Derivation of vertical aerosol profiles in lower stratosphere and upper troposphere using twilight photometry

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A photometer has been designed and developed for observation of the zenith sky illumination during the twilight period to monitor the vertical profile of aerosols. A red gelatin filter centered at 700 nm (50 nm half bandwidth) and RCA 6199 photomultiplier tube were selected for the photometer. The experimental set-up and method of data analysis to derive vertical profile of atmospheric aerosols have been presented. The observations were carried out at tropical location Pathardi (19° 9'N, 75° 10'E) during the period 1 Jan. 1993 - 31 Dec. 1994. The intensity parameter, \( -(\phi h' d\phi) \), was determined and its changes attributed to the variation in the structure of the atmosphere resulted due to changes in aerosol density. An algorithm is developed for the estimation of vertical profile of aerosols in terms of number density. The estimated vertical profile of aerosols compares well with other methods of observations.

1 Introduction

Atmospheric aerosols play an important role in radiation balance, global climate change and biogeochemical cycles of trace elements. They can affect radiative transfer directly by absorbing and scattering incoming and outgoing radiation and indirectly by influencing cloud microphysical and radiative properties and cloud lifetime. The twilight brightness measurement is an inexpensive but sensitive method to study the atmospheric aerosols and its detection along with tracing its progress through the atmosphere. The importance of twilight observations in detecting vertical discontinuities in the light intensity scattered by aerosols and molecules has been reported by various workers, and any change in the concentration of these species at the shadow height will alter the scattering intensity during the twilight period. The presence of a layer of particulate material in the lower stratosphere was deduced many years ago from twilight observations of purple light. The vertical aerosol profiles of the lower stratosphere can be obtained from twilight polarization measurements. The presence of the layer was directly verified with particle sampling equipment carried on high altitude balloons. The existence of the aerosol layer is an established fact. However, the role of aerosols in the interactions between stratosphere, troposphere and ground has not been fully elucidated. The continuous observations of aerosols vertical profile at this continent are scanty. Hence, it was decided to operate the twilight photometer at least for two consecutive years. An attempt is made, in this paper, to examine the association between the vertical profiles derived by this method and its comparison with others.

2 Twilight photometer

A convex lens of 20 cm diameter with focal length of 30 cm is used as telescopic lens. The red gelatin filter (peaking at 700 nm with 50 nm half bandwidth) of 2 cm diameter and an aperture of 0.5 cm diameter is placed at the focus of the convex lens. The aperture of 0.5 cm diameter provides approximately 1° field of view. Three different baffles at distance of 8, 16 and 24 cm from the focal plane of the lens are used to avoid internal scattering in the photometer. The photomultiplier tube (RCA6199) is placed at a distance of 1 cm from the filter. The gelatin filter is a wide bandwidth filter; hence a collimated beam is not essential. An amplifier (AD515) with variable gain of \( 10^2 \cdot 10^3 \) is used to provide the output signal. A change in gain is achieved by changing feedback resistance with a variable rotary switch. The above filter was selected to avoid absorption due to ozone. Detector
'look' angle of an order of few minutes has been used by some workers. However, the present photometer was used at 1° look angle in order to enhance the observable twilight radiation. For repeatability and convenience of operation, all the observations were carried out with the photometer pointed towards the zenith sky. The output of the photomultiplier was fed to a stable sensitive amplifier and the voltage was measured with digital multimeter. The twilight intensity observations were recorded manually at 30 s intervals using a digital multimeter during the time interval of sunset/sunrise to the time when the sun was at 7.3° below the horizon. The observation times were converted to the solar zenith angle, and from solar zenith angle, shadow heights were calculated. The clock was adjusted up to second using Indian Standard Time (IST). The measurement location is Pathardi (19° 9'N, 75° 10' E) in Maharashtra State of India.

The photometer's response function is derived from the transmission function of the filter used and the quantum efficiency curve of the photomultiplier tube. The overlapping of two functions is the response function of the photometer. As both the functions are at falling edge of response, the response function is poor. However, as the light gathering power of photometer is high, the measurable output signal is observed at the output of the photometer. The output of the photometer for observation during twilight period may be given as,

\[ \text{Output of photometer} = L \times QT \times EBP \times GPT \times E \times Sf \]

where,

\[ L = \text{Light gathering power of the photometer} = \frac{AT \times AA}{F^2} \]

\[ AT = \text{Area of telescopic lens} = \pi \times (\text{radius of lens})^2 = 3.14 \times (10 \text{ cm})^2 = 314 \text{ cm}^2 \]

\[ AA = \text{Area of aperture} = \pi \times (\text{radius of aperture})^2 = 3.14 \times (0.25 \text{ cm})^2 = 0.1962 \text{ cm}^2 \]

\[ F = \text{Focal length of convex lens} = 30 \text{ cm} \]

Therefore,

\[ L = \frac{314 \times 0.1962}{(30)^2} = 0.06845 \text{ cm}^2 \]

\[ QT = \text{Quantum efficiency of the photometer} \times \text{Transmission of filter} \]

The QT function is calculated for photometer which is in the order of $6 \times 10^{-3}$, when quantum efficiency is considered in percentage. However, in calculation, the absolute quantum efficiency is to be considered whose maximum value is 1. Hence, for calculation purpose QT is in the order of $6 \times 10^{-5}$ for this photometer.

\[ EBP = \text{The effective bandwidth of the photometer} \]

The filter is a wide bandwidth one. However, the combination of photomultiplier tube response curve and filter transmission is effective for photometer function. The effective bandwidth of this function is 300 Å (30 nm) for this photometer.

\[ GPT = \text{Gain of photomultiplier tube} \]

The maximum gain of photomultiplier tube is $5 \times 10^5$ at supply voltage of 1000 V. This is considered in calculations. However, during observations the supply voltage of photomultiplier tube was varied from 400 V to 1000 V. Therefore, the gain of the photometer was varied by $10^4$ by changing photomultiplier voltage.

\[ E = \text{Electronic charge in coulomb} = 1.6 \times 10^{-19} \text{ C} \]

\[ SI = \text{Sky intensity} = 5000 \text{ kR/Å} (\text{kR = Kilo Rayleigh}) \text{ to } 0.5 \text{ kR/Å} = \frac{5000 \times 10^9}{2\pi} \text{ photons/cm}^2/\text{str/s}/\text{s/Å} \text{ to } 0.5 \times 10^7/2\pi \text{ photons/cm}^2/\text{str/s}/\text{s/Å} \]

During evening time, sky intensity is 5000 kR/Å and at about 98° solar zenith angle during twilight, sky intensity is 0.5 kR/Å (Ref. 14).

The output current of the photometer varies from $10^{-4}$ A at the sunset to $10^{-8}$ A at 98° solar zenith angle. During observations it is dangerous to draw $10^{-4}$ A current through photomultiplier. Hence, during operation of photometer the supply voltage of photomultiplier was adjusted in such a way that the output current of photomultiplier was in the range of $10^{-4}$-$10^{-6}$ A. Also apertures of different diameters were used in such a way that the output current of photomultiplier was in this range.
3 Data collected at Pathardi

Pathardi (19°9'N, 75°10'E) is situated on Deccan Plateau, at an altitude of about 550 m in rural area. The population of Pathardi is approximately 25,000. The weather at Pathardi resembles that of Marathawada region. The station is free from industrial pollution.

3.1 The photometric observations of part of the zenith sky during twilight period

The photometric observations of part of the sky illumination during twilight period were carried out during the period 1 Jan. 1993-31 Dec. 1994 at the location Pathardi. The photometric observations were made under clear sky conditions only. Changes of illumination during the shielding of the sun by distant cloud masses, mountains, or by humid or hazy air are possible sources of erratic twilight curves, which are rejected in this experiment. The location is completely free from mountains and humid and hazy air in most of the days during the year. While taking observations, precautions are taken to avoid errors due to various parameters such as detector stability, recording of Indian Standard Time, etc. In each month a minimum of four and maximum of twelve twilight observation sets are available. There were a total of 185 twilight sets, out of which 124 were morning and 61 evening sets.

4 Method of data analysis

The data collected at the station Pathardi during 1 Jan. 1993-31 Dec. 1994 were used for the analysis. The methods of data analysis are also discussed.

4.1 Height of the earth’s shadow

The earth’s geometrical shadow height is defined as the vertical height, from the surface of the earth, of a point where the solar ray grazing the surface of the earth meets the line-of-sight. The geometry of aerosol layer surrounding the earth, illuminated by the solar radiation from the horizon \( H \) up to zenith \( Z \), as seen by an observer on the earth’s surface at \( O \) is shown in Fig. 1 (Ref. 15). In Fig. 1, \( C \) is the centre of earth, \( R \) the radius of earth, \( O \) is the point of observation, \( H \) is horizon, \( B \) is a point where sun ray passing near to the earth, \( Z \) is zenith, \( h \) is earth’s shadow height and \( \Delta \) is the sun’s depression. Using simple geometry, we can write

\[
h = R (\sec(\Delta) - 1)
\]

... (2)

The heights so derived may need:

(i) Corrections for refraction of the sun’s rays through the atmosphere. This is equivalent to reducing the solar depression by about half a degree\(^6\). Shah\(^6\) assumes the total deviation by refraction, for a tangential ray at a height of 6 km, to be 0.6 deg.

(ii) Due to the long air-path traversed by the grazing rays, the incident light will suffer attenuation in passing through the atmosphere owing to molecular scattering and the attenuation will depend on the wavelength. Moreover, the particles of dust and of condensed water which exist in varying quantities in the troposphere will add to further attenuation. So, some portion of the earth’s atmosphere will be almost opaque to the grazing rays and a correction has to be applied to the geometrical earth’s shadow height in order to get the effective height of the earth’s shadow. This is known as screening height. It is wavelength dependent, decreasing with increasing wavelength. The atmospheric dust including the Junge layer at 18-20 km causes a great increase in the intensity of red light at 20 km and below. Bigg\(^4\) assumed that if longer wavelengths are used, the earth’s shadow height will correspond to the geometrical height of the lowest grazing rays.
Megrlishvili\(^3\) assumed that due to extinction of the grazing rays during sunset, the least height accessible for investigation would be about 20 km and effective height of the base of the scattering layer would be \(h + 20\), where \(h\) is the geometrical shadow height without refraction. Volz and Goody\(^7\) have given argument to show that in a normal turbid atmosphere, the twilight comes from above 10 km for red light.

The calculation, by Shah\(^6\), of the intensity of primary scattered light in the sun's meridian plane at an angle of 70° from the zenith and from different levels of atmosphere when the centre of the sun is 1° below the horizon, shows that the maximum scattered light in the blue comes from a height of about 15 km and, in red, the maximum primary scattered light comes from a height of about 6 km above the surface of the earth. He assumes that the screening height for red light has to be 6 km.

When corrections (i) and (ii) are applied to Eq. (2), then we have:

\[
h = (R + h_s) \sec(\theta') - R \quad \ldots (3)
\]

where, \(h_s\) is the screening height and \(\theta' = \Delta - \bar{\omega}\).

In the present work screening height \(h_s\) has been assumed to be 6 km for red twilight, and \(\bar{\omega} = 0.5°\) is the total deviation by refraction for tangential ray at a height of 6 km.

Figure 2 provides curves for obtaining the height of an illuminated dust layer near sunrise and sunset. The solid curve gives the information about the height of illuminated layer with respect to the depression of the sun below the horizon when dust layer is illuminated to zenith without corrections [by Eq. (2)]. The dotted curve gives the information about height of illuminated layer with respect to the depression of the sun below the horizon when dust layer is illuminated to zenith with corrections [by Eq. (3)].

4.2 Method of raw data analysis

The effective height of earth shadow was calculated by Eq. (3). The shadow heights were computed for zenith sky observation, and the raw data were utilized for the analysis of \(-(\Delta h)^{-1}\) curve, where \(I\) is the observed intensity, \(dl\) and \(dh\) are the differences in intensities and shadow heights, respectively, observed at time \(t\) and \((t+dt)\). In this method, ratio of \(I\) and \(dl\) is taken which eliminates the unit of \(I\) measurement. During the twilight period, absolute measurement of intensity variations with time requires accurate and rapid readings to reveal change in the slope which are very small. A better approach would be the one emphasizing the discontinuities by instrumental differentiation of the brightness with respect to time as suggested by Bigg\(^4\).

Now,

\[
\frac{1}{I} \frac{dl}{dh} = - \frac{d \ln(I)}{dh} \quad \ldots (4)
\]

The effect of the Rayleigh scattering component on the value of \(-(\Delta h)^{-1}\) has been studied by Bigg\(^4\). Though the vertical profile of the molecular density varies exponentially with height, its effect on \(-(\Delta h)^{-1}\) curve is nearly constant and it does not give any structure on \(-(\Delta h)^{-1}\) curve. Hence, only irregular variations in \(-(\Delta h)^{-1}\) attributed to the particle number density. The lowest 6 km of the atmosphere does not allow the direct solar radiations during twilight period. The actual observation also shows major part of irregular variations. In this method the aerosol diameter which is comparable with wavelength of observations is only sensitive to this method. Hence, if there is change in aerosols size distribution, it will not be detected by this method. This is the case with single wavelength lidar system also. Thus, the intensity \(I\) can be assumed to be directly proportional to the particle number density, and so the twilight intensity measurement which determines the value of \(\ln\) (aerosol number density) may be used for obtaining the height profile of atmospheric aerosols. Thus,

\[
-dl(\Delta h)^{-1} = -\frac{d \ln\text{[aerosol number density]}}{dh} \quad \ldots (5)
\]
5 Result and discussion

To study the atmospheric aerosol particles and its detection along with tracing its progress through the atmosphere by twilight method, the observations of the rate of change of zenith sky illumination during the twilight period were carried out at tropical location Pathardi for continuous two years period, i.e. 1 Jan. 1993-31 Dec. 1994.

The photometer was operated during clear sky conditions. Other investigators\(^7,13,18-20\) used this technique for the event studies only; so, the annual cycle of variation is masked in their observations. Some interesting results obtained by using our continuous observations are discussed along with the physical interpretation wherever possible.

5.1 Intensity variations

The zenith sky intensities have been measured during twilight period to derive vertical profile of aerosol in the atmosphere. Figure 3 shows the plot of the log of intensity \(I\) (in arbitrary unit) versus solar depression angles observed in the present work and by Shah\(^19\) for comparison. The characteristic bend in the intensity curve near the solar depression angle \(-3^\circ-5^\circ\) is observed which is due to the changing of brightness of scattered sunlight. Figure 3 shows the similar trend in the intensity variation for both the curves. The logarithmic scale is used to draw this curve, hence small fluctuations in \((d\theta/dl)^{-1}\) curve are not reflected here as will be seen later in Figs. 5 and 6. This indicates that the method adopted here is in right direction. Some observers\(^7,21,22\) who have carried out photometry of the twilight sky, have been interested only in the brightness variation with respect to solar depression angle. Ljungahl\(^2\) attempted absolute measurements and interpreted results only in terms of molecular component. Several workers\(^3-7,19\) have specifically sought the dust component, but without measuring absolute intensities. In the present study, absolute calibration could not be attempted as the ratio of intensities is used in the analysis procedure. The trend of the observed intensities of sky during the twilight period is found to be the same as observed by other workers.

5.2 Vertical profile of \(-(d\theta/dl)^{-1}\)

The values of \(-(d\theta/dl)^{-1}\) are computed from observed value of \(I\) and \(dl\) at various solar zenith angles as explained in Sec. 4.2. Figure 4 shows the plots of \(-(d\theta/dl)^{-1}\) against shadow height \((h)\) for different selected day numbers. Day numbers 1-730 represent the dates between the observational period 1 Jan. 1993-31 Dec. 1994. The magnified views of the plots of \(-(d\theta/dl)^{-1}\) against shadow height \((h)\) for some selected observational days are shown in Fig. 5((a) and (b)).

It is noted from Figs 4 and 5 that the value of \(-(d\theta/dl)^{-1}\) decreases in the beginning which implies that the concentration of particles declines with height. The subsequent increase in the value of \(-(d\theta/dl)^{-1}\) is due to the rising of shadow across the dust layer. Again the value \(-(d\theta/dl)^{-1}\) decreases, which indicates a decrease in the concentration of particles with shadow height. The value of \(-(d\theta/dl)^{-1}\) is equivalent to the inverse of smoothed scale height of the actual atmosphere (dust scale height superimposed on molecular scale height) and, therefore, its vertical distribution will represent vertical distribution of aerosols in the atmosphere.

Figure 4 shows variation in the vertical distribution of aerosols for some selected days. Figure 4 is drawn by smoothing the small variations as seen in Figs. 5 and 6. It is found that the general trend of vertical
distribution of aerosol remains the same. There is close relation between morning and evening twilight 
\(-\langle d h \rangle^{-1} d l \) curves, as these are stratospheric observations and less evening-to-morning difference is expected. Similar type of vertical distribution of atmospheric aerosol had been reported by others \(^4,6,7,19\) using twilight photometric measurements. The vertical profiles obtained by Chagnon and Junge \(^12\) based on the direct measurements of large particles (\(0.1 < r < 10 \mu m\)) also show a similar structure of the atmosphere. A rapid decrease in concentration is noticed in the lower troposphere followed by a fairly constant concentration in the upper troposphere, and then, the aerosol layer in the stratosphere between 15 km and 25 km representing the dust layer.

In the present work, the observations were recorded during clear sky conditions. The effect of seasonal variation in solar radiations and their angular
distribution were included in the computations, since solar zenith angles were used to deduce the shadow height. The results of vertical distribution of atmospheric aerosol agree well with the results of Bigg\textsuperscript{15}, Shah\textsuperscript{6,19} and Jadhav and Londhe\textsuperscript{7}. The photometric observations of part of the sky illumination during twilight were made by Jadhav and Londhe\textsuperscript{7}. They determined the intensity parameter \( -\langle l\delta h\rangle^{-1}dI \) and concluded that the change in intensity parameter is due to variation in the atmospheric aerosol density. The optical evidence in favour of aerosol scattering near 20 km has also been presented by many others\textsuperscript{13,18,23,28}.

Though the existence of the aerosol layer is an established fact, the role of aerosols in the interactions between stratosphere, troposphere and ground has not been fully explored. The origin of the stratospheric aerosol during volcanic quiescence is not exactly known\textsuperscript{29}. There is a common source of sulphur compounds for the stratosphere in both the northern and southern hemisphere. The major precursor gas for the stratospheric aerosol is sulphur dioxide which comes mostly from volcanic activities. However, it also has a possibility of biogenic or anthropogenic origin. Carbonyl sulphide, an oxidation product of carbonyl disulphide with an atmospheric lifetime of about 1 year is another candidate for stratospheric sulphur aerosol\textsuperscript{30}. In the present work, an attempt has been made to examine vertical distribution of aerosols using inexpensive and sensitive twilight sky brightness measurement over the location Pathardi. The discontinuities in the slopes of intensity were observed at a shadow height of 13-28 km below the observer's horizon. These discontinuities were clearly observed as maxima on the curve of \( -\langle l\delta h\rangle^{-1}dI \) against \( h \). This implies that there is an existence of aerosol layer. The discontinuities in the intensity or maxima of curve \( -\langle l\delta h\rangle^{-1}dI \) against \( h \) are present throughout the period of observations over Pathardi.

5.3 Aerosol layer

The heights of the layer of aerosol maximum on measurement days were derived from vertical profiles of \( -\langle l\delta h\rangle^{-1}dI \). It is noticed from Fig. 6 that the mid-point of the aerosol layer was found to be moving either downward or upward. The mid-point of aerosol layer was found to be moving downward with average speed of the order of 6 km per month during the advent of the monsoon season, and found to be moving upward with average speed of the order of 2 km per month during the withdrawal phase of monsoon. The average height of aerosol layer maxima is found to be 20 km. This implies the existence of persistent dust layer at the height of about 20 km (Ref. 6) over the tropical location Pathardi. That means, the maximum concentrations of aerosols occur, on the average, at about 20 km and these are associated with maxima on the logarithmic gradient of intensity. In both the years (1993, 1994) lowest average height of
5.4 The aerosol number density

The variation in intensity parameter \(-\frac{dI}{dt}\) can be assumed mainly due to changes in aerosol density. However, with reasonable accuracy we may assume that the intensity \(I\) is directly proportional to particle number density, as lowest 6 km of atmosphere where maximum molecular density locates is opaque during the twilight period, i.e.,

\[
-\frac{1}{dI} = \frac{d\ln I}{dh} = -\frac{d\ln(n)}{dh}
\]

where, \(n\) is the aerosol number density. It may be possible to convert \(-\frac{dI}{dt}\) to aerosol number density per cm\(^3\) by integrating intensity parameter with respect to \(dh\) and multiplying it by appropriate factor. Using this concept an attempt has been made to derive vertical profile of aerosol number density by using the empirical relations. In order to derive number density profile the intensity parameter \(-\frac{dI}{dt}\) is multiplied by the factor 10 and antilog of this quantity has been taken, which is found comparable with vertical profile obtained by other workers. Figure 6 shows a height profile of intensity parameter and aerosol number density (per cm\(^3\)). It is noticed from Fig. 6(b) that the aerosol number density decreases from 100 particles per cm\(^3\) at 6 km to about 5 particles per cm\(^3\) at 9 km. The aerosol layer peak occurs at 17 km having concentration of the order of 9 particles per cm\(^3\). The aerosol concentration in stratosphere varies from 9 particle per cm\(^3\) at \(\sim 15\) km to 1 particle per cm\(^3\) at \(\sim 50\) km. Ramachandran et al.\(^{31}\) have reported that the particles concentration over Hyderabad, in volcanically quiescent period (22 Oct. 1985), varies from 80 particles per cm\(^3\) (at 6 km) to 9 particles per cm\(^3\) (at 32 km). The aerosol number density obtained using present approach is supporting the results of Ramachandran et al.\(^{31}\). Simultaneous observations are needed to arrive at the realistic estimates of aerosol number density. The present estimates, however, are very promising.

6 Conclusions

An analysis of twilight sky intensity measurement made over location Pathardi using twilight photometer presented in this work suggests the following:

(i) The twilight scattering methods which use less expensive observational set-up, yield a reasonable, qualitative picture of the vertical distribution of atmospheric aerosols.

(ii) The aerosol layer maxima shows downward drifting before cloudy days and upward drifting after cloudy days, and was found to be moving downward with average speed of 6 km per month during the advent of the monsoon season, and found to be moving upward with average speed of 2 km per month during the withdrawal phase of the monsoon.

(iii) The maximum concentrations of aerosols occur, on an average, at about 20 km and these are associated with maxima on the logarithmic gradient of intensity. The lowest average height of about 13 km was found in July and the highest of about 28 km in March.

(iv) The annual variation of aerosol number density is comparable with the annual variations derived by lidar method. This less expensive method of observations may be useful to study the vertical profile of aerosol number density on regular basis for long-term climate studies. The aerosol number density profile derived by empirical relation matches well with general trend of vertical profile of aerosols. However, more simultaneous observations are essential to arrive at definite conclusion.

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