Ozone Abundances in the Lower Mesosphere Deduced from Backscattered Solar Radiances

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ABSTRACT

Backscatter ultraviolet data obtained by the Explorer E satellite imply very large ozone column abundances above 56 km in the tropics during mid-day. The number of molecules in a vertical column decays by a factor of 2–3 after the solar zenith angle exceeds 75° in the evening. An increase of similar magnitude occurs after sunrise. Such behavior implies the presence of a greater source of odd oxygen than is included in current photochemical theories. Ozone profiles deduced between altitudes of 50 and 62 km when the solar zenith angle exceeds 80° are in reasonable agreement with past rocket results.

1. Introduction

Experimental studies of atmospheric ozone over the last decade have led to a greatly increased knowledge of the global distribution and natural variability of this important trace constituent (i.e., Heath *et al.*, 1974; London *et al.*, 1977). Most work, however, has emphasized total ozone or its vertical distribution at stratospheric heights. Since the O_3 concentration at the stratopause is nearly two orders of magnitude smaller than the peak values which occur between altitudes of 20 and 25 km, measurement techniques appropriate in the lower regions are often unsuited to the mesosphere.

Data on the O_3 abundance and temporal variation above 50–55 km are scarce, being attainable only from rocket and satellite platforms. Optical methods based on absorption of the ultraviolet solar irradiance at low to moderate solar zenith angles (SZA's) are not applicable above the 50–55 km region. The peak ozone absorption cross section of 1.12×10^{-21} m^2 at 255 nm (Ackerman, 1971) gives a vertical optical depth of unity between 40 and 45 km, and a value of 0.5 is reached below 50 km. Such methods, however, have provided high quality data in the stratosphere with a vertical resolution superior to that with remote sensing techniques (Krueger, 1973; Krueger and Minzner, 1976). When the sun is near the horizon rocketborne absorption methods can extend the O_3 profile to 70–80 km in altitude (Johnson *et al.*, 1952; Weeks and Smith, 1968; Weeks *et al.*, 1972). Temporal variations in the high altitude profile near sunrise and sunset may influence such results. Chemiluminescent techniques operate up to 70 km and are free from SZA restrictions (Hilsenrath *et al.*, 1969; Hilsenrath, 1971). Most data currently available in the mesosphere were obtained by optical methods.

Satellite measurements at night involving the occultation of bright stars by the earth's atmosphere allow the deduction of O_3 concentrations from 50 km to above 100 km. Although this method does not require an absolute calibration of the irradiance data, the height associated with a given concentration is very sensitive to errors in satellite attitude data. Such effects may be responsible for the large spread between the results of Hays and Roble (1973) and more recent measurements (Riegler *et al.*, 1976; Riegler *et al.*, 1977).

A disadvantage of the above techniques is their inability to be applied on a routine basis. Rockets provide a "snapshot" of the ozone distribution at the time of flight while the precise geometry required for stellar occultation measurements occurs infrequently. The backscatter ultraviolet (BUV) method, however, may be employed on a routine basis. The major disadvantages of the BUV method are that 1) the deduction of O_3 abundances requires an absolute calibration of the instrument and 2) the vertical resolution is limited by the finite width of the contribution functions. The first disadvantage could be eliminated if the instrument directly observed both the solar beam and the backscattered radiance. In practice, currently used instruments deploy a diffuser plate over the field of view when observing the sun. The scattering properties

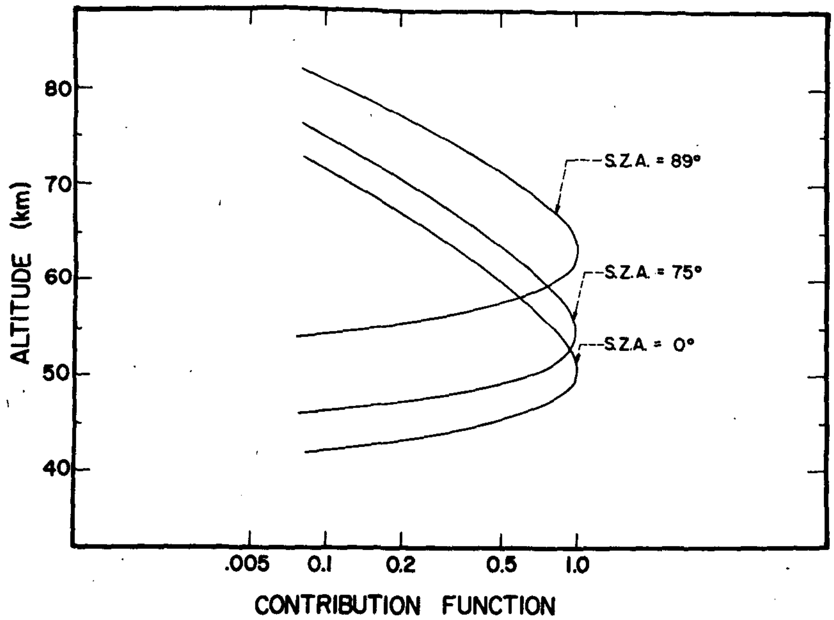

FIG. 1. Altitude variation of the contribution function for the 255.5 nm BUV signal with solar zenith angle. Curves are normalized to unity at the peak.

of this plate introduce a calibration factor not required when measuring a backscattered signal. The second limitation implies that BUV data are well suited to the study of spatial and temporal variations in ozone, but not to resolving finer details of the vertical profiles. Heath *et al.* (1973) have discussed the application of the BUV technique.

We here report an extension of the BUV procedure for monitoring stratospheric O_3 to operate in the lower mesosphere. We then apply the method to tropical data obtained by the Explorer-E satellite near sunrise and sunset to obtain O_3 column abundances above the 0.40 mb pressure surface ($z \approx 56$ km) and, at the highest solar zenith angles, vertical profiles between 50 and 62 km.

2. Data and analysis procedure

Several past analyses of BUV data which have appeared in the literature were based on plane parallel radiative transfer models (Anderson *et al.*, 1969; London *et al.*, 1977). This restricted the maximum SZA's to which the models could be applied to 65°–70°. Since the contribution function for a given backscattered wavelength rises in the atmosphere as the SZA increases, the plane parallel assumption effectively fixes the highest altitude from which reliable O_3 information can be obtained. This upper limit is roughly 55 km. This altitude restriction may be relaxed by generalizing the radiative transfer model. At very high SZA's (>80°) the backscattered radiance at the shortest wavelength, 255.5 nm, has a substantial contribution from the region near and above 60 km, allowing an ozone profile determination in the lower mesosphere. This assumes that the BUV instrument is observing the nadir radiance. In principle it is possible to observe radiation emerging from the atmosphere in directions other than in the vertical and thereby achieve longer absorption paths at smaller SZA's. However, the BUV field of view is quite large, 0.44 sr, and the laboratory calibration assumes a radiation input which is homogeneous across the instrument entrance. This precludes extracting reliable information from scattered radiances measured when viewing other than the nadir direction.

The radiative transfer equation appropriate to the observation of nadir radiances is

$$\frac{I_\lambda}{I_\lambda^0} = (3/16\pi)\beta_\lambda(1+\cos^2\theta)\int_0^{P^*} dP \exp\{-[1+\mathrm{Ch}(H_s,\theta)]\times\beta_\lambda P - [1+\mathrm{Ch}(H_a,\theta)]\sigma_\lambda N_a(P)\}, \quad (1)$$

where the symbols are defined as follows:

I_λ	vertically emergent radiance in the nadir direction at wavelength λ (W m^{-2} nm^{-1} sr^{-1})
I_λ^0	incident solar irradiance
β_λ	Rayleigh scattering coefficient in inverse pressure units
P	atmospheric pressure
θ	solar zenith angle
H_s	average scale height of bulk atmosphere
H_a	average scale height of the absorbing constituent
$\mathrm{Ch}(H,\theta)$	Chapman function for constituent of scale height H

σ_λ ozone absorption cross section (m^2)
P^* pressure at base of atmosphere
$N_a(P)$ number of ozone molecules in a vertical column of unit cross sectional area (m^{-2}) above pressure P.

Eq. (1) assumes that only single scattering occurs. This is an acceptable approximation at the altitudes considered for SZA's <85°. In practice the integrand of (1) becomes negligible at upper stratospheric pressures so that the emergent radiance is insensitive to the value of P^* used in the evaluation. In computing Chapman functions for (1) we use the analytic approximation given by Swider (1964). The 15°N annual model of the *U. S. Standard Atmosphere Supplements, 1966* was used to convert pressure to altitude.

Fig. 1 illustrates the altitude dependence of the contribution function $F_\lambda(\theta,P)$ at a wavelength of 255.5 nm for various solar zenith angles using the mid-latitude ozone model of Krueger and Minzner (1976). The emergent radiance is related to F_λ via

$$I_\lambda(\theta)=\int_{-\infty}^{\ln P^*} d(\ln P)F_\lambda(\theta,P). \quad (2)$$

The corresponding values at a wavelength of 273.5 nm peak 2–4 km lower than those shown, and at 283.0 nm the peak is 5–7 km lower. For SZA's >80° it is possible to obtain useful profile information up to 60–65 km. For all SZA's >60° we are able to report O_3 column abundances above 0.40 mb.

We deduce high altitude O_3 profiles ($z\sim$50–65 km) and column abundances by combining BUV data with calculations based on Eq. (1) and a parameterized ozone profile of the form

$$[O_3(P)]=KP^a, \quad (3)$$

where $[O_3(P)]$ is the ozone number density (m^{-3}) at pressure P. The column abundance above P is $N_a(P)=(H_sK/a)P^a$. The parameter a is the ratio of the bulk atmosphere scale height to that of ozone, i.e., $a=H_s/H_a$. Typical values of a range between 1.0 and 2.0 with the mid-latitude O_3 model of Krueger and Minzer (1976) giving $a\approx1.65$. The parameter K is numerically equal to the O_3 number density at a pressure of 1 mb. For a grid of solar zenith angles between 60° and 90° we compute emergent radiances at the three wavelengths, 255.5, 273.5 and 283.0 nm, for a range of parameters K and a. Comparison of the BUV data with the calculations leads to a set of parameters which reproduces the measured radiances.

A radiance measurement at one wavelength in the nadir direction does not supply sufficient information to deduce a unique O_3 profile. Fig. 2 presents schematic iso-radiance contours as a function of the parameter K in Eq. (3) and the ozone scale height H_a. A family of O_3 profiles for the parameter pairs defined by the curve is capable of reproducing the single-wavelength measurement. If measurements at two wavelengths are available a unique profile determination is possible within the restriction of the assumed analytic form

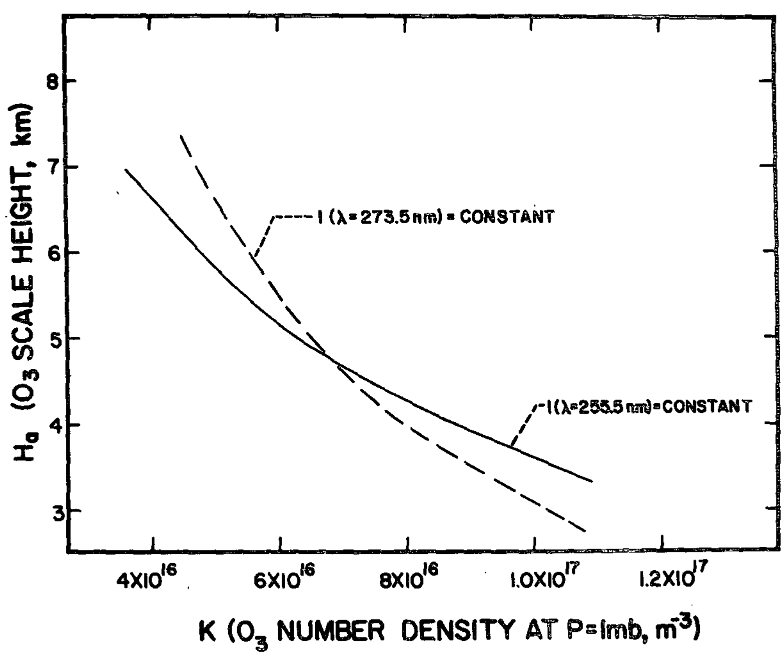

FIG. 2. Schematic iso-radiance contours as a function of O_3 scale height and the parameter K of Eq. (3), physically, the ozone concentration at a pressure of 1 mb. The solid line refers to 255.5 nm and the dashed line to 273.5 nm. The intersection of the two curves defines an ozone profile which reproduces both radiances.

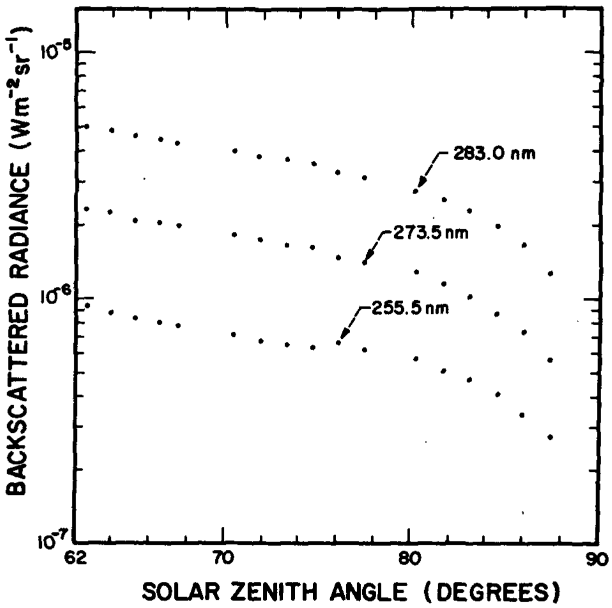

FIG. 3. Backscattered radiance data for wavelengths 255.5, 273.5 and 283.0 nm as a function of solar zenith angle for orbit 971.

[Eq. (3)]. Iso-radiance curves for the two wavelengths in $K-H_a$ (or $K-a$) space then intersect in one and only one point defining an ozone profile which reproduces both of the measured radiances. As a check on the accuracy of the method we have tested two wavelength pairs (255.5, 273.5 nm and 255.5, 283.0 nm) to assure that reasonably consistent results are obtained. Once a final profile is determined, it is used in the transfer equation to compute the emergent radiances. The BUV measurements were always reproduced to better than 2% in this procedure. A calculation of the contribution functions then fixes the altitude region over which the results apply.

During the period from which we take data, Atmosphere Explorer E was in an elliptical orbit with perigee near 150 km. The inclination to the equator was ~20°. Turn-ons of the BUV instrument were centered around perigee, which itself moved rapidly in local time. Hence, series of orbits are available which contain either sunset or sunrise data. Pertinent information for each orbit studied is given in Table 1. To avoid possible contamination from the radiation belts only data obtained when the satellite was below 700 km were analyzed. Fig. 3 presents a set of radiance values for solar zenith angles between 65° and 90°. Note that the points form a reasonably smooth curve indicating a good signal-to-noise ratio even at large SZA's.

TABLE 1. Orbits numbers of the Explorer-E satellite for which mesospheric ozone data have been analyzed.

Orbit	Date	Latitude of perigee	SZA at perigee* (deg)
971	5 February 1976	9.4°N	86.4E
975	5 February 1976	8.5°N	87.3E
977	5 February 1976	8.0°N	87.7E
1503	17 March 1976	0.8°S	138.0M
1680	30 March 1976	13.7°S	83.2M
1686	30 March 1976	12.5°S	85.8M

* M = morning, E = evening.

TABLE 2. Ratios of backscattered radiances measured on Explorer-E to those computed with the Krueger-Minzner ozone model for all solar zenith angles.*

SZA (deg)	$I(\lambda=255.5\text{ nm})\vert_m / I(\lambda=255.5\text{ nm})\vert_c$	$I(\lambda=283.0\text{ nm})\vert_m / I(\lambda=283.0\text{ nm})\vert_c$
60	0.72	0.92
75	0.77	0.97
89	1.01	0.92

* m denotes radiance measured on orbit 971; c denotes radiance computed using the Krueger and Minzner (1976) mid-latitude model.

The raw data consist of currents which must be corrected by a background subtraction and converted to absolute radiances using laboratory derived calibration factors. A dark current measurement is not routinely done in flight since it represents a minor correction at small SZA's where most data are acquired. However, at the large SZA's of interest here it is non-negligible. We have determined dark currents from data taken when the sun is well below the earth's limb and find that they are approximately 20–25% of the total current measured at 255.5 nm at SZA's >85°. To test the sensitivity of the deduced O_3 abundance to this background we performed two calculations for each orbit, one using radiances corrected for the dark current and one using the raw radiances with no subtraction. For SZA's between 75° and 80° the ozone column abundances derived with the correction were 10–13% larger than when the background was ignored. This difference increased to 20–25% at SZA's near 85°.

3. Results

Liu *et al.* (1976) have discussed the present status of photochemical theories in the stratosphere and mesosphere. Current models predict substantially more O_3 above 60–65 km at night and such behavior has been confirmed by rocket experiments (Heath *et al.*, 1974). Prior to sunset computed ozone distributions are essentially time invariant. However, analysis of the short-wavelength BUV data indicates greater temporal variations in the mesosphere than predicted by theory. We have used the mid-latitude O_3 model of Krueger and Minzner (1976) to compute the solar zenith angle variation of the backscattered radiances at 255.5 and 283.0 nm for comparison to the BUV results. Absolute values of the solar irradiance were taken from the measurements of Broadfoot (1972) as compiled with a 1 nm resolution by Donnelly and Pope (1973). This matches the resolution of the BUV instrument. Ozone absorption cross sections were taken from Ackerman (1971). The 283.0 nm contribution function peaks between 40 and 50 km for SZA's between 60° and 90°, while the corresponding altitudes for the 255.5 nm data are 54 to 64 km. Table 2 presents the ratio of the radiance observed on orbit 971 to that computed for various SZA's. At 283.0 nm the radiances derived from theory and measurement are in good agreement and show no obvious variation with SZA. At SZA's < 75° the measured radiance at 255.5 nm is less than the predicted; however, with increasing SZA the measurement becomes comparable to theory based on a time-invariant O_3 profile. Although the absolute values of the ratios depend both on the instrument calibration and on absolute solar irradiance values, the variability of the 255.5 nm signal (lower mesosphere) compared to the 283.0 nm results (upper stratosphere) cannot be altered. The agreement between calculations and BUV at 283.0 nm suggests that the various input parameters are quite accurate. The results of Table 2 imply larger O_3 values in the tropical lower mesosphere during mid-day than predicted by photochemical theories

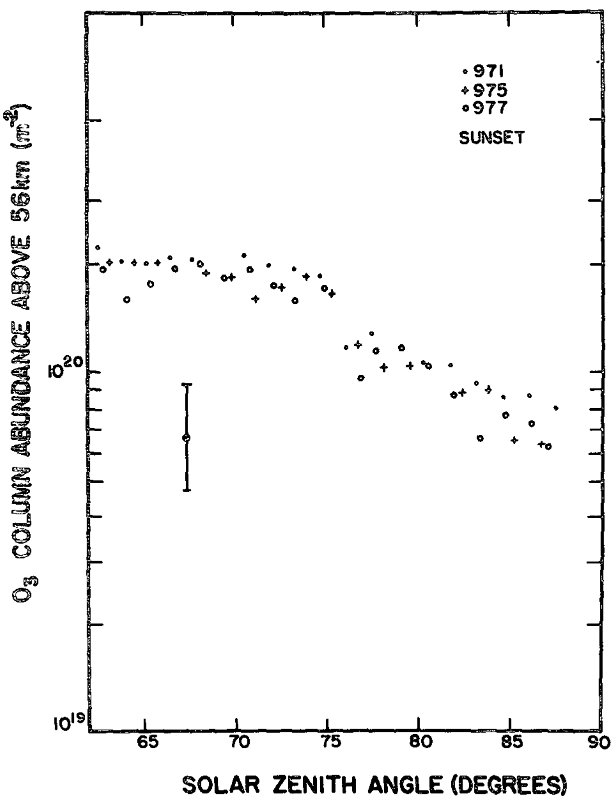

FIG. 4. Ozone column abundances above the 0.40 mb level (~56 km altitude) for evening orbits 971, 975 and 977. The point with error bars is the result of the mid-latitude O_3 model of Krueger and Minzner (1976) and should be compared with the BUV results only at large solar zenith angles.

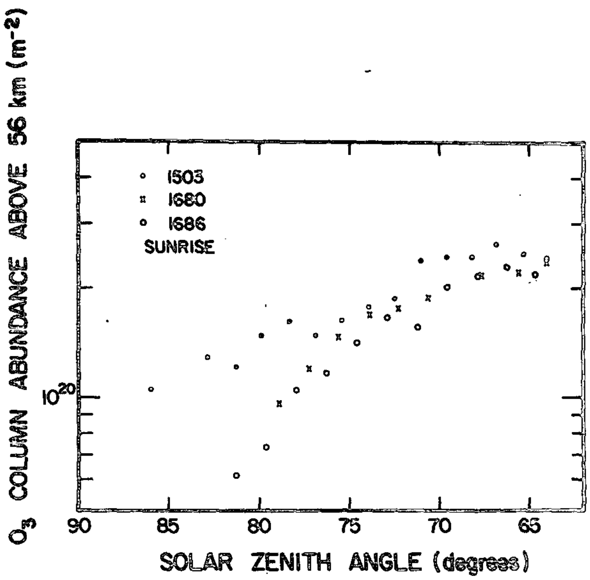

FIG. 5. Ozone column abundances above the 0.40 mb level for morning orbits 1503, 1680 and 1686.

(Liu *et al.*, 1976) and the mid-latitude rocket model (Krueger and Minzner, 1976). As sunset approaches a decrease in the O_3 abundance occurs.

Fig. 4 presents the O_3 column abundance above the 0.40 mb pressure surface (~56 km) as a function of solar zenith angle for orbits 971, 975 and 977. These are representative of measurements near sunset. The results were derived using the wavelength pair (255.5, 273.5 nm) which allows resolution near the 0.40 mb level for any solar zenith angle. The number of O_3 molecules in a column remains constant up to SZA's > 75° where a substantial decrease takes place which continues until sunset. The O_3 decrease is readily visible in the raw radiance data at 255.5 nm but is not obvious in the longer wavelengths (see Fig. 3). On all evening orbits there is a local maximum in the 255.5 nm backscattered radiance despite the increasing SZA near 75°. The signal decreases toward higher SZA's, but less rapidly than predicted for a constant O_3 abundance in the lower mesosphere. The number of O_3 molecules in a column above 56 km at SZA's < 75° is roughly a factor of 3 larger than that given by the mid-latitude rocket model of Krueger and Minzner (1976), denoted by the point with error bars in Fig. 4. However, as noted previously, mesospheric ozone data have come primarily from optical methods applied near sunset. The proper comparison of the BUV results and the rocket model should be made at large SZA's, where they are in good agreement. Fig. 5 presents morning O_3 column abundances above 0.40 mb for orbits 1503, 1680 and 1686. The behavior with solar zenith angle is qualitatively the same as that in the evening. Note, however, that the results for orbit 1503, which were obtained 13 days before the

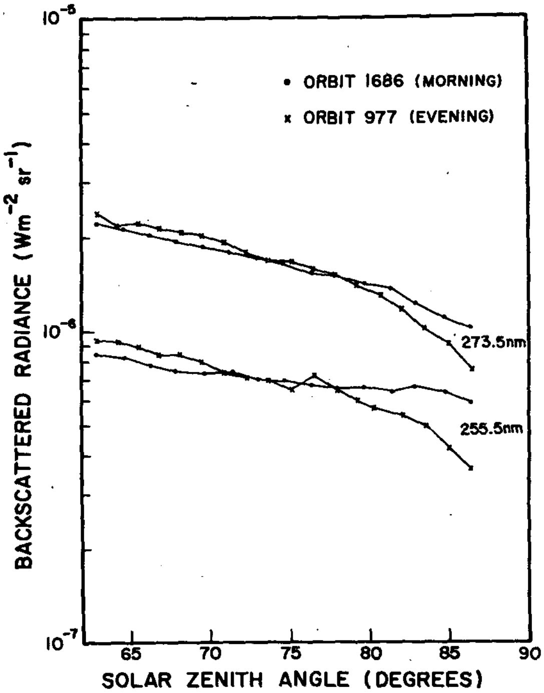

FIG. 6. Comparison of backscattered radiances at 255.5 and 273.5 nm for morning orbit 977 and evening orbit 1686.

other two, show less variation prior to reaching a constant value.

Column abundances derived with the (255.5, 283.0 nm) wavelength pair were approximately 15% less than those reported for the set (255.5, 273.5 nm). This difference is probably due in part to relative calibration uncertainties in the backscattered radiances and solar irradiances, however, a nonconstant ozone scale height would produce a similar result. The (255.5, 283.0 nm) pair predicts a smaller O_3 scale height than the (255.5, 273.5 nm) wavelength set until SZA's near 82° are reached. At larger SZA's the scale heights predicted by the two wavelength pairs are in good agreement. Since the inversion technique assumes a constant absorber scale height, the quantity H_a in Eq. (1) must be interpreted as an average scale height over the altitudes resolved by the wavelength pair being used. Thus, the BUV data imply a larger average O_3 scale height in the mid-day lower mesosphere ($z \approx 55$–60 km) than exists in the upper stratosphere. Data at SZA's >75° produce values of the parameter K in agreement with those obtained at smaller SZA's. The average O_3 scale height is the major variable, decreasing as the SZA increases.

Fig. 6 compares the backscattered radiances of one morning (orbit 1686) and one evening (orbit 977) data set at the two shortest wavelengths monitored. The radiances from different orbits are similar until SZA's between 75° and 80° are reached. The evening data than fall below those in the morning implying more O_3 in the late evening than at comparable SZA's in the early morning for these orbits. At wavelengths >283.0 nm no significant difference between morning and evening exists. Such behavior is consistent with a zenith angle dependent ozone production. After the evening depletion of the unknown source a residual amount of O_3 remains which will decay with some characteristic time constant. In the morning the reappearance of the source leads to a rapid O_3 rise. We note, however, that radiances from the morning orbit 1503 are similar to the evening data so that the morning-evening asymmetry is variable.

For SZA's >80° it is possible to deduce ozone profiles between 50 and 60–65 km. This is the period of temporal change so the results are not representative of mid-day conditions; however, a comparison with twilight rocket data provides a measure of the accuracy of the BUV method under these conditions. Fig. 7 presents the average profile derived from evening data and compares it to the Krueger-Minzner rocket model. The parameters defined by Eq. (3) are $K=(8.26\pm0.24)\times10^{14}$ and $a=1.66\pm0.05$. The error bars are the standard deviations of the mean for the 16 data sets used. The agreement between the two profiles is remarkably good.

The temporal variations found in the BUV data are independent of the instrument calibration and absolute values of the solar irradiance. We have tested the sensitivity of the numerical results to uncertainties in the above quantities by varying the solar irradiance within reasonable limits, ±15%. The mid-day O_3 column abundances changed by no more than 20% in this procedure. We note, however, that use of the Chapman function in (1) implicitly assumes a hor-

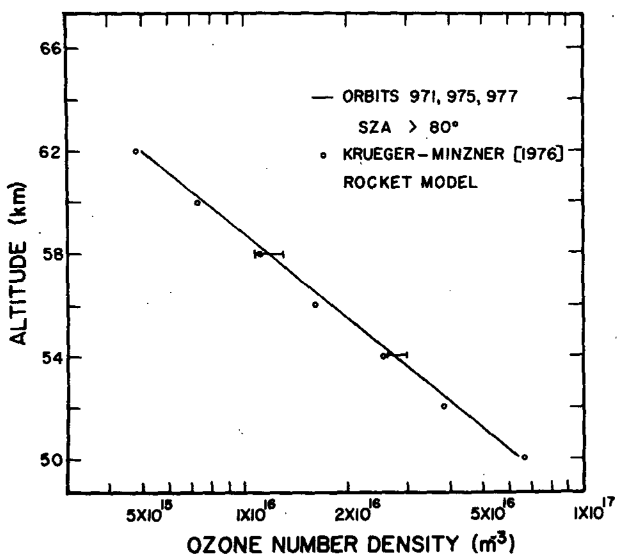

FIG. 7. Average O_3 profile deduced from 16 spectrometer scans at solar zenith angles >80° in the evening. Error bars denote the values deduced from the parameters K and a of Eq. (3) ±1 standard deviation of the mean. Open circles give the mid-latitude rocket model results of Krueger and Minzner (1976).

izontally homogeneous ozone distribution, a condition that is clearly violated by the results of Figs. 4 and 5. The O_3 abundance at a given altitude is larger along the path of the incident solar beam than on the vertical path up to the BUV instrument. The deduced O_3 abundance is therefore characteristic of a slightly smaller SZA than that at the scattering altitude. The accuracy of Eq. (1) depends on the magnitude of the variation in SZA along the path of the incoming radiation. The bulk of the absorption occurs within one to two ozone scales heights of the point of scattering, a vertical distance of less than 10 km. However, the SZA at the scattering point differs from that 10 km higher in altitude, measured along the path of the incoming solar beam, by less than 1° until values above 85° are reached. Hence, negligible error is introduced by the use of Eq. (1). At SZA's >85° the error is such as to dampen the actual O_3 variation. In addition, the effects of multiple scattering become significant under these observing conditions.

4. Summary and conclusions

Backscatter ultraviolet observations of the ozone column abundance above 0.40 mb reveal larger values than predicted by current photochemical models when the solar zenith angle is less than 75° (Liu *et al*., 1976; Liu and Cicerone, 1977; Sze, 1977). At higher sun angles the column density decays, being a factor of 2–3 smaller at sunset than at earlier times of the day. An increase of similar magnitude occurs after sunrise. Such behavior is contrary to theoretical predictions (Park and London, 1974). This suggests the presence of a very large production of odd oxygen in the mesosphere. The results of this study require a source which is very sensitive to the solar zenith angle, being inoperable at night.

As a potential explanation of the large daytime O_3 concentrations we propose the following. A major odd oxygen source in the mesosphere is the predissociation of O_2 in the Schumann-Runge bands. While a calculation of the dissociation rate is complicated by the highly structured cross sections (Ackerman *et al*., 1970), an additional problem concerns the fine structure of the solar spectrum between 175.0 and 200.0 nm. Inspection of high-resolution spectra reveals irradiance variations of a factor of 2–3 over wavelength intervals of order 0.01 nm wide (Moe *et al*., 1976). This latter difficulty has not been accounted for in aeronomic calculations but could significantly alter the numerical results. We therefore suggest that the inclusion of a detailed solar spectrum would lead to a larger mesospheric O_2 dissociation rate. We are presently investigating this possibility. Alternate odd oxygen sources may involve reactions of metastable species. The BUV results from the Explorer E are restricted to the tropical regions; however, the same photochemical mechanisms surely operate elsewhere, although perhaps with altered magnitudes. For the hypothesized mechanism, the O_3 increase is a maximum in the tropics.

Note added in proof: Since the acceptance of this paper we have carried out a series of atmospheric opacity calculations for the wavelength region 182.5–184.5 nm, in the Schumann-Runge bands, using both low- and high-resolution solar spectra. We find that the two sets of results differ by less than 10% even though the solar irradiance at different wavelengths varied by more than a factor of 2. It is reasonable to suppose that a similar result would be obtained for the O_2 dissociation rate including the entire wavelength range of the Schumann-Runge bands.

An alternate source of odd oxygen involving $O_2(^1\Delta_g)$ has been suggested to the principal author by R. J. Cicerone. If the photodissociation frequency of $O_2(^1\Delta_g)$ at 60 km altitude were $\sim 5\times10^{-5}$–1×10^{-4} s^{-1} the resulting odd oxygen source would be comparable to that derived from dissociation of ground state O_2. In view of its potential importance, a laboratory investigation of this process is warranted.

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