (2012a) have reported SWDRF over Pune for SUR, TOA and ATM for December 2006 and April 2007 as -43, -6 and 35 W/m^2 for December -59, -6 and 55 W/m^2 for April months

Heating rates

The average heating rate of the atmosphere due to aerosols is defined as :

$$\partial T / \partial t = (g / C_p) * (\Delta F_{atm} / \Delta P)$$

where, $\partial T/\partial t$ is the heating rate (K/day), Cp is the specific heat capacity of air at constant pressure, g is the acceleration due to gravity and ΔP is the atmospheric pressure difference between top and bottom boundary of each layer (Liou, 2002). The mean heating rates during pre-monsoon, monsoon, post-monsoon and winter seasons were 1.43, 2.78, 0.94 and 0.97 K/day, respectively. Heating rates showed good positive correlation with AOT during all the seasons ($R^2 = 0.79$, $p = 0.0001$) whereas Angstrom exponent showed significant negative correlation with heating rates ($R^2 = 0.59$, $p = 0.003$). This feature indicates towards impact of coarse size aerosols mainly dust particles on the warming of the atmosphere over Pune.

4. Conclusions

Present study examined the aerosol optical characteristics using the MICROTOPS II Sun photometer observations for the years 2003 to 2013 and Sun-sky radiometer observations for the year 2013 over Pune. Also MICROTOPS II data was cross checked with Sun Sky Radiometer data. The main findings of the investigation are:

The daily mean AOT_{500} for the period February 2003 to March 2013 was 0.77 ± 0.22 and the mean Angstrom exponent was 0.92 ± 0.24.

Seasonal variation of daily AOT_{500} and Angstrom exponent showed three characteristic features. In post monsoon to winter seasons highest frequency of low values of AOT_{500} were associated with highest frequencies of high values of Angstrom exponent indicating dominant anthropogenic aerosols. In the pre-monsoon season, highest frequency of high AOT_{500} values corresponded with lowest frequencies of high Angstrom exponent showing dominance of coarse mode soil dust aerosols. In the monsoon season moderate frequency of high AOT_{500} was associated with moderate frequency of high Angstrom exponent indicating mixed aerosols over Pune which is attributed to scavenging of coarse particles due to downpour.

Percentage diurnal departure of AOT_{500} from daily mean showed peak at 1230pm and dip at 0530pm in all the seasons which is attributed mainly to lifting loose soil and other particulates and formation of clouds all caused by surface heating and vertical convective mixing and increase in local anthropogenic activity from morning till noon and decrease thereafter. Percentage diurnal departure of α from daily mean exhibited highest value at about 0800am and reached lowest by noon and again increased thereafter till 0530pm for pre-monsoon and monsoon seasons a reverse behavior of AOT variation. But for post-monsoon and winter seasons it decreased throughout the day from morning high positive departures. The varying diurnal variations between winter and pre-monsoon months can be attributed to change in meteorological processes and source regions.

Diurnal variation of AOT_{500} tend to follow the temperature variations in all seasons but the magnitude of departure is found to be complex due to the prevalence of meteorological factors that have contrasting influence on AOT in any season.

Comparison of AOT_{500} observations between MICROTOP II and Sun-sky radiometer for the period May 2011 to February 2013 indicated correlation of 0.83 suggesting a good agreement.

Spectral variation of AOT, SSA and ASP at seven wavelengths derived from Sun-sky radiometer showed similar trend in all wavelengths. High AOT and ASP and low SSA values were witnessed in all wavelengths during April/May to August and vice versa during September/ October to March/April. High SSA at higher wavelengths indicates towards the presence of scattering type particles like sulphate, dust and sea salt, especially during September/October to March /April and low SSA values during April / May to August signifies presence of more absorbing type particles such as black carbon and hematite dust.

The ARF at SUF and TOA was negative during all the seasons indicating dominance of scattering type aerosols. However, the negative forcing at TOA in pre-monsoon was comparatively less which denotes possible presence of absorbing dust particles as well as impact of combustion activities in the local surroundings. The ATM forcing was positive in all the seasons indicating heating of the atmosphere and it is more during monsoon. ARF at ATM was more than double during June to September than that in rest of the year. A high degree of correlation for ARF at ATM and SUF with AOT indicates that ARF is a strong function of AOT. The radiative forcing efficiency indicates scattering nature of aerosols at SUF and TOA which indicates cooling at surface and top of the atmosphere whereas, there is warming of the atmosphere in between.

The mean heating rates during pre-monsoon, monsoon, post-monsoon and winter seasons were 1.43, 2.78, 0.94 and 0.97 K/day, respectively. Heating rates showed good positive correlation with AOT during all the seasons (R^2 = 0.79, p = 0.0001) whereas Angstrom exponent showed significant negative correlation with heating rates (R^2 = 0.59, p = 0.003).

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References

Alam, K., Trautmann, T., Blaschke, T. and Majid, H., 2012, "Aerosol optical and radiative properties during summer and winter season over Lahore and Karachi", *Atmospheric Environment*, **50**, 234-245.

Devara, P. C. S., Saha, S. K., Raj, P. E., Sonbawne, S. M., Dani, K. K., Tiwari, Y. K. and Maheskumar, R. S., 2005, "Four year climatology of total column tropical urban aerosol, ozone and water vapor distributions, over Pune, India", *Aerosol Air Qual. Res.*, **5**, 107-120.

Dani, K. K., Raj, P. E., Devara, P. C. S., Pandithurai, G., Sonbawne, S. M., Maheskumar, R. S., Saha, S. K. and Jaya Rao, Y., 2012, "Long-term trends and variability in measured multi-spectral aerosol optical depth over a tropical urban station in India", *International Journal of Climatology*, **32**, DOI: 10.1002/joc.2250, 153-160.

Dey, S. and Tripathi, S. N., 2008, "Aerosol direct radiative effects over Kanpur in the Indo-Gangetic basin, northern India : Long-term (2001-2005) observations and implications to regional climate", *J. Geophys. Res.*, **113**, D04212. DOI: 10.1029/2007JD009029.

Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., Van der Linden, P. J., Dai, X., Maskell, K. and Johnson, C. A., 2001, "Climate Change 2001: The Scientific Basis", Cambridge University Press, Cambridge, UK.

Intergovernmental Panel on Climate Change (IPCC), 2007, "Climate change 2007 : The physical science basis: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change", Chapter 2, p129.

Kumar, Sumit, Devara, P. C. S. and Manoj, M. G., 2012a, "Multi-site characterization of tropical aerosols: Implications for regional radiative forcing", *Atmospheric Research*, **10**6, 71-85.

Kumar, Sumit and Devara, P. C. S., 2012b, "A long term study of aerosol modulation of atmospheric and surface solar heating over Pune", *Tellus B*, ***64***, doi.org/10.3402/tellusb.v64i0.18420

Liou, K. N., 2002, "An Introduction to Atmospheric Radiation", Academic Press, San Diego.

Moorthy, K. K., Babu, S. S. and Satheesh, S. K., 2005, "Aerosol characteristics and radiative impacts over the Arabian Sea during the inter-monsoon season: results from ARMEX field campaign", *Journal of the Atmospheric Sciences*, **62**, 1, 192-206.

Morys, M., Mims III, F. M., Hagerup, S., Anderson, S. E., Baker, A., Kia, J. and Walkup, T., 2001, "Design, Calibration, and Performance of MICROTOPS II hand-held Ozone Monitor and Sun Photometer", *J. Geophys. Res.*, **106**, 14573-14582.

Nakajima, T., Tonna, G., Rao, R., Boi, P., Kaufman, Y. and Holben, B. N., 1996, "Use of sky brightness measurements from ground for remote sensing of particulate poly dispersions", *Appl. Opt.*, **35**, 2672-2786.

Pandithurai, G., Pinker, R. T., Devara, P. C. S., Takamura, T. and Dani, K. K., 2007, "Seasonal Asymmetry in Diurnal Variation of Aerosol Optical Characteristics over Pune Western India", *J. Geophys. Res.*, **112**, D08208, doi:10.1029/2006JD007803.

Pandithurai, G., Dipu, S., Dani, K. K., Tiwari, S., Bisht, D. S., Devara, P. C. S. and Pinker, R. T., 2008, "Aerosol radiative forcing during dust events over New Delhi, India", *J. Geophys. Res.*, **113**, D13209, doi:10.1029/2008JD009804.

Panicker, A. S., Pandithurai, G., Safai, P. D. and Kewat, S., 2008, "Observations of enhanced aerosol longwave radiative forcing over an urban environment", *Geophysical Research Letters*, **35**, L04817, doi: 10.1029/2007GL032879.

Panicker, A. S., Pandithurai, G. and Dipu, S., 2010, "Aerosol indirect effect during Successive contrasting monsoon years over Indian sub continent: using MODIS data", *Atmospheric Environment*, **44**, 1937-1943, doi:10.1016/j.atmosenv.2010.02.015.

Pant, P., Hedge, P., Dumka, U.C., Ram Sagar., Satheesh, S.K. and Krishna Moorthy, K., 2006, "Aerosol characteristics at high altitude location in central Himalayas : Optical properties and radiative forcing", *Journal of Geophysical Research*, **111**, Article ID D17206, 2006.

Pawar, G. V., Devara, P. C. S., More, S. D., Pradeep Kumar, P. and Aher, G. R., 2012, "Determination of aerosol characteristics and direct radiative forcing at Pune", *Aerosol Air Qual. Res.*, **12**, 1166-1180.

Ramanathan, V., Crutzen, P. J. and Lelievld, J., 2001, "Indian Ocean Experiment: An Integrated Analysis of the Climate Forcing and Effects of the Great Indo-Asian Haze", *J. Geophys. Res.*, **106**, 28371-28398.

Ranjan, R. R., Nandita D Ganguly., Joshi, H. P. and Iyer, K. N., 2007, "Study of aerosol depth and precipitable water vapour content at Rajkot, a tropical semi-arid station", *Indian Journal of Radio & Space Physics*, **36**, 27-32.

Ricchiazzi, P., Yang, S., Gautier, C. and Sowle, D., 1998, "SBDART : a research and teaching software tool for plane parallel radiative transfer in the Earth's atmosphere", *Bull. Am. Meteorol. Soc.*, **79**, 2101-2114.

Santos, D., Costa, M. J. and Silva, A. M., 2008, "Direct SW aerosol radiative forcing over Portugal", *Atmospheric Chemistry and Physics*, **8**, 19, 5771-5786.

Satheesh, S. K. and Ramanathan, V., 2000, "Large Differences in the Tropical Aerosol Forcing at the Top of the Atmosphere and Earth's Surface", *Nature*, **405**, 60-63.

Singh, S., Soni, K., Banoi, T., Tanwar, R. S., Nath, S. and Arya, B. C., 2010, "Clear-sky direct aerosol radiative forcing variations over mega-city Delhi", *Ann. Geophys*., **28**, 1157-1166, doi:10.1029/2005GL023062.

Smirnov, A., Holben, B. N., Eck, T. F., Slutsker, I., Chatenet, B. and Pinker, R. T., 2002, "Diurnal Variability of Aerosol Optical Depth Observed at AERONET Sites", *Geophys. Res. Lett.*, **29**, 2115, doi: 10.1029/2002GL016305.

Srivastava, M. K., Singh, S., Saha, A., Dumka, U. C., Hegde, P., Singh, R. and Pant, P., 2006, "Direct solar ultraviolet irradiance over Nainital, India, in the central Himalayas for clear-sky day conditions during December 2004", *J. Geophys. Res*., **111**, D08201, doi:10.1029/2005JD006141.

Srivastava, A. K., Devara, P. C. S., Jaya Rao, Y., Bhavanikumar, Y. and Rao, D. N., 2008, "Aerosol optical depth, ozone and water vapor measurements over Gadanki, A Tropical Station in Peninsular India", *Aerosol and Air Quality Research*, **8**, 4, 459-476.

Srivastava, A. K., Soni, V. K., Singh, S., Kanawade, V. P., Singh, N., Tiwari, S. and Attri, S. D., 2014, "An early South Asian dust storm during March 2012 and its impacts on Indian Himalayan foothills : A case study", *Science of the Total Environment*, **493**, 526-534.

Verma, S., Payra, S., Gautam, R., Prakash, D., Soni, M., Holben, B. and Bell, B., 2013, "Dust events and their influence on aerosol optical properties over Jaipur in northwestern India", *Environ Monit Assess*, DOI 10.1007/s10661-013-3103-9.

Wang, J., Xia, X., Wang, P. and Christopher, S. A., 2004, "Diurnal variability of dust aerosol optical thickness and Angstrom exponent over dust source regions in China", *Geophysical Research Letters*, **31**, L08107, doi: 10.1029/2004GL019580.