

# VALIDATION OF TROPOSPHERIC SPECIES MEASURED BY SCIAMACHY USING THE AMAXDOAS INSTRUMENT ON BOARD THE DLR FALCON

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## ABSTRACT/RESUME

The AMAXDOAS (Air Borne Multi Axis Differential Optical Absorption Spectroscopy) experiment was successfully operated on board the German research aircraft DLR Falcon in two connected major campaigns in September 2002. It consists of two spectrometers (covering the UV and visible spectral range), each connected to several telescopes directed in different viewing angles above and below the aircraft. This set-up makes it possible to separate the tropospheric and stratospheric trace gas columns for a significant number of the observed species (e.g. O<sub>3</sub>, NO<sub>2</sub>, OClO, BrO, H<sub>2</sub>O, O<sub>4</sub>, SO<sub>2</sub>, and HCHO). The selection of trace gases and the separation of the troposphere and stratosphere makes this instrument well suited for the validation of SCIAMACHY on ENVISAT. Together with the OLEX- (Lidar) and the ASUR (Microwave) sensor the AMAXDOAS was operated during two campaigns ranging from Munich to Spitsbergen and Greenland and back and from Munich to Africa and the Seychelles (see also [1]).

This article reports on the first AMAXDOAS results for the troposphere. Because of the limited time for data analysis the focus of this study is only on two species, H<sub>2</sub>O and the oxygen dimer O<sub>4</sub>. In addition, also the measured intensities from zenith and nadir direction were investigated. From the O<sub>4</sub> absorptions and the measured intensities information on aerosols, clouds and the ground albedo can be derived, which is of great importance for the validation of simultaneous SCIAMACHY observations.

The H<sub>2</sub>O vertical column density (VCD) observed by the AMAXDOAS instrument for cloud free scenes ranges from 0.6 to  $1.7 \cdot 10^{23}$  molec/cm<sup>2</sup> over land and from 0.5 to  $1.0 \cdot 10^{23}$  molec/cm<sup>2</sup> over ocean. The H<sub>2</sub>O VCDs above clouds are usually significantly lower. The first stratospheric AMAXDOAS data sets are presented in a separate contribution [2].

## 1 INTRODUCTION

The SCIAMACHY instrument aboard the European research satellite ENVISAT will for the first time provide stratospheric profiles and tropospheric column densities of many atmospheric parameters and constituents on a global scale [3,4].

For the validation of the SCIAMACHY trace gas products the AMAXDOAS measurements are particularly well suited because of several reasons:

- Similar spectral properties of the AMAXDOAS instruments (wavelength range: 300 - 570 nm; spectral resolution: 0.5 - 1.2 nm FWHM) allow the observation of a large variety of SCIAMACHY target species (O<sub>3</sub>, NO<sub>2</sub>, OClO, BrO, H<sub>2</sub>O, O<sub>4</sub>, SO<sub>2</sub>, and HCHO) with similar sensitivity.
- The simultaneous measurements of different viewing directions (nadir, zenith and several elevation angles above and below the aircraft) allows to separate the stratospheric and tropospheric columns (see e.g. [5,6]).
- Aircraft measurements have the advantage to measure over great distances, covering different climatic zones, solar zenith angles, etc. with a single instrument. In particular remote regions can be reached.
- The spatial variability across a SCIAMACHY ground pixel can be observed. This is in particular important for tropospheric observations which are typically strongly affected by a changing cloud coverage.
- Because of the great flexibility of aircraft measurements it is possible to combine the AMAXDOAS measurements with simultaneous observations from different platforms. The information on the spatial variability derived from the AMAXDOAS measurements can be combined with information on the diurnal variation and the vertical distribution derived from ground based and balloon borne observations, respectively.

Since the first measurement campaign in September 2002 only part of the observed spectra could be analysed. Here we give an overview on the first tropospheric H<sub>2</sub>O and O<sub>4</sub> results for a flight track from Nairobi to the Seychelles on September, 19. The results indicate that the AMAXDOAS measurements can resolve the high variability of tropospheric trace gas distributions. In addition, also the rapidly varying cloud influence can be well captured. Both information is of great importance for the validation of the collocated SCIAMACHY measurement which represents an average across the whole ground pixel. Unfortunately, so far no SCIAMACHY data of O<sub>4</sub> and H<sub>2</sub>O

for the comparison with the AMAXDOAS observations were available. Thus the actual validation exercise has to be done in the future.

## 2 INSTRUMENT AND DATA ANALYSIS

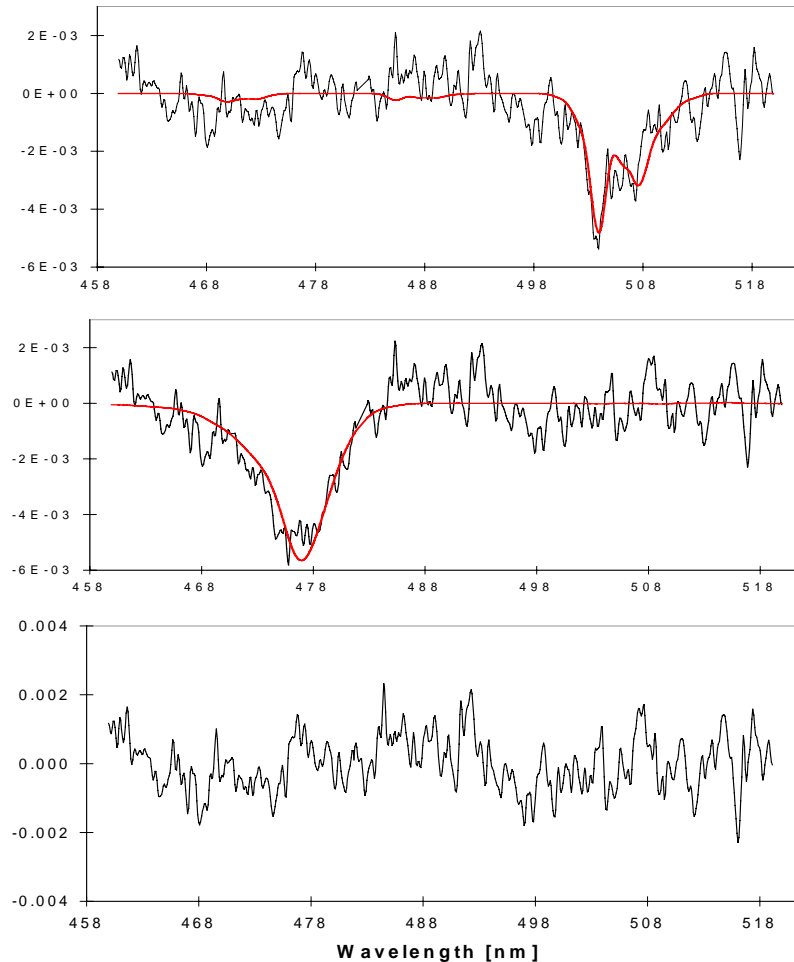


Fig. 1. DOAS fitting result for a spectrum from the zenith viewing direction (6:24 UTC). The laboratory spectra of  $\text{H}_2\text{O}$  and  $\text{O}_4$  (thick lines) are scaled to the absorptions found in the AMAXDOAS measurement. Please note that the absorptions found in zenith directions are typically much weaker compared to the nadir spectra (see also Fig. 2).

Since the instrumental set-up is already described in a previous publication [5] we give only a short overview here. Two separate DOAS instruments one for the UV (300 to 440 nm) and one for the visible (400 to 570 nm) are employed. The detectors consist of two-dimensional CCD arrays, which allow the simultaneous measurement of up to ten distinct spectra of different viewing angles (in the September 2002 campaigns we used 4 viewing direction). In contrast to many Multi Axis DOAS measurements from the ground [7, 8, 9] which scan different elevation angles sequentially, this set-up allows a higher temporal resolution (several measurements per minute) which is especially important for a fast flying aircraft like the Falcon ( $\approx 800$  km/h). For a typical integration time of 30 s the spatial resolution of the AMAXDOAS on board the DLR Falcon is about  $\approx 6.5$  km.

The measured spectra were analysed using the DOAS technique [10]. Several reference spectra of the trace gases which show structured absorptions in the respective wavelength regions are fitted to the logarithm of the measured spectrum using a non linear least squares algorithm [11]. For the analysis of  $\text{H}_2\text{O}$  and  $\text{O}_4$  absorptions the wavelength region between 460 and 519 nm was taken into consideration. For  $\text{O}_4$  and  $\text{H}_2\text{O}$  absorption cross sections from [12] and from the HITRAN data base [13] were used, respectively. Also the cross sections for  $\text{NO}_2$  [14] and  $\text{O}_3$  [15] were included into the fitting routine. In Fig. 1 results of the spectral analysis for  $\text{O}_4$  and  $\text{H}_2\text{O}$  are shown. The strong Fraunhofer structures of the measured spectra are removed using a solar reference spectrum usually taken at low solar zenith angle (SZA) and high flight altitude. In contrast to satellite observations where the solar reference spectrum (direct sun light) contains no atmospheric absorption structures, the AMAXDOAS analysis yields no absolute atmospheric column density (SCD) but the difference in the column densities between

the measurement and the solar reference spectrum (DSCD). For H<sub>2</sub>O and O<sub>4</sub>, however, solar reference spectra could be identified which only contain negligible atmospheric absorptions. These spectra were recorded at high flight altitude above a high cloud layer (between 7:15 and 7:25 UTC on September, 19). Thus we consider the AMAXDOAS O<sub>4</sub> and H<sub>2</sub>O fitting results as SCDs (instead of DSCDs). Usually the SCD is converted into the more appropriated VCD using the air mass factors (AMF) concept [16, 17]. For fine structured absorptions like those of H<sub>2</sub>O also the non linearity (often also referred to as saturation effect) between the true SCD and the measured absorption has to be corrected [18,19]. In this preliminary analysis we mainly focus on trace gas SCDs without conversion into VCDs and without a correction of the non-linearity. Both corrections will be the subject of a more detailed analysis. Nevertheless, also for the uncorrected SCDs several important conclusions can be drawn. Moreover, the uncorrected trace gas SCDs are particularly well suited for the validation of the 'raw' SCIAMACHY trace gas SCDs.

### 3 RESULTS

In Fig. 2 the time series of the O<sub>4</sub> and H<sub>2</sub>O measurements as well as the detected intensities are shown for the zenith direction (filled symbols) and nadir direction (open symbols). Also shown is the altitude profile of the Lidar backscatter ratio between 13 and 28 km (taken from [21]). The flight consists of three major parts: measurements on ground before the start, ascent to 33000 ft, and measurements at flight altitude (33000 - 37000 ft). The solar zenith angle (SZA) decreases from 70° before the start to 20° at the end of the flight.

All three measured quantities (intensity, O<sub>4</sub> SCD, H<sub>2</sub>O SCD) depend in particular on two quantities: the flight altitude, and the cloud cover (clouds below the aircraft). They also depend to a lesser degree on the SZA, the ground albedo, and on the cloud layer above the aircraft. The H<sub>2</sub>O SCD also depends on the tropospheric water vapour concentration. In the following the results are discussed in more detail:

#### 3.1 Influence of clouds below the aircraft

Clouds typically cause a strong and rapid variation of the nadir intensities because they are typically much brighter than the albedo of the surface. Since clouds (especially at high altitudes) shield significant parts of the total tropospheric column, the nadir O<sub>4</sub> and H<sub>2</sub>O SCDs are typically decreased when increased nadir intensities indicate clouds. This shielding effect of clouds on the nadir observations can be seen for several observations during the whole flight; a pronounced event is around 6:23 UTC and at the end of the flight, when strongly enhanced nadir intensities correspond with significantly decreased nadir SCDs of O<sub>4</sub> and H<sub>2</sub>O (see also Fig. 3).

A significant cloud influence can be also seen for the zenith measurements before the start. However, for these cases the cloud influence is different [20].

For nadir observations at flight altitude cloud free scenes can be identified by low intensities; between 5:15 and 6:45 UTC about 50% of all nadir measurements can be identified as cloud free. From these clear sky nadir measurements the total atmospheric column can be derived.

Although during most parts of the flight an anticorrelation between the nadir intensity and the trace gas SCDs is seen (see Fig. 3) there are also some exceptions; the most pronounced at about 6:50 UTC. These observations are most probably related to very low clouds which overcompensate the shielding effect by an enhancement of the absorption path due to multiple Mie-scattering inside the clouds.

These complex findings indicate the importance of an sophisticated cloud correction. Nevertheless, for a direct comparison between AMAXDOAS and SCIAMACHY measurements both sensors should be similarly affected by cloud influences. This is a strong advantage of the validation by AMAXDOAS.

#### 3.2 Height profiles of O<sub>4</sub> and H<sub>2</sub>O; Influence of the flight altitude

Immediately after the start a rapid decline of the H<sub>2</sub>O and O<sub>4</sub> SCDs for the zenith direction is observed. This decrease is caused by the decreasing fraction of the total atmospheric column above the aircraft. From the knowledge of the flight altitude height profiles of O<sub>4</sub> and H<sub>2</sub>O can be directly retrieved, which can in particular serve as input for radiative transport calculations (AMF determination).

#### 3.3 Influence of the ground albedo

It is aimed to apply an absolute radiance calibration to the AMAXDOAS instruments [22]. Absolute radiances are an important information for the validation of SCIAMACHY level 1 data. Even without this absolute radiance calibration the intensity measurements from nadir and zenith can yield important information on clouds (see above) or on the ground albedo. For the flight from Nairobi to the Seychelles the aircraft crossed the African coast around 5:38 UTC. In Fig. 2 it can be seen that at this time the minimum intensities (indicating cloud free scenes) significantly decrease (for 520 nm, where the albedo for land is larger than for ocean).

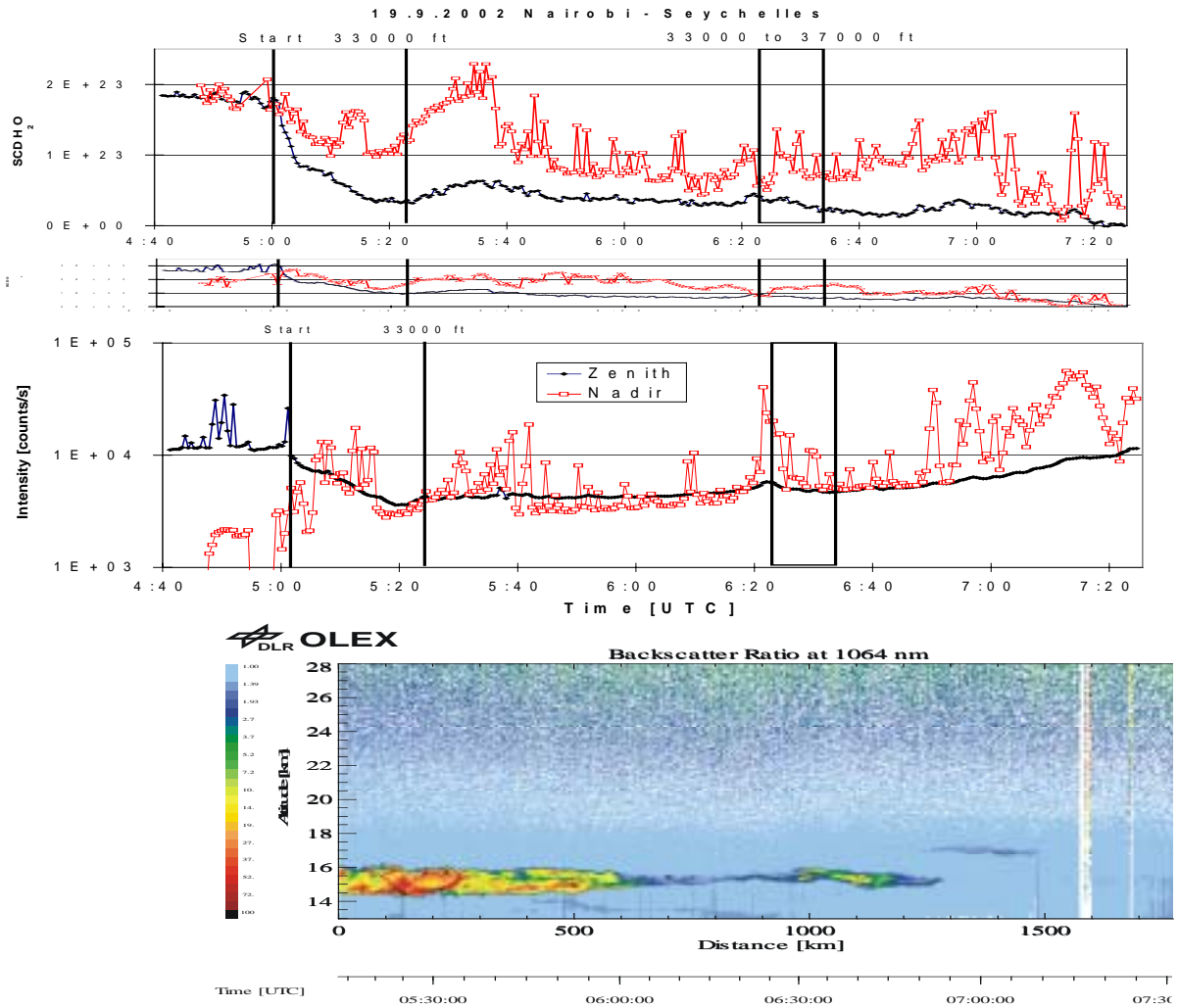


Fig. 2 Trace gas SCDs (for H<sub>2</sub>O, top, and O<sub>4</sub>, below) as well as the observed intensities during the selected flight from Nairobi to the Seychelles on September, 19, 2002. Red open symbols indicate nadir observations, black filled symbols zenith observations. At the bottom the altitude profile of the backscatter ratio measured by the OLEX Lidar is displayed [21]. For the first half of the flight an extended week cloud around 15 km was observed. The vertical lines indicate the time of the start, the arrival at flight level (33000 ft) as well as the ascent from 33000 to 37000 ft.

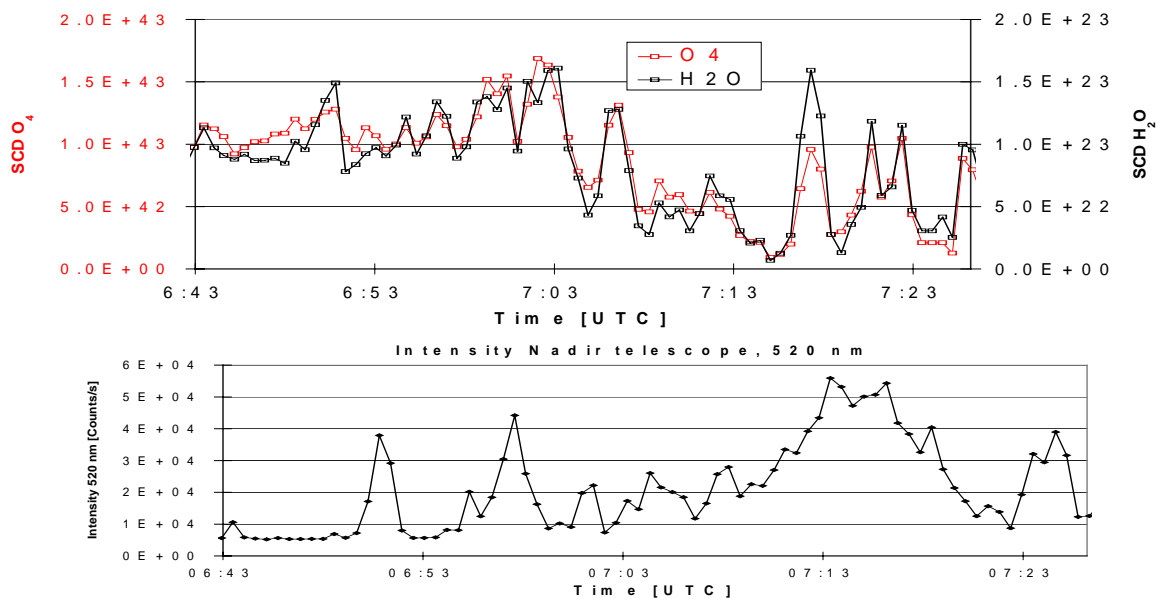


Fig. 3 Nadir trace gas SCDs and intensities for the last part of the flight. High intensities indicate clouds below the aircraft. An anticorrelation between the trace gas measurements and the intensities is found for almost all measurements (shielding effect).

### 3.4 Selection of solar reference spectra

As discussed in section 2) the choice of suitable solar reference spectra is important for the determination of absolute SCDs. After about 6:55 UTC increased nadir intensities indicate a continuous cloud coverage (this is also confirmed from the visual observations during the flight). The nadir O<sub>4</sub> and H<sub>2</sub>O SCDs show the lowest values during the whole flight which are likely caused by high clouds. Especially for H<sub>2</sub>O with its surface near profile (similar also for O<sub>4</sub>) we conclude that nearly the total atmospheric column is shielded by the clouds. We selected the measurements during these period as solar reference spectra for the DOAS analysis (see above).

The choice of these solar reference spectra can be tested considering the zenith and nadir measurements before the start. For these conditions both directions should yield similar results because they both receive light which has penetrated the whole atmosphere (for the nadir direction via reflection on the ground). For H<sub>2</sub>O similar values can indeed be observed for nadir and zenith, but for O<sub>4</sub> differences occur. These might be caused by differences in the altitude profiles for O<sub>4</sub> and H<sub>2</sub>O, since the absorption paths through the atmosphere are different for zenith (field of view  $\approx 1^\circ$ ) and nadir observations (field of view via reflection on the ground  $\approx 180^\circ$ ). However, we can't rule out measurement errors because of the very small light intensity for the nadir telescopes before the start.

### 3.5 Radiative transfer considerations for zenith measurements at clear and cloudy sky

Because of the complex observing geometry AMAXDOAS observations of trace gas absorptions and intensities are an excellent possibility for the test of radiative transfer models. One interesting finding is that the zenith measurements of H<sub>2</sub>O and O<sub>4</sub> at flight altitude are systematically larger than expected for the small trace gas concentrations above the aircraft (only less than a percent of the total atmospheric column for H<sub>2</sub>O and a few percent for O<sub>4</sub> are located above flight altitude). This indicates that a significant fraction of the detected light in zenith direction has already 'seen' the atmosphere below the aircraft (where higher concentrations of O<sub>4</sub> and H<sub>2</sub>O exist. This finding is also confirmed by radiative transport modelling [6] and by the intensity enhancement for the zenith telescope when the nadir intensities indicate a bright cloud below the aircraft. Especially around 6:23 UTC also enhanced zenith SCDs for O<sub>4</sub> and H<sub>2</sub>O also indicate that a higher percentage of the zenith photons have seen higher concentrations of O<sub>4</sub> and H<sub>2</sub>O from altitudes below the aircraft. Please note also that the zenith SCDs of O<sub>4</sub> and H<sub>2</sub>O show very similar variations during the whole flight (see Fig. 2).

### 3.6 Influence of clouds above the aircraft

The OLEX Lidar observations (see [21]) indicate an extended layer of enhanced Mie-scattering at about 15 km. Especially around 5:25 and 6:40 UTC large backscatter ratios were detected. For these times also in the zenith intensities slight variations are found. Modelling of these intensities (and also the respective trace gas absorptions) could yield additional information about this cloud influence.

### 3.7 Results for H<sub>2</sub>O

Useful atmospheric H<sub>2</sub>O information can be retrieved from the zenith measurements during the ascent (see section 3.2) and from nadir measurements for cloud free scenes. From the decrease of the zenith H<sub>2</sub>O SCD after the start we find that the H<sub>2</sub>O SCD within the boundary (below  $\approx 2$  km) is about  $1 \cdot 10^{23}$  molec/cm<sup>2</sup>. From 2 km up to the flight altitude at 11 km the H<sub>2</sub>O SCD is about  $0.5 \cdot 10^{23}$  molec/cm<sup>2</sup>. A rough conversion into VCDs (see section 2) yields a H<sub>2</sub>O VCD of about  $0.7 \cdot 10^{23}$  molec/cm<sup>2</sup> for the boundary layer and of about  $0.3 \cdot 10^{23}$  molec/cm<sup>2</sup> for the free troposphere.

From nadir observations at flight level highly varying H<sub>2</sub>O SCDs are observed (even for cloud free scenes). In general, the H<sub>2</sub>O SCDs over land (before 5:38 UTC) are higher than over ocean; the highest values are found around 5:35 UTC (about  $2 \cdot 10^{23}$  molec/cm<sup>2</sup> for cloud free conditions). A rough conversion into H<sub>2</sub>O VCDs yields values ranging from  $0.6$  to  $1.7 \cdot 10^{23}$  molec/cm<sup>2</sup> over land and from  $0.5$  to  $1.0 \cdot 10^{23}$  molec/cm<sup>2</sup> over the ocean. The H<sub>2</sub>O VCDs above clouds are usually significantly lower.

Unfortunately no SCIAMACHY data for H<sub>2</sub>O and O<sub>4</sub> are available so far for validation. However, we compared the AMAXDOAS H<sub>2</sub>O SCDs to those measured by GOME over the flight track over the ocean (from 43° to 52° longitude) at 6:60 UTC. The GOME H<sub>2</sub>O SCDs are between  $1 \cdot 10^{23}$  molec/cm<sup>2</sup> and  $1.4 \cdot 10^{23}$  molec/cm<sup>2</sup> (see [23]) which is in good agreement with the nadir H<sub>2</sub>O SCDs measured by AMAXDOAS.

### 3.8 Results for O<sub>4</sub>

Similarly to H<sub>2</sub>O also O<sub>4</sub> information can be retrieved from the zenith measurements during the ascent and from nadir measurements for cloud free scenes. From the total decrease of the nadir O<sub>4</sub> SCD during ascent ( $3.1 \cdot 10^{43}$  molec<sup>2</sup>/cm<sup>5</sup>) a O<sub>4</sub> VCD of  $0.9 \cdot 10^{43}$  molec<sup>2</sup>/cm<sup>5</sup> is derived. This is lower than the total atmospheric column derived from the atmospheric pressure and temperature profile (about  $1.25 \cdot 10^{43}$  molec<sup>2</sup>/cm<sup>5</sup>). This apparent discrepancy results mainly from the overestimated O<sub>4</sub> values at flight altitude (see section 3.5).

The clear sky nadir O<sub>4</sub> SCDs at flight altitude decrease during the flight because of the decreasing SZA. A rough conversion into O<sub>4</sub> VCDs yield about  $1 \cdot 10^{43}$  molec<sup>2</sup>/cm<sup>5</sup> for the first part and about  $0.6 \cdot 10^{43}$  molec<sup>2</sup>/cm<sup>5</sup> for the last part of the flight. Again, the total O<sub>4</sub> VCD seems to be underestimated, most likely because the O<sub>4</sub> absorption for the selected solar reference spectra is not negligible (see section 3.5). More precise O<sub>4</sub> (and also H<sub>2</sub>O) results will be derived in future analysis with more detailed radiative transfer calculations.

## 4 CONCLUSIONS

We presented the first tropospheric analysis from the novel AMAXDOAS observations during a major campaign during September 2002. Results for O<sub>4</sub>, H<sub>2</sub>O and the measured intensities were determined for a flight from Nairobi to the Seychelles on September, 19. Because of the limited time we show only preliminary data (mainly trace gas SCDs). We also applied a rough conversion into trace gas VCDs, but more precise data will be obtained in future analysis. It should be pointed out that already trace gas SCDs are well suited for the validation of similar 'intermediate products' of SCIAMACHY.

From the selected AMAXDOAS measurements tropospheric profiles of H<sub>2</sub>O and O<sub>4</sub> during the start can be derived. For H<sub>2</sub>O we found that  $\approx 0.7 \cdot 10^{23}$  molec/cm<sup>2</sup> reside in the boundary layer. During the flight the total VCD of H<sub>2</sub>O ranged from  $0.6$  to  $1.7 \cdot 10^{23}$  molec/cm<sup>2</sup> over land and from  $0.5$  to  $1.0 \cdot 10^{23}$  molec/cm<sup>2</sup> over ocean.

From the measured intensities as well as the retrieved O<sub>4</sub> (and H<sub>2</sub>O) absorptions important information on the ground albedo and the cloud cover could be retrieved. The latter is in particular important because clouds have a strong influence on satellite and aircraft observations of tropospheric species. For the selected flight about 50% of all measurements were influenced by clouds.

We found that the AMAXDOAS measurements are well suited to resolve both the variability of the atmospheric trace gases and those of clouds. Thus AMAXDOAS observations can yield spatial cross sections of trace gas absorptions and cloud information across the ground pixel of SCIAMACHY. From these cross sections average values can be derived which are needed for the validation of SCIAMACHY measurements.

Unfortunately, so far no tropospheric SCIAMACHY data of H<sub>2</sub>O and O<sub>4</sub> are available. Thus, the actual validation can only be performed in the future. However, we analysed GOME observations over the flight track over the ocean (from 43° to 52° longitude) at 6:60 UTC. The GOME H<sub>2</sub>O SCDs are between  $1 \cdot 10^{23}$  molec/cm<sup>2</sup> and  $1.4 \cdot 10^{23}$  molec/cm<sup>2</sup> which is in good agreement with the nadir observations of AMAXDOAS (see Fig. 2).

Future AMAXDOAS analysis will concentrate on further trace gases and on further flights. In particular more detailed radiative transport calculations will be applied to derive more precise tropospheric VCDs.

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