

# Ozone profile retrieval from limb scatter measurements in the HARTLEY bands: further retrieval details and profile comparisons

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**Abstract.** SCIAMACHY limb scatter radiance measurements at selected wavelengths in the HARTLEY bands have been used to retrieve ozone profiles in the upper stratosphere and lower mesosphere. Comparisons with profiles measured by a ground based radiometer in Norway, MIPAS on board ENVISAT, HALOE on UARS and MLS on AURA indicate an agreement within 15% between 40 and 55 km and show that the retrieval provides reliable ozone profiles at these altitudes. Above 55 km, an increasing overestimation is observed. Beside the profile comparisons, further retrieval features of the current retrieval (version 1.26) are described.

## 1 Introduction

Mesospheric ozone profiles are of interest, e.g. for investigations of Sun–Earth interactions like SPE s (Rohen et al., 2005) or of solar cycle effects. They are also required for modeling atmospheric dynamics, e.g. for climate forecasts.

First successful retrievals of mesospheric ozone profiles from limb-scattered solar radiation employed measurements from the UV-spectrometer on the Solar Mesosphere Explorer (SME) satellite (Thomas et al., 1980; Rusch et al., 1983). A routine retrieval of ozone in the HUGGINS bands from spectra measured by the Shuttle Ozone Limb Sounder Experiment (SOLSE) (McPeters et al., 2000) was difficult to establish due to a bad signal-to-noise ratio but introduced more

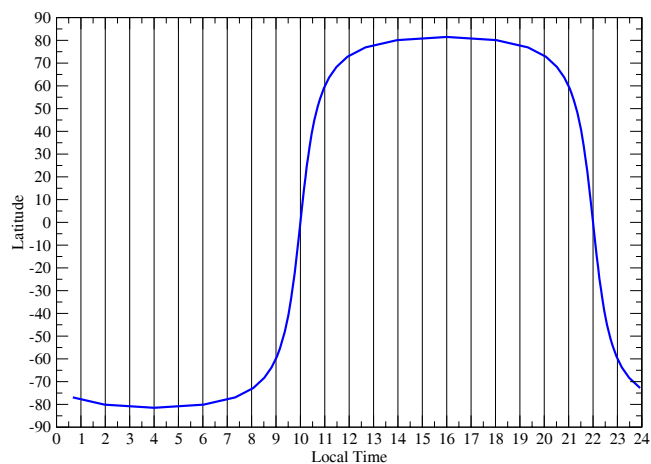
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sophisticated retrieval techniques which are now commonly used for retrievals from limb scatter measurements, e.g. doublet and triplet retrieval vector composition (Flittner et al., 2000). This paper provides further details on the retrieval algorithms and shows comparisons with profiles from a ground based radiometer, MIPAS on board ENVISAT, HALOE on UARS and MLS on AURA.

## 2 SCIAMACHY limb measurements

SCIAMACHY's (Bovensmann et al., 1999) local over-flight times vary mostly around 10:00 a.m. during the day (see Fig. 1).

The spectrometer measures limb-scattered solar radiation from the Earth's surface up to 95 km tangent height in steps of about 3.3 km (Schwab et al., 1996), its instantaneous field of view in limb mode is 110×2.6 km (horizontal×vertical) at the tangent point. The horizontal *across* track coverage is 960 km with a spatial resolution of 240 km at best, depending on the channel. For the spectral range used in this study only a single detector read-out is performed at each tangent height step leading to an across-track spatial resolution of 960 km. One single vertical limb scan takes 60 s. Within this period of time ENVISAT moves about 447 km. Moreover, the tangent point moves towards the spacecraft during an upward scan. This effect corresponds to a horizontal distance of about 145 km. Both effects lead to an apparent movement of the tangent point of about 300 km in flight direction.



**Fig. 1.** ENVISAT local overflight times.

SCIAMACHY consists of eight channels covering the spectral range between 214 and 2384 nm. The first channel covers the lowest spectral region up to 314 nm with a spectral resolution of 0.21 nm whose measurements are used for this presented retrieval.

### 3 Retrieval methodology and algorithm

The ozone profile retrieval methodology was originally proposed for air-borne limb measurements (e.g. Cunnold et al., 1973; Aruga and Heath, 1982). It is based on the fitting of measured and modeled limb radiance profiles at discrete wavelengths. The inversion technique applied to SME measurements, up to now the only successful ozone retrieval from satellite-borne limb scatter measurements in the UV-B, exploits radiances at 265 and 296.4 nm (Rusch et al., 1983); ozone profile retrievals at wavelengths in the HARTLEY bands from SOLSE observations (McPeters et al., 2000) have also been done using this method, but had difficulties due to a low signal-to-noise ratio. For their attempt of a routine profile retrieval, radiances at eleven discrete wavelengths between 300 and 355 nm have been used (McPeters et al., 2000). For this retrieval, the maximum of ozone absorption cross section in the HARTLEY bands (around 255 nm) (Wayne, 1987; Burrows et al., 1999; Bogumil et al., 2003) is used for ozone profile retrievals at highest possible altitudes (see Rohen et al. (2005); Rohen (2006); Rohen et al. (2006) for a description of the algorithm).

Thirteen wavelengths were selected at 250, 252, 254, 264, 267.5, 273, 283, 286, 288, 290.5, 305, 307, and 310 nm, avoiding emission lines while covering the full range in the HARTLEY bands (see Rohen (2006) for a detailed description of the potential contamination of limb scatter measurements by airglow emissions in the HARTLEY bands). Slight adjustments of the wavelengths have been done empirically.

The inversion of the measurement vector is performed by a nonlinear *Optimal Estimation* iteration scheme (Rodgers, 1976). For further details of the retrieval methodology and algorithm we refer to previous publications (Rohen et al., 2005; Rohen, 2006; Rohen et al., 2006). However, the reached sensitivity of the retrieval to ozone is proposed to be between about 35 and 60 km, sometimes 65 km.

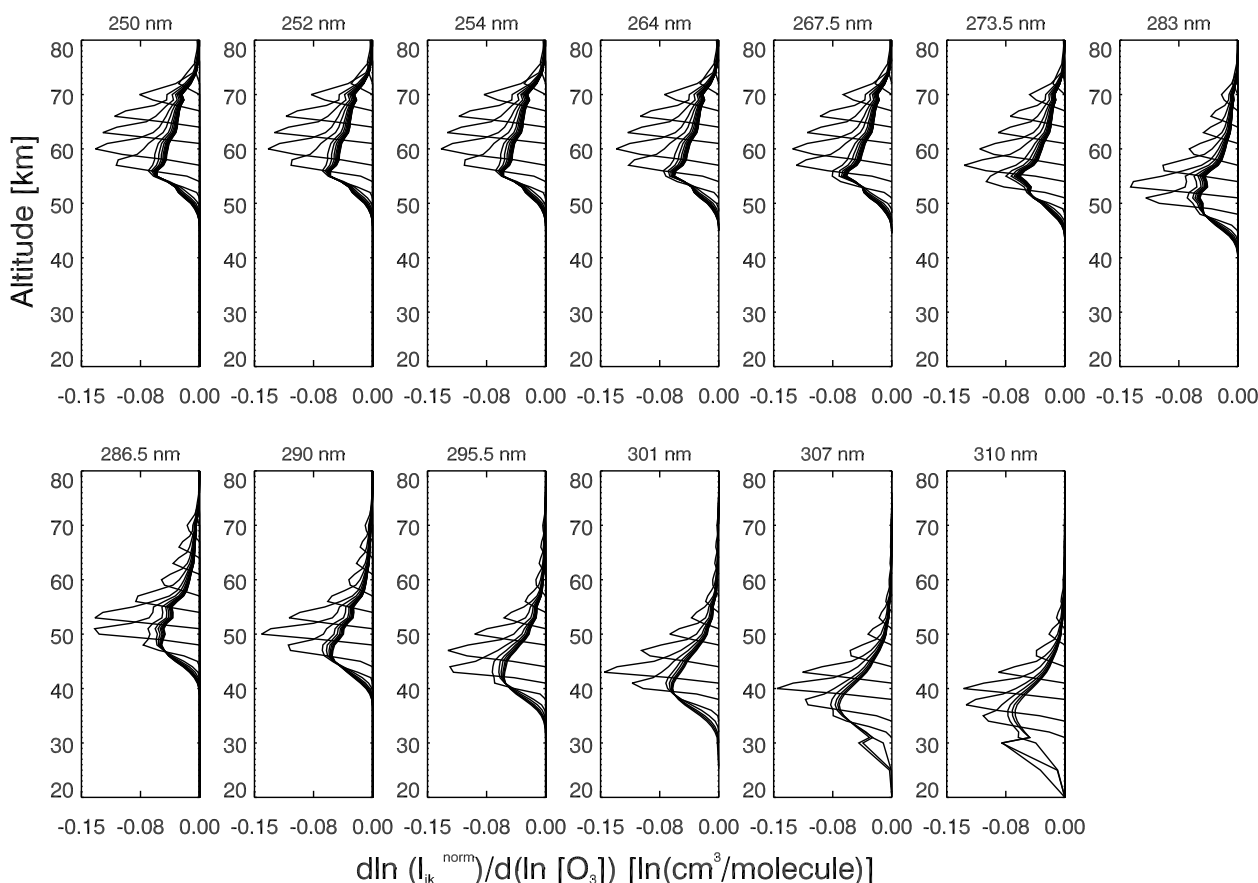
Weighting functions (Fig. 2) indicate that a continuous coverage of the sensitivity at these altitudes is given properly. Figure 3 shows fits and residuals for all thirteen wavelengths (sample fit). A bias can be seen at 250 and 254 nm.

Averaged residuals between 35 and 65 km are below 3% (Rohen et al., 2005).

In terms of a priori, the retrieval is relatively insensitive with respect to the assumed a priori (see averaging kernels in Fig. 3 of Rohen et al., 2006). Retrieved profiles based on different a priori (50% difference) differ by less than 4% for all relevant altitudes (Fig. 4). The multiple scattering contribution to the total limb radiance is relatively small, at least in the HARTLEY bands. The multiple scattering contribution for wavelengths below 300 nm is less than 1% (Oikarinen et al., 1999; Loughman et al., 2005, e.g.). Figure 4 shows the relative double scattering contribution, derived with SCIA-RAYS.

At 60 km tangent height, this contribution corresponds to about 5% at 310 nm and decreases to 1% below 290 nm. Sample inversions show that the induced differences in the retrieval due to the neglect of double scattering is below 2% at 40 km and decreases strongly with altitude. Therefore, only single scattering is considered in the retrieval.

One of the largest problems with SCIAMACHY limb data processing is still the tangent height misalignment (von Savigny et al., 2003). Many techniques have been suggested in order to improve the tangent height registration, e.g. using the shape of the radiance profiles (Janz et al., 1996; McPeters et al., 2000; Sioris et al., 2003; von Savigny et al., 2003). For this retrieval, tangent heights are corrected by tangent height offsets retrieved with the TRUE (Tangent height Retrieval by Ultraviolet-B Exploitation) VERSION14 technique (Kaiser et al., 2004; von Savigny et al., 2005). TRUE is based on an ozone-knee approach using the wavelength range between 295 and 305 nm. The tangent height offsets – tropical mean values averaged over the  $-20^{\circ}$  and  $20^{\circ}$  latitude range – are applied to all limb measurements of a corresponding orbit. The tangent height accuracy is estimated to be about 500 m after TRUE. Figure 5 shows the effect of tangent height shifts of the limb radiances of  $-2.5$  km and  $-0.5$  km on a sample ozone profile retrieval. A tangent height error of 2.5 km leads to differences in the ozone concentrations of up to 90%, and a 0.5 km tangent height inaccuracy results in errors of up to 17%. The reason why this error increases with altitude is still unclear. At least, a connection to the decreasing ozone scale height is unlikely due to the relatively small change of the scale height.



**Fig. 2.** Weighting functions for all thirteen wavelengths and all tangent heights from a sample profile retrieval. Solar zenith angle is  $79^\circ$ .

External stray light is another potential error source which is referred to as the contamination from outside the FOV, e.g. from clouds and bright Earth. SCIAMACHY stray light was estimated at 310 nm by subtraction of the modeled from the measured radiances (Fig. 5); radiances smaller than 310 nm are less affected; van Soest, 2005). Stray light at 76 km reaches 120% of the in-field signal whereas the impact on the profile retrieval itself is negligible. Induced differences never exceed 1% between 35 and 65 km; the normalization of the radiance profiles may help reducing the stray light effects. In summary, stray light has a large impact on the measurements only above around 70 km, and hence has almost no effect on the retrieval in the region of interest between 40 and 60 km.

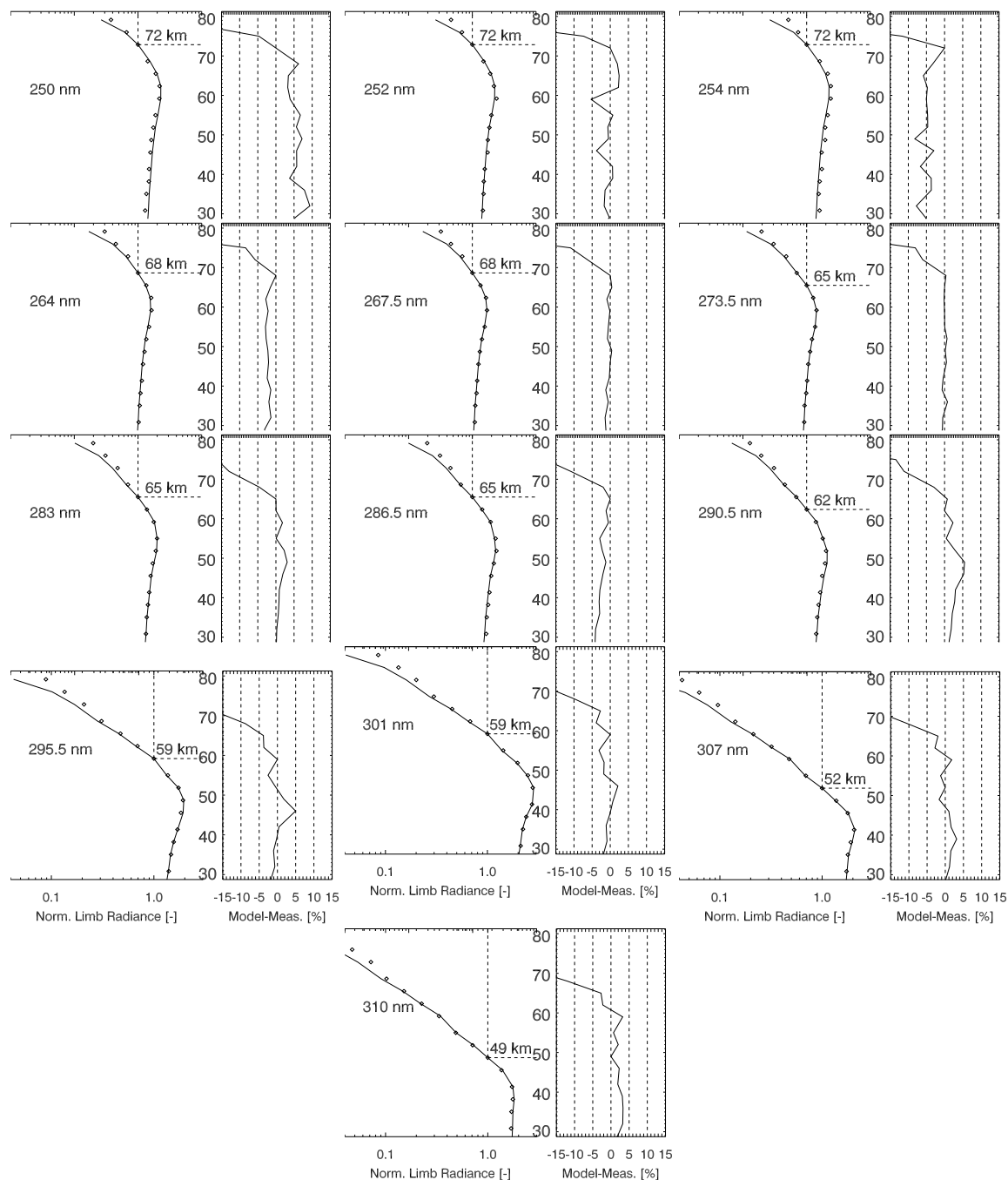
#### 4 Profile comparisons

Profile comparisons have been done with profiles measured by MIPAS on ENVISAT, the Radiometer for Atmospheric Measurements (RAM) at Spitsbergen, the HALogen Occultation Experiment (HALOE) on the Upper Atmosphere Re-

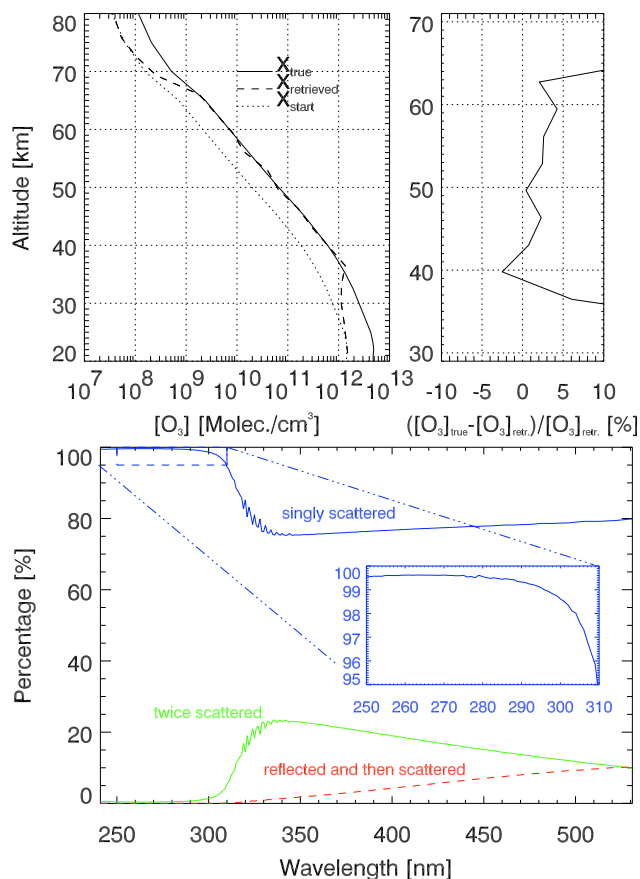
search Satellite (UARS) and the Microwave Limb Sounder (MLS) on AURA. SCIAMACHY Level 1 VERSION504 and TRUE VERSION14 have been used. Figure 6 shows the overall statistics for all instruments which are discussed in the next sections. The respective coincidence criteria are shown by the annotation in the respective figures. Measurements from the other instruments have been taken mostly in 2003 and 2004 covering the entire latitude bands.

The mean difference is below about 15%, depending on the altitude, with mean  $1\sigma$  deviation of below 20%. Comparisons with MLS and MIPAS profiles show an overestimation of the SCIAMACHY ozone concentrations at altitudes above 55 or 50 km.

Figure 7 shows a statistics of 432 comparisons between retrieved ozone profiles and the used a priori profiles from a sample data set in 2003 and 2004. The mean agreement between the compared profiles is within 20%. The standard deviations are obviously larger compared to those shown in Fig. 6, e.g. 30% at 40 km or 60% at 50 km. This shows the value added by the retrieval.



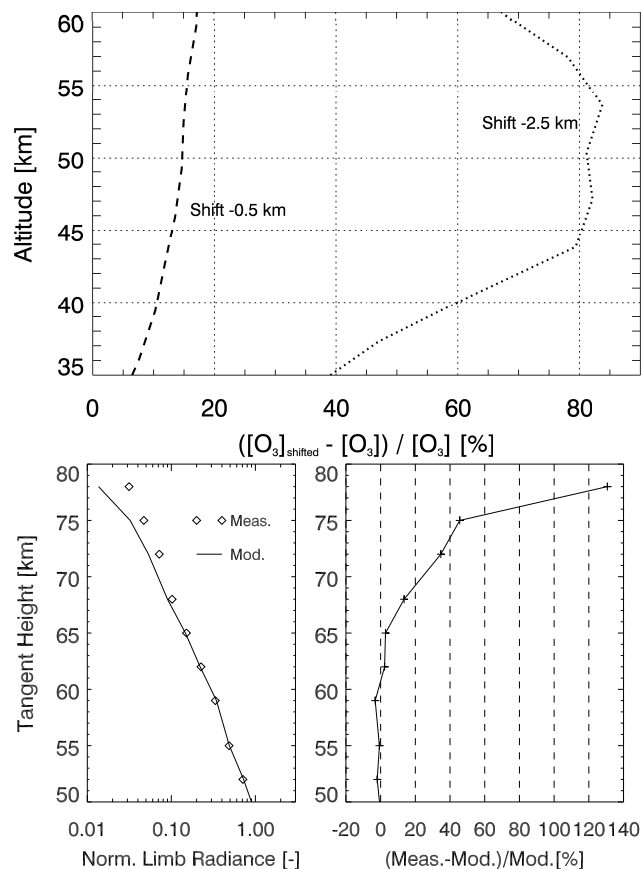
**Fig. 3.** Fits of limb radiance profiles for all thirteen wavelengths (sample profile retrieval, orbit 10945, number 1521, state 3). Dots in the respective left panels denote the measurement, the solid lines the model. Radiance profiles are normalized at a certain altitude, respectively, normalization points are denoted. Residuals are depicted in the right panel of the figures. The biases of the fits at 250 and 254 nm have not been found in every other sample retrieval. Above 70 km, contamination of the measurements by stray light is the reason for the general underestimation of the model.



**Fig. 4.** Retrieval features: Differences caused by different a priori which distinguish by 50%.  $x_{\text{start}}$  denotes the a priori. Right: Fraction of scattered and reflected photons at 60 km tangent height as modeled by SCIARAYS. Ozone absorption is considered, albedo=0.5. The fraction of double scattering at 30 km altitude is only slightly enlarged by a few percent relatively to that at 60 km. The inner box shows an enlargement for wavelengths between 250 and 310 nm.

Observations of ozone depletion during the Halloween solar storm in fall 2003 – using profiles from the presented profile retrieval – agree fairly well with observations of e.g. Global Ozone Monitoring by Occultation of Stars (GOMOS), or the Solar Backscatter Ultraviolet Instrument (SBUV II) (Rohen et al., 2005). Those comparisons also show the reliability of the profile retrieval in the altitudes between 40 and 60 km, beside the fact that the retrieval even works under extreme conditions like during a solar storm.

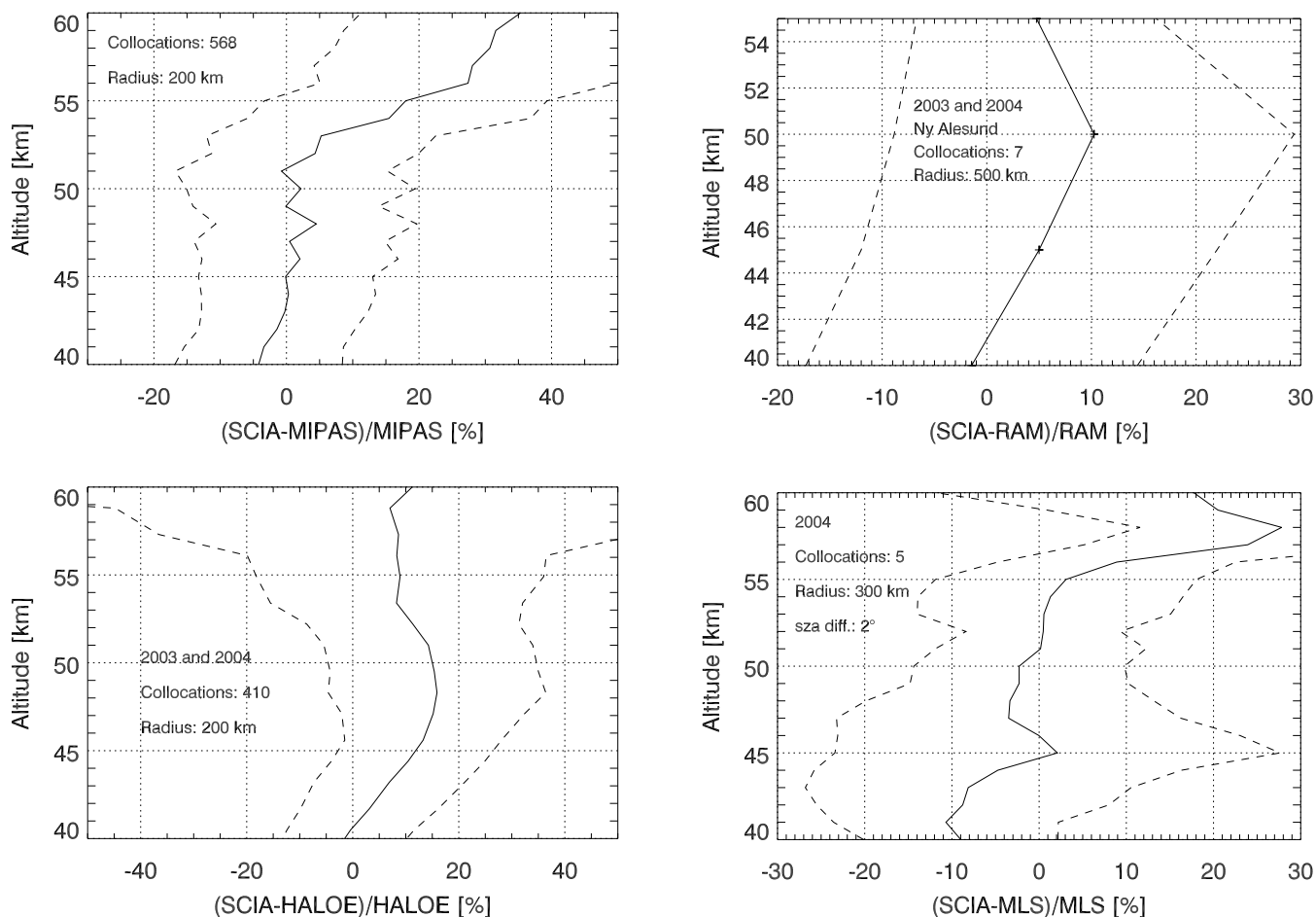
The MIPAS ozone profiles used for this comparison were retrieved at the Institut für Meteorologie und Klimaforschung (IMK) (von Clarmann et al., 2001, 2003; Glatthor et al., 2005; Glatthor et al., 2006, and references therein) and provide a vertical coverage from 6 km to 68 km. MIPAS's FOV is 30 km horizontally and 3 km vertically. The IMK MIPAS ozone profiles were recently shown to agree with POAMIII



**Fig. 5.** Sensitivity studies: (a) differences in sample ozone profiles caused by  $-0.5$  km and  $-2.5$  km tangent height shifts. (b) Stray light contamination: simulated and measured limb radiances at 310 nm from a sample retrieval and their difference in percent.

(Lumpe et al., 2003), LIDAR and ozone sonde measurements to within 10% for altitudes between 10 km and 60 km (Steck et al., 2007). MIPAS profiles have also been previously compared to SCIAMACHY stratospheric ozone observations and those of Global Ozone Monitoring by Occultation of Stars (GOMOS) (Bracher et al., 2005). In terms of GOMOS, the agreement of the compared profiles is also within 10% up to 60 km altitude. SCIAMACHY ozone profiles retrieved from an earlier processor VERSION216 have been compared already with MIPAS profiles in earlier studies (Rohen et al., 2006), but those have been retrieved with the operational ESA processor; they showed a slight underestimation. For the comparisons of our retrieved profiles with MIPAS profiles retrieved at the IMK, the following coincidence criteria were used: (a) the sza of the measurements do not differ by more than  $2^\circ$ , (b) co-location radii of 100 km or 200 km. We find good general agreement, but also increasing differences with altitude, particularly at altitudes above about 50 km.

An inappropriate a priori ozone profile is not a likely candidate as a reason for the observed differences because the



**Fig. 6.** Statistics from comparisons of SCIAMACHY (retrieval version 1.26) with profiles from MIPAS, RAM, HALOE, and MLS. Solid lines denote the mean deviation and dashed lines the  $1\sigma$  standard deviation. Only five collocations have been found in the case of MLS due to the strongly different local overflight times of the respective spacecrafts.

retrieved ozone concentrations are still fairly insensitive to the a priori at 55 km altitude. Furthermore, stray light was shown to have a small impact on the retrieval.

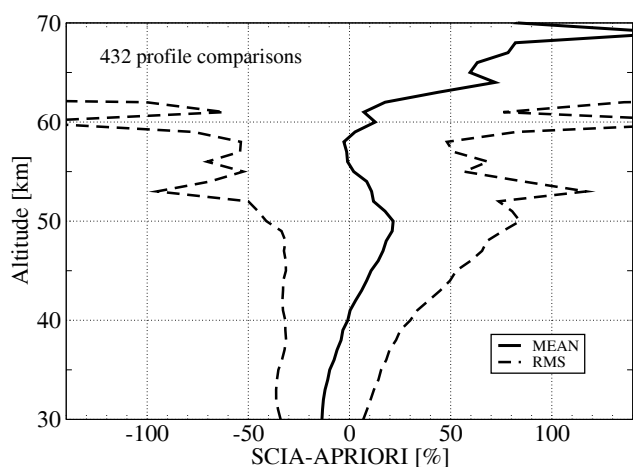
Comparisons of SCIAMACHY and MIPAS ozone profiles for different latitude bands and sza show no significant changes in the comparisons.

The current HALOE (Russell et al., 1993) ozone retrieval VERSION19 provides profiles up to 70 km. These were validated against numerous instruments, including ozone sondes, LIDARs, balloons, rocketsondes, and satellites (Brühl et al., 1996; Rusch et al., 1997). HALOE ozone profiles are in good agreement with SAGEII measurements up to 50 km altitude. Above 50 km, HALOE observations were found to overestimate ozone by up to 20% due to incorrect photochemical corrections (Natarajan et al., 2005; Nazaryan et al., 2005). This finding requires confirmation by a more extensive statistical analysis since the Natarajan et al. (2005) and Nazaryan et al. (2005) analyses are only based on sample cases.

Solar occultation measurements like those of HALOE are performed at local times which are characterized by minimum ozone concentrations (see Fig. 8).

In order to compare HALOE observations with SCIAMACHY limb observations, a one dimensional version of the Single Layer Isentropic Model of Chemistry And Transport (SLIMCAT) model (Chipperfield, 1999) photochemical model has been used in order to derive HALOE ozone profiles to the SCIAMACHY sza. Figure 8 shows exemplarily the ozone content at  $90^\circ$  and  $74^\circ$  sza which differ by up to 20% below 60 km and up to several hundred percent above 60 km (see also Rohen (2006) for details).

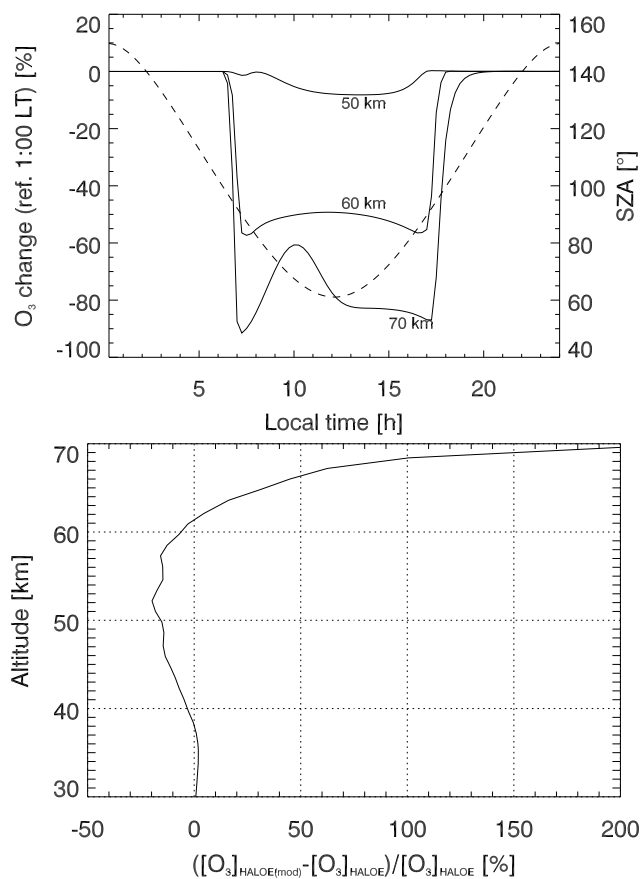
The comparison between HALOE and SCIAMACHY profiles indicates a fairly good agreement in terms of the statistical mean (Fig. 6). But the modeled HALOE ozone profiles have large errors and make it therefore difficult to conclude on the quality of the SCIAMACHY profiles, at least above 50 km. Below 50 km, the agreement is fairly good, similar to the one observed with MIPAS.



**Fig. 7.** Statistics from 432 comparisons between retrieved profiles and the corresponding used a priori profiles from a sample data set in 2003 and 2004.

Comparisons have also been done with ground based measurements from the Radiometer for Atmospheric Measurements (RAM) over Ny Ålesund (78° N, 11° E), Norway (Langer, 1999). Retrieved ozone profiles cover 15 to 55 km and exhibit a vertical resolution between 8 km and 20 km (Palm et al., 2005). RAM profiles were validated by ozone sondes between 18 km and 24 km within 10%, and with LIDAR instruments between 16 km and 34 km within 11% (Palm et al., 2005). MLS observations have been used for validation of RAM profiles between 20 and 55 km which also show an agreement within 10% (Langer, 1999). In order to compare SCIAMACHY and RAM ozone, SCIAMACHY ozone profiles were convolved with averaging kernels of the RAM ozone inversion. In spite of the poor vertical resolution, collocated ozone profiles indicate an agreement within 10% (Fig. 6). Only a slight overestimation is observable above 50 km, but this smaller differences above 50 km – as seen in the MIPAS comparisons – may be due to the poor vertical resolution of the RAM profiles. Again, a reliable statement of the quality of the retrieved profiles above 50 km through this comparison is difficult to give.

The last comparison has been done with profiles from the MLS instrument on AURA (VERSION15 (Froidevaux et al., 2006)). Validation has been done up to altitudes of 0.1 mbar (about 48 km) with ozone profiles measured by HALOE, Stratospheric Aerosol and Gas Experiment II (SAGEII), POAMIII, and Atmospheric Chemistry Experiment (ACE). MLS ozone profiles agree well within 5% with those profiles in the lower stratosphere but an underestimation in the upper stratosphere and an overestimation towards the mesosphere is has been observed. Although an upper limit of 0.46 mbar (about 54 km altitude) was recommended for the ozone product, good sensitivity in the upper mesosphere is also proposed (Livesey et al., 2005).



**Fig. 8.** SLIMCAT simulation of diurnal variability of ozone at 50, 60 and 70 km on 11 August 2002. The dashed line denotes the sza and the solid lines denote the ozone changes at the respective altitudes. Right: Relative differences between a HALOE sample ozone profile – photochemically modeled to 74° – and HALOE profile at 90° sza

A statistics from collocated SCIAMACHY and MLS profiles within 300 km radius (Fig. 6, right, bottom) now shows a good agreement up to 55 km, but an overestimation of the SCIAMACHY profiles above that height. In contrast to the MIPAS comparisons, large differences are found above approximately 56 km, whereas such large differences are found already at 50 km in the case of MIPAS. Like in the comparisons with MIPAS profiles, increasing differences with altitude can be observed.

## 5 Conclusions

SCIAMACHY/ENVISAT limb scatter measurements in the HARTLEY bands have been used to retrieve ozone profiles in the upper stratosphere and lower mesosphere (retrieval version 1.26) which show sensitivity between 35 and 65 km altitude, but in general, the retrieval provides reliable ozone profiles within about 15% accuracy between 40 and 55 km.

Above this altitude, retrieved profiles overestimate the true ozone content more and more with altitude.

Comparisons with ozone profiles from MIPAS on ENVISAT, HALOE on UARS, a ground based radiometer in Norway, and MLS on board AURA confirm this, differences are of about 15% up to 55 km and an increasing overestimation above 55 km can be seen. In the future, the findings from this profile retrieval together with those from the ozone retrievals in the HUGGINS and CHAPPUIS bands will help to provide ozone profiles between 10 and 55 km from SCIAMACHY limb measurements.

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