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Key Points:

- New data set of atomic oxygen at the mesopause from SCIAMACHY green line
- Differences to other methods show need for photochemical/NLTE model improvement
- Energy balance and retrieval of other species likely affected by results

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Global distribution of atomic oxygen in the mesopause region as derived from SCIAMACHY O(¹S) green line measurements

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Abstract A new data set of atomic oxygen abundance in the upper mesosphere and lower thermosphere is presented. The data are derived from the nighttime atomic oxygen green line limb emission measurements of the SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) instrument on the European Environmental Satellite. The temporal coverage is October 2002 until April 2012, and the latitudinal extent is 50°S to 80°N at 10 P.M. local time. This data set is compared to other satellite data sets, in particular to recently published data of SABER (Sounding of the Atmosphere using Broadband Emission Radiometry) and the Mass Spectrometer and Incoherent Scatter model. SCIAMACHY atomic oxygen peak abundances are typically $3\text{--}6 \times 10^{11}$ mol/cm³ at the atomic oxygen maximum region, depending on latitude and season. These values are similar to previous values based on chemiluminescence measurements of the atomic oxygen three-body recombination reaction but at least 30% lower than atomic oxygen abundances obtained from SABER.

1. Introduction

Atomic oxygen plays a crucial role in the photochemistry and energy balance of the upper mesosphere and lower thermosphere (UMLT) [e.g., *Brasseur and Offermann*, 1986]. It is the most abundant reactive species in this part of the atmosphere. Its lifetime is several weeks or longer and a significant amount of absorbed solar energy is redistributed in the upper atmosphere through the transport of atomic oxygen atoms.

The atomic oxygen abundance is a key parameter in several chemical reactions and collisional processes in the UMLT. Several methods for deriving temperature [e.g., *López-Puertas and Taylor*, 2001; *Kutepov et al.*, 2006; *Gusev et al.*, 2006], ozone [e.g., *Kaufmann et al.*, 2003; *López-Puertas and Taylor*, 2001; *Smith et al.*, 2008], and other constituents depend on the knowledge of this species.

The measurement of ground state atomic oxygen in the UMLT is very difficult and has been made by rocket-borne mass spectrometers at selected locations only (see, e.g., the review of *Gumbel* [1998]). These measurements have been used in combination with airglow observations (O(¹S), O₂ A-Band, OH*) to develop photochemical models that provide the basis for the derivation of atomic oxygen from satellite remote sensing measurements. Since the early 1970s, several satellites measured airglow emissions to derive atomic oxygen. More recent ones are WINDII (WIND Imaging Interferometer) [*Russell et al.*, 2005] on the Upper Atmosphere Research Satellite, SABER (Sounding of the Atmosphere using Broadband Emission Radiometry) [*Mlynchak et al.*, 2013a] on the TIMED (Thermosphere Ionosphere Mesosphere Energetics Dynamics) satellite, OSIRIS (Optical Spectrograph and Infrared Imager System) [*Sheese et al.*, 2011] on the Odin satellite, and the Imager of Sprites and Upper Atmospheric Lightning instrument on board the FORMOSAT-2 satellite [*Gao et al.*, 2012]. The WINDII atomic oxygen data set was derived from O(¹S) green line emission data at high altitudes and from OH* emissions below. SABER nighttime oxygen was derived from OH* emissions as well. The OSIRIS data were derived from O₂ A-band emission data [*Sheese et al.*, 2011]. Other satellite-borne remote sensing measurements of atomic oxygen are based on daytime ozone measurements [see, e.g., *Kaufmann et al.*, 2003; *Gil-Lopez et al.*, 2005; *Mlynchak et al.*, 2013a] or in the lower thermosphere on the measurement of the 63 μm fine structure emission of atomic oxygen [*Grossmann et al.*, 2000].

This work presents the 10 year data set of atomic oxygen abundance as derived from green line emission data of the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument on Envisat. SCIAMACHY was in operation from 2002 to April 2012. Although Envisat is in a different orbit than the TIMED satellite, the asynoptic sampling of the two satellites exhibits several coincidences

Table 1. Rate Constants/Fitting Parameters and Their Uncertainties as Used in the Simulation of O(¹S) Green Line Volume Emission Rates^a

Symbol	Rate Constant/Fitting Parameter	References
k'_{OOM}	$4.7 \times 10^{-33} \times \left(\frac{300}{T}\right)^2 (\text{cm}^6 \text{mol}^{-2} \text{s}^{-1})$	<i>Campbell and Gray</i> [1973]
$k'_{1\text{SO}2}$	$(2.32 \pm 0.94) \times 10^{-12} \times \exp\left(\frac{-812 + 1.82 \times 10^{-3} T^2}{T}\right) (\text{cm}^3 \text{mol}^{-1} \text{s}^{-1})$	<i>Capetanakis et al.</i> [1993]
$A_{1\text{D}}$	$(1.260 \pm 0.095) (\text{s}^{-1})$	<i>Nicolaides et al.</i> [1969]
$A_{1\text{S}}$	$(1.394 \pm 0.105) (\text{s}^{-1})$	<i>Nicolaides et al.</i> [1969]
C_{O}	211 ± 10	<i>McDade et al.</i> [1986]
C_{M}	15 ± 2	<i>McDade et al.</i> [1986]

^aFollowing *Sheese et al.* [2011], we assume a 10% uncertainty for the k'_{OOM} rate constant.

in latitude and local time. This allows for the comparison of data sets from the SCIAMACHY and SABER instruments at the same phase of the migrating diurnal tide, which has a large effect on the atomic oxygen abundance [e.g., *Smith et al.*, 2010].

Since ongoing retrieval activities of the SABER and Michelson Interferometer for Passive Atmospheric Sounding temperature and ozone data sets rely on the provision of reliable atomic oxygen densities, this work aims to provide an independent and additional data set of this quantity.

Moreover, the 10 year data set of SCIAMACHY presented in our paper allows further insight into our understanding of the energy budget of the mesopause region and its dependence on solar activity.

2. Modeling

Airglow green line radiances at 557.7 nm are spontaneous emissions of the O(¹S) excited state. The process for producing O(¹S) is called the two step “Barth Mechanism” [*Barth and Hildebrandt*, 1961] and involves the following two reactions:

where M describes a molecule of the background atmosphere and O₂^{*} is an electronic state of molecular oxygen, most likely O₂(c¹Σ_u[−]) [*McDade and Llewellyn*, 1988]. Assuming steady state conditions, the green line emission rate $V_{1\text{S}}$ is given by the ratio of the O(¹S) production and loss terms multiplied by the O(¹S → ¹D) Einstein coefficient ($A_{1\text{D}}$)

$$V_{1\text{S}} = A_{1\text{D}} \frac{k'_{\text{OOM}} k'_{\text{O}2\text{O}} [\text{O}]^3 [\text{M}]}{(A_{1\text{S}} + k_{1\text{SO}2} [\text{O}_2] + k_{1\text{SO}} [\text{O}]) (A_{\text{O}2^*} + k_{\text{O}2\text{O}2} [\text{O}_2] + k_{\text{O}2\text{N}2} [\text{N}_2] + k_{\text{O}2\text{O}} [\text{O}])} \quad (3)$$

$A_{1\text{S}}$ and $A_{\text{O}2^*}$ are the O(¹S) and O₂^{*} inverse radiative lifetimes, k'_{OOM} is the rate for reaction (1) forming O₂^{*}, and $k'_{\text{O}2\text{O}}$ is the rate for reaction (2) forming O(¹S). The quantities $k_{\text{O}2\text{O}2}$, $k_{\text{O}2\text{N}2}$, $k_{\text{O}2\text{O}}$, and $k_{1\text{SO}2}$, $k_{1\text{SO}}$ are the rates for collisional deactivation of O₂^{*} and O(¹S) by O₂, N₂, and O, respectively. Concentrations of these species are marked by squared brackets. A compilation of the rate constants for these processes is given, e.g., in *Khomich et al.* [2008]. *McDade and Llewellyn* [1986] fitted a reduced set of parameters neglecting some higher-order terms in equation (3) to the ETON rocket measurements:

$$V_{1\text{S}} = A_{1\text{D}} \frac{k'_{\text{OOM}} [\text{O}]^3 [\text{M}]}{(A_{1\text{S}} + k_{1\text{SO}2} [\text{O}_2]) (C_{\text{M}} [\text{O}_2] + C_{\text{O}} [\text{O}])} \quad (4)$$

The derived fitting parameters C_{M} and C_{O} were later confirmed by *Murtagh et al.* [1990] to give a consistent picture of atomic oxygen green line and O₂ atmospheric band emission profiles. For the analysis of SCIAMACHY data, we apply this set of rate constants and fitting parameters (Table 1) as well, with some minor updates according to *Gao et al.* [2012].

Figure 1. SCIAMACHY monthly average zonal mean green line spectra for October 2005 at 35°N–40°N. About 1100 individual spectra were averaged. The noise level of individual spectra is about $6 \cdot 10^8$ photons/(cm² sr s). The tangent altitude of the spectra is given in the legend (in kilometers).

applying difference operators of zero and first orders [Twomey, 1977]. A zero-order Tikhonov regularization constrains the absolute value, while the first-order minimizes the derivative in order to smooth the solution. Fitting parameters are the atomic oxygen density, a spectrally constant radiative offset, the green line spectral peak position, and the width of the instrument spectral line shape. The latter are kept constant at the different altitudes, whereas the regularization for atomic oxygen and the radiative offset were adjusted in such a way that the altitude resolution of the retrieved values is of the order of the vertical field of view of the instrumental data (about 3 km). A priori profiles of atomic oxygen as well as other atmospheric parameters (temperature, N₂, and O₂ abundance) needed in the model are taken from the Mass Spectrometer and Incoherent Scatter (MSIS) model [Picone et al., 2002] or from colocated SABER measurements (see below).

The retrieval altitude grid is similar to the line of sight tangent altitudes. The regularization term is adjusted in such a way that the influence of the a priori atomic oxygen profiles is virtually absent in the 90–105 km altitude regime. The accuracy of the retrieved atomic oxygen densities is determined by uncertainties in the forward model rate constants (Table 1) and the background atmosphere. The largest source of uncertainty is temperature: a 10% uncertainty in this quantity maps nearly linearly into the atomic oxygen uncertainty.

Figure 2. Temporal evolution of atomic oxygen abundance at 99 km.

3. Measurements and Results

SCIAMACHY measurements of nighttime green line limb emissions exhibit a signal-to-noise ratio of the order of 1, which is too small to make a single profile retrieval. Therefore, we calculate monthly and 5° latitudinal mean spectra first (Figure 1). The number of spectra averaged in each latitude bin is a few hundred at high latitudes and more than 1000 at low latitudes. In this step, we also apply the empirical auroral model of Zhang and Paxton [2008] to reject all profiles, which were exposed to an average electron density of more than 0.1 keV.

In the next step, we fit an entire vertical profile of atomic oxygen to a sequence of limb spectra. Mathematically, this fitting is a nonlinear inversion problem. It is solved by minimizing a cost function containing the difference between the model of O(¹S) emissions and the real measurements, weighted by the inverse measurement error. In addition, a regularization term is added to the cost function. In our case, this is a Tikhonov-type matrix

Uncertainties in the abundance of O₂ and N₂ are negligible. The rate constant k'_{OOM} is the largest source of uncertainty of the forward model parameters. A 10% uncertainty affects retrieved atomic oxygen by about 8%. Uncertainties of all other forward model parameters together affect atomic oxygen by another 5% so that the overall (root sum square) error due to the rate constants is of the order of 10%. Adding temperature uncertainties results in an overall uncertainty of 15% for a 10% uncertainty in temperature.

The latitudinal coverage of the nighttime limb spectra is 50°S to 80°N before the auroral filter is applied and about 50°S to 60°N afterward

Figure 3. Latitude-altitude cross sections of atomic oxygen abundance for 2007 retrieved from SCIAMACHY green line emission data. (top) MSIS background atmosphere. (bottom) SABER background atmosphere. Each plot represents monthly mean data.

instruments, which measure atomic oxygen from a precessing satellite orbit at various local times, observed a similar latitudinal structure at 10 P.M. local time [Smith *et al.*, 2010; Russell *et al.*, 2005].

The tidal structures as well as the absolute values of atomic oxygen might be biased by the background atmosphere utilized in the model of $O(^1S)$. To investigate this effect, we repeat the retrieval of atomic oxygen utilizing colocated SABER measurements of the background atmosphere instead of MSIS model data.

(Figure 2). The restriction of the latitudinal coverage in comparison to the ground track of a near-polar satellite is caused by the seasonality of solar illumination and various calibration measurements made at the night-side of the satellite orbit. The amount of nighttime limb measurements increased significantly in 2004 due to a change in the SCIAMACHY instrument operations [e.g., Kaufmann *et al.*, 2010]. The typical number of profiles per month and 5° latitude is 500–1000; at outermost latitudes, it is 100–400.

The temporal evolution of the data shows annual and semiannual oscillations with typical amplitudes of 5–15%. Largest atomic oxygen values are observed during equinox at midlatitudes, which is a consequence of the semiannual variation of the amplitude of the migrating diurnal tide [Smith *et al.*, 2010] transporting atomic oxygen up and down. A multilinear regression analysis reveals a pronounced correlation with $F_{10.7}$ solar flux data as well, which is difficult to identify visually in Figure 2. Annual mean differences of atomic oxygen densities between the maximum (2002) and minimum (2008) of the 23rd solar cycle reveal variations of 5% at 90 km and 25% at 102 km being in phase with the solar flux. von Savigny and Lednyts'kyy [2013] derived a similar distribution of SCIAMACHY atomic oxygen data when they studied the quenching of OH^* by atomic oxygen.

The latitudinal distribution of atomic oxygen density (Figure 3, top) shows smallest values at the equator (at low altitudes) and largest values at $30^\circ N$ – $60^\circ N$, depending on season. This is a local time effect caused by atmospheric tides. The local time at which the satellite observations are taken is such that oxygen is displaced upward over the equator and downward in midlatitudes. The WINDII and SABER

Figure 4. (left) Zonal mean atomic oxygen abundance for (top left) fall 2005 and (bottom left) winter 2005/2006 at 0° – 20° N. WINDII data are for 1993 and 1993/1994, respectively. The WINDII data were derived from green line emission data, OSIRIS from O_2 A-band emissions, and SABER from OH^* . (right) Zonal mean atomic oxygen abundance for September 2005 at (top right) 20° – 40° N and (bottom right) 40° – 60° N.

The coincidence criteria are 2.5° latitude and 1 h local time. The latitudinal structures in SABER temperature and total density (not shown here) are more pronounced than in MSIS data. The utilization of this data amplifies atomic oxygen tidal patterns generally, particularly due to the temperature dependency of the rate constants. (Figure 3, bottom). However, the global mean absolute values are very similar in both cases; differences are typically less than 20%.

As mentioned in the introduction, the absolute values of atomic oxygen at the mesopause region are affected by large uncertainties and have been a matter of debate for several decades. A comparison between the SCIAMACHY atomic oxygen data set as derived in this study and other experimental data sets (from WINDII, OSIRIS, and SABER) is illustrated in Figure 4 (left) for low latitudes (0° – 20° N) and for fall (September, October, and November) as well as for winter conditions (December and January). All data sets are for 10 P.M. local time, except for OSIRIS, which is for 6:30 P.M. The vertical resolution of the data varies between 1 km to about 3 km. All data were collected during fall 2005 and winter 2005/2006, respectively, except for WINDII data, which is for 1993 and 1993/1994, respectively. The OSIRIS and WINDII data were extracted from Figure 9 in Sheese *et al.* [2011] and Figures 4 and 5 in Russell *et al.* [2005], respectively. All data sets were recorded at the same 11 year solar cycle phase.

The comparison of these data sets reveals the following: All measurements which rely on emissions resulting from the atomic oxygen recombination reaction (1), namely, from SCIAMACHY, OSIRIS, and WINDII, show rather similar values, which are typically 3 – 5×10^{11} mol/cm³ at atomic oxygen peak altitudes. The MSIS model data at low latitudes are similar (fall) to 60% larger (winter) than SCIAMACHY data at atomic oxygen peak altitudes. Largest atomic oxygen values are observed by the SABER instrument, which show more than

6×10^{11} mol/cm³ for the two seasons shown in Figure 4. SABER atomic oxygen mean values were calculated by screening out individual data points showing more than 12.5×10^{11} mol/cm³ as recommended by Mlynczak *et al.* [2013a]. On a daily average (not shown), SABER values are somewhat smaller than the 10 P.M. data, in accordance with the data presented by Mlynczak *et al.* [2013a].

Figure 4 (right) shows a comparison between SCIAMACHY, SABER, and MSIS atomic oxygen abundances for 20°–40°N and 40°–60°N in September 2005. In both latitude bands, SCIAMACHY peak values are of the order of 6×10^{11} mol/cm³, whereas MSIS values are about 30% lower and SABER data about 30% higher, respectively. Latitudinal and seasonal variations in the differences between MSIS and measured data should be considered with some caution, because MSIS accounts for the density effect associated with tidal temperature variations but neglects vertical motions of the oxygen related to the tides [Russell *et al.*, 2005]. Since vertical motions dominate the tidal behavior of the oxygen [Ward, 1998], this affects the diurnal as well as the seasonal distribution of MSIS atomic oxygen.

4. Conclusions

SCIAMACHY atomic oxygen nighttime green line limb emission data provide the basis for a new data set of atomic oxygen in the UMLT region. The absolute values of these data are similar to other satellite data sets also using the chemiluminescence of O(¹S) and O₂^{*}, but significantly smaller than SABER data. A possible reason could be the usage of different emissions for the derivation of atomic oxygen. However, WINDII atomic oxygen densities derived from simultaneously measured O(¹S) and OH^{*} emissions agreed fairly well where they overlapped in altitude. The agreement between SCIAMACHY, WINDII, and OSIRIS might be inherent in the system, because the photochemical models for the O(¹S) green line and O₂(b¹Σ_g⁺) A-band emissions rely, at least in part, on the same processes for the formation of the emitting states—so these measurements are not independent in a photochemical sense. Widely used MSIS atomic oxygen abundances are similar or larger at low latitudes (depending on season) and smaller at higher latitudes than SCIAMACHY data.

In view of the energy budget of the mesopause region, SCIAMACHY atomic oxygen densities indicate lower chemical heating than SABER. To balance the radiative cooling by carbon dioxide, as, i.e., measured by the SABER instrument [Mlynczak *et al.*, 2013b], this would leave more space for other processes such as heat deposition by gravity wave dissipation [e.g., Medvedev and Klaassen, 2003].

Seeing from another perspective, the difference between SCIAMACHY and SABER atomic oxygen measurements might also indicate deficits in key rate coefficients applied in the underlying photochemical/nonlocal thermodynamic equilibrium models.

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