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NEAR-INFRARED SPECTROMETER, TWILIGHT, ATMOSPHERE, PROFILES, LATITUDE, NIGHT, BAND

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Mesospheric ozone concentration at an equatorial location from the 1.27-μm O_2 airglow emission

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Abstract. The vertical emission profile of the O_2 ($a^1\Delta_g$) airglow at 1.27 μm has been measured during the evening twilight near the equator (Alcântara, Brazil; 2.5°S, 44.2°W) using a rocket-borne infrared photometer. The profile is used to derive the vertical distribution of ozone between 60 and 86 km. The ozone profile shows a minimum concentration at 77 km and a secondary maximum of 1.34×10^8 cm^{-3} at 82 km. The height of the secondary maximum is lower than observed at midlatitudes, and the 77-km minimum is sharper than that which has been observed at low latitudes by satellite measurements.

Introduction

Measurements of the ozone distribution in the troposphere and stratosphere have received increased attention in recent years because of concern about ozone destruction by anthropogenic clorofluorocarbon (CFC) gases. In spite of the much lower density of ozone in the mesosphere and lower thermosphere, its measurement is also important because of the fundamental role it plays in the aeronomy of this region. Mesospheric ozone profiles have been measured up to 60 km by the stellar occultation technique with instruments on board sounding rockets [*Miller and Ryder*, 1973; *Weeks et al.*, 1978]. This technique does not work at greater heights because of the low level of solar UV absorption. The photolysis of ozone by the solar ultraviolet radiation in the Hartley band (200-350 nm) yields O_2 excited in the $a^1\Delta_g$ state which decays to the $X^3\Sigma_g$ state in the vibrational levels $v' = 0$ and 1, producing emissions in the infrared atmospheric system (IAS) at 1.27 μm and 1.58 μm, respectively. The first measurements of the IAS from ground-based photometers were made using the 1.58-μm band by *Vallance-Jones and Harrison* [1958], and the first measurement of the height distribution of the IAS at 1.27 μm was made during daytime by *Evans et al.* [1968] by means of a rocket launched at White Sands (33° N). These rocket-borne measurements of the 1.27-μm emission have been used to infer the ozone density profile during daytime and twilight from 60 to 100 km [*Evans and Llewellyn*, 1970, 1972; *Evans et al.*, 1988]. Most of these rocket-borne measurements of the ozone height distribution have been obtained at middle and high latitudes [*Llewellyn and Witt*, 1977; *Makino et al.*, 1979; *Yamamoto et al.*, 1983]. Also, satellite-borne long-path limb measurements of the 1.27-μm emission and the corresponding ozone profile have been obtained over a wide range of latitudes [*Thomas et al.*, 1983,1984]. In this paper, we report the first rocket-borne measurement of the 1.27-μm IAS emission at an equatorial latitude and determine the corresponding ozone altitude profile from 60 to 86 km.

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Instrumentation

A single-channel infrared photometer of the kind described by *Llewellyn and Witt* [1977] was mounted with its optical axis parallel to a Sonda II rocket. This radiometer used an uncooled chopped lead sulfide detector with phase-sensitive detection, equipped with an optical interference filter centered on the O_2 IR atmospheric (0-0) band at 1.27 μm with 17 nm full width at half maximum.

The experiment was part of a cooperative program between the Brazilian National Space Research Institute (INPE) and the Aeronautics and Space Institute (IAE). The rocket was launched on December 9, 1991, from Alcântara (2.5°S, 44.2°W) at 1805 LT (2105 UT). Sunset time at the apogee was at 1756 LT, and when the rocket reached this altitude, the solar depression angle was 2.5°. Relevant parameters for the rocket flight are shown in Table 1. Absolute laboratory calibration of the radiometer was carried out before the flight, and an in-flight check was performed near apogee by means of an internal lamp in the photometer field of view. Data were obtained only during the ascent, since the rocket started to tumble shortly after reaching apogee at 89 km.

Data Reduction

As a first step in the data reduction, the data received at a high transmission rate were averaged in 0.4-s intervals. With this process, the height interval is about 300 m at 60 km and less than 100 m above 85 km. The overhead intensity was corrected to the zenith by a simple cosine factor. The result is shown as individual points plotted in Figure 1. The continuous curve shown in this figure was obtained using the incremental straight-line fitting technique of *Murtagh et al.*[1984], with a 7-km fitting length. This height interval was chosen because it provides good noise reduction while preserving the real structure in the profile. As seen in the

Table 1. Data for the Rocket Flight

Parameter	
Launch site	Alcântara,Brazil(2.5°S,44.5°W)
Time (UT)	2105
Date	December 9, 1991
Apogee	89 km
Solar zenith angle	92.5°

figure, an overhead intensity of 3000 kR is obtained at 60 km for the IAS (0-0) band.

To obtain the volume emission profile, the curve in Figure 1 was differentiated at 1-km intervals using the slopes provided by the incremental straight-line fitting method. This volume emission profile is shown in Figure 2. Error bars represent the standard errors of the estimate in the least mean square's determination of the slope of each straight line. The ozone concentration can be obtained from the measured volume emission rates using a simplified photochemical model described by *Thomas et al.* [1984]. Recently, *Mlynczak et al.* [1993] have considered a more complete kinetic model for the daytime steady state O_2 ($a^1\Delta_g$), taking into account the photolysis of O_2. They conclude that inferred daytime profiles using only the O_3 photolysis may be overestimated by as much as 10% in the lower mesosphere and underestimated by as much as 20% in the lower thermosphere, depending on solar cycle, latitude, and season. In this paper, we will use the same kinetic model and constants as *Thomas et al.* [1984]. The difference between the results of this analysis and that of *Mlynczak et al.* [1993] will subsequently be shown.

The production of O_2 ($a^1\Delta_g$) during daytime is mainly through ozone photodissociation.

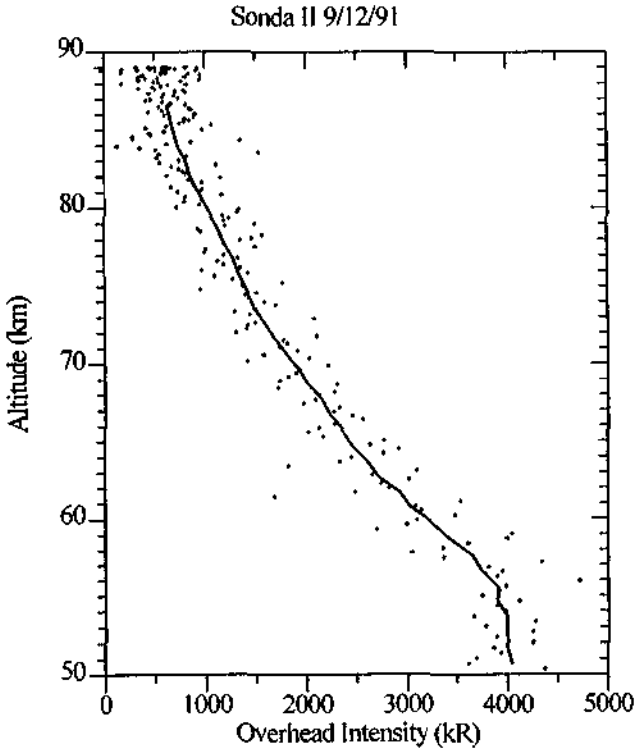

Figure 1. The $O_2(a^1\Delta_g)$ overhead emission corrected to zenith.

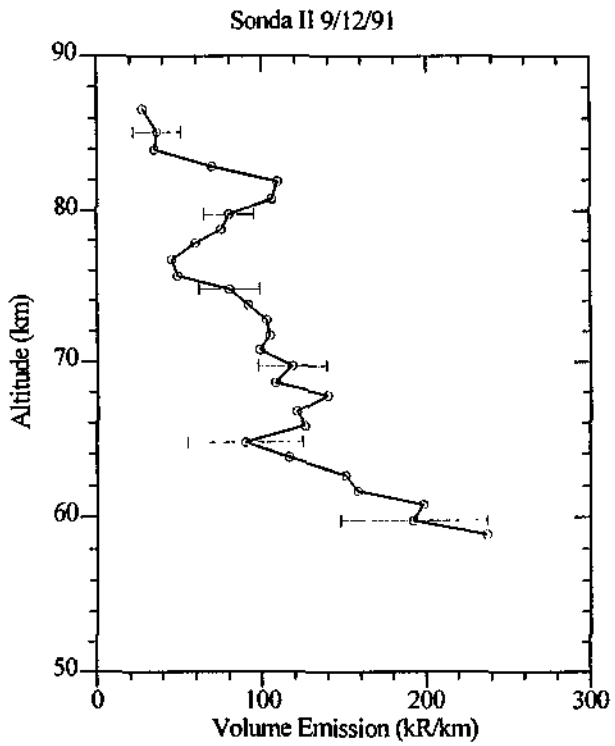

Figure 2. The $O_2(a^1\Delta_g)$ volume emission height profile derived from Figure 1.

$$O_3 + h\nu\ (210\text{-}310\ nm) \rightarrow O_2(a^1\Delta g) + O(^1D),\quad \varepsilon J_3 \quad (1)$$

However, for more accurate derivation of ozone densities, other sources of O_2 ($a^1\Delta_g$) should be taken into account. These are the deexcitation of the O_2 ($^1\Sigma$) molecules,

$$O_2(^1\Sigma) + M \rightarrow O_2(a^1\Delta_g) + M, \qquad k_s \qquad (2)$$

which are partially produced by transfer from the $O(^1D)$ state

$$O(^1D) + O_2 \rightarrow O(^3P) + O_2(^1\Sigma), \qquad k_0 \qquad (3)$$

or, mainly at high altitudes, by the resonance absorption of sunlight,

$$O_2 + h\nu\ (762\ nm) \rightarrow O_2(^1\Sigma), \qquad J_s \qquad (4)$$

The $O(^1D)$ atom can also be quenched by N_2,

$$O(^1D) + N_2 \rightarrow O(^3P) + N_2, \qquad k_n \qquad (5)$$

and the O_2 ($a^1\Delta_g$) can either be quenched by O_2 or radiate,

$$O_2(a^1\Delta_g) + O_2 \rightarrow 2\,O_2, \qquad k_D \qquad (6)$$

$$O_2(a^1\Delta_g) \rightarrow O_2 + h\nu\ (1.27\ \mu m), \qquad A_D \qquad (7)$$

Therefore, for the sunlit atmosphere, steady state can be assumed, and the emission rate is given by

$$V(h) = \left\{\frac{P_s\ k_s\ [M]}{A_s + k_s\ [M]} + J_3\varepsilon\ [O_3]\right\} \times \frac{A_D}{A_D + k_D\ [O_2]} \quad (8)$$

where A_D and A_s are the Einstein coefficients for O_2 ($a^1\Delta_g$) and O_2 ($^1\Sigma$), respectively. P_s is the production rate of the O_2 ($^1\Sigma$), given by

$$Ps = J_s[O_2] + J_3\,\varepsilon\,[O_3]\,k_0/(k_0 + 3.76\,k_n). \quad (9)$$

For the conditions of our experiment (low depression angle of 2.5^0), full illumination is still a good approximation for heights above ≈75 km, and the steady state formula (8) can be used to infer $[O_3]$. For lower altitudes, the reduction of solar flux and deviation from steady state should be taken into account. Complete assessment of the changing solar flux over the $O_2(a^1\Delta_g)$ and O_3 concentration can only be made with a complete time-evolution model [*López-Gonzáles et al.*, 1989, 1992], but for altitudes below ≈75 km and low depression angle a much simpler photochemical scheme, as that used by *Llewellyn and Witt* [1977], can be used. In this approximation, only equations (1), (6), and (7) are considered, and a simple linear equation for the change of the metastable oxygen concentration is obtained:

$$d/dt\,[O_2(a^1\Delta_g)] = \varepsilon J_3[O_3] - (k_D[O_2] + A_D)[O_2(a^1\Delta_g)]. \quad (10)$$

To solve equation (10), it is assumed that the $[O_3]$ profile does not change with time. In this case, the equation can be integrated knowing the time variation of J_3. Time variations for these photodissociation coefficients were calculated for the launch conditions at each height. For integration purposes, a trapezoidal approximation [*Plemel*, 1974; *Llewellyn and Witt*, 1977] was fitted to the calculated $J_3(t)$ at each height, J_{30} being the constant daytime value, t_0 the time at which J_3 starts to decrease from its constant daytime value, and t_1 the time when J goes to zero. When this trapezoidal approximation is used for J_3 at each height, a simple analytical solution is obtained for $[O_2(a^1\Delta_g)]$ for the interval $t_0 \le t \le t_1$.

$$[O_2(a^1\Delta_g)](t) = [O_2(a^1\Delta_g)](t_0) \times \left(\frac{t_1 - t + \tau\,(1 - e^{-(t-t_0)/\tau})}{t_1 - t_0}\right) \quad (11)$$

where $\tau = (k_D[O_2] + A_D)^{-1}$. For this calculation, the atmosphere was divided in 1-km shells, and a ray path calculation was used to evaluate the O_2 and O_3 column concentrations. Solar flux values were from *Mount and Rothman* [1983], and the cross sections for O_3 and O_2 are from *Ackerman*[1971]. The required vertical distributions of O_2 and O_3 are from *Barnett and Corney* [1985] and *Keating and Young* [1985], respectively. It should be noted that the O_3 distribution assumed in order to calculate the J_3's has only a second-order effect on the derived ozone profile.

The second factor in the right-hand side of equation (11) permits us to correct the $[O_2(a^1\Delta_g)]$ (or the corresponding V(h)) obtained at a time after the sunset to its daytime value. This factor is 1 above ≈73 and decreases quickly to a value of 0.2 at 60 km. Using (8), after correcting V(h) by the factor in (11), the result for the O_3 profile is shown in Figure 3 (solid line). Rate coefficients and transition probabilities are those given by *Thomas et al.* [1984]. The error bars reflect the uncertainty in the measured $[O_2(a^1\Delta_g)]$ profile. Minimum ozone density occurs at 77 km, and a local maximum is observed at 82 km.

The effect of using the updated $O_2(a^1\Delta_g)$ model given by *Mlynczak et al.* [1993] in the O_3 profile retrieval is shown as a dotted line in Figure 3. It can be seen that use of the *Mlynczak et al.* [1993] model results in only a very small change in the ozone profile, a change which is in fact much smaller than the experimental error.

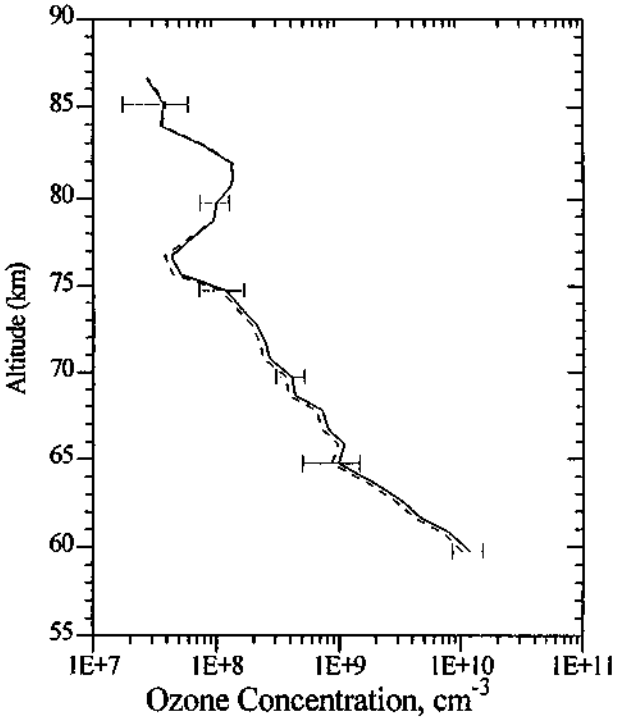

Figure 3. Ozone height profile derived from the $O_2(a^1\Delta_g)$ profile of Figure 2. The solid curve uses the kinetic scheme of *Thomas et al.*[1983,1984], and the dashed curve uses *Mlynczak et al.*'s [1993] updated model (see text).

Discussion

In Figure 4, our measurement is compared with other data obtained at different latitudes, seasons, and solar zenith angles. Below 74 km, a range for the ozone concentration at middle and high northern latitudes is given in the model of *Krueger and Minzner* [1976], based on a large number of rocket measurements. It is seen that most of our data points fall within their 1-σ limit. The profiles of *Evans and Llewellyn* [1973] and *Llewellyn and Witt* [1977], obtained at high-latitude stations (Churchill (58.7^0 N) and Kiruna (67.9^0 N)) at solar zenith angle of 93^0 and 94.1^0, respectively, are also shown in the figure. The main difference between these measurements and ours is that for our data, the minimum occurs at 77 km and the maximum at 82 km, whereas for the high-latitude measurements, the minima are around 80 km and the maxima 85 km. This difference is not surprising in view of the large separations in latitude and time between the profiles.

Mesospheric ozone profiles at lower latitudes have been obtained by limb scanning of the 1.27-μm dayglow with an instrument on board the Solar Mesosphere Explorer (SME) satellite [*Thomas et al.*, 1983, 1984]. Monthly means of these ozone measurements for several latitudes are given by *Keating and Young* [1985]. These measurements for December and the equator are also plotted in Figure 4. It is interesting to observe that the SME measurements for low latitudes also show the ozone minimum occurring below 80 km and the local maximum slightly above 80 km. In our data, the minimum and maximum structures are more pronounced. This is not unexpected, since the rocket measurement represents an instantaneous picture of the ozone profile, while

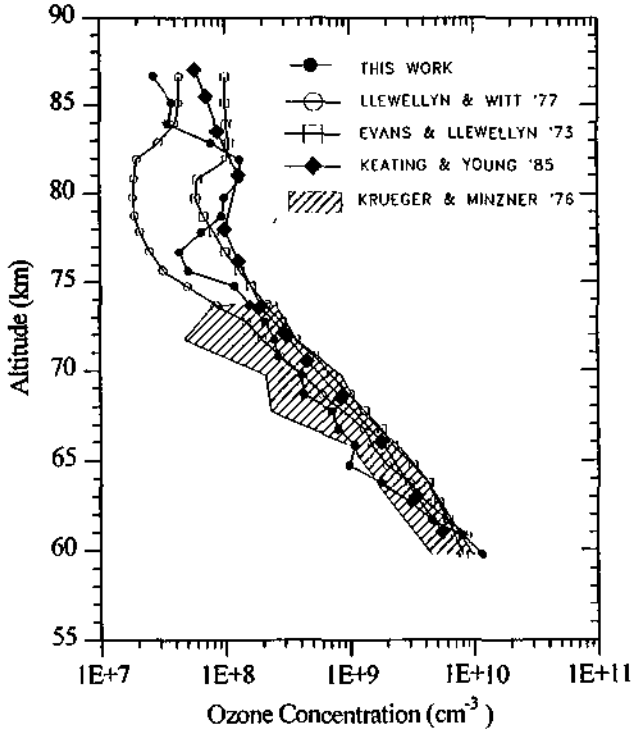

Figure 4. The deduced ozone concentration profile compared with other observations.

the satellite data represent a monthly mean integrated over a long horizontal path.

The decrease in height of the secondary ozone peak observed at low latitudes is worthy of discussion. Since the secondary peak results from a balance between the vertical transport of atomic oxygen from above and atomic hydrogen from below, the present results indicate that this balance is different at low latitudes. Our ground-based airglow observations carried out near the equator (Fortaleza; 4°S, 45°W) show a strong semiannual oscillation of the mesospheric atomic-oxygen-related O I 557.7 nm, NaD and OH emissions [*Takahashi et al.*, 1994], with equinoctial maxima and minima at the solstices. This suggests that there is increased vertical transport of atomic oxygen at the solstices, provoking more rapid chemical loss of atomic oxygen, with the result that atomic oxygen is depleted at heights above about 85 km [*Garcia and Solomon*, 1985]. A rocket observation of the O I 557.7 nm emission profile carried out from Natal (5.8°S, 35.2°W) on December 9, 1985 [*Gobbi et al.*, 1992], indicated a height of 96 km for the peak of the atomic oxygen distribution, appreciably lower than that measured at midlatitudes. This supports the idea of strong downward transport of atomic oxygen during solstices in the equatorial region. The present measurement was carried out close to the December solstice. The lowering of the secondary peak of ozone could be a consequence of this increased vertical transport.

The absolute values of the derived ozone concentration depend, of course, on the kinetic and spectroscopic constants used in the analysis. *Mlynczak and Olander* [1995] suggest that the present uncertainties in these constants lead to an uncertainty of about 15%. The Einstein A-coefficient used in the present work was derived by *Badger et al.* [1965]. *Mlynczak and Nesbitt* [1995] reanalyzed recent measurements of the absorption strength published by *Hsu et al.* [1992] and arrived at the conclusion that the Einstein A-coefficient should be a factor of 1.75 smaller than that of *Badger et al.* [1965]. In the same paper, however, *Mlynczak and Nesbitt* [1995, p.1384] comment that "Since acceptance of this paper, the existence of unpublished laboratory and atmospheric absorption data which suggests an absorption band strength in closer agreement with B-value reported by Badger et al. has been brought to our attention, thus underscoring the need for additional in-depth study of this system." In view of the doubt which this statement throws on the new value for the A-coefficient, in the present work, for the purpose of comparison with other measurements, we preferred the use of the earlier value. If new laboratory measurements confirm significant changes in the kinetic and spectroscopic constants, the same airglow intensities can always be reprocessed in order to update the data basis.

Summary and Conclusions

The vertical profile of the $O_2(a^1\Delta_g)$ infrared atmospheric emission at 1.27 μm was measured at evening twilight near the equator using a rocket-borne photometer. This emission was used to infer the ozone profile from 60 to 86 km. The derived O_3 densities are similar to those observed at higher latitudes at heights up to 75 km. At greater heights, the measurement showed a local minimum of 4.31×10^7 cm^{-3} at 77 km and a local maximum of 1.34×10^8 cm^{-3} at about 82 km, both heights being several kilometers lower than those observed at higher latitudes. The SME ozone profiles for low latitudes show maxima and minima much less pronounced than those visible in our profile, but at similar heights.

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