

Use of O₂ airglow for calibrating direct atomic oxygen measurements from sounding rockets

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Abstract. Accurate knowledge about the distribution of atomic oxygen is crucial for many studies of the mesosphere and lower thermosphere. Direct measurements of atomic oxygen by the resonance fluorescence technique at 130 nm have been made from many sounding rocket payloads in the past. This measurement technique yields atomic oxygen profiles with good sensitivity and altitude resolution. However, accuracy is a problem as calibration and aerodynamics make the quantitative analysis challenging. Most often, accuracies better than a factor 2 are not to be expected from direct atomic oxygen measurements. As an example, we present results from the NLTE (Non Local Thermodynamic Equilibrium) sounding rocket campaign at Esrange, Sweden, in 1998, with simultaneous O₂ airglow and O resonance fluorescence measurements. O number densities are found to be consistent with the nightglow analysis, but only within the uncertainty limits of the resonance fluorescence technique. Based on these results, we here describe how better atomic oxygen number densities can be obtained by calibrating direct techniques with complementary airglow photometer measurements and detailed aerodynamic analysis. Night-time direct O measurements can be complemented by photometric detection of the O₂ ($b^1\Sigma_g^+ - X^3\Sigma_g^-$) Atmospheric Band at 762 nm, while during daytime the O₂ ($a^1\Delta_g - X^3\Sigma_g^-$) Infrared Atmospheric Band at 1.27 μm can be used. The combination of a photometer and a rather simple resonance fluorescence probe can provide atomic oxygen profiles with both good accuracy and good height resolution.

1 Introduction

Atomic oxygen is the major carrier of chemical energy in the Earth's mesosphere and lower thermosphere (MLT) and is thus a key component for the aeronomy of this region. It is formed by O₂ photolysis mainly in the Schumann-Runge continuum and Schumann-Runge bands in the thermosphere and mesosphere, respectively, and at lower altitudes in the Herzberg continuum (Brasseur and Solomon, 2005). In the thermosphere the chemical lifetime of O is long (months, even years) and the local abundance is governed by dynamic control. Here O can be transported horizontally or vertically and the deposition of the stored chemical energy can take place at large spatial or temporal distances from the sources (Rees and Fuller-Rowell, 1988). In the mesosphere the local abundance of atomic oxygen is governed by chemical control through the O_x and HO_x cycles. Here the lifetime of O is only of the order of hours because of the relatively high molecular collision and reaction rates in this denser part of the upper atmosphere. Around the mesopause, in-between these two regions, the chemical and transport lifetimes are of similar magnitude and the local abundance depends on the interplay of the different control mechanisms. The transport processes involved in redistributing O in the mesopause region are long-distance transport by the global meridional circulation with ascending and descending motion in summer and winter, respectively, variations due to tidal and wave motions on various scales, as well as exchange by turbulent diffusion (Liu et al., 2008; Russell et al., 2005)

Accurate knowledge about the vertical distribution of O is crucial for any comprehensive study of chemical and dynamical processes in the MLT region. Measurements need to be both accurate and of good height resolution. Absolute profiles of atomic oxygen have been obtained in situ by means of rocket-borne experiments since the early 1960's. In the beginning, mass spectrometry was the dominant method for atomic oxygen measurements but since the early 1970's



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the resonance fluorescence technique using the O (³S–³P) transition at 130 nm has taken over this role and is the only technique in use today (Dickinson et al., 1980; Sharp, 1991). Much debate has been going on about the reliability of in situ measurements by means of rocket-borne techniques (Gumbel, 1999). The existing measurements reported by different research groups show consistency in the general shape of the profiles, but little consistency in the absolute densities. The variability in the measurements is thought to be mostly of instrumental nature and not necessarily geophysical (e.g. Gumbel, 1997). Possibly the most accurate technique applied so far are mass spectrometers based on cryogenic removal of shock effects, with a stated accuracy of better than 50% (Offermann et al., 1981). Unfortunately, this technique is expensive and no longer in use today. A general review of direct atomic oxygen measurements in the mesosphere and lower thermosphere is in preparation. Major challenges to a quantitative analysis are the calibration and aerodynamics of the experiments. It is therefore desirable to define complementary calibration procedures that are independent of the instrument characteristics of the specific direct O measurement. In this paper, we discuss such calibration procedures in terms of photometric airglow measurements.

In the mesopause region the recombination of atomic oxygen generates molecular oxygen in a number of metastable states. Through a number of energy transfer and quenching processes the excited O₂ then gives rise to nightglow emissions covering the spectral range from the ultraviolet to the infrared (Meriwether, 1989; Wayne, 2000). These processes cause a complex and strongly altitude dependent photochemistry between about 85 and 105 km. Also energy transfer to atomic oxygen (Barth and Hildebrandt, 1961) and reactions between ozone and hydrogen forming vibrationally excited OH molecules (Bates and Nicolet, 1950) contribute to the nightglow emissions with different peak altitudes depending on the details of the photochemistry.

It is generally accepted that the excitation energy originates from the recombination of O, but it took long time to establish the exact mechanisms and reaction rates, and some have still not been established. There have been several rocket experiments in the past with the objective to simultaneously study atomic oxygen density and oxygen airglow intensity (e.g. Sharp, 1980; Thomas, 1981; McDade et al., 1984). However the most extensive series of such multi-instrumented rocket studies was the set of campaigns with OXYGEN/S35 in March 1981 (Witt et al., 1984) followed by the ETON (Energy Transfer in the Oxygen Nightglow) venture conducted in March 1982 from South Uist, UK (Greer et al., 1986) and OASIS (Oxygen Atom Studies In Space) conducted from White Sands in June 1983 (Murtagh et al., 1990). The ETON campaign was a multi-rocket campaign in which a series of seven payloads was launched to measure altitude profiles of emission features and atmospheric composition. The analysis of the ETON database resulted in detailed knowledge about nightglow excitation and quench-

ing mechanisms (Greer et al., 1986; McDade et al., 1986a, 1986b, 1987a, 1987b, Murtagh et al., 1986), and a numerical model has been developed based on this (Murtagh, 1989; Murtagh et al., 1990).

Attempts have been made to determine the altitude profiles of atomic oxygen from ground based measurements of different O related nightglow emissions (e.g. Haley et al., 2001; Stegman and Murtagh, 1991). This technique takes advantage of the differences in the molecularities of specific nightglow emission features to recover a parameterised atomic oxygen density profile. This way O densities close to the peak of the layer can be recovered to within 20%. However the detailed vertical structure of the profile cannot be recovered.

The plan for this paper is to first present results on the O₂ nightglow and direct atomic oxygen measurements obtained during the NLTE rocket campaign. In the discussion in Sect. 3 the inverted nightglow emissions are compared to the direct O measurements and also comparisons to other measurements are made. We conclude in Sect. 4 with a discussion of the reliability of O number densities retrieved from the resonance fluorescence technique and the possibility to improve these by calibration with simultaneous O₂ nightglow or dayglow measurements.

2 NLTE – techniques and results

The Swedish-led NLTE (Non Local Thermodynamic Equilibrium) sounding rocket campaign was conducted at the Esrange rocket range near Kiruna in northern Sweden in March 1998. It was an international rocket campaign (involving also groups from Norway, Germany, Austria and the USA) with the prime purpose to investigate processes contributing to the energy balance of the MLT region during polar winter. Rocket-borne and ground-based measurements investigated dynamic conditions and the distribution of neutral and charged species between 70 and 130 km.

The central aim of the Stockholm group was the study of processes controlling the distribution of atomic oxygen. This was done by directly measuring atomic oxygen using the resonance fluorescence and absorption technique simultaneously with photometric measurements of the O₂ Atmospheric Band at 762 nm and the N₂⁺ (0–0) 1st Negative band at 391 nm. The O₂ Atmospheric Band is a product of O atom recombination and can be used as an alternative way to retrieve atomic oxygen densities. The N₂⁺ (0–0) 1st Negative band is a sensitive indicator for the presence of precipitating (auroral) electrons. In addition, the passband of the N₂⁺ photometer comprises the (5–3) band of the O₂ Chamberlain system, which also is a result of atomic oxygen recombination. Additional experiments on the NLTE payloads addressed temperature, neutral and plasma densities as well as turbulence (Lübken et al 1999; Friedrich et al, 1999). By obtaining profiles of total density, absolute O densities can

be converted into mixing ratios, which helps to distinguish dynamic and chemical influences on the O distribution.

Two identical rocket payloads were launched as the main events during the campaign. NLTE-1 was launched on 3 March at 22:33 UT into geomagnetically very quiet conditions and NLTE-2 three days later on 6 March at 21:26 UT into geomagnetically extremely quiet conditions. On NLTE-2 the 391 nm photometer showed no sign of electron precipitation thus ensuring a study of the atmospheric energetics and chemistry in the absence of auroral influence. In addition, this made it possible to study the weak O₂ Chamberlain emission in the 391 nm filter band. The electron density measurement onboard the payload (Langmuir probe calibrated by a radio wave propagation experiment; Jacobsen and Friedrich, 1979) showed very low concentrations (see Fig. 1) and the ground-based O (¹S) Green Line photometer showed emissions of 250R during the first and 120R during the second launch indicating low and no auroral emissions during the two flights, respectively. The a_p index of geomagnetic activity showed low, but not extremely low, values of 3 and 5 during the first and second launch, respectively.

2.1 Resonance fluorescence and absorption

The resonance fluorescence technique is based on the resonance transition of O (³S₁–³P_{2,1,0}) at 130.4 nm. This triplet is emitted using an oxygen discharge lamp and the radiation can be resonantly absorbed and scattered by O atoms in the vicinity of the instrument. A suitable detector collects part of this scattered light as a direct measure of the atomic oxygen abundance. The use of the resonance transition at 130.4 nm to measure atomic oxygen in the atmosphere was originally proposed by Young (1961). For a detailed description of this measurement technique see e.g. Dickinson et al. (1980), Sharp (1991), or Gumbel et al. (1998). The O (³S₁–³P_{2,1,0}) absorption cross section of about $2 \times 10^{-13} \text{ cm}^{-2}$ exceeds the Rayleigh scattering cross section by 11 orders of magnitude and the absorption cross section of molecular oxygen by 6 orders of magnitude. Thus this technique provides both a sensitive and selective measurement of the atmospheric atomic oxygen abundance in the entire altitude range where the measurements were performed.

2.1.1 Calibration

The resonance fluorescence measurement represents a very sensitive but not very accurate measurement as it is not easy to calibrate. The relationship between the fluorescence signal and the atmospheric O density is determined by the intensity and emission line shapes of the lamp, detector sensitivity, measurement geometry, atmospheric temperature and aerodynamic flow properties in the measurement volume. Different calibration techniques exist and include numerical (e.g. Ulwick et al., 1994; Gumbel and Witt, 1997), laboratory (e.g. Sharp, 1991) and in situ absorption methods (e.g. Dickinson et al., 1980; Gumbel et al., 1998). Knowledge about the

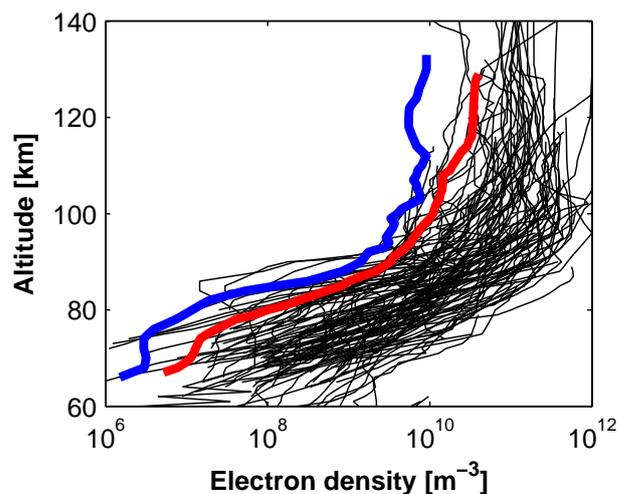


Fig. 1. Electron density measurements from NLTE-1 (red profile) and NLTE-2 (blue profile) together with other measurements north of 60° N latitude (thin black profiles) (M. Friedrich, personal communication, 2007).

line shapes of the lamp's resonance emission is essential for all three calibration techniques, and indirect information of the line shapes can be obtained from curve-of-growth measurements (e.g. Rawlins and Kaufman, 1977). The only direct measurement of resolved lamp emission line shapes have been reported by Jenkins et al. (1985).

For NLTE; in situ absorption calibrations supported by numerical simulations were utilised in a similar way as described by Gumbel et al. (1998) for an earlier rocket measurement. The in situ absorption measurement on the other hand is independent of absolute instrument parameters as it presents a relative measurement and thus provides a direct way of calibrating the atmospheric data. On NLTE, booms were deployed near the top of the payload thus positioning the absorption detectors opposite the O lamp, at distances of 33 and 65 cm, to measure the relative amount of the 130.4 nm triplet transmitted by the oxygen atoms. Although improving the measurement accuracy, the absorption calibration is intrinsically uncertain since the absorption process is very sensitive to the relationship between atmospheric absorption line shapes and emitted lamp line shapes (Gumbel and Witt, 1997). While the atmospheric absorption line shape is essentially determined by Doppler broadening at the atmospheric temperature, the emitted lamp line shape is only known with limited accuracy. Our NLTE analysis is based on the line shape results reported by Jenkins et al. (1985) from laboratory measurements with various lamp parameters. The stability of the lamp output before and during flight is an additional source of uncertainty. In total, we estimate the overall uncertainty of the direct atomic oxygen measurements during NLTE to be a factor of 2–3, in consistency with earlier investigations of the resonance fluorescence and absorption technique (Gumbel, 1999).

2.1.2 Aerodynamics

For most rocket-borne measurements also aerodynamic effects have to be considered very carefully. An exception is the accurate but complex and expensive cryo-cooled mass spectrometer that avoids aerodynamical effects by “freezing out” the shock front (Offermann and Tatarczyk, 1973; Offermann et al., 1981). As the payload moves through the atmospheric region of interest, conditions change from continuum flow via the transition regime to free molecular flow. The high velocity of the rocket payload can produce many different perturbations related to aerodynamics. These include compression and rarefaction of the atmospheric density, composition changes due to mass-dependent species separation, chemical reactions in the shock region and on payload surfaces, excitation of atoms and molecules (i.e. payload glow), Doppler influence on optical measurements, and contamination of the measurement volume by outgassing and desorption from the payload itself. A discussion of atomic oxygen measurements from sounding rockets under such flow conditions has been given by Bird (1988). For studying the aerodynamic effects in these changing flow conditions we use the DSMC (Direct Simulation Monte Carlo) model DS2V v.4.5 (Bird, 1994). With this model we can simulate two-dimensional and axially symmetric problems from continuum flow conditions to free molecular flow.

In Fig. 2a,c the simulated number density field around the NLTE payload is shown for two different altitudes. Figure 2b,d shows the O mixing ratio normalised to the undisturbed O mixing ratio in the flow field. The deployed absorption boom in the plots indicates the position of the absorption path. At 90 km altitude (Fig. 2a–b) there is a very pronounced shock region around the payload and the absorption detector is positioned outside the disturbed region. In panel b it can be seen that there is a depletion of O close to the payload but an increase further out at the edge of the shock region. This aerodynamic separation effect is caused by the lower mass of the O atom with respect to the N₂ and O₂ molecules. At 120 km (Fig. 2c–d) the shock region is smoothed out and the absorption detector is well inside the disturbed region. Panel d shows here a depletion of O relative to the undisturbed atmosphere along the entire absorption path.

Figure 3 shows the mean relative change of the total air density and the O density in the absorption path for different altitudes from 80 to 125 km. During the ascent of the payload, the air density is higher in the measurement region than in the undisturbed atmosphere. Without correction, the measurements will overestimate the O density. It is however not sufficient to investigate only the total air density. Rather, the additional effect on the mixing ratio of the O atoms needs to be included. For the NLTE measurements, the O mixing ratio in the absorption path up to 95 km is slightly higher than in the surrounding atmosphere, whereas above this altitude the fraction is significantly lower. The reason is that at the

lower altitudes the detector is outside the shock region and the absorption path includes both the depleted region near the payload and the O-enriched region near the outer edge of the shock. Above 95 km the shock grows and the entire absorption path is inside the perturbed region with depleted O.

The velocity of the payload and the temperature in the perturbed region are not high enough for chemical reactions, or excitation of atoms and molecules. Neither are Doppler influences on the resonance measurement significant as the absorption path is largely perpendicular to the payload velocity vector. Contamination of the measurement volume by outgassing and desorption from the payload is evident during the upleg part of the flight. Effects on O absorption measurements have been discussed by Gumbel et al. (1998).

2.1.3 Direct atomic oxygen results

The result of the O fluorescence measurement during NLTE-2 is shown in Fig. 4. This profile has been calibrated with the absorption measurement and corrected for aerodynamic effects as described in the two previous sections. The measurement shows a structured profile with a peak density of 2.7×10^{11} atoms per cm³ at 97.5 km and a very sharp cut-off at the lower ledge below 90 km. The data is sampled at 150 Hz which results in a data point every 5–6 m in the altitude interval from 90 to 105 km. The profile in Fig. 4 is smoothed to a vertical resolution of 250 m and the error bars show 1 standard deviation. A comparison to the NLTE-1 results will be given in Sect. 3.

2.2 Nightglow photometry

The molecular oxygen band systems are, together with the O (¹S–¹D) Green Line and the OH Meinel band system, the main contributors to the terrestrial nightglow spectrum. As mentioned earlier, atomic oxygen is mainly formed through O₂ photolysis during the day and can then recombine to form excited oxygen molecules during the night. The recombination reaction has an exothermicity of 5.12 eV which mainly resides in the newly formed O₂ molecule, the so called precursor state of the molecular and atomic oxygen nightglow emissions. With this available energy high vibrational levels of the ground state or any of the five bound states, may be formed (Slanger and Copeland, 2003). The energy levels of these states are shown in Fig. 5. Another weakly bound state, ⁵Π_g, has been proposed as a likely precursor to the above lower-lying light-emitting states. The excited oxygen molecule may then decay through emission of a photon, energy transfer to O, or quenching by other atoms or molecules (e.g. O₂, N₂, O or CO₂).

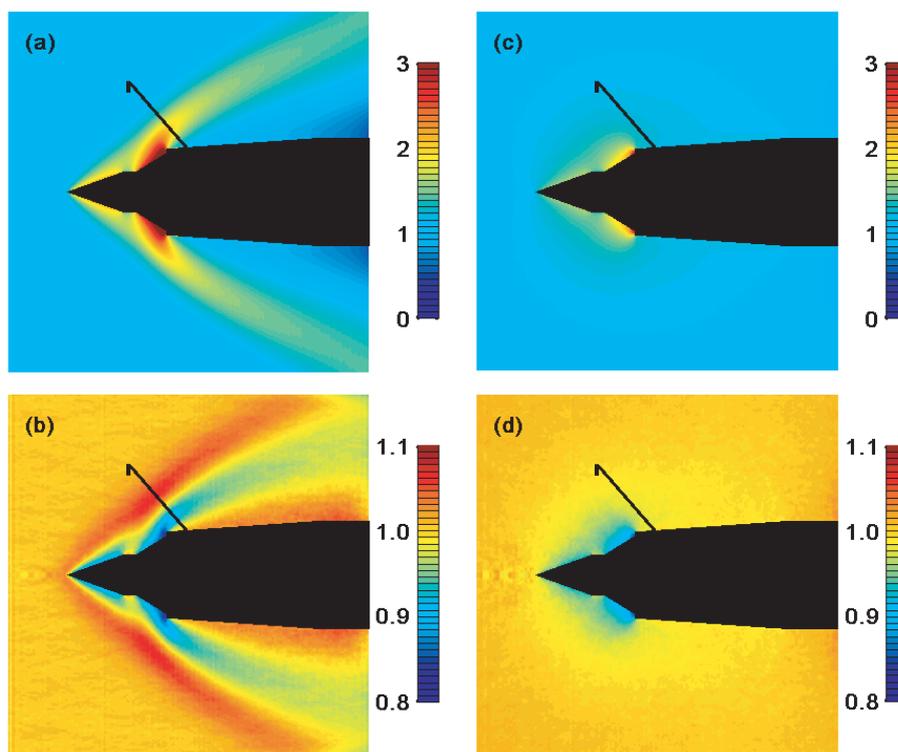


Fig. 2. Direct Simulation Monte Carlo results of the interaction between the NLTE rocket payload and the surrounding atmosphere. Panel (a) shows the total air density normalised with the unperturbed air density, and panel (b) shows the normalised O mixing ratio, both at 90 km altitude. Panel (c) and (d) shows the same properties as (a) and (b) but for 120 km altitude. The boom is not included in the flow simulations.

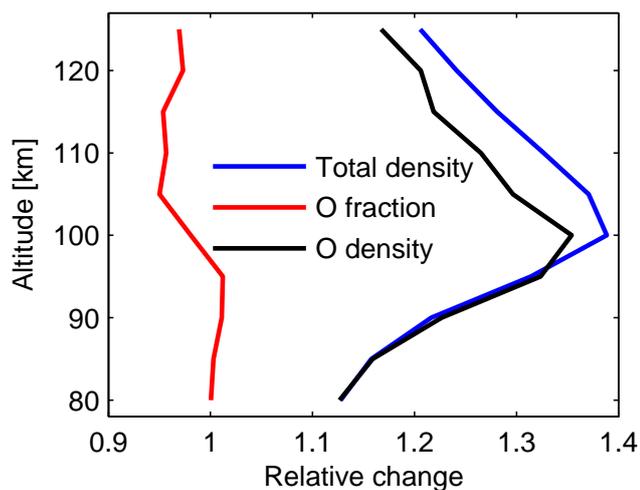


Fig. 3. Aerodynamic simulation of the interaction of the NLTE rocket payload with the atmosphere and the effect on the total air density and O mixing ratio at different altitudes. The profiles show the relative change in the absorption path between the lamp and the absorption experiment on the boom.

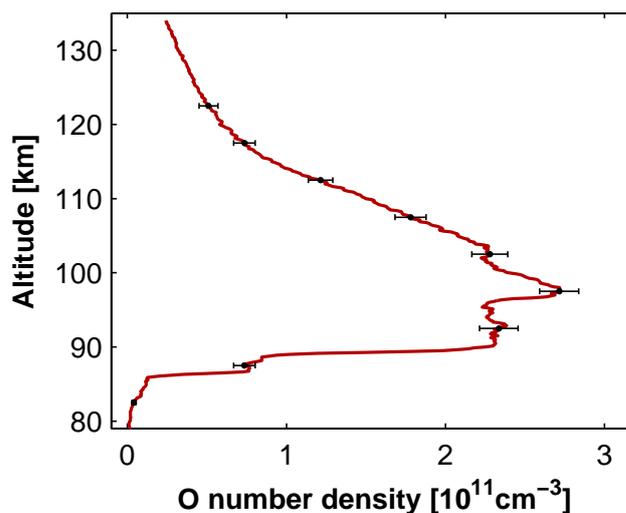


Fig. 4. The atomic oxygen number density measured with the resonance fluorescence and absorption technique during the NLTE-2 rocket launch. Error bars indicate 1 standard deviation around the mean. The vertical resolution of the smoothed profile is 250 m.

2.2.1 Nightglow measurements during NLTE

The second strongest O₂ nightglow emission feature is the Atmospheric Band system ($b^1\Sigma_g^+ \rightarrow X^3\Sigma_g^-$) dominated by the (0–0) and (0–1) bands at 762 nm and 864 nm, respectively. The (0–0) band cannot be studied from the ground because of self absorption in the atmosphere between the emitting layer and the observer. It is however easy to measure with filter photometers from sounding rockets.

Optical measurements of the upper atmosphere from sounding rockets have been made since the 1950's (Heppner and Meridith, 1958) both for understanding airglow chemistry in itself and for using airglow as a tool to understand atmospheric (e.g. dynamical) processes. For airglow measurements in general, a filter photometer is positioned under the nose cone viewing along the rocket axis. During ascent, after the nosecone ejection, the photometer then counts the incoming photons from the overhead column. When the rocket passes through the layer the measured photon flux drops and above the emission layer only weak background emissions from the zodiacal and galactic light are present (unless there are auroral emissions in the wavelength region defined by the photometer passband). After the profile has been corrected for background emissions and attitude (van Rhijn effect) it is converted from counts to radiance (or Rayleigh) using pre-flight laboratory calibrations. The profile can then be smoothed (Fig. 6a) and numerically differentiated to yield the volume emission rate of the emitting layer (Fig. 6b). Ideally one should first differentiate the radiance profile and then smooth the resulting volume emission profile since this retains more of the original structure and noise. In practise, however, this is not realistic because of the noise, and some smoothing is necessary before the radiance profile can be differentiated (Murtagh et al., 1984). The data were sampled with 100 Hz on both photometers which results in an altitude resolution of about 9 m during the passage of the airglow layer. However, because of the noise, the profiles need to be averaged to a vertical resolution of 1 km for the Atmospheric band and 2 km for the Chamberlain band in order to get satisfactory results after the differentiation. The dotted lines in panel a of Fig. 6 shows a 1 standard deviation around the mean profiles.

The nightglow model used in the following is a numerical model based almost exclusively on the findings of the ETON rocket campaign. It is a self-consistent model of the most common nightglow emissions including the O(¹S) Green Line, OH Meinel and several O₂ band emissions (Murtagh, 1989; Murtagh et al., 1990). For the O₂ Atmospheric band, the emitting state ($b^1\Sigma_g^+$) is assumed to be excited via an energy transfer mechanism from the precursor state with O₂ as the transfer agent (Greer et al., 1986). The volume emission rate of the O₂ Atmospheric band may be expressed as

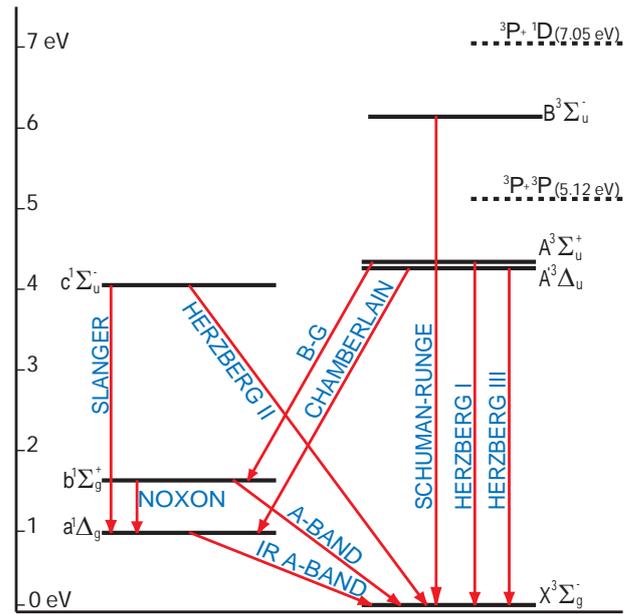


Fig. 5. Energy level diagram of O₂ transitions in the nightglow (after Greer et al., 1987).

(Murtagh, 1989)

$$V_{\text{at}} = \frac{A_1 \cdot k_1 [\text{O}]^2 \{[\text{N}_2] + [\text{O}_2]\} [\text{O}_2]}{\left\{ A_2 + k_2^{\text{O}_2} [\text{O}_2] + k_2^{\text{N}_2} [\text{N}_2] + k_2^{\text{O}} [\text{O}] \right\}} \cdot \frac{1}{\{C^{\text{O}_2} [\text{O}_2] + C^{\text{O}} [\text{O}]\}} \quad (1)$$

Equation (1) comprises the excitation and emission processes as well as quenching of the precursor state and O₂($b^1\Sigma_g^+$) by various atmospheric constituents. A_1 is the (0–0) band transition probability, A_2 is the inverse radiative lifetime of O₂($b^1\Sigma_g^+$, $v=0$), k_1 is the temperature-dependent rate coefficient for the three-body recombination of atomic oxygen, and $k_2^{\text{O}_2}$, $k_2^{\text{N}_2}$ and k_2^{O} are the rate coefficients for the quenching of O₂($b^1\Sigma_g^+$) by O₂, N₂ and O and are all determined by laboratory investigations. The coefficients C^{O_2} and C^{O} represents the quenching of the precursor state and were derived empirically from the ETON measurements. The values of all coefficients can be found in Table 1. The O₂ Atmospheric band profile measured during NLTE-2 is the black profile shown in Fig. 6.

The second photometer on the NLTE payloads measured the emission from the N₂⁺ 1st Negative band at 391.4 nm. This emission is a sign of precipitating auroral electrons and thus a sensitive indicator of auroral activity. Included in the passband of this photometer is also the (5–3) band of the O₂ Chamberlain system at 390.5 nm. This is not a well studied emission feature since it is very faint and easily dominated by the presence of even weak auroral emissions. Emissions from the Chamberlain band system are also in general blended in the much stronger Herzberg I and II band systems. During the NLTE-2 launch the auroral activity was extremely low and the O₂ Chamberlain band could thus be studied. The

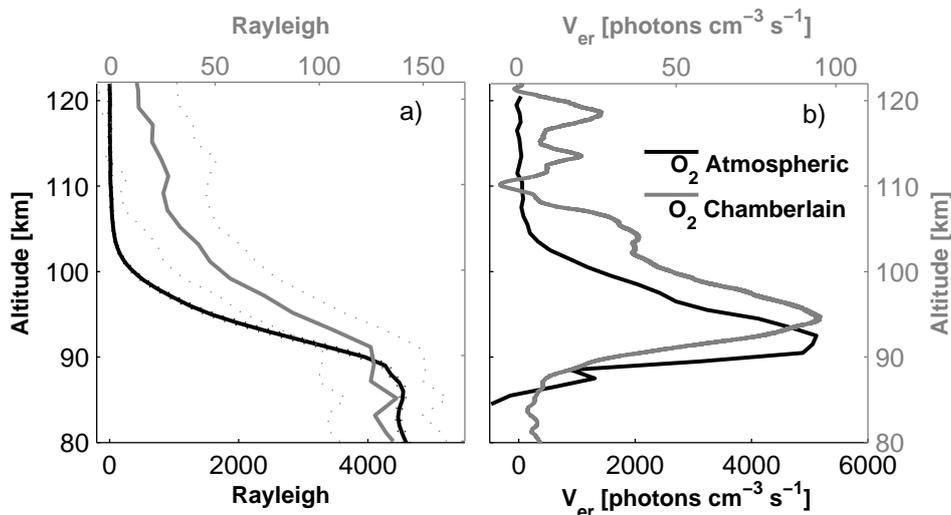


Fig. 6. Measured nightglow profiles during the NLTE-2 rocket launch. The black profiles are the (0–0) O₂(*b*¹Σ) Atmospheric Band at 762 nm and the gray profiles are the (5–3) band of the O₂(*A*¹Δ) Chamberlain system at 391 nm. Panel (a) shows the measured column emission in Rayleigh (the radiance in photons cm⁻² str⁻¹ s⁻¹ multiplied by 4π × 10⁻⁶). The dotted lines show a 1 standard deviation around the mean indicating the signal-to-noise ratios for the two measurements. The Atmospheric band was interval averaged with 1 km vertical resolution, while the Chamberlain band has a vertical resolution of 2 km in order to get satisfactory volume emission profiles after differentiation. Panel (b) shows the volume emission rate of the airglow layers, i.e. the differentiated column emission profiles from panel (a).

Table 1. Adopted rate coefficients and excitation parameters. All coefficients are in the usual units in terms of molecules, cm³, and seconds. *T* is the atmospheric temperature, and γ is the production efficiency of the emitting state, O₂(*A*¹Δ_u), in the recombination reaction (see Eq. 2 and the text).

Coeff.	Value	Reference
k_1	$4.7 \times 10^{-33} (300/T)^2$	Campbell and Gray, 1973
$k_2^{O_2}$	4.0×10^{-17}	Martin et al., 1976
$k_2^{N_2}$	2.2×10^{-15}	Martin et al., 1976
k_2^O	8.0×10^{-14}	Slanger and Black, 1979
k_2^O	0	McDade et al., 1986a
k_3^O	$2.4 \times 10^{-10} \times \gamma$	Stegman and Murtagh, 1991
$k_3^{O_2}$	$9.8 \times 10^{-11} \times \gamma$	Stegman and Murtagh, 1991
A_1	0.079	Vallance Jones, 1974
A_2	0.083	Vallance Jones, 1974
A_3	0.85	Stegman and Murtagh, 1991
Excitation parameters for use with $k_2^O = 8 \times 10^{-14}$:		
C^{O_2}	6.6 ± 0.4	McDade et al., 1986a
C^O	19 ± 2	McDade et al., 1986a
Excitation parameters for use with $k_2^O = 0$:		
C^{O_2}	7.5 ± 0.5	McDade et al., 1986a
C^O	33 ± 4	McDade et al., 1986a

emitting state is thought to be directly produced in the three-body recombination reaction of atomic oxygen and the loss processes are quenching by O and O₂. The volume emission rate of the Chamberlain system can thus be expressed as

$$V_{\text{ch}} = \frac{\gamma \cdot A_3 \cdot k_1 [\text{O}]^2 \{[\text{N}_2] + [\text{O}_2]\}}{k_3^{\text{O}} [\text{O}] + k_3^{\text{O}_2} [\text{O}_2]} \quad (2)$$

where k_1 is again the temperature-dependent rate coefficient for the three-body recombination of atomic oxygen, and k_3^{O} and $k_3^{\text{O}_2}$ are the rate coefficients for the quenching of the emitting state by O and O₂, respectively, γ is the production efficiency of the emitting state, O₂(A'³Δ_u), in the recombination reaction, and A_3 is the inverse of the radiative lifetime (see Table 1 for values). Chamberlain band emissions are not available from the ETON campaign, but they were measured before (Stegman et al., 1983; Stegman and Murtagh, 1991) and are included in the model. The Chamberlain profile from NLTE-2 is the gray profile shown in Fig. 6. The volume emission rate of the Chamberlain band system is about 50 times weaker than that of the Atmospheric band system. Also, as can be seen in Fig. 6a, the signal-to-noise ratio is much better for the Atmospheric band, as indicated by the dotted lines showing a 1 standard deviation around the smoothed airglow emission profiles. As mentioned earlier, the vertical resolution of the Atmospheric band volume emission profile is 1 km, while it is 2 km for the Chamberlain band profile. The difference in peak altitudes is consistent with the photochemistry of the two emitting states where the Chamberlain band emission is directly produced in the recombination reaction without an intermediate energy transfer step, and thus radiates at higher altitudes.

2.2.2 Atomic oxygen retrievals

Equations (1) and (2) can readily be inverted to derive the atomic oxygen densities from any Atmospheric band or Chamberlain emission profile using the reaction coefficients derived from the ETON measurements. A necessary input to this retrieval is the temperature and neutral density of the background atmosphere. The atomic oxygen profiles derived from the NLTE-2 nightglow profiles are shown in Fig. 7. The gray line is the profile derived from the Chamberlain band emission and the solid black line is the profile derived from the Atmospheric band emission. In this paper, the retrieved oxygen profiles from the Atmospheric band were derived using the rate coefficients and constants for the case of no quenching of the emitting state by atomic oxygen (see Table 1). Slanger and Black (1979) suggested that under nighttime conditions the rate coefficient for this quenching, k_2^{O} , may be much smaller than was determined in the lab. McDade et al. (1986a) took this into consideration and adopted the measured value by Slanger and Black (1979) as an upper limit and zero as a lower limit. The omission of quenching of the emitting state by atomic oxygen has little effect on the retrieved oxygen profile. The dashed black lines indicate the

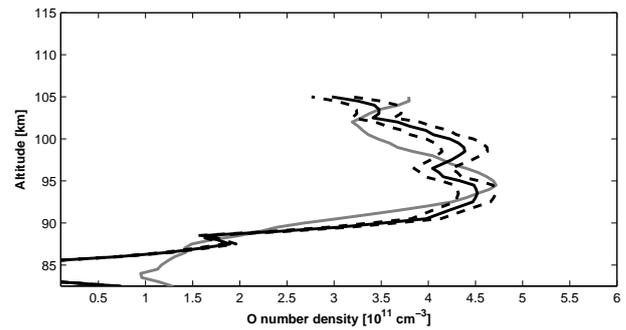


Fig. 7. Atomic oxygen number density derived from the nightglow measurements during NLTE-2. The gray line is the O profile retrieved from the Chamberlain nightglow emission profile. The solid black line is the O profile retrieved from the Atmospheric band emission profile without quenching of the precursor state by atomic oxygen. The dashed black lines show the possible error due to uncertainties in the rate constants for the Atmospheric band inversion.

uncertainties in the retrieval due to uncertainties in the reaction rates. For this analysis the atmospheric temperature and density have been taken from MSIS-E90 (Hedin, 1991) since on NLTE-2 the onboard neutral density and temperature measurement failed. It can be noted that for the night of the launch lidar measurements up to 85 km, well below the emission layer, indicate that MSIS is ~30 K too warm. Using the temperature climatology of Rauthe et al. (2008) and atmospheric density climatology of Rapp et al. (2001) the sensitivity of the nightglow inversion to atmospheric temperature and density variability can be estimated. At 95 km (close to the peak altitude of the oxygen profile) the temperature variability in March is typically ± 6 K which yields a variability in the O density inversion of $\pm 4\%$. At the same altitude the total density variability is typically $\pm 12\%$ for the winter months (January to March) which translates into a variability of the inverted O density of $\pm 10\%$. Above ~105 km the possible errors in the measured Atmospheric band emission rates are large (low signal to noise ratio) and the inversion procedure used to determine the atomic oxygen number density becomes unreliable (Murtagh et al., 1990).

The retrieval of atomic oxygen from the nightglow emissions yields O profiles with reliable absolute values (peak density). However, the altitude resolution is limited and the detailed structure of the derived profiles, especially from the weak Chamberlain band emission, may not be geophysical. The O profile retrieved from the Atmospheric band indicates a double peak structure with peak densities of $4.5 \times 10^{11} \text{ cm}^{-3}$ at 94 km and $4.4 \times 10^{11} \text{ cm}^{-3}$ at 99 km. The main peak of the profile derived from the Chamberlain band shows an atomic oxygen density of $4.7 \times 10^{11} \text{ cm}^{-3}$ at 95 km.

3 Discussion

The atomic oxygen profiles derived from the A-band and Chamberlain emissions are in good agreement with each other when it comes to absolute peak density and altitude. Consistency is also found with the ground-based photometer measurements of the O (¹S→¹D) Green Line at 557.7 nm. The nightglow model provides a direct relationship between Atmospheric band and Green Line emission rates, based on the rate coefficients from ETON. For the Atmospheric band measurement from NLTE-2 the nightglow model predicts a Green Line emission of 125–130 R, which is in very good agreement with the 120–125 R seen by the ground-based Green Line photometer. However, as can be seen in Fig. 8, the atomic oxygen number density profile derived from the direct resonance fluorescence and absorption technique (dashed red line) is not consistent with the number densities derived from the nightglow emissions (solid gray and black lines) and differs by a factor of about 1.7 in the peak density. As discussed in Sect. 2.1, this difference is most likely due to difficulties in achieving a good calibration of the direct O measurement. It can be noted that the direct measurement of O is consistent with the nightglow analysis within the uncertainty of the measurement technique. For comparison, also the O profile from the MSIS-E90 model (dashed gray line) is shown in Fig. 8.

In order to improve the uncertainty and compatibility of rocket-borne atomic oxygen measurements, we now suggest using the Atmospheric band emission measurement to calibrate the direct measurement. This is best done in terms of the entire atmospheric O column and the undifferentiated photometer measurement. Starting out from the directly measured O profile from NLTE-2, using the model we first calculate the corresponding volume emission rate of the Atmospheric band and then integrate this profile to obtain a column emission. This can be compared directly to the rocket-borne photometer measurements. The ratio between these two column radiances is then determined and used as a calibration factor. The atomic oxygen profile measured directly is then multiplied by the square root of this factor (since the volume emission rate is proportional to (O)², see Eq. 1). The resulting calibrated atomic oxygen profile with a peak density of $4.45 \times 10^{11} \text{ cm}^{-3}$ is the solid red profile in Fig. 8. While the airglow measurement provides a reliable absolute peak density, the very sensitive resonance fluorescence measurement provides the detailed structure of the atomic oxygen profile over the entire measured altitude range including the very low values below 80–85 km and at the apogee above 130 km.

The corresponding results from the NLTE-1 flight are shown in Fig. 9 with the direct O measurement and the O profile retrieved from the Atmospheric band emission. As opposed to NLTE-2, on this flight the 390 nm photometer detected a weak variable N₂⁺ auroral emission and thus the O₂ Chamberlain band emission could not be retrieved. The di-

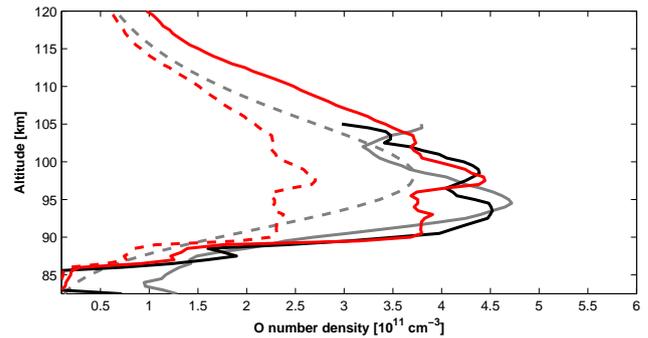


Fig. 8. The solid gray and black lines as in Fig. 7. The dashed red line is the measured atomic oxygen profile from the direct technique from Fig. 4. The solid red line is the nightglow-calibrated direct measurement and the dashed gray line is the MSIS-E90 atomic oxygen profile from the time of the NLTE-2 launch.

rect measurement yielded a peak density of $5.6 \times 10^{11} \text{ cm}^{-3}$ at 97 km, whereas the nightglow yielded a lower value. On NLTE-1 the measurement of the neutral density and temperature during the descending part of the rocket flight by the CONE instrument (Lübken et al., 1999) gave good background information for the O retrieval from the Atmospheric band. The structure in the retrieved oxygen profile below ~ 100 km are mainly due to the fact that the Atmospheric band emission profile measured during this rocket flight was more structured than that measured during NLTE-2. Also, as stated earlier, at higher altitudes (above ~ 100 – 105 km) errors in the measured Atmospheric band emission rates can grow large due to the low signal-to-noise ratio and the retrieval procedure becomes less reliable. Also the use of the more structured temperature profile influences the oxygen retrieval, but to a lesser extent. The solid blue line in Fig. 9 shows the airglow-calibrated O profile with a peak density of $4.6 \times 10^{11} \text{ cm}^{-3}$. This airglow-calibrated peak value is 31% lower than obtained by the direct technique (dashed blue line), as compared to NLTE-2 where the calibrated peak value was 72% higher. Since the NLTE-1 and -2 payloads were essentially identical, this difference between them underlines the calibration challenges with the resonance fluorescence technique described earlier.

Figure 10 shows a comparison of the atomic oxygen profiles derived from the NLTE resonance fluorescence and O₂ Atmospheric band measurements with earlier measurements featuring simultaneous resonance fluorescence and airglow measurements. The thick lines indicate atomic oxygen profiles from resonance fluorescence measurements and the thin lines are the results from the O₂ Atmospheric band inversion. The black profiles were obtained during the SOAP/WINE (the Selective Optical Atmospheric Probe/Winter In Northern Europe, solid thick and thin lines) and USU (Utah State University, dashed line, 72 min after SOAP, resonance fluorescence only) measurements during the MAP/WINE

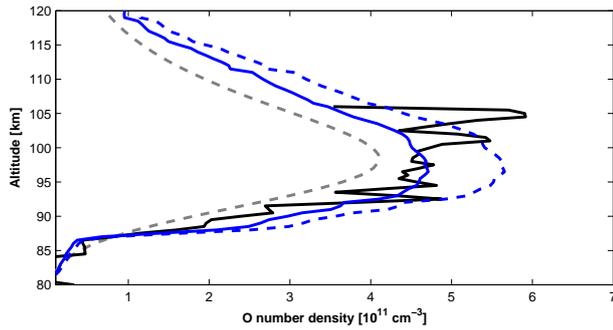


Fig. 9. Atomic oxygen profiles from NLTE-1. The black line shows the O profile retrieved from the Atmospheric band emission without quenching of the precursor state by atomic oxygen. The dashed blue line is the measured atomic oxygen profile from the direct technique. The solid blue line is the nightglow-calibrated direct measurement and the dashed gray line is the MSIS-E90 atomic oxygen profile from the time of the NLTE-1 launch.

(Middle Atmosphere Program/Winter In Northern Europe) sounding rocket campaign (Dickinson et al., 1987). The blue lines are the NLTE-1 profiles (resonance fluorescence profile without nightglow calibration) and the red lines are NLTE-2 profiles (again resonance fluorescence profile without nightglow calibration). The green profiles are from measurements during the OXYGEN/S35 sounding rocket campaign (Witt et al., 1984). Note that the resonance fluorescence measurements vary by a factor of 50 while there is only a spread by a factor of 3 in the nightglow inversion profiles. Since the photometric airglow measurements are simple, they allow for a straight-forward comparison between different measurement occasions. Hence, the results in figure 10 further stress the variability and uncertainty in the resonance fluorescence instruments and the measurement process.

4 Conclusions

Rocket-borne resonance fluorescence at 130.4 nm in combination with in situ absorption provides an accurate way of measuring atomic oxygen if a detailed characterisation of the instrument is available. In particular, sufficient knowledge about the O resonance lamp and its emitted line shapes is needed. This was the basis for the direct O measurements during the ETON project, comprising e.g. highly resolved laboratory studies of the lamp output. Consequently, the ETON results on atomic oxygen and nightglow rate coefficients are the best available and have come in use as a standard. This way of performing O measurements with detailed instrument calibration before flight and in-flight is in principle the most accurate, but it is generally difficult and too expensive to achieve for every single rocket flight. A simple way of calibrating atomic oxygen measurements is therefore highly desirable.

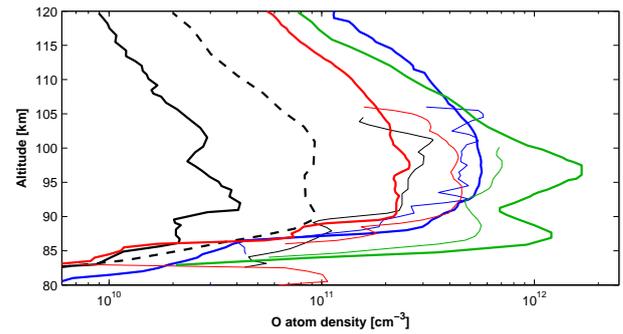


Fig. 10. Comparison between the NLTE results and other earlier rocket campaigns with simultaneous resonance fluorescence and airglow measurements. Heavy lines denote atomic oxygen profiles obtained from direct measurements while thin lines denote oxygen profiles retrieved from O₂ Atmospheric band emission measurements. The black lines are from SOAP/WINE (solid thick and thin lines) and USU (dashed line, 72 min after SOAP, resonance fluorescence only); the blue lines are the profiles from NLTE-1 (without nightglow calibration); red lines are the profiles from NLTE-2 (again without nightglow calibration); and the green lines are the profiles obtained during the OXYGEN/S35 sounding rocket campaign. Note the spread by a factor of 50 in the direct measurements and only by a factor of 3 in the profiles retrieved from the nightglow measurements.

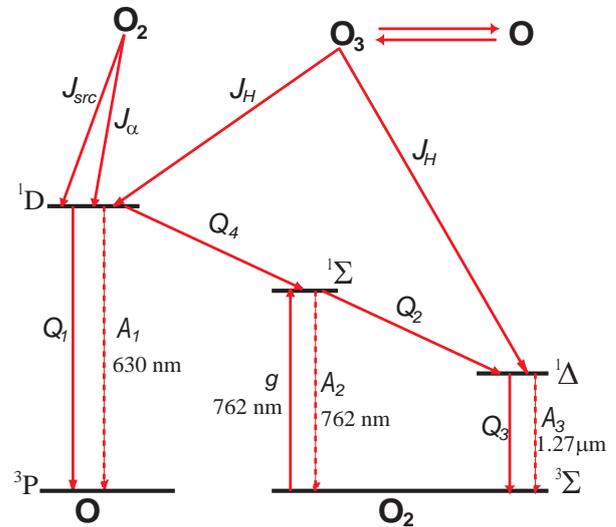


Fig. 11. The O₂(*a*¹Δ_g) dayglow production mechanisms (after Mlynczak et al., 1993). *J* indicate solar photolysis, *Q* collisional quenching, *A* spontaneous emission, and *g* solar absorption in the Atmospheric band.

Therefore, the calibration of the direct O measurement by simultaneous airglow measurements, combined with a detailed aerodynamic analysis, is often preferable. Airglow photometers are relatively easy to calibrate in the laboratory. In this way knowledge of the exact performance of the resonance fluorescence instrument, and expensive and complicated absorption measurements, are not needed. Major

sources of error are no longer the instrument calibration, but only the uncertainties in the airglow reaction coefficients and the background atmosphere density and temperature. Good neutral density and temperature measurements are desirable for a good analysis considering the limited ability of model atmospheres to represent the local atmospheric conditions at the time of measurement.

For daytime measurements the Atmospheric band is not adequate as a calibration of O measurements because of the dominance of high background illumination due to direct solar resonance excitation. Instead the (0–0) band of the O₂ IR Atmospheric system at 1.27 μm can be used (e.g. Mlynczak et al., 2001). This emission is related to the photolysis of ozone which, during the day, is in steady state with atomic oxygen (see Fig. 11) and a retrieval of atomic oxygen is thus possible. Mlynczak et al. (2001) discuss combined rocket-borne measurements of the IR-Atmospheric Band and the Atmospheric Band during daytime.

In summary, we suggest future rocket-borne measurements of atomic oxygen profiles to be based on combinations of a relatively simple “uncalibrated” 130.4 nm resonance fluorescence probe and a calibrated airglow photometer. While the resonance fluorescence probe provides the sensitivity and altitude resolution, the photometer provides the necessary calibration of the atomic oxygen data. This approach would also largely improve the compatibility between various rocket-borne O measurements as inter-comparisons between photometers are much more straight-forward than inter-comparisons between complex resonance fluorescence techniques. The airglow inversions presented in the present paper are based on nightglow rate coefficients from the ETON database. If the relevant rate coefficients are changed as a result of future investigations, a re-calibration of O profiles will be straight-forward for retrievals that are based on airglow photometry.

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