Study of aerosols in the atmosphere by twilight scattering

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ABSTRACT

Photometric measurements of the light scattered from the twilight sky, in the direction of the sun's vertical at angle an of 70 deg. from the zenith, were made during IGY–IGC period 1957–59, at Mt. Abu, India, with a photometer, the telescope of which covered a circular field of sky of about 1 deg. in the diameter.

Observations have revealed that the discontinuities in the slopes of intensity curves are observed when the centre of the sun was between 5 and 6 deg. below the observer's horizon. These discontinuities are clearly displayed as maxima on the curves of logarithmic gradients of intensity represented by $d \ln I/dh$ (h is the effective height of the earth's shadow with proper allowance for atmospheric refraction and screening by the atmosphere near the earth) against δ, the solar depression below the observer's horizon; and these are attributed to changes in the structure of the atmosphere due to changes in the dust content of the atmosphere.

At Abu, the average height of maximum dust accumulation is about 20 km with the lowest of 14 km being found in June and highest of 23 km in Nov.–Dec.

1. Introduction

Part of the sky illumination during the day and during the twilight is due to the scattering of sun-light by large particles of condensed water and/or dust. There is a more or less permanent but variable haze layer in the lower layers of the troposphere, whose intensity normally decreases upward, and there is evidence that there are also other layers at higher levels.

Particles of linear dimensions larger than the wavelength of light, scatter light much more in the forward than in the backward direction and it may be expected that the intensity of the twilight glow will depend largely on the size, nature and distribution of dust and water particles present in the atmosphere. The dust will also have the effect of reducing the intensity of the incident primary light illuminating the upper atmosphere.

Changes in the intensity of sky light at a fixed angle above the horizon where the earth's shadow traverses different levels of the atmosphere, can give information about the vertical distribution of larger particles (aerosols) in the atmosphere.

Many observers have measured the intensity of twilight at various zenith angles and wavelengths. A comprehensive work is that of Lunghall (1949) who also summarised the earlier work. One of the aims of the earlier work was to determine the air density as a function of height; in recent years experiments have been designed specifically to study the aerosols.

E. K. Bigg (1956, 64), in Australia, studied this problem of detecting dust layers in the atmosphere by measuring the variation in the brightness of a small part of the sky during twilight by a photoelectric photometer with a small aperture. He carried out the measurements at an angular distance of 70 deg. from the zenith in the sun's meridian plane for solar elevations 0 to 14 deg. below the horizon and in the spectral region 6000–8000 Å. He concluded from his results that the twilight method yields a reasonable qualitative picture of the dust distributions in the atmosphere and that a widely spread sulphate layer at a height of 15–25 km

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can be detected. Bigg assumed that the lowest height corresponded to rays grazing the earth's surface (allowing for refraction).

In the U.S.S.R., Megrilishvili (1958) made intensity measurements of the twilight glow in a direction 70 deg. from the zenith in the sun's meridian plane, on two different wavelengths, one at 9400 Å and the other at 5270 Å. He did not observe any maximum on the curves of $-d \ln I/dh$ against h for 5270 Å but observed two maxima, one at about 47 km and the other at about 100 km for 9400 Å. He assumed that due to extinction of grazing rays during sunset, the least height accessible to investigation would be about 20 km, so that the effective height of the layer directly illuminated by solar radiation would be $h + 20$ where h is the geometrical shadow height of the earth.

Volz & Goody (1962) in a comprehensive study describe the twilight observations made by them at the Blue Hill observatory, Boston, Mass., at 20 deg. above the horizon in the sun's meridian plane using a photometer and filters in different spectral bands from red to ultraviolet.

With the aid of model computations based on single scattering they have constructed profiles of turbidity which show maximum turbidity between 15–30 km and this they have associated with the maxima in the logarithmic gradients of intensity and in the intensity ratio of Red to Green light.

Other optical evidence in favour of aerosol scattering near 20 km has also been presented by Rosenberg (1960), Elterman & Campbell (1964), Fiocco & Grams (1964, 1966), Grams & Fiocco (1967), Clemesha *et al.* (1966), and Collins *et al.* (1966).

The atmospheric aerosol concentrations and size distributions measured by direct sampling from balloons and aircraft up to 30 km (Junge, Chagnon & Manson, 1961) also indicate a maximum density of particles in the range 0.1-μ to 1-μ radius at about 20 km.

With the idea of detecting the presence and levels of dust layers and their day-to-day changes, by the method of twilight scattering, twilight intensity measurements were started at Mt. Abu (24° N and 4000 ft above sea-level) in India from 1957–59. A twilight photometer in conjunction with a telescope of small aperture and a colour filter transmitting in red (centred at 6000 Å) was used.

2. Experimental technique

For obtaining a good resolution of the different dust layers, one should use (*a*) the longest convenient wavelength in order to reduce the scattering contributions by air molecules and (*b*) the smallest possible beam-width in the light-gathering equipment in order that the position of the discontinuity may be clearly defined. Hence a telescope was designed which covered a circular field of sky about 1 deg. in diameter and a Chance filter OR1 which, when used in conjunction with the photomultiplier RCA 1P22 as a light detector, covered a spectral-band with an effective wavelength of 6600 Å with a half band-width of about 500 Å. The filter used had a sharp short wavelength cut-off at about 6200 Å and therefore most of the Chappius band of ozone absorption with a maximum intensity at 6100 Å would remain in the cut-off region of the filter.

The output of the photomultiplier was fed to a stable sensitive D.C. amplifier and the current was measured with an Avometer. The amplified current varied linearly with the incident illumination. The meter readings I were recorded every 30 seconds from the time of sunset to the time when the centre of the sun was 14 deg. below the horizon. The observation times were converted to the solar zenith angles Z. The intensity I (in arbitrary units) and solar depression $\delta(=Z-90)$ in deg. constituted the raw data.

3. Direction of observation

Bearing in mind that there will be a preponderance of light in the forward direction in the light scattered by particles whose linear dimensions are large in comparison with the wavelength, and that there will also be an increasing attenuation of light with approach to the horizon, a compromise had to be made in deciding the direction of observation. Therefore the telescope of the photometer was allowed to remain at a fixed altitude (70 deg. from the zenith) and sunset (sunrise) azimuth.

4. 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 (see Fig. 1).

EE is the section of the earth's surface and *C* is its centre, *SS'* the sun's grazing rays and *P'M'* the height of the earth's geometrical shadow at the point where the direction of vision from *o* meets the tangential ray from the sun, δ the depression of the centre of the sun below the observer's horizon, θ the depression of the sun at the point *M'* on the earth vertically below the point where the grazing solar rays intersect the direction of observation which is 70 deg. from observer's zenith towards the sun, β the depression of the sun at the point *M* on the earth vertically below the point where the direction of observation intersects the solar rays tangential to the earth's surface at a height of h_0 km vertically above the earth's surface, *a* denotes the earth's radius and α the angle between the observer's zenith and the direction of observation.

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 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.

This screening h_0 will raise the base of the scattering layer. Besides, a tangential ray passing completely through the lower atmosphere with closest approach at height h_0 km from the earth's surface would be deviated by refraction through *w* deg.

Thus, PM (Fig. 1), the height *h'* of the earth's geometrical shadow at the point where the direction of vision from 0 cuts the tangential ray from the sun at a height h_0 km from the earth's surface will be modified to account for refraction

$$h = (a + h_0) \sec \theta' - a, \quad (1)$$

where *h* is the effective height of earth's shadow and

$$\theta' = \beta - w \quad (2)$$

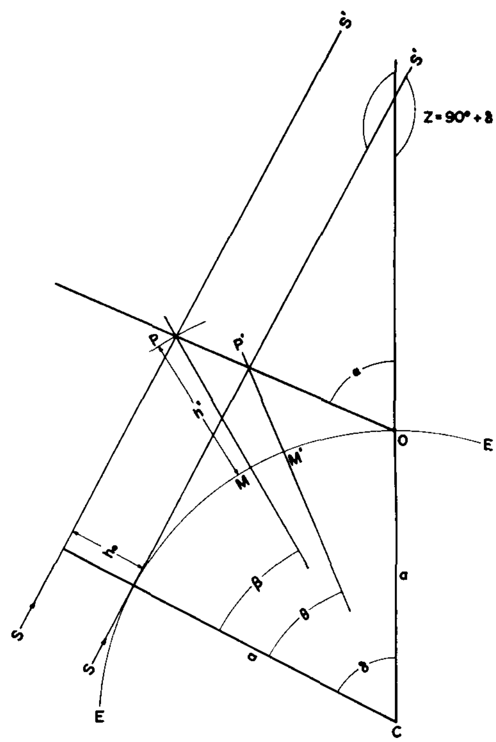

Fig. 1. Schematic diagram of the solar grazing rays illuminating the twilight sky.

(*w* is the assumed constant), β can be calculated by the formula

$$\tan \beta = \tan \delta - \frac{\sin \alpha}{\cos(\alpha - \delta)} \left\{ \sec \delta - \frac{a}{a + h_0} \right\}; \quad (3)$$

h is the effective height of the earth's shadow, i.e. height of the base of the directly illuminated regions in the atmosphere for different depressions of the sun (corrected for screening height and atmospheric refraction), can be calculated by eq. 1 provided h_0 is known.

Bigg (1956) assumed that if longer wavelengths are used, the earth's shadow height will correspond to the geometrical height of the lowest grazing rays.

Megrilishvili (1958) 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 earth's geometrical shadow height without refraction.

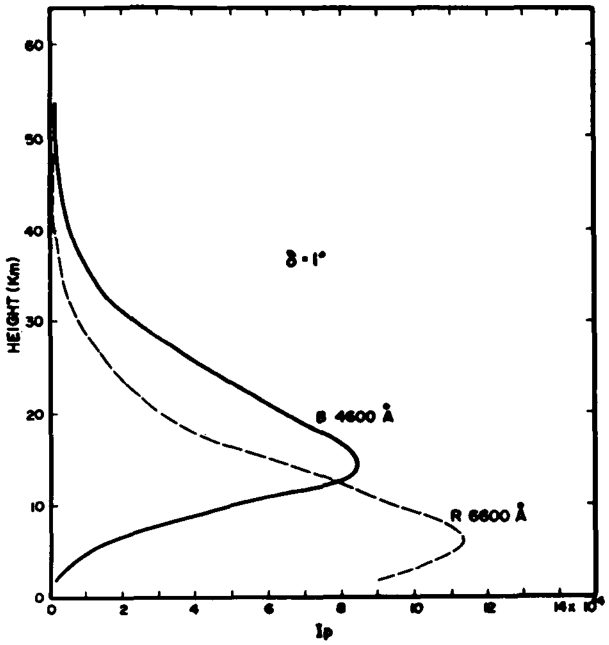

Fig. 2. Intensity of primary scattered light from different levels in the atmosphere in the direction 20 deg. above horizon in the sun's meridian plane.

Volz & Goody (1962) have given arguments to show that in a normal turbid atmosphere, we can take it that the twilight comes from above 10 km for red light.

The author has also made calculations of the intensity of primary scattered light in the sun's meridian plane at an angle of 70 deg. from the zenith, from different levels in the atmosphere when the centre of the sun is 1 deg. below horizon (without correcting for screening height or refraction). The treatment is similar to that used by Dave (1956). Curve R in Fig. 2 is the height-intensity curve for red light (6600 Å) and curve B for blue light (4600 Å).

It is seen from the curves that the maximum scattered light in the blue comes from a height of about 15 km and falls off rapidly at lower heights. In the red, the maximum primary scattered light comes from a height of about 6 km above the surface of the earth. Remembering that there will always be some dust in the lowest of the atmosphere, the screening height for red light has been assumed to be 6 km.

Table 1 gives the heights of the directly illuminated region in the atmosphere for different depressions, δ of the sun below the horizon, in a direction of 70 deg. from the zenith towards the sun. θ' is the depression of the sun below the horizon for the observer situated on the surface of the earth vertically below the point where the direction of observation meets the

Table 1

h = Effective height of the earth shadow in the direction at 70 deg. from the zenith in the sun's meridian. δ = Solar depression below observer's horizon. θ' = Solar depression below horizon for an observer on the earth vertically below the point where the direction of observation (70 deg. from the zenith) meets the refracted grazing rays with closest approach to the earth at 6 km (w-as in eq. 2)

Depression of the sun			Depression of the sun		
δ, deg.	θ', deg	h, km	δ, deg.	θ', deg.	h, km
1	0.2	6.0	7	5.4	35.0
2	1.2	8.0	8	6.2	44.0
3	2.1	11.0	9	6.9	53.0
4	3.0	15.0	10	7.7	64.0
5	3.8	20.0	11	8.4	75.0
6	4.6	27.0	12	9.1	87.0

refracted grazing rays with closest approach to the earth at 6 km, are also given in Table 1, for different solar depressions δ. w, the total deviation by refraction, for a tangential ray at a height of 6 km, is assumed to be 0.6 deg. As the photometer does not follow the sun in the azimuth, heights determined in the latter parts of twilight will be underestimated.

5. Analysis

In a pure dust-free atmosphere, if the intensity of sky light is assumed to be due mainly to the primary scattered light, the intensity I will be proportional to the pressure p at the level of the effective shadow. But, as a matter of fact, there is a certain amount of secondary and multiply scattered light added to the primary scattered radiation and moreover, the atmosphere contains some particles of haze and condensed water vapour which cause additional brightness in certain directions and also attenuate the direct radiation.

If we consider only primary scattered light from a defined direction in a dust-free Rayleigh atmosphere, we have

$$\frac{dI}{I} = \frac{dp}{p}, \quad (4)$$

where I is the brightness of the sky in that direction and p is the pressure at the effective base of the layer directly illuminated by the solar radiations.

Since the change of pressure with height in the atmosphere is given by the equation

$$dp = -g\varrho dh, \quad (5)$$

where dp is the fall of pressure corresponding to an increase in height dh

and
$$p = \frac{RT\varrho}{M}, \quad (6)$$

where p is the pressure, ϱ the density, T the temperature, R the universal gas constant and M the mean molecular weight.

From (5) and (6) we get

$$-\frac{dp}{p} = \frac{Mg}{RT}\cdot dh$$

(assuming T to be constant within dh),

i.e.
$$d \ln p = -\frac{Mg}{RT}\cdot dh. \quad (7)$$

Now from eq. 1

$$\frac{a+h}{a+h_0} = \sec\theta',$$

i.e.
$$\frac{1+h/a}{1+h_0/a} = \sec\theta'$$

expanding both sides in series we get

$$\left(1+\frac{h}{a}\right)\left(1-\frac{h_0}{a}\right) = 1+\frac{\theta'^2}{2}$$

or
$$\frac{h}{a}-\frac{h_0}{a} = \frac{\theta'^2}{2} \quad (8)$$

($h_0 h/a^2$ = negligibly small), differentiating (8) with respect to h

$$\frac{dh}{a} = \theta'\cdot d\theta',$$

i.e.
$$\frac{d\theta'}{dh} = \frac{1}{\theta'\cdot a}. \quad (9)$$

Equation 4 can be rewritten as

$$\frac{d\ln I}{d\ln p} = 1; \quad (10)$$

also
$$\frac{d\ln I}{d\ln p} = -\frac{d\ln I}{dh}\left(\frac{RI}{Mg}\right), \quad \text{from (7)}$$

$$= -\frac{d\ln I}{dh}\cdot H(h).$$

Therefore
$$-\frac{d\ln I}{dh} = H^{-1}(h)$$

in a dust-free Rayleigh atmosphere. However, in an actual atmosphere scatterers (dust) other than molecules are also present. Therefore the molecular scale height $H(h)$ will be superimposed with a dust scale height.

Since there is a certain amount of uncertainty

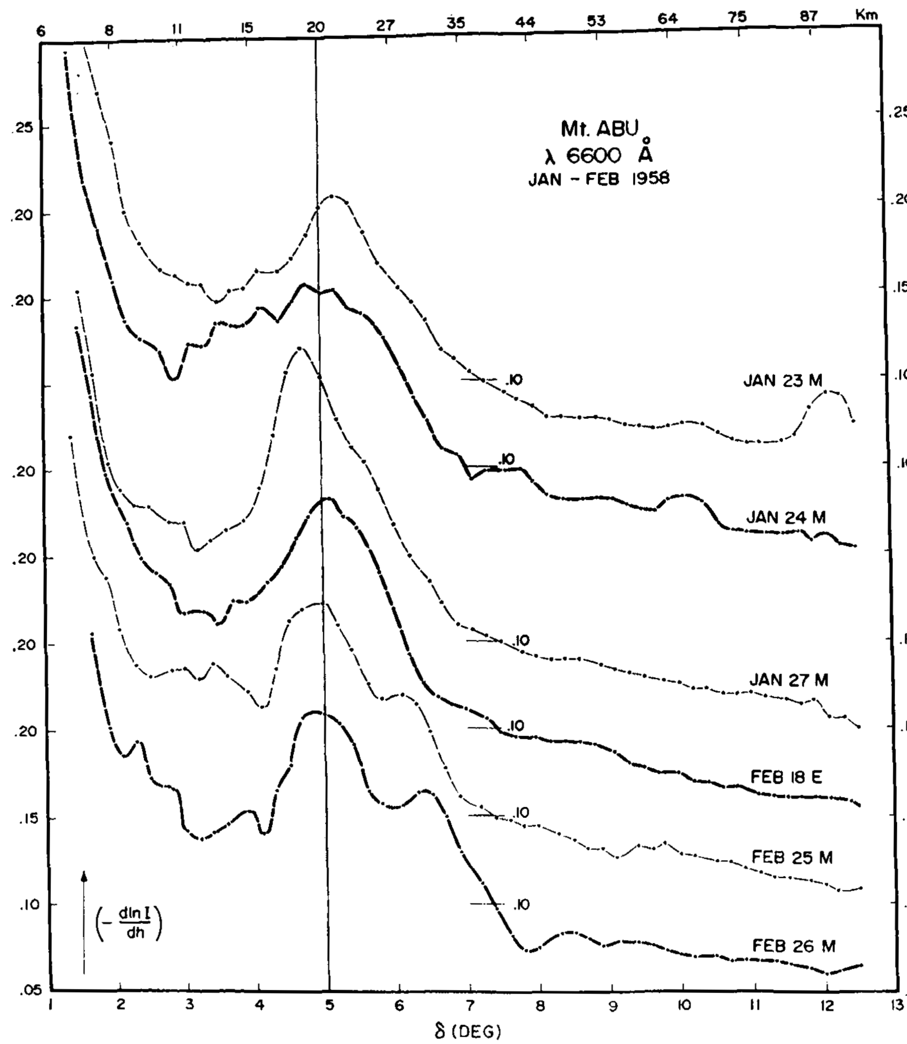

Fig. 3. Variation of $-d \ln I/dh$ for different solar depressions δ. M, Morning twilight; E, Evening twilight.

in defining the effective height of the earth's shadow, i.e. the height of the base of the region directly illuminated by solar radiation, we considered it desirable to plot $d \ln I/dh$ against δ.

$$\text{Now} \quad -\frac{d \ln I}{dh} = -\frac{d \ln I}{d\delta} \cdot \frac{d\delta}{dh}, \tag{11}$$

where dh is the change in vertical height in the direction of observation corresponding to change of $d\delta$ in solar depression. The quantity $-d \ln I/d\delta$ along the direction of observation is computed from the raw values of I corresponding to δ, for different mean δ, and $d\delta/dh$ is calculated with help of eqs. 2, 3 and 9 for each mean δ.

Thus, the values of $-d \ln I/dh$ were computed for different values of δ. The quantity $-d \ln I/dh$ calculated from the actual observations also takes into account the contribution by particles other than molecules so that the plot of $-d \ln I/dh$ against δ represents the conditions existing in an actual atmosphere.

6. Results

Figs. 3–7 present the plots of values of $-d \ln I/dh$ against δ in different months of 1958. The values were smoothed by taking three-point running means. The effective height

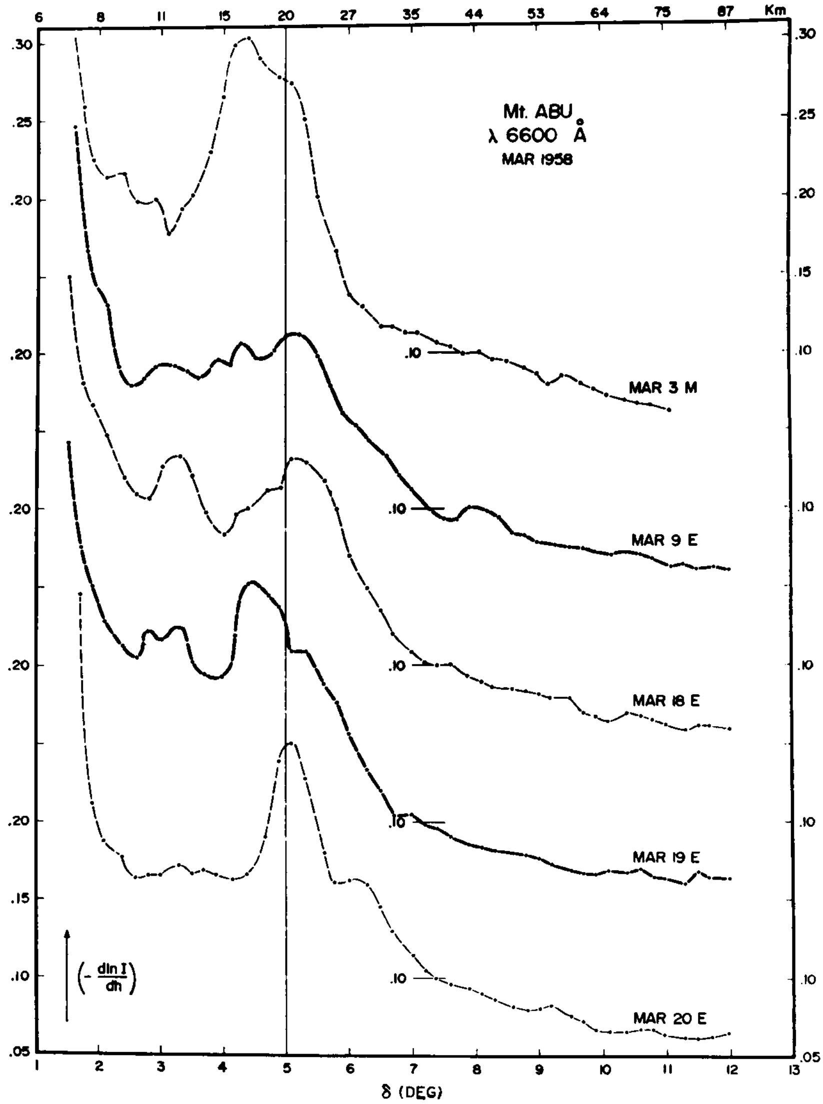

Fig. 4

Figs. 4–7. (Explanations as in Fig. 3.)

of the base of the region directly illuminated by solar radiations, in the direction of observation corresponding to different solar depressions are also given at the top scale of the figures. As the sun sinks below the horizon, the effective height of the earth's shadow rises and scattering takes place from higher and higher levels. Most of the light received at the ground will be the primary scattered light by the particles (molecular + large particles) for solar depressions less than 6–7 deg. Therefore

$$-\frac{d \ln I}{dh} \simeq -\frac{d \ln (n)}{dh},$$

where n is particle number density.

It is noted from the figures that the value of $-d \ln I/dh$ decreases as the solar depression below horizon increases, implying decrease of concentration with height in the lower troposphere. When the centre of the sun is between 3 and 4 deg. (corresponding height of the effective earth shadow between 11 and 15 km) the in-

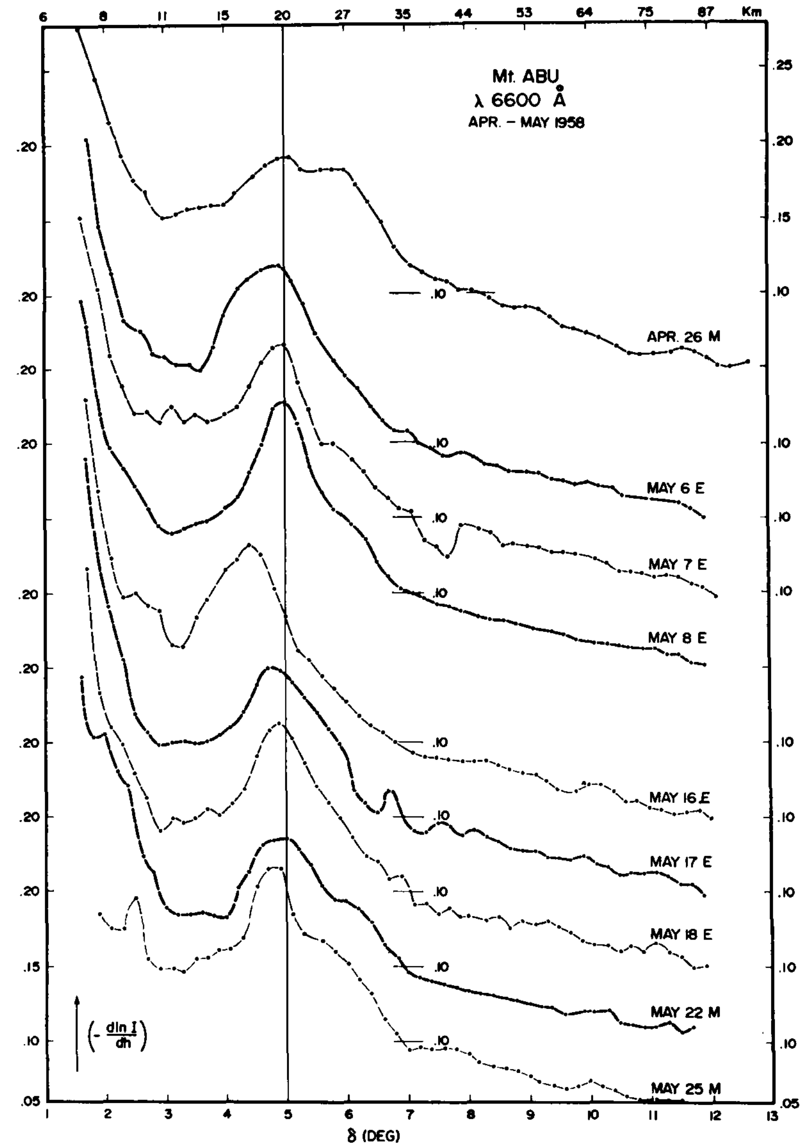

Fig. 5

tensity seems to be changing very little, implying constant concentration at that level.

The subsequent increase in the logarithmic gradient is due to the rising of the shadow across the dust layer when the centre of the sun is between 4 and 6 deg. This corresponds to the height between 15 and 25 km. The maximum occurs, on the average, at the height of 20 km. The occurrence of this maximum is more or less a regular feature of the curves, implying the existence of a persistent dust layer at the height of about 20 km.

The vertical profiles obtained by Chagnon & Junge (1961) based on the direct measurements of large particles ($0.1 < r < 10\ \mu$) shows a similar structure of the atmosphere consisting of three main layers. First, a rapid decrease in concentration in the lower troposphere, second, a

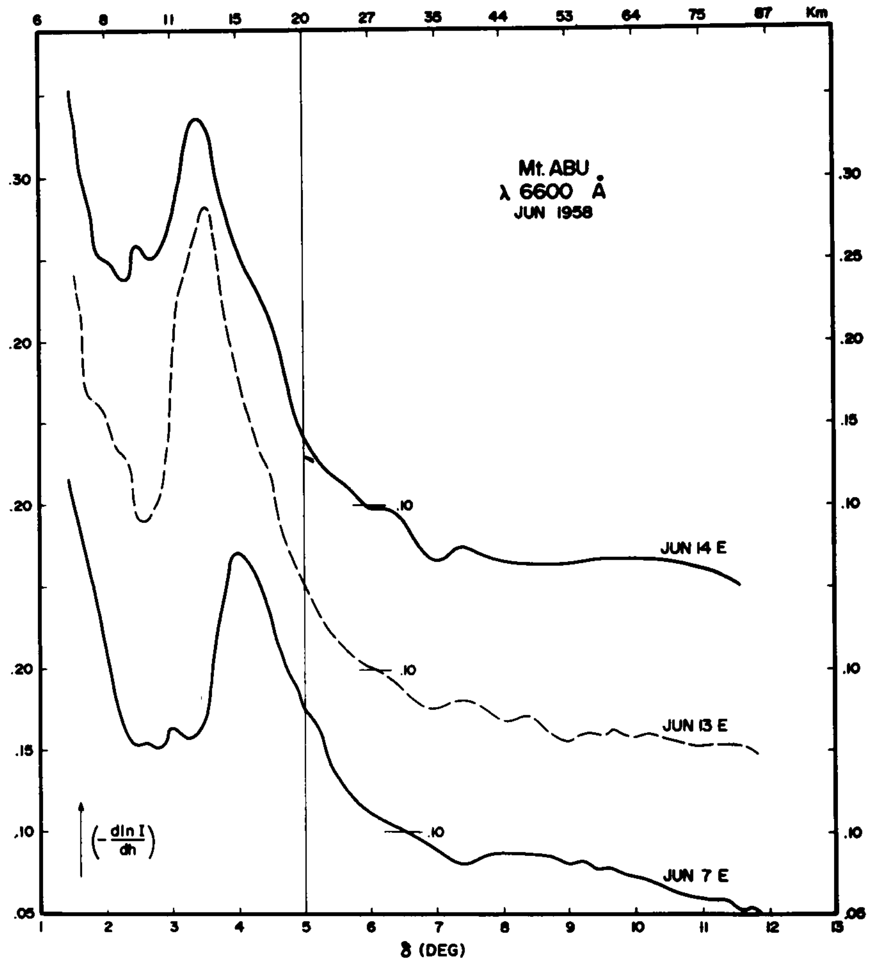

Fig. 6

fairly constant concentration in the upper troposphere, and third, the aerosol layer in the stratosphere between 15 and 25 km representing the dust layer.

From January–May, the average height of this maximum was about 20 km, while in November–December it was about 23 km, excepting Dec. 7 and 17 curves which are unusual, the highest in any month. The lowest height of about 14 km was observed in June (only 3 sets of observations). This suggests that in the summer months the layer occurs at a lower height than in winter months, but more observations are required to establish this.

The variation of logarithmic gradient of intensity is very similar after 7 deg. solar depression, irrespective of season.

Sometimes a secondary peak is observed near 30 km with the main peak still generally located at about 20 km (except on December 7). Examples of this were found on Feb. 25, Feb. 26, March 20, May 17 and Dec. 7. A secondary peak around 13 km was also observed, in few cases for instance on March 18, March 19 and Dec. 9 with the main peak still located at around 20 km. On Dec. 17, the main peak was shifted to about 12 km while the second broad but less prominent peak was located at about 20 km, but such cases are very rare. When main peak shifts to the lower height, the second layer of constant concentration also shifts to the lower levels, as shown in June curves.

Fig. 8 presents the height distribution of the average values of $-d \ln I/dh$ for the winter

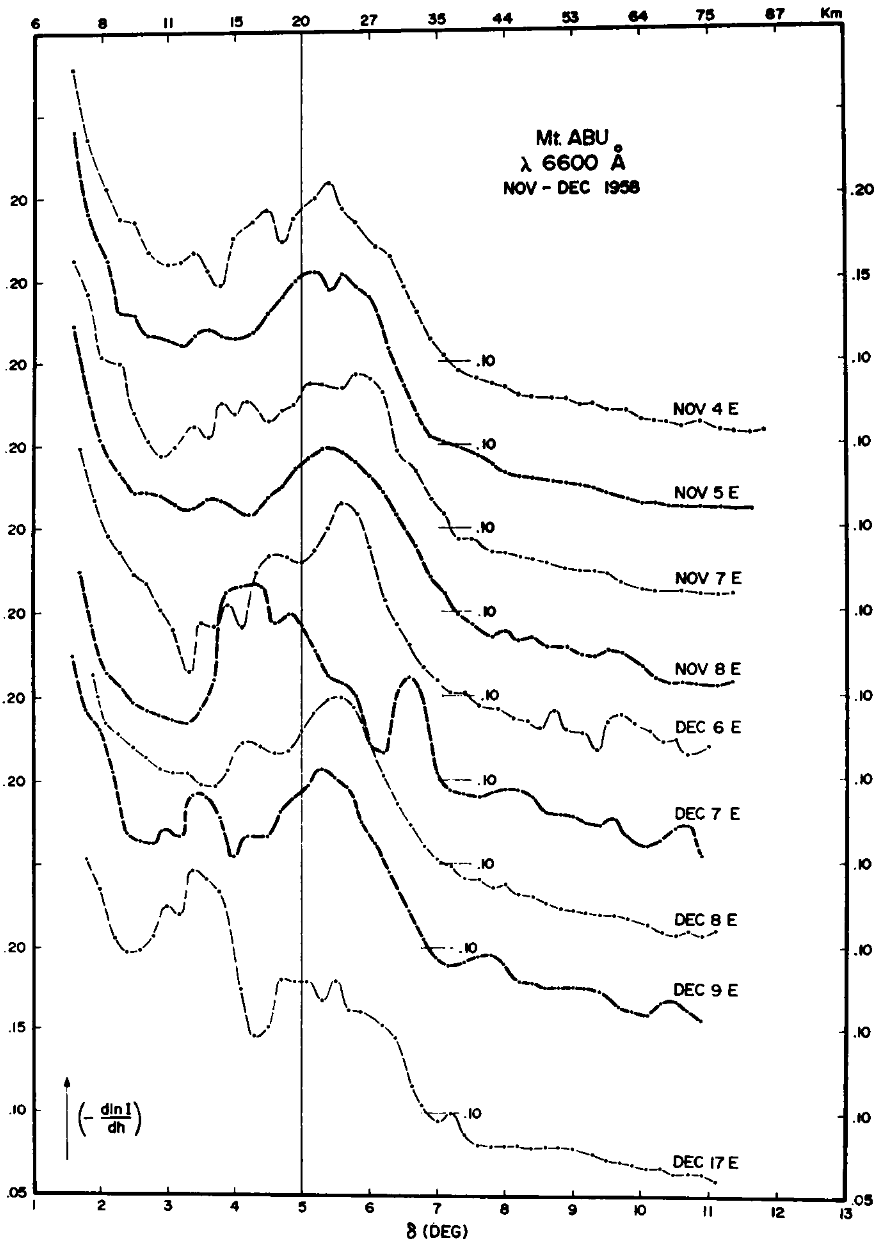

Fig. 7

months (Jan.–Feb. 1958, 6 sets). The average curves for winter months given by Bigg (1964) and height distributions of particles determined by Junge *et al.* (1961) by direct methods are also plotted for comparison. These two latter curves are reproduced from Bigg's (1964) paper.

Fig. 8 suggests that the mean curves yield a reasonable, qualitative picture of the dust distributions at least in the stratosphere, and confirms the existence of a dust layer at about 20 km.

The existence of a dust layer near 20 km was first suggested by Gruner (1942) to explain the purple light above the sunset (sunrise) point.

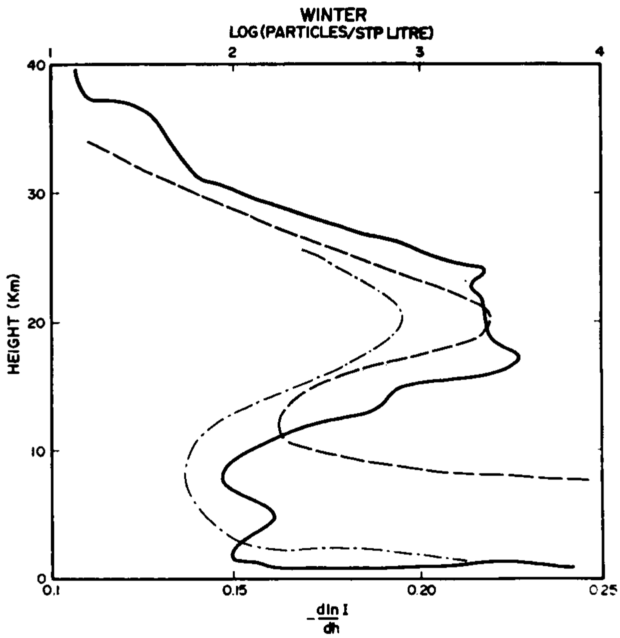

Fig. 8. Height distribution of average values of $-d\ln I/dh$ for the winter months. ---, January–February 1958, Mt. Abu (24° N); ——, June–July 1960, Canowindra (34° S) from Bigg (1964); -·-, Junge *et al.* (1961) from Bigg (1964).

7. Conclusions

The twilight scattering method yields a reasonable, qualitative picture of the vertical distributions of dust particles at least in the lower stratosphere. At Abu, the maximum concentrations occur, on the average, at about 20 km and these are associated with maxima on the logarithmic gradient of intensity. The lowest average height of about 14 km was found in June while the highest (23 km) was found in November–December.

The Abu measurements together with the measurements of Bigg (1964) and of Junge *et al.* (1961) confirm the existence of a stratospheric dust layer at around 20 km.

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ИЗУЧЕНИЕ АЭРОЗОЛЯ В АТМОСФЕРЕ МЕТОДОМ СУМЕРЕЧНОГО РАССЕЯНИЯ

В течение МГГ-МГГС в период 1957–59 гг. на г. Абу в Индии были проведены фотометрические измерения света, рассеянного сумеречным небом в направлении солнечного вертикала при угле в 70° от зенита. Измерения проводились с помощью фотометра, телескоп которого покрывал круговой участок неба с диаметром около 1°. Наблюдения показали, что разрывы в наклонах кривых интенсивностей имеют место, когда центр Солнца находится между 5 и 6° ниже горизонта для наблюдателя. Эти разрывы ясно проявляются в виде максимумов на кривых логарифмических градиентов интенсивности — $d \ln I/dh$ (h — эффективная высота тени Земли с учетом атмосферной рефракции и затенения атмосферной вблизи земли) в зависимости от б — угла депрессии Солнца под горизонтом для наблюдателя. Эти максимумы связываются с изменениями структуры атмосферы благодаря изменения концентрации атмосферной пыли. Вблизи г. Абу средняя высота слоя максимальной аккумуляции пыли порядка 20 *км*, причем наименьшая высота 14 *км* наблюдалась в июне, а наибольшая — 23 *км* в ноябре–декабре.