

RETRIEVAL OF STRATOSPHERIC TRACE GASES FROM SCIAMACHY LIMB MEASUREMENTS

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ABSTRACT

Stratospheric profiles of various trace gases can be retrieved from limb measurements performed by SCanning Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) instrument on the ENVISAT satellite. A two-step method is used to retrieve stratospheric distribution of NO₂, BrO and OCIO. In the first step, slant column densities (SCDs) of the trace gases are derived from the SCIAMACHY limb spectra by Differential Optical Absorption Spectroscopy (DOAS). Second, the measurement geometry is simulated by applying the full spherical radiative transfer model "Tracy-II". The inversion is performed by an optimal estimation method. The Monte Carlo RTM method implements completely statistical approach of light paths and light scattering and absorption in the atmosphere, however it is time consuming and therefore cannot be repeated for every iteration that requires reasonable linearization to an a-priori atmosphere. We demonstrate the current status of the retrieval and the results. They agree well with climatological expectations and results acquired from other instruments.

1. INTRODUCTION

Nitrogen and halogen compounds are playing a major role in chemical processes of the atmosphere being involved in processes responsible for ozone loss e.g. [1].

Long term and global observations of atmospheric trace gases like O₃, NO₂, BrO and OCIO have been realized with GOME, the first satellite instrument using DOAS technique (e.g. [2], [3], [4], [5], [6], [7]). It operates in nadir observation mode that provides total column data with global coverage achieved in 3 days at equatorial region [8] at high latitudes even in one day.

In recent years besides nadir observational geometry, measurements in limb geometry are providing further opportunities, namely, to extract moderate resolution profile information by measuring backscattered light viewing air masses at different elevation angles. Satellite instruments such as the Optical Spectrograph and Infrared Imager System (OSIRIS) on Odin satellite [9], SCIAMACHY on ENVISAT [10], also the Stratospheric Aerosol and Gas Experiment (SAGE III) on the Meteor 3 have limb observation possibility [11].

The SCIAMACHY instrument on the ENVISAT satellite is flying in a near polar Sun synchronous orbit having inclination from equatorial plane approximately 98.5° and is performing one orbit in 100 minutes with equator crossing time of 10:00 in descending mode. The satellite probes atmosphere in the day side of Earth in alternating sequences of nadir and limb measurements. Limb scans are performed with approximately 3.3 km elevation with swap across flying direction of 960 km at tangent point (TP) consisting of 4 pixels. (In this study DOAS retrieval is performed for each of four pixels but measured SCDs are co-added in order to increase error statistics.) The field of view (FOV) is 0.045° in elevation and 1.8° in azimuth that corresponds approximately 2.5 km and 110 km at TP respectively. The SCIAMACHY is measuring in the UV-vis-NIR spectral range 240 – 2380 nm having spectral resolution of approximately 0.25 – 0.55 nm in UV-vis range. For more instrumental details please refer to [10].

In addition to existing limb retrievals of O₃, NO₂ and OCIO from OSIRIS [12], [13], [14] and retrieval possibility of O₃ and NO₂ from SAGE III [11], SCIAMACHY for the first time allows retrieval also of BrO stratospheric profiles.

The aims of this study is to describe in detail a new and relatively simple retrieval of the NO₂, BrO and OCIO vertical profiles from the SCIAMACHY limb measurements developed at the Institute of Environmental Physics at the University of Heidelberg (IUP Heidelberg). It allows efficient acquisition of the trace gas profiles, and shows good agreement of the retrieved BrO and NO₂ profiles with balloon measurements [15] and [16].

2. RETRIEVAL ALGORITHM

In this work NO₂ and BrO retrieval from SCIAMACHY measured limb spectra is done in two blocks as illustrated in fig. 1. In the first part, slant column densities (SCDs) for the trace gases are derived from the SCIAMACHY limb spectra by Differential Optical Absorption Spectroscopy (DOAS). Second, the trace gas SCDs are converted into vertical concentration profiles applying radiative transfer modeling and the optimal estimation method.

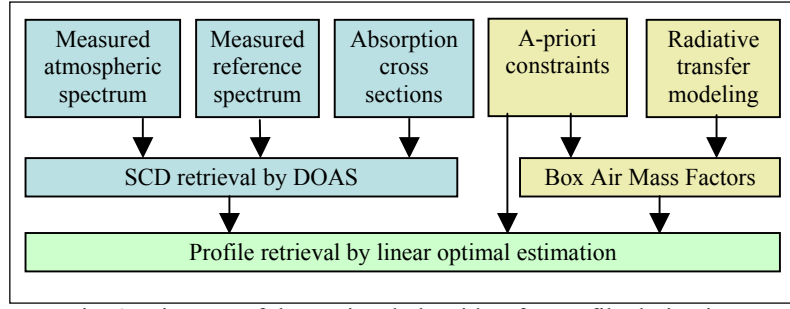


Fig. 1. Diagram of the retrieval algorithm for profile derivation.

2.1. Slant column density retrieval

The SCD of the considered absorber is deduced by Differential Optical Absorption Spectroscopy [17]. The DOAS retrieval for NO₂ is performed in the 420–450nm, for BrO in 337 – 357 nm and for OCIO in 363 – 391 nm spectral range. The measured and calibrated spectral information from SCIAMACHY is analyzed with respect to a pseudo top-of-the-atmosphere (TOA) reference spectrum taken as average of SCIAMACHY measurements at tangent heights between 40 to 46 km for NO₂, 36 km for BrO and 33 km for OCIO to infer the SCDs. Considered trace gas absorption cross sections are NO₂ at 223K from [18], O₃ at 241 K [18], BrO at 228 K [19], H₂O at 273 K from [20], O₄ at 298 K from [21] and 213 K from [22] for OCIO. In addition we account for the Ring effect [23], instrumental straylight and broadband spectral features by considering a calculated Ring spectrum and the inverse of the TOA reference at 36 km for BrO and 43 km for NO₂ as fitting parameters.

Fig. 2 demonstrates examples of the DOAS fit for BrO and OCIO.

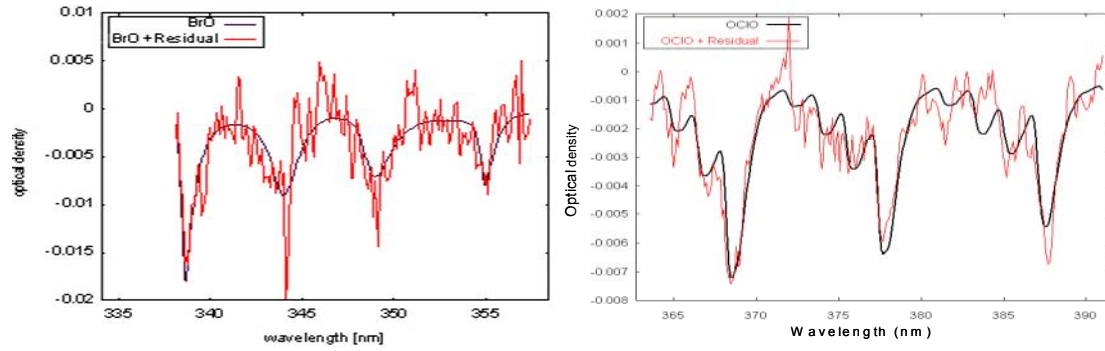


Fig.2. Examples of BrO (left) and OCIO(right) DOAS fits.

2.2. Profile retrieval

To retrieve the profile of a trace gas the functional connection between SCDs and profile should be determined:

$$y = F(x, b) + \varepsilon \quad (1)$$

where y – measurement vector (a SCDs set retrieved for different tangent heights of one SCIAMACHY limb scan state),

x – state vector (trace gas profile)

ε - measurement error

F – forward model that according our knowledge i.e. radiative transfer model, transforms state vector into the measurement vector

b – auxiliary parameters not intended to be retrieved.

Assuming a linearized problem with respect to the a-priori atmosphere equation (1) can be written:

$$y = Kx + \varepsilon \quad (2) \quad K_{ij} = A_{ij} \cdot \Delta h_j \quad (3)$$

Here each K_{ij} element of the weighting function matrix \mathbf{K} expresses how large the impact of a change in concentration at altitude level j on measured SCDs at tangent height i will be. In this study in the place of weighting

function matrix consisting of elements K_{ij} the box air mass factor matrix \mathbf{A} is retrieved by a radiative transfer modeling. A discussion about radiative transfer modeling please find in chapter 3. h is the thickness of layer j . The trace gas profile vector \mathbf{x} is retrieved using Bayesian approach described by [24]. A-priori assumes concentration variation in 100% range.

Since SCIAMACHY has tangent height miss-pointing from values given in the dataset [25], a tangent height correction was performed according the month average pointing errors given in [26].

After correction for the miss-pointing the retrieved SCDs are interpolated to a tangent height grid of 3 km and also retrieval is constrained into a similar grid having 3 km altitude steps.

3. RADIATIVE TRANSFER MODELLING

We use 3D full spherical Monte Carlo radiative transfer model “Tracy-II” to calculate box AMFs. The model “Tracy-II” is an up-to-date version of the RTM “Tracy” [27]. The largest advantage of Monte Carlo models in the limb geometry is that they assume not only 3D shaping of Earth but also provide the possibility to simulate to a high degree an inhomogeneous atmosphere. They deal with microscopic processes in the atmosphere in a probabilistic way.

The detector sees either photons directly scattered into the line of sight (LOS) of the instrument or photons already scattered by the atmosphere or the ground below. The limb geometry is characterized by relatively long paths of photons along the LOS after the last scattering event in comparison to the paths before the last scattering.

A limiting factor for the retrieval at low altitudes is high Rayleigh scattering probability i.e. the atmosphere is optically thick. In the case when the atmosphere is optically transparent nearly symmetrical distribution of scattering of photons across tangent point (TP) scattering into the LOS can be observed. But at low elevations having optically dense atmosphere most of the photons visible by the instrument are contributing to the LOS at sites far from the TP, that means that the measured spectra practically do not contain information about the lower part of atmosphere.

Fig. 3 shows an example of box AMFs calculated for limb geometry. The low sensitivity for altitudes below 12-15 km can be realized. In fact, as will be seen later this fact allows to retrieve trace gases starting from above this altitude. In return for high elevations for optically thin atmosphere the box AMFs are tending to the geometrical estimation of box AMFs:

$$AMF_{geom} \approx \frac{l_{LOS}}{h} + \frac{b}{\cos(SZA)} \quad (4)$$

where l_{LOS} refers to the part of LOS through an atmospheric layer of interest, h – vertical thickness of that layer, b is some constant mostly depending on the difference between the altitude of elevation and the altitude of box of interest. SZA means solar zenith angle. The maximum retrieval altitude therefore obviously is not determined by the sensitivity regarding AMFs but low intensities that can be measured at high elevations (fig. 4) giving lower signal to noise ratio.

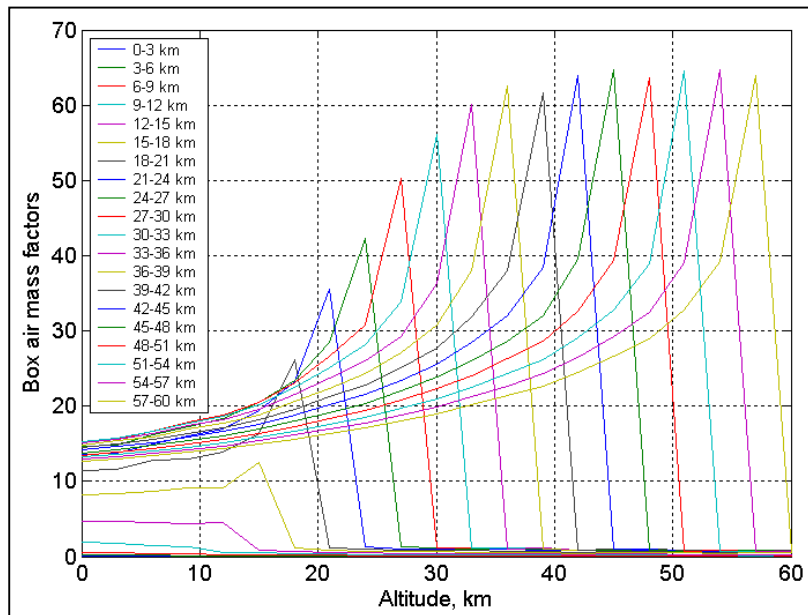


Fig. 3. Box AMFs plotted for 3 km thick boxes as function of the tangent height. The peak value usually is located at the tangent height equal to altitude of the “box” simulated.

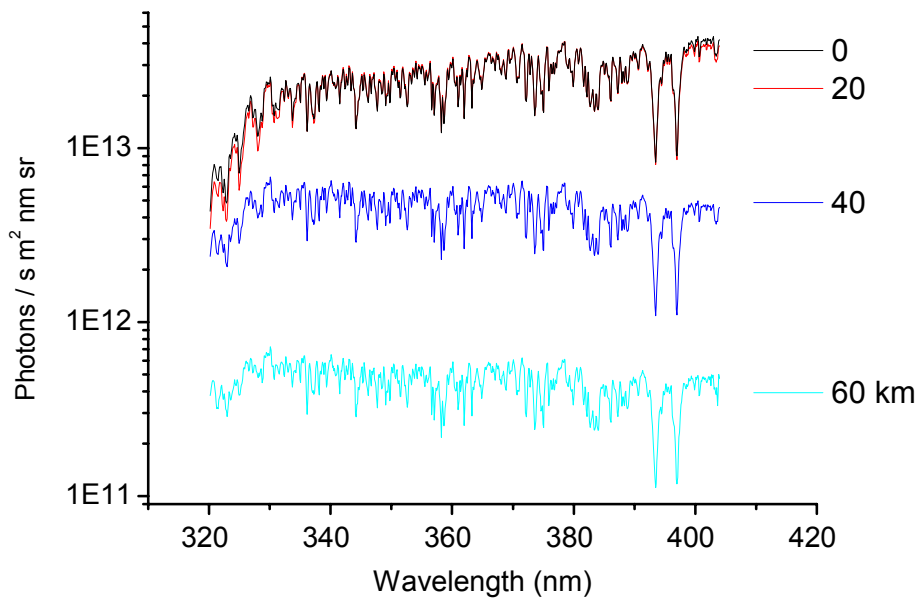


Fig. 4. Example of measured intensities by SCIAMACHY instrument at different tangent heights in the wavelength range 320 to 410 nm.

4. RESULTS

Fig. 5 shows latitudinal cross sections i.e. concentration as function of latitude and altitude for sample orbits at austral winter – spring for BrO, NO₂ and OClO and corresponding averaging kernels. From the corresponding averaging kernels (right panel) one can realize that for altitudes from 10 to 15 km the retrieved profiles consist mainly of the a-priori information. For altitudes above it seems largely independent from a-priori, see example for OClO retrieval in fig. 6. Fig. 5 illustrates nicely the variety of issues on stratospheric chemistry that can be investigated with the SCIAMACHY limb observations: Denoxification and denitrification in polar winter, bromine monoxide showing only little dependence on season (compared to OClO), chlorine activation in winter and deactivation in spring.

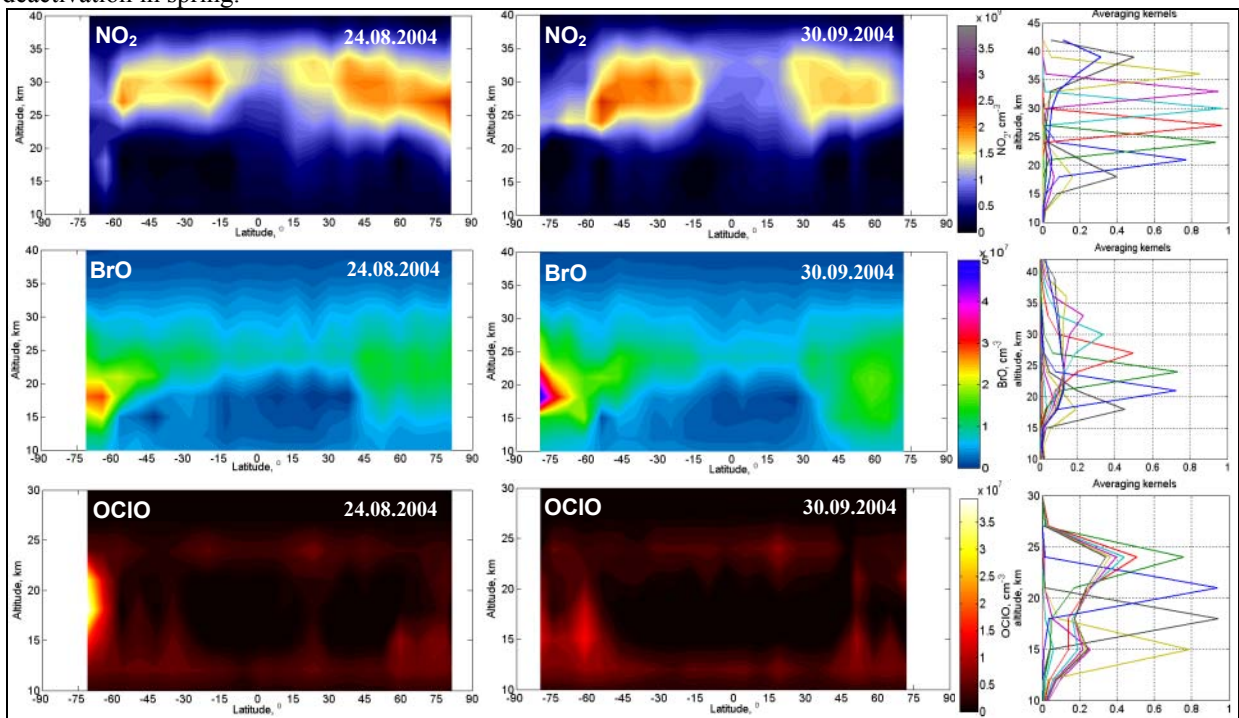


Fig. 5. Left side: Latitudinal cross sections of retrieved trace gases – NO₂, BrO and OClO for selected orbits from 24.08.2004 and 30.09.2004. Right side: Characteristic averaging kernels. The retrieval is more sensitive to the real state of the atmosphere at altitudes where the averaging kernels are more close to unity.

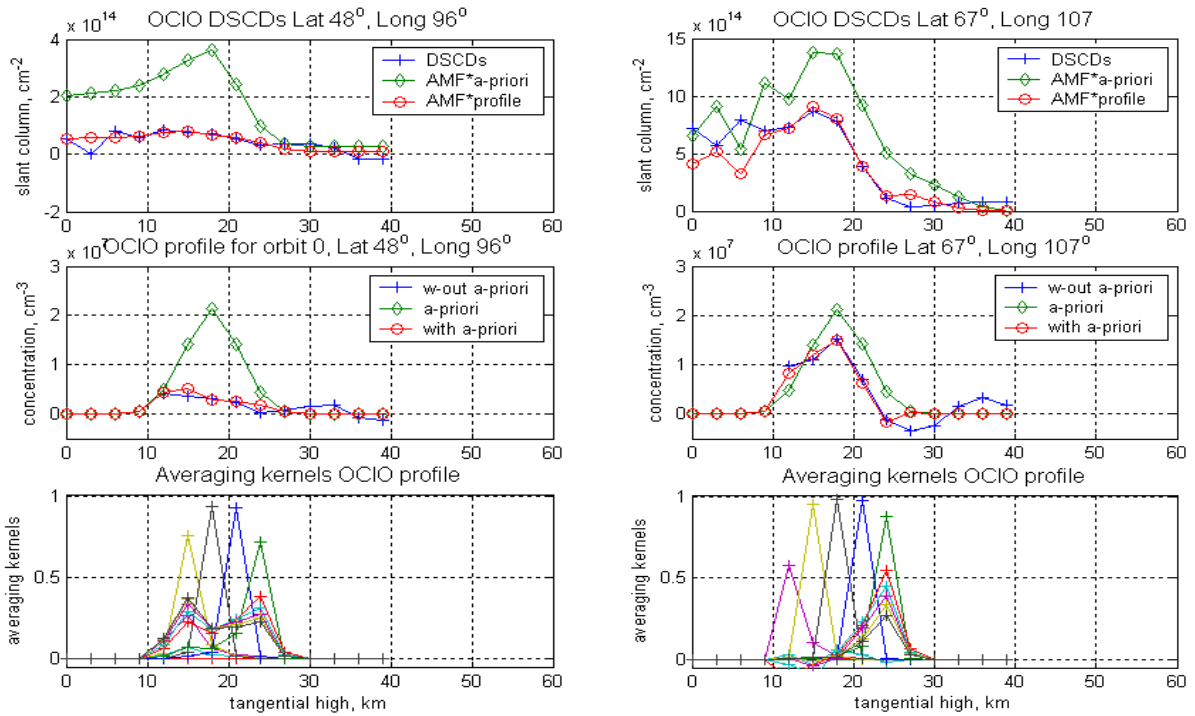


Fig. 6 Two retrievals of OCIO for different latitudes (left) with negligible chlorine activation and (right) with large chlorine activation. The same a-priori profile was used in both cases. In addition to the results using an inversion with a-priori profile, also results from a direct inversion (without a-priori) are plotted. They are only retrieved for the altitude range 12-39 km. For this range they agree well with the results using the a-priori profile. The independency of retrieval from a-priori can be seen from 15 to 25 km altitude range.

4. VALIDATION

Comparison of BrO profiles with photochemically corrected balloon measurements probing the same air masses as SCIAMACHY given in [15] was performed and shows good agreement (Fig. 7). Also the NO₂ profiles have been validated and show good agreement, see [16] for details.

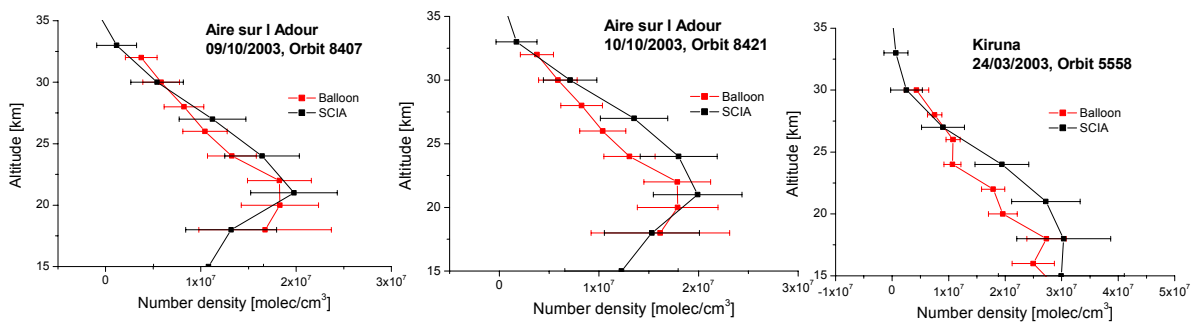


Fig. 7. Comparison of SCIAMACHY measured BrO profiles with that from balloon measurements. Balloon profiles are taken from [15].

5. CONCLUSIONS

SCIAMACHY provides scattered light measurements in limb geometry from which atmospheric trace gas profiles can be successfully retrieved. A two step retrieval for NO₂, BrO and OCIO is performed for selected orbits of SCIAMACHY limb measurements enabling to study separately the impact of spectroscopy and radiative transfer. It is possible to retrieve trace gas concentrations for altitudes 15 - 36 km (BrO), 15 - 39 km (NO₂), 15 - 24 km (OCIO) with a vertical resolution of ~3 km. The obtained global dataset is forming an unique time series of measurements for investigating atmospheric chemistry. The retrieved profiles are in accord with expectations from stratospheric chemistry. The acquired vertical distribution of NO₂ and BrO from SCIAMACHY agrees well with data from

balloon instruments. Further validation especially of OCIO and sensitivity studies are needed, as well as the investigation of effect of horizontal gradients effect on the retrieval.

6. REFERENCES

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