RETRIEVAL OF CO, H_2O, CH_4, CO_2, AND N_2O COLUMNS FROM SCIAMACHY/ENVISAT BY WFM-DOAS: CURRENT STATUS

M. Buchwitz, R. de Beek, S. Noël, H. Bovensmann, and J. P. Burrows

*Institute of Environmental Physics (iup) / Institute of Remote Sensing (ife), University of Bremen FB1,
Otto Hahn Allee 1, 28334 Bremen, Germany, E-mail: Michael.Buchwitz@iup.physik.uni-bremen.de*

ABSTRACT

The spectra measured by SCIAMACHY contain information on a number of important atmospheric trace gases. The near-infrared nadir spectra enable the retrieval of total column amounts of CO, H_2O, CH_4, CO_2, and N_2O. We are developing a modified DOAS algorithm (WFM-DOAS) to accomplish this task. Here we give an overview about the current status of this activity (using WFM-DOAS Version 0.4). We show that encouraging higher level data products can be obtained despite the preliminary calibration of the operational spectra. For example, accurate water vapour columns can been retrieved over ocean and land, plumes of CO can be detected, the relatively small (few percent) inter-hemispheric difference of the methane mixing ratio is visible in single day data, and there are strong indications that for the first time a CO_2 source/sink region has been identified from space. We highlight a number of issues that need further study and, if solved, are expected to result in improved data products.

1. INTRODUCTION

SCIAMACHY [1] is one of the three atmospheric chemistry instruments onboard ENVISAT. As the other two instruments, i.e. MIPAS and GOMOS, SCIAMACHY observes the Earth limb. Additionally, however, nadir observations are being performed which are sensitive also to the lower troposphere. Satellite remote sensing of the chemical composition of the troposphere is a new area - in particular measurements of Greenhouse gases (e.g., CH_4, CO_2, and N_2O). In the following we give an overview of what we have achieved within the first two years of the ENVISAT mission concerning the retrieval of total column amounts of H_2O, CO, CH_4, CO_2, and N_2O from the SCIAMACHY nadir spectra using the WFM-DOAS retrieval algorithm [3]. All results have been obtained with WFM-DOAS Version 0.4.

2. SCIAMACHY INSTRUMENT

SCIAMACHY [1] is a spectrometer that measures reflected, scattered and transmitted solar radiation in the spectral region 214-2380 nm at moderate spectral resolution (0.2-1.6 nm). SCIAMACHY typically performs a sequence of alternating nadir and limb measurements. On the dayside about 20 nadir "states" are executed during each orbit. Each state lasts about one minute and covers a region of about 960 km across track

times 450 km along track (in between two nadir states is a gap of similar size due to the limb measurements). A state is a sequence of individual measurements performed with identical instrument parameters (such as integration time and, therefore, horizontal resolution). The horizontal resolution of the nadir measurements depends on orbital position and spectral interval but is typically 60 or 120 km across track times 30 km along track.

The in-flight optical performance of SCIAMACHY is overall as expected from the on-ground calibration and characterization activities [2]. One exception is the time dependent optical throughput variation in the SCIAMACHY near-infrared (NIR) channels 7 and 8 due to ice build-up on the detectors. This effect is minimised by regular heating of the instrument [2]. This „ice issue" is relevant for most of the data products discussed in this study as channels 7 and 8 cover absorption bands of CO_2, CH_4, N_2O, H_2O, and CO. For this and a number of other reasons the SCIAMACHY spectra are currently limited in quality. In order to improve the quality of our data products we pre-process the operational Level 1 data products mainly with respect to a better dark signal calibration [7,8]. As the calibration of the SCIAMACHY solar spectra is also preliminary we use a solar spectrum with an improved calibration (provided by J. Frerick, ESA/ESTEC).

3. RETRIEVAL ALGORITHM

The Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval algorithm [3] is an unconstrained linear-least squares method based on scaling (or shifting) pre-selected vertical profiles. The fit parameters for the trace gases are directly the desired vertical columns. The logarithm of a linearized radiative transfer model plus a low-order polynomial is fitted to the logarithm of the ratio of a measured nadir radiance and solar irradiance spectrum, i.e., observed sun-normalized radiance.

The WFM-DOAS reference spectra are the logarithm of the sun-normalized radiance and its derivatives. They are computed with a radiative transfer model [4]. Multiple scattering is fully taken into account. In order to avoid time-consuming on-line radiative transfer simulations, a fast look-up table scheme has been implemented [6]. *A priori* information about the atmosphere is only used to get a reasonable linearisation point for the radiative transfer simulations but not to constrain the retrieval. In order to make sure that all observed variability is entirely

due to the SCIAMACHY spectral measurements, a single model atmosphere is used (exception: surface topography changes are considered using a coarse grid (0, 1, 2, 3 km; next neighbour approach, i.e., no interpolation)). Clouds are an issue for nadir measurements of tropospheric gases. In order to identify cloud-contaminated pixels we currently use a simple threshold algorithm based on sub-pixel information as provided by the SCIAMACHY Polarization Measurement Devices (PMDs) [7,8]. There are indications that the in-orbit slit function of SCIAMACHY is different from the one measured on-ground (H. Schrijver, SRON, personal communication). We use a slit function that has been determined by applying WFM-DOAS to the in-orbit nadir measurements. We selected the one that resulted in best fits, i.e., smallest fit residuum [7,8].

4.   TRACE GAS RESULTS

4.1   Water vapour (H$_2$O)

Water vapour columns have been retrieved from the spectral region 685-710 nm covering H$_2$O and O$_2$ absorption lines [15]. A visible channel has been used for water retrieval, rather than a near-infrared channel, because of higher signal-to-noise ratios, especially over the ocean. O$_2$ has been used for air mass factor correction of the initially retrieved H$_2$O columns to compensate for errors introduced by, e.g., clouds, aerosols, and surface reflectivity variations. Figure 1 shows the retrieved water columns for January 27, 2003. Clearly visible are well known features of atmospheric water vapour, e.g., a band of high concentrations in the tropics and lower concentrations at higher latitudes. The WFM-DOAS water vapour retrievals have been compared with independent SSM/I microwave satellite measurements (which are only available over ocean) and ECMWF analysed meteorological fields (over ocean and land) and very good agreement has been found [15]. For example, the linear correlation coefficients r is larger than 0.9, the standard deviation of the difference is about 0.5 g/cm^2 (this includes contributions from the temporal and spatial variability of water vapour), and the mean difference is ~0.2 g/cm^2.

The WFM-DOAS results discussed in [15] were scaled with a constant factor of 1.1 to compensate for a low bias relative to the SSM/I and ECMWF reference data. In [15] not only WFM-DOAS retrievals are discussed but also results from a different algorithm called AMC-DOAS. For AMC-DOAS the same scaling factor has been used. In [15] it is shown that both algorithms are essentially equivalent as they give nearly identical results. AMC-DOAS has recently been optimised, e.g., by using a SCIAMACHY slit function different from the one used in [15]. The new results are presented in [16,17] and are similar as the ones shown in [15] but have been obtained without application of a scaling factor.
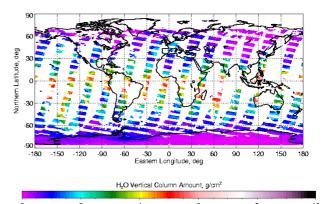

Figure 1: *Water vapour total columns as retrieved by WFM-DOAS using the SCIAMACHY nadir measurements from January 27, 2003. The regular gaps along the orbits are due to the limb measurements. The irregular gaps (mostly in the tropics) are due to clouds.*

4.2   Carbon monoxide (CO)

Carbon monoxide has been retrieved from a small spectral fitting window around 2365 nm covering four CO absorption lines [6,7]. The CO lines in this spectral region are weak compared to the absorption features of methane and water vapour present in this region. The depth of a CO absorption line for relatively low CO concentrations is typically on the order of the instrument noise level. Therefore, and because of the preliminary calibration and possibly other reasons, it is difficult to accurately fit the CO lines. The WFM-DOAS fit residuals are not yet noise limited but dominated by rather stable spectral features on the order of the weak CO lines. In order to compensate for a still to be investigated apparent overestimation, the WFM-DOAS Version 0.4 CO columns have been scaled with a constant factor of 0.5. Our future work will focus on (i) using a larger fitting window covering more CO lines to improve the precision, (ii) achieving better fits, and (iii) removing the need for scaling the columns. Despite these difficulties it has been shown that SCIAMACHY can detect atmospheric CO variability [7]. Figure 2 shows enhanced CO concentrations as measured by SCIAMACHY on October 26, 2003, around San Diego. They result from heavy fires in southern California in October/November 2003. A quantitative comparison with the CO measurements of MOPITT aboard EOS-Terra is not possible for this day due to lack of MOPITT data. In [7] plumes of CO due to biomass burning in Africa are shown which are in good agreement with MOPITT. We quantitatively compared the SCIAMACHY CO columns for three days within the time period January-October 2003 with the Version 3 CO column data product of MOPITT [11,13]. The days have been selected because of (i) good overlap with the MOPITT daytime measurements and (ii) availability of (nearly) all 14 daily orbits of SCIAMACHY. For the measurements over land the linear correlation coefficient is ~0.6. The standard deviation of the difference is in the range 0.4-0.6x10^{18} molecules/cm^2 (~20%) and the mean

differences for the daily data are between +0.1% and +10.3% (positive bias). For 10 degree latitude averages agreement is typically better than 30% with a positive bias of the SCIAMACHY data relative to MOPITT over large parts of the southern hemisphere.
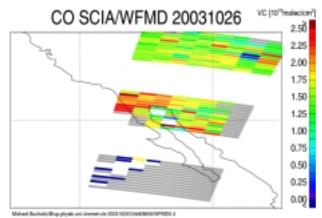

Figure 2: *Vertical columns of carbon monoxide as retrieved by WFM-DOAS using SCIAMACHY nadir measurements over southern California around San Diego on October 26, 2003, where large areas were burning. Cloud contaminated pixels are shown in grey.*

Several days of SCIAMACHY CO data have been compared with ground based Fourier Transform Spectrometer (FTS) measurements at various stations. Agreement is typically within 30% [12,18,19] but more studies are needed (more data need to be compared; many stations are located on top of mountains (only observing overhead columns) or near the coast (with many pixels over water where the measurements are less reliable)). Figures 3-6 show bi-monthly averages of the cloud free SCIAMACHY data over land for the time period March-October 2003. The interpretation of these figures is somewhat difficult due to persistent cloud cover over many regions, especially in the tropics. Clearly visible are enhanced concentrations of CO especially over large parts of the northern hemisphere in May-August (e.g., eastern USA and Canada, Europe, Russia) relative to March/April and September/October. This is in reasonable qualitative agreement with MOPITT (a detailed comparison has not yet been made). Enhanced concentrations are visible over Portugal in July/August 2003 where large areas were burning. The high columns measured over South America and Africa in September/October 2003 agree reasonably well with the MOPITT measurements (the plumes as measured by SCIAMACHY appear to be extended somewhat further to the North). There are, however, also significant differences relative to MOPITT, for example, over South America in May/June where SCIAMACHY sees an extended region with high concentrations not seen by MOPITT. May/June is not the main biomass burning season for this region but the ATSR Fire Atlas shows a significant number of fires in the region where the high columns are detected by SCIAMACHY. More investigations are needed to clarify the differences relative to MOPITT.
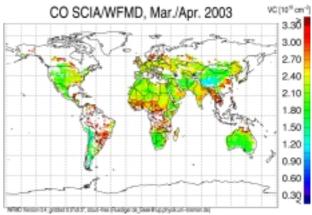

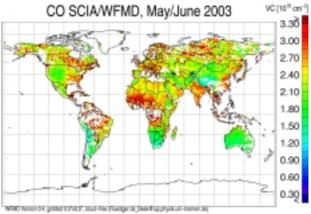**Figure 3:** CO columns for March/April 2003.
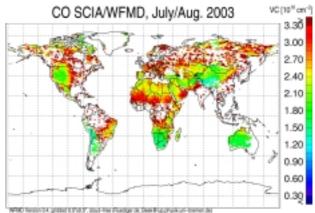

Figure 4: CO columns for May/June 2003.
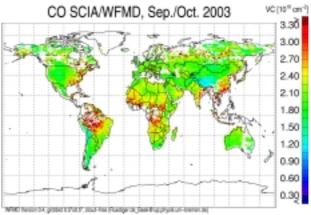

Figure 5: CO columns for July/August 2003.

Figure 6: CO columns for September/October 2003.

4.3 Methane (CH$_4$)

The retrieval of the well-mixed gases CH$_4$, CO$_2$, and N$_2$O is extremely challenging as basically only small variations on top of a large background are of relevance in order to obtain information on the (surface) sources and sinks of these important Greenhouse gases (this is the objective of the EC 5th FP research project EVERGREEN, see http://www.knmi.nl/evergreen).

Methane columns are retrieved from a small spectral fitting window (2265-2280 nm) located in SCIAMACHY channel 8 [6,8]. Several days of SCIAMACHY spectra from the time period January to October 2003 have been processed by WFM-DOAS and compared with global models of chemistry and transport [8]. Comparisons have been made for the absolute methane columns (in molecules/cm^2) but also for the dry air column averaged methane mixing ratios XCH$_4$ (in ppbv). XCH$_4$ is the retrieved methane column divided by the air column. The measured air column is the retrieved oxygen (O$_2$) column divided by the well known dry air mixing ratio of O$_2$ (which is 0.2095). The O$_2$ columns have been retrieved from the O$_2$-A-band spectral region also observed by SCIAMACHY. Within EVERGREEN it is planned to use the XCH$_4$ measurements rather than the absolute columns for inverse modelling of methane sources and sinks because the division by the air column eliminates disturbing methane column variations due to surface topography and/or pressure changes. The WFM-DOAS Version 0.4 O$_2$ columns have been scaled by 0.85. The methane columns are not scaled.

Figure 7 shows a comparison of XCH$_4$ as retrieved from SCIAMACHY and corresponding results from the global model TM5 of the Joint Research Centre (JRC) in Ispra, Italy (data provided by P. Bergamaschi, see [8] and references given therein). The hemispheric mean values of the mixing ratios and their difference, i.e., the inter hemispheric difference (IHD) which is only a few percent, agree very well. As can be seen, the scatter of the measurements is significantly larger (~100 ppbv or 6%) than the variability of the model values (~10 ppbv).

In [8] it is shown that for the four analysed days the measured IHD is in the range 30-110 ppbv in reasonable agreement with the model data (TM5 model of JRC/Ispra and TM3 model of KNMI (data provided by J. F. Meirink)), which are in the range 48-71 ppbv. For the individual measurements, the standard deviation of the difference with respect to the model data is in the range 100-200 ppbv (5-10%) for XCH$_4$. The mean difference between the measured and modelled columns is less than 2% for two of the days but 10% and 15% for the two other days. This indicates that there is a time dependent bias, which is most probably related to calibration problems, e.g., the ice issue mentioned in Section 2 (for the days investigated the bias increases with transmission loss). More studies are needed to confirm this also for other days. We also plan to retrieve methane from SCIAMACHY channel 6, which is not affected by the ice issue.

Several days of SCIAMACHY methane data have been compared with ground based FTS measurements at various stations. Agreement is typically within 5-15% [12,19].
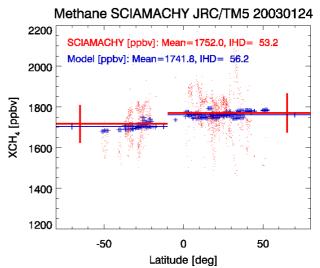

Figure 7: *Comparison of the dry air column averaged CH$_4$ mixing ratio (XCH$_4$) as retrieved by WFM-DOAS from the SCIAMACHY measurements over land on January 24, 2003 (red dots) with corresponding results of the global TM5 model of JRC/Ispra (blue crosses). The red horizontal lines show the hemispheric averages of the SCIAMACHY measurements. The blue horizontal lines show the corresponding averages for the TM5 model. The vertical lines indicate the standard deviation of the individual data points. The mean value of the SCIAMACHY measurements is 1752 ppbv (TM5: 1742 ppbv). The inter hemispheric difference (IHD) as measured by SCIAMACHY is 53 ppbv (TM5: 56 ppbv).*

4.4 Carbon dioxide (CO$_2$)

Absorption bands of CO$_2$ are covered by the SCIAMACHY channels 7 (~2 μm) and 6 (~1.6 μm). Because channel 7 is affected by the ice issue (see Section 2) we currently retrieve CO$_2$ from channel 6 (fitting window: 1558-1594 nm). The WFM-DOAS Version 0.4 CO$_2$ data products are the absolute CO$_2$ columns (in molecules/cm^2) and the dry air column averaged mixing ratios XCO$_2$ (in ppmb; defined analog to XCH$_4$, see Section 4.3). The absolute columns are scaled by a constant factor of 1.27.

Figure 8 shows SCIAMACHY XCO$_2$ measurements over Africa end of January to beginning of February 2003. The most prominent feature is the extended region of low CO$_2$ mixing ratios around Zambia/Congo (shown in blue). We compared our measurements with results from the global model TM3 of the Max Planck Institute (MPI) in Jena, Germany (data provided by S. Körner, see [8] and references given therein). The model also shows an extended region of low CO$_2$ mixing ratios over Zambia/Congo (not shown here). The depth of the "CO$_2$ hole" in the model is however less deep (~5 ppmv) than observed by SCIAMACHY (~20 ppmv). For the

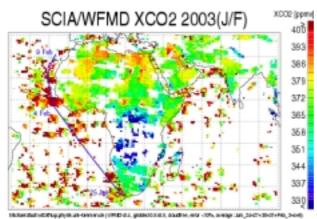SCIAMACHY measurements a significant uncertainty is introduced due to large cloud cover in this region.

Figure 8: *Dry air column averaged CO_2 mixing ratios (XCO2) as retrieved by WFM-DOAS from the SCIAMACHY measurements end of January to beginning of February 2003. Gaps are mainly due to clouds. The blue line along the west coast of Africa shows the track of the German research vessel Polarstern with an FTS on board for the validation of the SCIAMACHY measurements [19]. In [19] it is shown that the FTS measurements agree with the nearby SCIAMACHY measurements over land within about 5%.*

For January 24, 2003, the SCIAMACHY measurements over land have been quantitatively compared with the results of the MPI Jena TM3 model [8]. For the absolute CO_2 columns the mean difference is 0.16×10^{21} molecules/cm^2 (2.3%) and the standard deviation of the difference is 0.50×10^{21} molecules/cm^2 (7.7%). For XCO2 the standard deviation of the difference is 14.5 ppmv (3.9%).

4.5 Nitrous oxide (N_2O)

Currently, N_2O is only retrieved as a by-product of methane retrieval (see Section 4.2). The N_2O lines detected by SCIAMACHY are weak. They are significantly weaker than the absorption structures of methane. The difficulties in accurately fitting the N_2O lines are similar as for CO (see Section 4.2). The N_2O columns have been scaled with a constant factor of 0.66. So far only a very limited number of comparisons with independent measurements have been performed. In [12,19] it is shown that the WFM-DOAS Version 0.4 N_2O columns agree with ground based FTS measurements within typically 30% (positive and negative biases have been observed).

5. CONCLUSIONS AND OUTLOOK

We have shown that in spite of a number of still to be investigated issues related to, e.g., the preliminary calibration of the SCIAMACHY spectra in the near-infrared, encouraging higher-level data products can be obtained. For example, good water vapour columns have been retrieved over ocean and over land which agree well with SSM/I satellite microwave measurements over

ocean and ECMWF analysed meteorological fields over ocean and land. Plumes of CO due to, e.g., biomass burning in Africa, are detectable with SCIAMACHY and show good agreement with MOPITT. The retrieval of the well-mixed gases CH_4, CO_2, and N_2O is also possible but extremely challenging. This is because only small variations on top of a large background are of relevance in order to obtain information on the (surface) sources and sinks of these important Greenhouse gases. This is one of the major applications for these measurements.

In order to estimate the accuracy and precision of the SCIAMACHY/WFM-DOAS data products they have been compared with independent measurements and global models. So far only a limited number of data have been analysed. Table 1 summarizes our current precision and accuracy estimates. The precision has been estimated from signal-to-noise considerations, retrieval algorithm sensitivities, and from the standard deviation of the difference of the individual columns and the reference data (measurements and models). The accuracy has been estimated from mean differences with respect to the reference data (e.g., by considering all measurements for a given day close to a given ground station).

Gas	Horiz. resol. [km x km]	Estimated precision (scatter) [%]	Estimated accuracy (bias) [%]	Ref.
	Preliminary estimates of precision and accuracy of SCIAMACHY / WFM-DOAS Version 0.4 vertical column data products			
H_2O	30 x 60	**< 0.5 g/cm^2** *(includes large natural variability)*	**~0.2 g/cm^2** *(mostly negative bias)*	[15]
CO	30 x 120	**10-20**	**10-30** *(mostly positive bias)*	[3,6,7, 12,18,19]
CH_4	30 x 120	**1-6**	**2-15** *(time dependent bias due to ice issue; mostly negative)*	[3,6,8,12]
CO_2	30 x 60	**1-4**	**2-5** *(mostly negative bias)*	[3,6,8]
N_2O	30 x 120	**10-20**	**+/- 30**	[3,12]

Table 1: *Preliminary estimates of precision and accuracy of the SCIAMACHY/WFM-DOAS Version 0.4 total column data products based on a limited amount of measurements (several days of the year 2003) compared with reference data (ground based, satellite, models). The estimates refer to measurements over land (for water vapour: ocean and land). These estimates have been determined from the typical scatter (for the precision) and the typical mean difference (for the accuracy) of the measurements relative to the reference data for given time/space intervals (e.g., ground station overpass). Therefore, the biases are not the overall (globally/full-year) bias of the measurements.*

Our goal is to achieve nearly bias free retrievals with a single measurement precision close to the theoretical limit as determined by instrument noise (i.e., ~1% for the strong absorbers CH_4 and CO_2 and ~10% for the weak absorbers CO and N_2O [3]). Table 1 indicates that this goal has not yet been achieved. A number of issues have been identified that need further study and, if solved, are expected to result in improved data products. First of all more data need to be processed and analysed to improve the statistics, to investigate seasonal effects, and to better discriminate between random errors and biases and to determine their time dependence (if any). Several calibration issues need to be better addressed (e.g., ice issue, solar spectrum, slit function). For still to be determined reasons the fit residuals are not yet signal-to-noise limited but dominated by rather stable spectral features. This needs to be improved. The columns of most gases are scaled (with a constant factor) which is unsatisfactory. Also the cloud detection algorithm needs optimisation. At present it cannot clearly discriminate between clouds and surfaces covered by snow or ice. The WFM-DOAS look-up table scheme also can be improved to reduce errors related to, e.g., the variability of pressure profiles and surface reflectivity. Last but not least it has to be pointed out that so far only narrow spectral fitting windows have been analysed which cover only a small subset of the absorption lines detected by SCIAMACHY. We expect a significant improvement from using a larger fitting window for CO and from retrieving CH_4 from channel 6 which is not affected by the ice issue.

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