

Measurement of Total Atmospheric Ozone Using Sky Radiation¹

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ABSTRACT

Instead of measuring total ozone by observing the direct sun (if visible) or at the zenith, photometric measurements of sky radiation were made in the solar vertical. The newly developed spectrophotometer uses filters and a photomultiplier as the light sensor.

Applying a theoretical model of the atmosphere as a primary reduction parameter, the measured spectral intensities of skylight are compared to deduce total ozone amount. Preliminary results show fair correlation with the ozone values obtained by the Dobson spectrophotometer, the standard instrument.

1. Introduction

Observations of total atmospheric ozone have been made at an increasing number of places during the last 50 years. At the present time, the network of total ozone observations includes about 90 stations, with a greater density in the Northern Hemisphere. It is a remarkable fact that despite extensive use of new measurement techniques for ozone observations, the Dobson spectrophotometer has remained the approved standard instrument for total ozone measurements for the last two decades. The sensitivity and accuracy of this double monochromator instrument is very high. However, its maintenance, calibration and manual operation are somewhat complicated and have been discussed elsewhere (Paetzold, 1961; Paetzold and Zschörner, 1967; Dobson, 1957).

For the important purpose of continuity of measurements and ease of operation a filter spectrophotometer was developed at the Institute for Geophysics and Meteorology, University of Cologne (Ghazi, 1968). It is a small, simple and automatic recording instrument which operates by remote control.

2. Experimental technique

Fig. 1 shows the optical system of the filter spectrophotometer. A beam of sky radiation falls through the quartz window on a plane mirror A which rotates with uniform angular velocity. The mirror is mounted at an angle 45° to the surface. The radiation beam which falls on the mirror is converged by a quartz lens C and reflected rectangularly by another plane mirror D. It is then limited by a shutter E in such a manner that a view angle of only about 1° is covered. The radiation beam is then made parallel by the lens F. These parallel

rays, after passing through a combination of filters J, fall ultimately on a photomultiplier.

The mirror A makes one vertical circular scan of the sky during a rotation. A slow movement of the system, over the vertical axis B, in a horizontal plane from 0° – 180° , permits scanning over all vertical circles, so that the radiation intensity of the entire hemisphere can be registered by the instrument in a few minutes for all the filters used. The spectral transmission maxima of the filters are as follows:

- Filter 1: 3200 Å in the Hartley absorption bands of ozone
- Filter 2: 3600 Å in the region of no O₃ absorption
- Filter 3: 3900 Å in the region as filter 2 (tentative use)
- Filter 4: 4700 Å in the Chappuis absorption bands of ozone

Using the above technique, readings can be made from every point on the sky and the radiation intensity recorded by the instrument. The output of the multiplier is fed to the recording instruments. To eliminate all possible variations in the spectral sensitivity of the photomultiplier, a calibration check is made through a lamp H at each measurement. The photoelectric voltages are plotted in an analogous measuring process by a sensitive oscillograph. In this manner it is possible to record the variations in the radiation intensity over each of the scanned vertical cross sections. Fig. 2a presents the measured intensity curves in a vertical plane from horizon to horizon through the sun (the solar vertical) for a cloudy and a clear day, while Fig. 2b shows the responses for each of the four filters on a cloudy day. The time difference between each of the filter scans is 12 sec.

3. Analysis

Generally, the total amount of ozone (in a vertical column) is deduced from the measured reduction in the

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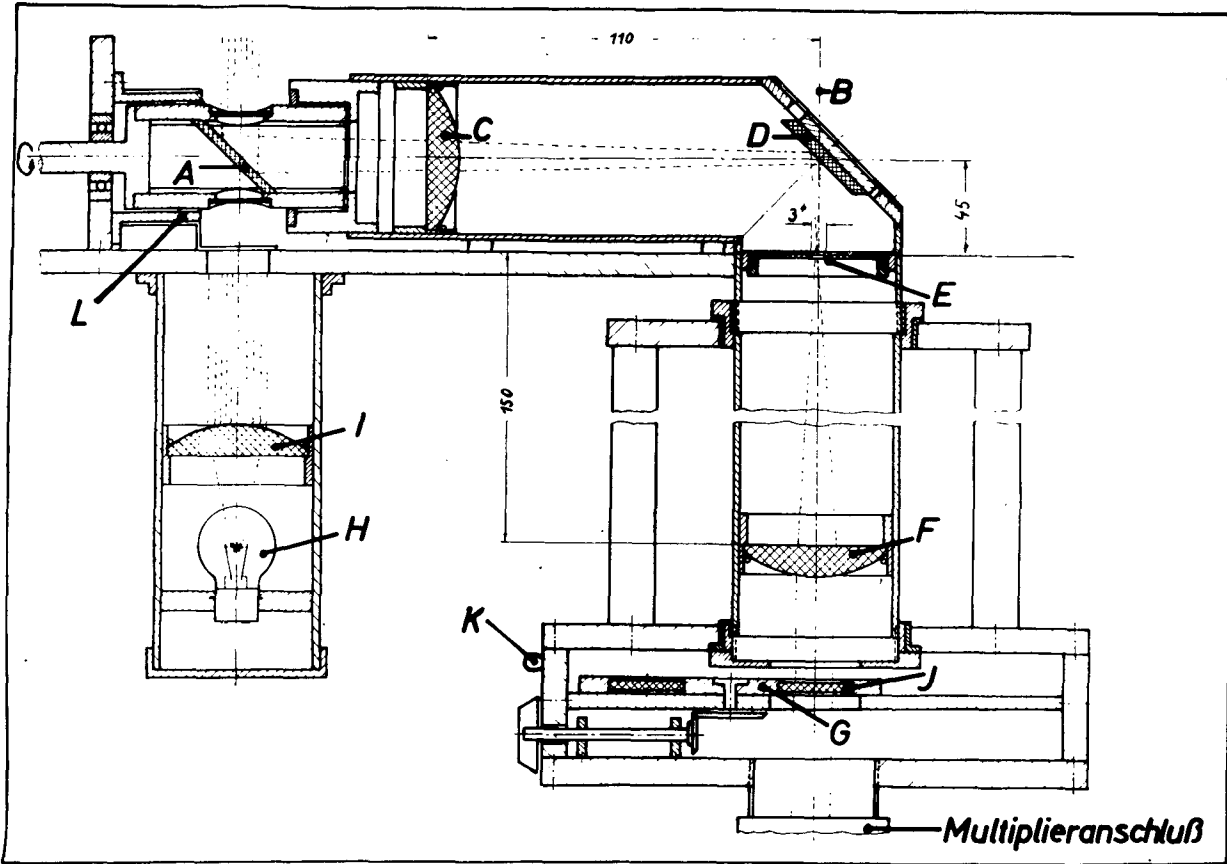


FIG. 1. Schematic view of the optical system of the filter spectrophotometer: A, D, plane mirrors; B, vertical axis; C, F, quartz lenses; E, shutter; G, filter wheel; J, spectral filters; H, calibration lamp; K, filter position indicators.

intensity of sunlight of certain chosen wavelengths during their passage through the atmosphere. Measurements are usually made either with the instrument

pointed directly at the sun or at the zenith. Since the climatic conditions prevailing in European latitudes reduce the time during which direct solar observations

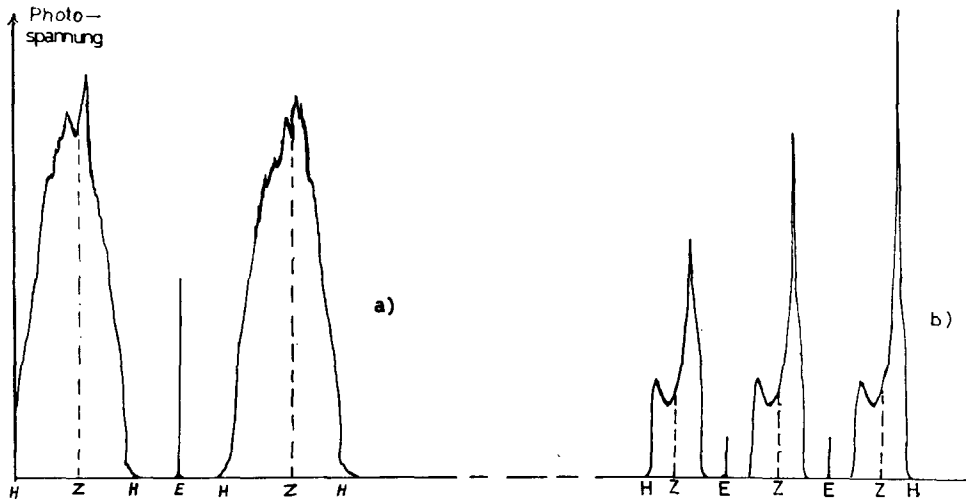


FIG. 2a. Schematic curves representing the spectral radiance along vertical cross sections of the sky for filter 1 for an overcast sky, a), and a cloudless sky near the sun, b): H, horizon; Z, zenith, E, calibration value.

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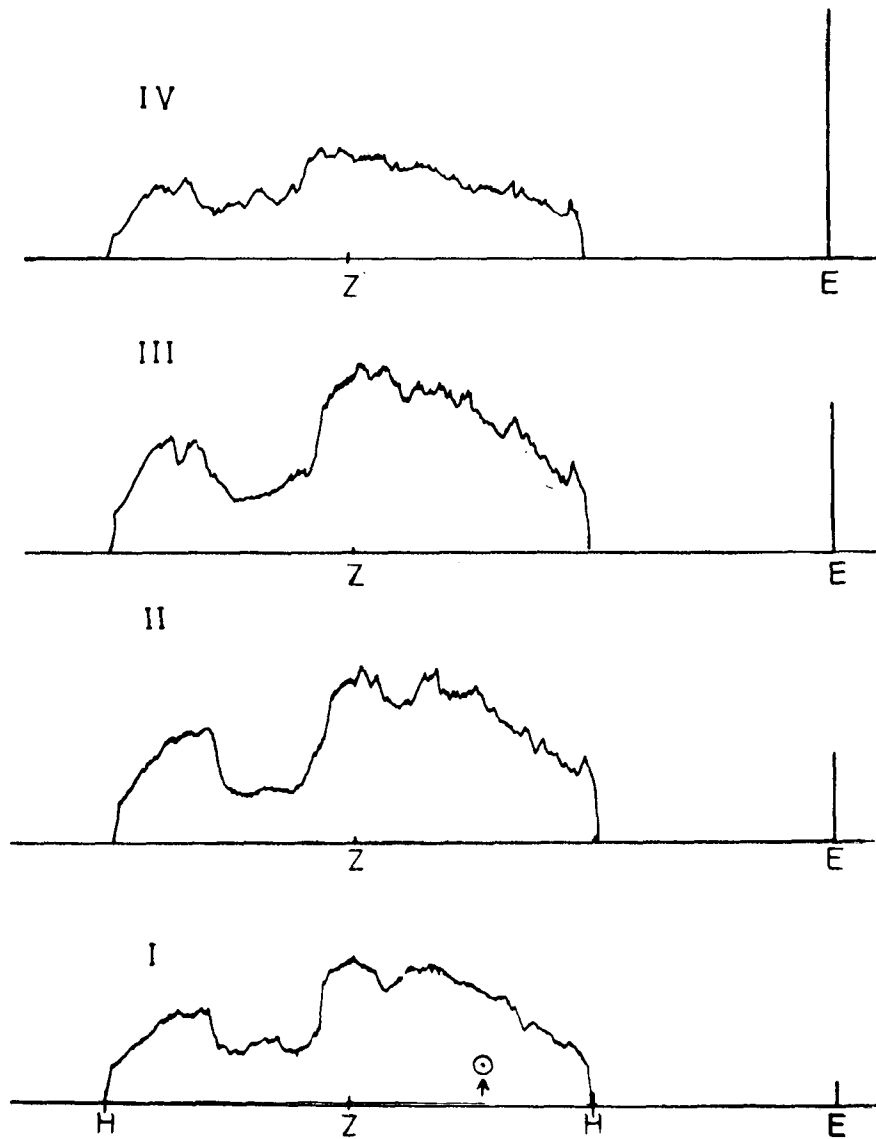


FIG. 2b. Measured spectral radiance curves (schematic) for a cloudy day in the solar vertical for filters 1-4; scan time for each of the filters is 0.1 sec from horizon to horizon. Arrow denotes the sun's elevation. Time difference between each filter scan is 12 sec.

can be made, and zenith values of radiance are not always reliable, there was a need for taking measurements at other sky positions.

Instead of measuring solar radiation with the photometer pointed directly at the sun, skylight measurements were made as mentioned above in the solar vertical. The reduction of intensity is determined by measuring the sky radiation at $\lambda_1 = 3200 \text{ \AA}$ where ozone absorbs with a decadian absorption coefficient $\alpha = 0.32 \text{ cm}^{-1}$ and at $\lambda_2 = 3600 \text{ \AA}$ where there is almost no absorption by atmospheric ozone.

To determine the total amount of ozone it is necessary to make relative measurements. The radiation flux

passing through the filters is registered as a photoelectric voltage U and is given by

$$U = ITPE, \tag{1}$$

where I is the intensity of incident radiation on the earth's surface, T the transmission factor of the filter, P the sensitivity of the photomultiplier, and E the calibration value at each measurement.

If we consider the extraterrestrial intensity at the wavelength to be $I_0(\lambda)$, then the attenuation during the passage of radiation through the earth's atmosphere (according to Beer's law) would be

$$I(\lambda)d(\lambda) = I_0(\lambda)10^{-k(\lambda)d(\lambda)}. \tag{2}$$

In the above equation $k(\lambda)$ can be expressed as a function of three different components, i.e., those due to Rayleigh scattering k_R , particle or Mie scattering k_M , and gaseous absorption k_A . In our limited case k_A can be written as

$$k_A = \alpha(\lambda)\mu\Omega, \tag{3}$$

where $\alpha(\lambda)$ is the absorption coefficient of ozone, μ the traversed ozone mass $[f(z)]$, and Ω the total ozone. Therefore Eq. (2) can be expressed as

$$I(\lambda)d(\lambda) = I_0(\lambda)10^{-[k_R+k_M+\alpha(\lambda)\mu\Omega]}d(\lambda). \tag{4}$$

As mentioned above the observations are made at two wavelengths λ_1 and λ_2 , so that (4) can be solved for Ω as

$$\Omega = (\alpha\mu)^{-1} \log \left[\left(\frac{I_2 I_{01}}{I_1 I_{02}} \right) 10^{k_{R2}-k_{R1}} 10^{k_{M2}-k_{M1}} \right]. \tag{5}$$

As $\alpha(\lambda_2) = 0$, Eq. (5) becomes

$$\Omega = (\alpha\mu)^{-1} \log \left[\left(\frac{I_2 I_{01}}{I_1 I_{02}} \right) \frac{R_1 M_1}{R_2 M_2} \right], \tag{6}$$

where $R_{1,2}$ are the Rayleigh radiation intensities at the earth's surface for λ_1 and λ_2 , $M_{1,2}$ the Mie radiation intensities at the earth's surface for λ_1 and λ_2 , $I_{01,2}$ the extraterrestrial intensities at λ_1 , λ_2 , and $I_{1,2}$ the intensities of sky radiation for λ_1 and λ_2 at the earth's surface. Since we wish to transform $I_{1,2}$ into voltages $U_{1,2}$ [on the basis of Eq. (1)], we find, assuming $I_{01,2}$ to be constant, that

$$\Omega = (\alpha\mu)^{-1} [A + \log(U_2/U_1) - \log(R_2/R_1) - \log(M_2/M_1)], \tag{7}$$

where A is the instrument constant given by

$$A = (E_1 T_2 P_2 \log I_{01} - E_2 T_1 P_1 \log I_{02}).$$

Following Chandrasekhar (1950), Coulson *et al.* (1960), and Dave and Furukawa (1966), the Rayleigh radiation intensity at the earth's surface can be defined as

$$R = (1/\mu) \int_0^\tau R_0(t; \mu, \zeta; -\mu_0 \zeta_0) \times \exp[-(\tau-t)/\mu] \omega(t) d(t), \tag{8}$$

where R_0 is the source matrix; ω the albedo of single scattering; τ the optical thickness of the atmosphere,

$$\int_{z_0}^\infty k_v(z) \rho(z) dz,$$

where $\rho(z)$ is the density at height z and $k_v(z)$ the mass scattering coefficient ($\sim \lambda^{-4}$); μ, ζ the direction parameters of the scattered radiation; and μ_0, ζ_0 the direction parameters of the incident radiation. Rayleigh's theory (1899) pertains to a dark blue sky. This condition is generally not fulfilled, since dispersion of the radiant

energy takes place due to the presence of very many discrete particles such as haze, dust, water droplets, etc., in the atmosphere. This dispersion accounts for the visibly lighter blue color of the sky. Thus, the implication of the theory of atmospheric scattering would require consideration of complete size distributions of scattering particles. However, for our limiting case the use of the Rayleigh atmosphere as the primary reduction parameter seems a plausible compromise, at present.

4. Atmospheric model

A plane-parallel atmosphere is assumed in which the radiation is scattered according to Rayleigh's law, without any other absorption than that of ozone, in the spectral region $3000 < \lambda < 5000 \text{ \AA}$. Based upon Chandrasekhar's (1950) ingenious solution to the problem of determining the intensity of diffused radiation emerging from the bottom of the atmosphere, Coulson *et al.* (1960) have presented extensive tables of intensity parameters of Rayleigh radiation. Dave and Furukawa (1966) have calculated these Rayleigh intensities with multiple scattering affected by ozone absorption for different solar elevations, wavelengths, directions of observation, albedo and pressure levels. These results were adapted for our calculations after a selective examination of radiation parameters.

Assuming a mean vertical distribution of atmospheric ozone (0.341 atm-cm total ozone) representative of the middle latitudes of the Northern Hemisphere, a spectral distribution of scattered Rayleigh radiation in the solar vertical for $\lambda = 3200 \text{ \AA}$ is derived as shown in the Fig. 3; the lower curve includes the absorption by ozone. It is evident that the scattered radiation is attenuated by a constant factor due to the absorption of ultraviolet light by ozone. Nevertheless, comparison of the curves shows that the variation in the structure of the Rayleigh radiation due to total ozone is substantially negligible.

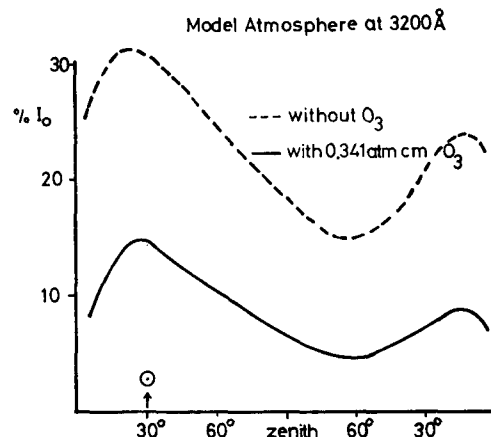


FIG. 3. Theoretical Rayleigh atmosphere in the solar vertical with (solid curve, $\Omega = 0.341 \text{ atm-cm}$) and without (dashed curve) ozone absorption derived for filter 1 (3200 \AA). Arrow denotes sun elevation angle h .

In view of this apparently minor variation, an estimate of the effects of absorption due to the vertical distribution of atmospheric ozone in a Rayleigh atmosphere could be made. For this reason, it is interesting to consider the Rayleigh intensities, in the solar vertical, at different pressure levels above the earth's surface up to the altitude of maximum ozone concentration. As can be seen in Fig. 4, the intensity of Rayleigh radiation at the level of the ozone maximum (even in the regions near the horizon) is less than 5% of the corresponding extraterrestrial intensity I_0 . The structure of the Rayleigh atmosphere shows only a minor variation with altitude until the radiation traverses the lowest part of the atmosphere. Thus, in analogy to the total ozone amount, the vertical distribution of ozone does not affect the structure of the relative distribution of Rayleigh intensity. The depletion of Rayleigh radiation is therefore proportional to the total amount of ozone. Due to this fact it is possible to consider the attenuation of radiation by ozone separately from other molecular scattering processes. In general, two points should be emphasized concerning the altitude dependance of Rayleigh radiation:

- 1) The intensity of radiation increases with the depth of the traversed atmosphere.
- 2) Radiation is absorbed near the horizon at the earth's surface. As will be discussed later, this absorption is a function of the wavelength.

5. Results

The theoretical structure of the Rayleigh atmosphere in the solar vertical due to multiple scattering and ozone absorption, for spectral regions used in the filter instrument, is shown in Fig. 5. These curves are computed for a solar zenith distance² $z=30^\circ$, total ozone $\Omega=0.341$ atm-cm, and ground albedo $a=0.2$. The significant results are 1) the increase of intensity with wavelength at the horizon and the corresponding decrease at the zenith, 2) minima of scattered radiation for all wavelengths at the anti-solar side, and the fact that 3) maxima of intensities do not correspond to the position of the sun.

In order to compare the measured and computed radiation in the solar vertical, for the same ozone amount, the total ozone measurements were made by a Dobson spectrophotometer using direct sunlight (i.e., using AD wavelengths). Fig. 6 shows the measured intensity curves for conditions similar to those used in the computations for Fig. 5.

The structure of the theoretical Rayleigh atmosphere is evident only at the anti-solar side of the solar vertical. It is tempting and logical to speculate that this scattering on the solar side is chiefly due to Mie radiation. To support this reasoning, measurements were made on clear days for different solar elevations. Figs. 7 and 8

² Solar zenith distance (or angle) z and its complementary angle, the solar elevation angle h , are used interchangeably.

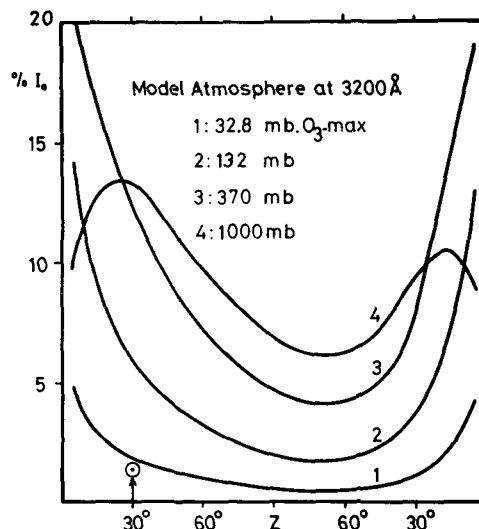


FIG. 4. The Rayleigh atmosphere in the solar vertical at different pressure levels as derived for filter 1, for 0.341 atm-cm, $z=60^\circ$: curve 1, 32.8 mb (i.e., the O_3 maximum); 2, 132 mb; 3, 370 mb; 4, 1000 mb.

present the theoretical angle measured radiation structures, respectively, at a low solar elevation angle (i.e., $z=72^\circ$). In general, the structure of Rayleigh atmosphere is retained better for lower sun elevations. Several measurements in this respect showed that the discrepancy between the measured radiation and that from the theoretical model increases with longer wave-

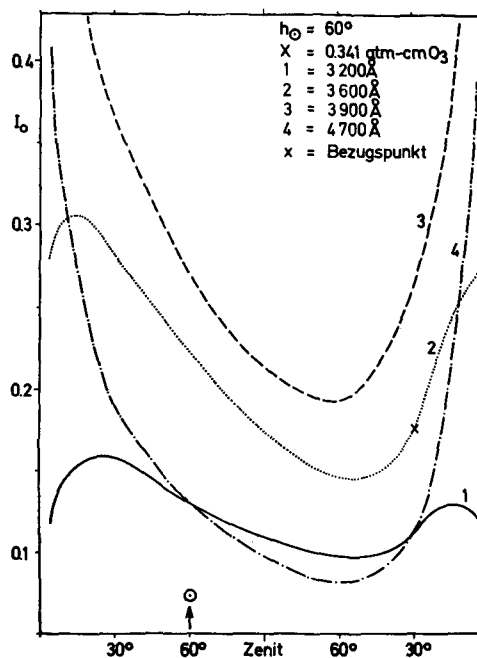


FIG. 5. Computed curves for various spectral regions representing the theoretical model of the Rayleigh atmosphere with multiple scattering and ozone absorption in the solar vertical. I_0 is the extraterrestrial intensity, assumed as unit. The sun elevation angle is 60° and $\Omega=0.341$ atm-cm.

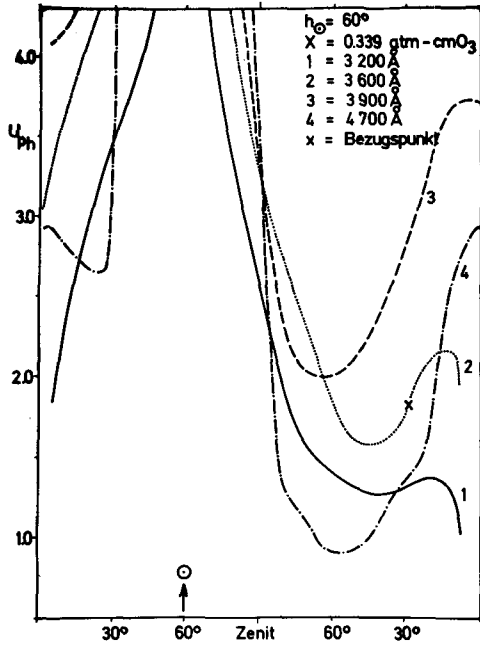


FIG. 6. Measured spectral radiance for $\Omega=0.341$ atm-cm in the solar vertical expressed in photoelectric voltage units. The point x is the reference point to Fig. 5.

length, in spite of the fact that the ratio of intensities between the wavelength regions used does not vary significantly. No linear relation could be found between the intensity value of zenith light as a function of solar elevation (Becker, 1970).

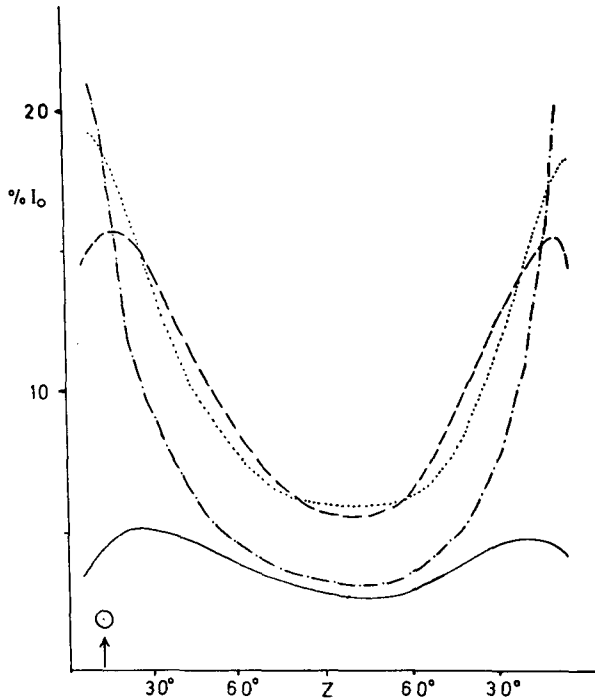


FIG. 7. The Rayleigh atmosphere in the solar vertical for the same conditions and definitions as mentioned in Fig. 5. The sun elevation angle h is 19° .

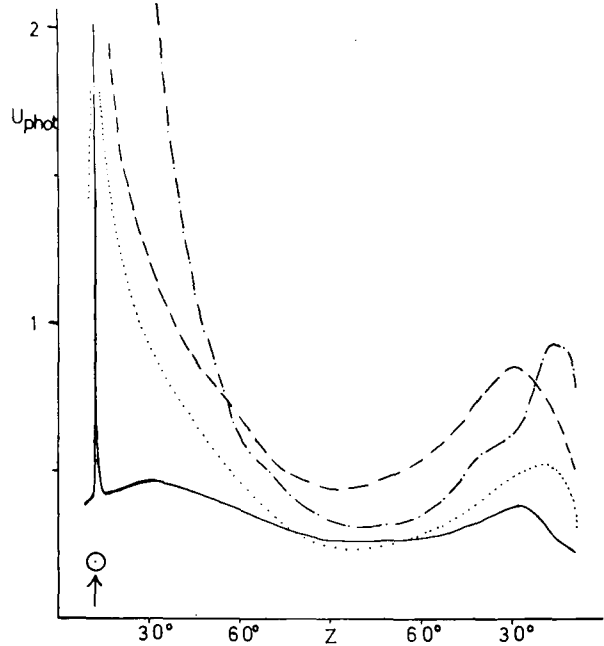


FIG. 8. Measured spectral radiance in the solar vertical expressed in photoelectric voltage units. Curve definitions are same as in Fig. 6 with weather parameters as in Fig. 7.

Fig. 9 presents the structure of sky radiation at 3200 \AA in the solar vertical measured on a clear day with a visibility > 25 km, solar zenith distance $z=49.3^\circ$ and total ozone $\Omega=0.336$ atm-cm (solid curve). The dashed curve represents the measured radiation for the same weather conditions and sun elevation, but for a different ozone amount (0.302 atm-cm). It is interesting

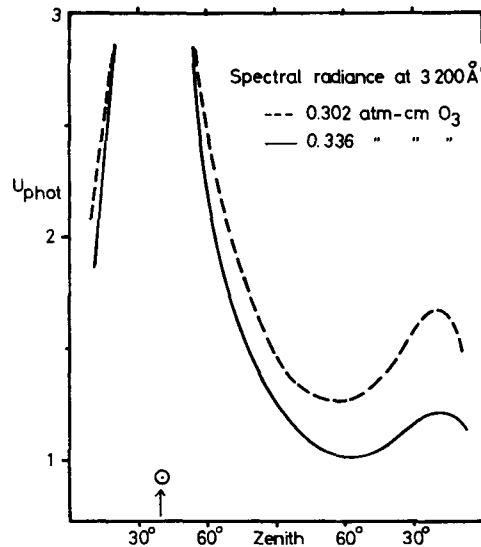


FIG. 9. Measured structure of radiance in the solar vertical for filter 1 (3200 \AA) accounting for different amounts of total ozone on clear days. Solid curve represents the spectral radiance for $\Omega=0.336$ atm-cm and the dashed curve the radiance for $\Omega=0.302$ atm-cm. The solar zenith angle $z=49.3^\circ$, visibility ~ 25 km.

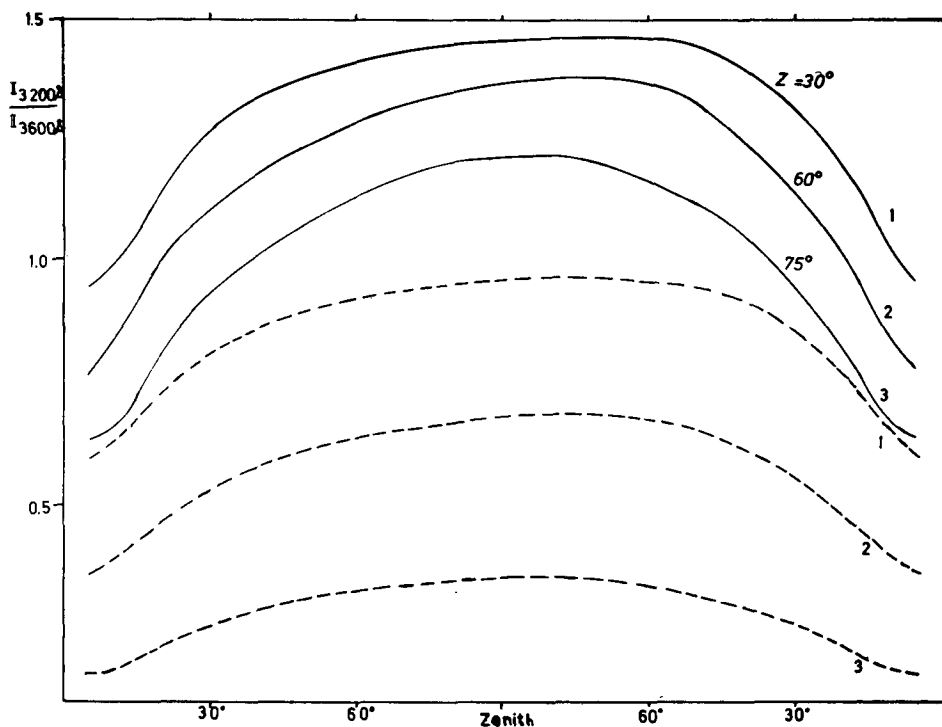


FIG. 10. Computed intensity ratios (I_{3200}/I_{3600}) in the solar vertical for different solar zenith angles z . The solid curves are computed without ozone absorption and the dashed curves include absorption by ozone (0.341 atm-cm).

to note the variation of intensity, especially at the anti-solar side.

Since relative measurements are needed for the deduction of total atmospheric ozone, it is appropriate to consider the ratios of the theoretical Rayleigh radiation intensities as compared to the ratios of measured radiation for the spectral regions involved.

Fig. 10 illustrates the ratio of the theoretical intensities, $I_1:I_2$, as derived for different zenith angles of the sun in the solar vertical. The solid curves represent the pure Rayleigh atmosphere with multiple scattering and without ozone absorption, while the dashed curves include absorption by ozone for 0.341 atm-cm total O_3 .

Significantly, the ozone amount becomes more effective with increasing solar zenith angle due to the increasing length of the ozone path. In other words, the difference between the Rayleigh radiation with and without ozone absorption increases with decreasing solar elevation. This relationship to ozone amount, however, is not apparent in the ratio of the theoretical intensities, $I_{4700\text{\AA}}:I_{3600\text{\AA}}$, as shown in Fig. 11, perhaps as a result of the comparatively much smaller ozone absorption in the Chappuis bands. Strikingly, the ratio becomes convex at the zenith in contrast to the concave nature of the curve in Fig. 10. In predicting total ozone, the ratio $I_1:I_2$ is preferable to the ratio of other wavelengths used, since the transmission maxima of filters 1 and 2 are so close to each other that the effect of scattering is essentially identical for both spectral

regions. These filters have a bandwidth of 180 Å. Furthermore, measurable differences in intensities exist due to large difference in the corresponding absorption coefficients of ozone.

For various amounts of ozone the ratio curves shown in Figs. 10 and 11 can be calculated by relating the different mass absorption coefficients $\tau(z)$ in (8) to the respective ozone densities. As mentioned earlier, the vertical distribution of ozone has little if any effect on the structure of the Rayleigh atmosphere at stratospheric and upper tropospheric levels. Therefore, the calculations could be made applying a simple multiple-layer model of the atmosphere. The balloonborne ozone-sondes ascents made at the Institute of Geophysics and Meteorology, University of Cologne, are being used to parameterize this model; a recent result shows a secondary maxima with high values of total ozone especially during the spring (Paetzold, 1969).

Figs. 12 and 13 compare measured and computed ratios of intensities for constant sun elevations on a clear and cloudless day with visibility > 40 km. Curve 1 is computed without ozone absorption and curve 2 includes ozone absorption (0.341 atm-cm). Curve 3 illustrates the structure of the measured radiation ratio for $\Omega=0.339$ atm-cm. As mentioned above, simultaneous measurements were made with the Dobson instrument to account for the total ozone amount.

Figs. 14a-c show the measured intensity ratios $I_1:I_2$ as compared to those computed for different sun eleva-

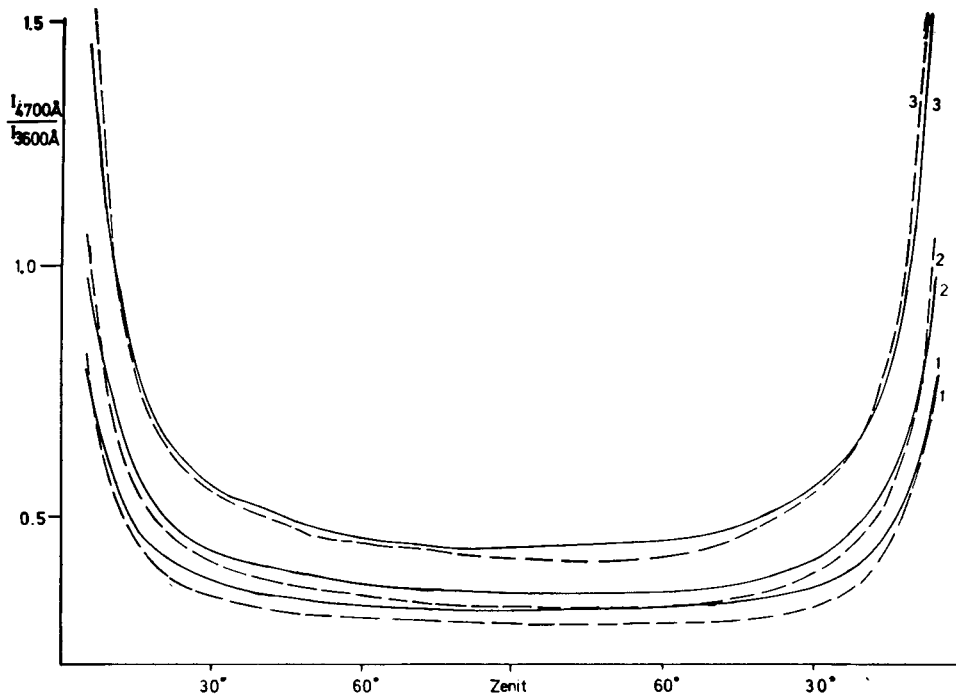


FIG. 11. Theoretical intensity ratios ($I_{4700\text{\AA}}:I_{3600\text{\AA}}$) in the solar vertical for different sun elevations: curve 1, 60° ; 2, 30° ; 3, 15° . Solid curves are for pure Rayleigh atmosphere without ozone absorption and dashed curves include absorption by ozone (0.341 atm-cm).

tions on a clear day with a ozone daily mean value of 0.337 atm-cm. The definitions of the curves are the same as in Fig. 12. It can be seen that the difference between curves 1 and 2 increases with increasing solar zenith distance. A series of such measurements were made for diverse weather conditions. In general, the difference between the measured and computed ratios of the spectral intensities varies with the amount of ozone, cloudiness and the sun elevation. For $35^\circ < z < 60^\circ$ on clear or homogeneously cloudy days, the measured and computed ratios are in good agreement for the same

amount of ozone. The use of filters does not allow satisfactory measurement at greater zenith distances because of the displacement of transmission maxima toward longer wavelengths within the filter band (Bojkov, 1966).

Measurements in the solar vertical for clear days showed that the scattering effects around the sun apparently decrease with increasing solar zenith angle. On the other hand, Mie scattering increases at the anti-

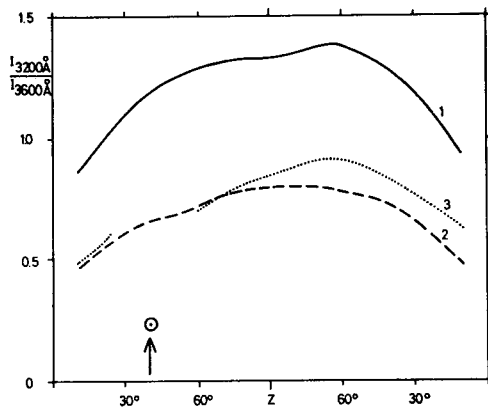


FIG. 12. Theoretical and measured ratio of intensities ($I_{3200\text{\AA}}:I_{3600\text{\AA}}$) in the solar vertical for sun elevation $h=38^\circ$. Curve 1 is computed without ozone absorption, curve 2 includes absorption by 0.341 atm-cm O_3 , while curve 3 presents measured intensity ratio on a clear day with total ozone $\Omega=0.339$ atm-cm.

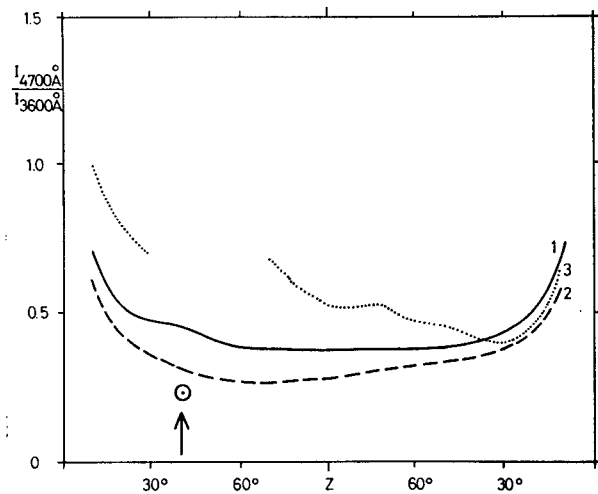


FIG. 13. Theoretical and measured ratio of spectral intensities $I_{4700\text{\AA}}:I_{3600\text{\AA}}$ in the solar vertical for $h=38^\circ$. Curve definitions are the same as in Fig. 12.

solar side. It was found that the intensity value at the zenith is not characteristic of the variation of radiation intensity in the solar vertical.

On cloudy days, comparison of measured ratio $I_1:I_2$ with the computed Rayleigh ratio (each including the same total ozone absorption) shows a discrepancy which could be accounted for by Mie scattering due to clouds. To investigate these effects, one has to assume that the cloud albedo is independent of the wavelength, at least

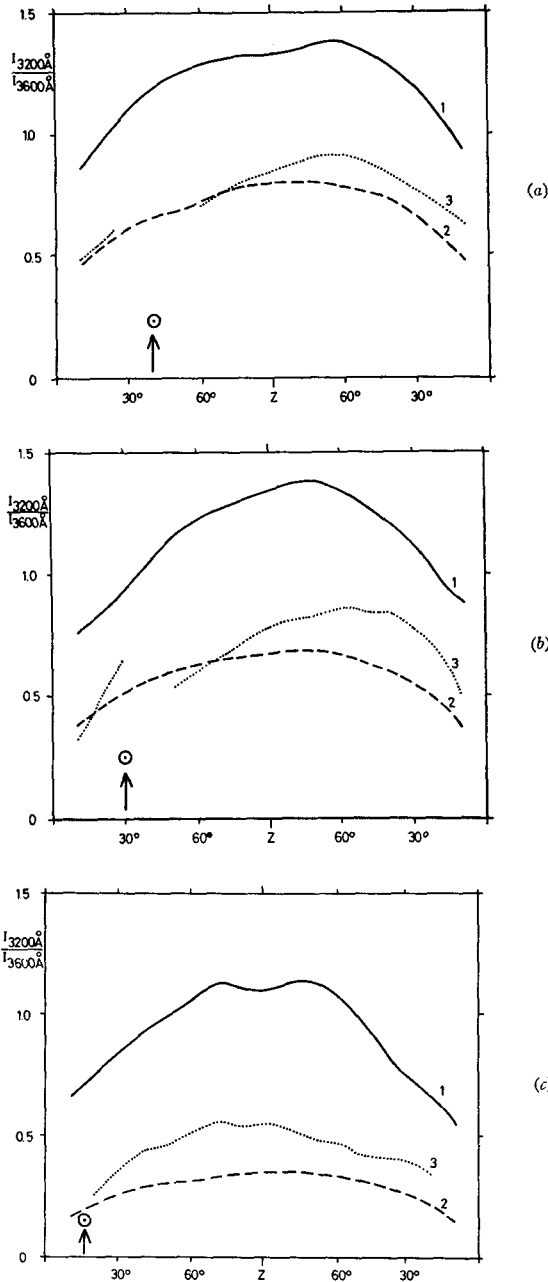


FIG. 14. Theoretical and measured ratio of spectral radiance $I_{3200\text{Å}}:I_{3600\text{Å}}$ in the solar vertical for different sun elevations: a, 38°; b, 30°; c, 17°. Curve definitions are same as in Fig. 12.

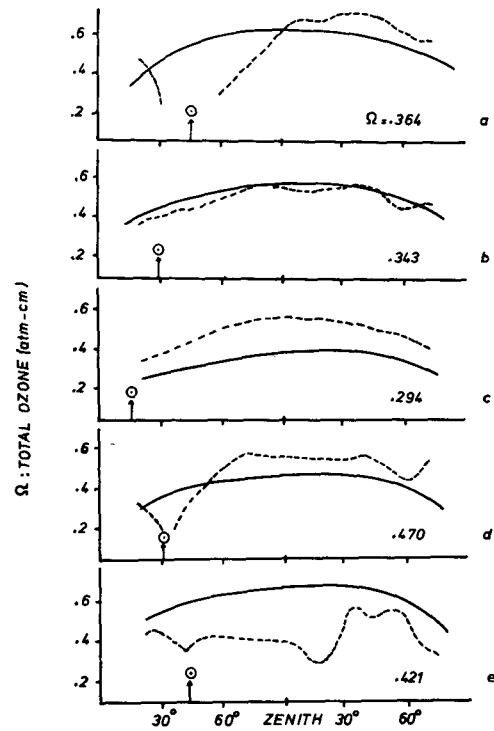


FIG. 15. Some examples of the measured (dashed lines) and reduced intensity ratios $I_{3200\text{Å}}:I_{3600\text{Å}}$ (solid lines) accounting for different amounts of total ozone (see Table 1).

for our limited case. It is then possible to classify the effects of Mie scattering on the measured spectral radiance for different weather conditions. In this manner a measured Rayleigh atmosphere with multiple scattering and ozone absorption can be derived through correction. This would then differ from the theoretical model only by the respective value of total ozone. The ratio of $I_{4700\text{Å}}:I_{3600\text{Å}}$ are also used to determine the referred scattering effects qualitatively. A detailed study in this respect for a series of measurements will be published elsewhere.

Thus, to deduce the total amount of atmospheric ozone, the set of curves as shown in Fig. 10 are used as a basic reduction parameter. The measured ratio is modified according to the sun elevation and cloudiness. Smoothed curves are then compared with those computed for the same zenith angle of the sun. The calculated difference gives the actual amount of ozone in the atmosphere. Although the measured ratio of intensities could be unsteady and irregular in structure, a mean distribution of spectral intensity ratio is derived. Fig. 15 presents some selected examples of the measured and reduced (corrected) intensity ratios $I_1:I_2$ for different amounts of total ozone. The data are given in Table 1 along with the associated cloud conditions.

By comparing our calculated values with the measured total ozone values obtained by a Dobson spectrophotometer (Table 1), we find a reasonable degree of

TABLE 1. Comparison between calculated and Dobson ozone values.

Fig.	Date	Cloud conditions	Total ozone (atm-cm)	
			Calculated	Measured*
15a	13 June 1969	1/8 ci toward SW, blue sky, visibility >20 km	0.364	0.359
15b	5 September 1969	hazy, visibility <6 km	0.343	0.308
15c	19 December 1969	cc, ripples, visibility <5 km	0.294	0.278
15d	4 March 1970	2/8 cs, fibrous clouds	0.470	0.463
15e	25 April 1970	8/8 ns, rain	0.421	0.396

* Using a Dobson spectrophotometer.

correlation. Deviations of the calculated ozone from the standard Dobson values depend on the horizontal visibility. This may be due to a certain dependence of visibility on the wavelength of scattered radiation and the relative changes of visibility for different origins of aerosol as observed by Fimpel *et al.* (1968).

At present, we find the mean error between the calculated and measured total ozone values lies, in general, within $\pm 7\%$ of the average Dobson ozone values. An upper limit can be set to this deviation when a digitized set of such measurements is available.

6. Conclusions

Preliminary measurements and their evaluation have shown that it is possible to measure the total amount of atmospheric ozone with appropriate accuracy on clear and homogeneously overcast days, for solar zenith angles $30^\circ < z < 60^\circ$. To eliminate subjective effects, the measurement and evaluation procedure should be completely digitized. Thus, averaging, smoothing and ratio making, etc., could all be done by computer. In this way the accuracy and density of measurements would be increased and the effects of haze and clouds parameter-

ized. Such measurements would then be of significance in synoptic meteorology.

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