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columns from  
SCIAMACHY**

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# Carbon monoxide, methane and carbon dioxide columns retrieved from SCIAMACHY by WFM-DOAS: year 2003 initial data set

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## Abstract

The near-infrared nadir spectra measured by SCIAMACHY on-board ENVISAT contain information on the vertical columns of important atmospheric trace gases such as carbon monoxide (CO), methane (CH<sub>4</sub>), and carbon dioxide (CO<sub>2</sub>). The scientific algorithm WFM-DOAS has been used to retrieve this information. For CH<sub>4</sub> and CO<sub>2</sub> also column averaged mixing ratios (XCH<sub>4</sub> and XCO<sub>2</sub>) have been determined by simultaneous measurements of the dry air mass. All available spectra of the year 2003 have been processed. We describe the algorithm versions used to generate the data (v0.4; for methane also v0.41) and show comparisons of monthly averaged data over land with global measurements (CO from MOPITT) and models (for CH<sub>4</sub> and CO<sub>2</sub>). We show that enhanced concentrations of CO resulting from biomass burning have been detected which are in reasonable agreement with MOPITT. The measured XCH<sub>4</sub> is enhanced over India, south-east Asia, and central Africa in September/October 2003 in line with model simulations where they result from surface sources of methane such as rice fields and wetlands. In qualitative agreement with model simulations the XCO<sub>2</sub> measurements over the northern hemisphere show the lowest mixing ratios around July due to uptake of CO<sub>2</sub> by the growing vegetation of the land biosphere. We also identified potential problems such as a too low inter-hemispheric gradient for CO, a time dependent bias of the methane columns on the order of a few percent, and a few percent too high CO<sub>2</sub> over parts of the Sahara.

## 1. Introduction

Knowledge about the global distribution of carbon monoxide (CO) and of the relatively well-mixed greenhouse gases methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>) is important for many reasons. CO, for example, plays a central role in tropospheric chemistry (see, e.g., [Bergamaschi et al., 2000](#), and references given therein) as CO is the leading sink of the hydroxyl radical (OH) which itself largely determines the oxidizing capacity of

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the troposphere and, therefore, its self-cleansing efficiency and the concentration of greenhouse gases such as CH<sub>4</sub>. CO also has large air quality impact as a precursor to tropospheric ozone, a secondary pollutant associated with respiratory problems and decreased crop yields. Satellite measurements of CH<sub>4</sub> and CO<sub>2</sub> in combination with inverse modeling have the potential to help better understand their surface sources and sinks than currently possible with the very accurate but rather sparse data from the network of surface stations (see Houweling et al., 1999, 2004; Rayner and O'Brian, 2001, and references given therein). A better understanding of the sources and sinks of CH<sub>4</sub> and CO<sub>2</sub> is important for example to accurately predict the future concentrations of these gases and associated climate change. Monitoring of the emissions of these gases is also required by the Kyoto protocol.

The first CO results from SCIAMACHY retrieved by WFM-DOAS have recently been presented in Buchwitz et al. (2004a), and first results on CH<sub>4</sub> and CO<sub>2</sub> have been presented in Buchwitz et al. (2004b), both papers focusing on a detailed analysis of single day data (except for CO<sub>2</sub> for which also time averaged data have been discussed). Here we present the first large data set of the above mentioned gases obtained by processing nearly a year (year 2003) of nadir radiance spectra using initial versions (v0.4 and v0.41) of the WFM-DOAS retrieval algorithm. WFM-DOAS is a scientific retrieval algorithm which is independent of the official operational algorithm of DLR/ESA.

The SCIAMACHY/WFM-DOAS data set has been compared with independent ground based Fourier Transform Spectroscopy (FTS) measurements. These comparisons, which are limited to the data close to a given ground station, are described elsewhere in this issue (e.g., Dils et al., 2005; Sussmann et al., 2005). Initial comparison for a sub-set of the data can be found in de Maziere et al. (2004); Sussmann and Buchwitz (2005); Warneke et al. (2005). Here we focus on the comparison with global reference data.

Accurate retrieval of the columns of the gases from the SCIAMACHY spectra is a challenging task for many reasons, e.g., because of calibration issues (related to the high and variable dark signals in the near-infrared), because the weak CO lines are

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difficult to detect, and because of the challenging accuracy and precision requirements for CO<sub>2</sub> and CH<sub>4</sub>. When developing the retrieval algorithm many choices have to be made (selection of spectral fitting window, inversion procedure including definition of fit parameters and use of a priori information, radiative transfer approximations, etc.) to process the data in an optimum way such that a good compromise is achieved between processing speed and accuracy of the data products. In this context it is important to point out that other groups are also working on this using quite different approaches (see [Gloudemans et al., 2004](#); [Frankenberg et al., 2005, 2004](#)).

This paper is organized as follows: In Sect. 2 the SCIAMACHY instrument is introduced followed by a description of the WFM-DOAS retrieval algorithm in Sect. 3. Section 4 gives an overview about the processed data mainly in terms of time coverage. The main sections are the three Sects. 5–7 where the results for CO, CH<sub>4</sub>, and CO<sub>2</sub> are separately presented and discussed. The conclusions are given in Sect. 8.

## 2. The SCIAMACHY instrument

The SCanning Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) instrument ([Burrows et al., 1995](#); [Bovensmann et al., 1999, 2004](#)) is part of the atmospheric chemistry payload of the European Space Agencies (ESA) environmental satellite ENVISAT, launched in March 2002. ENVISAT flies in sun-synchronous polar low Earth orbit crossing the equator at 10:00 a.m. local time. SCIAMACHY is a grating spectrometer that measures spectra of scattered, reflected, and transmitted solar radiation in the spectral region 240–2400 nm in nadir, limb, and solar and lunar occultation viewing modes.

SCIAMACHY consists of eight main spectral channels (each equipped with a linear detector array with 1024 detector pixels) and seven spectrally broad band Polarization Measurement Devices (PMDs) (details are given in [Bovensmann et al., 1999](#)). For this study observations of channel 6 (for CO<sub>2</sub>) and 8 (for CH<sub>4</sub>, CO, and N<sub>2</sub>O), and Polarization Measurement Device (PMD) number 1 (~320–380 nm) have been used. In

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5 addition, channel 4 has been used to determine the mass of dry air from oxygen (O<sub>2</sub>) column measurements using the O<sub>2</sub> A band. Channels 4, 6 and 8 measure simultaneously the spectral regions 600–800 nm, 970–1772 nm and 2360–2385 nm at spectral resolutions of 0.4, 1.4 and 0.2 nm, respectively. For SCIAMACHY the spatial resolution  
10 depends on the spectral interval and orbital position. For channel 8 data, the spatial resolution, i.e., the footprint size of a single nadir measurement, is 30×120 km<sup>2</sup> corresponding to an integration time of 0.5 s, except at high solar zenith angles (e.g., polar regions in summer hemisphere), where the pixel size is twice as large (30×240 km<sup>2</sup>). For the channel 4 and 6 data used for this study the integration time is mostly 0.25 s corresponding to a horizontal resolution of 30×60 km<sup>2</sup>. SCIAMACHY also performs direct (extraterrestrial) sun observations, e.g., to obtain the solar reference spectra needed for the retrieval.

SCIAMACHY is one of the first instruments that performs nadir observations in the near-infrared (NIR) spectral region (i.e., around 2 μm). In contrast to the ultra violet (UV) and visible spectral regions where high performance Si detectors have been manufactured for a long time, no appropriate near-infrared detectors were available when SCIAMACHY was designed. The near-infrared InGaAs detectors of SCIAMACHY were a special development for SCIAMACHY. Compared to the UV-visible detectors they are characterized by a substantially higher pixel-to-pixel variability in quantum efficiency and dark (leakage) current. Each detector array has a large number of dead and bad pixels. In addition, the dark signal is significantly higher compared to the UV-visible mainly because of thermal radiation generated by the instrument itself. The in-flight optical performance of SCIAMACHY is overall as expected from the on-ground calibration and characterization activities (Bovensmann et al., 2004). One exception is the time dependent optical throughput variation in the SCIAMACHY NIR channels 7 and 8 due the build-up of an ice layer on the detectors (“ice issue”). This effect is minimised by regular heating of the instrument (Bovensmann et al., 2004) during decontamination phases. The ice layers adversely influence the quality of the retrieval of all gases discussed in this paper as they result in reduced throughput (transmission) and, therefore,

reduced signal and signal-to-noise performance. In addition, changes of the instrument slit function have been observed which introduce systematic errors. All these issues complicate the retrieval.

### 3. WFM-DOAS retrieval algorithm

5 The Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval algorithm and its current implementation is described in detail elsewhere (Buchwitz et al., 2000a, 2004a,b). In short, WFM-DOAS is an unconstrained linear-least squares method based on scaling pre-selected trace gas vertical profiles. The fit parameters are 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 the 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 taking into account line-absorption and multiple scattering (Buchwitz et al., 2000b). A fast look-up table scheme has been developed in order to avoid time consuming on-line radiative transfer simulations.

15 In order to identify cloud-contaminated ground pixels we use a simple threshold algorithm based on sub-pixel information as provided by the SCIAMACHY Polarization Measurement Devices (PMDs) (details are given in Buchwitz et al., 2004a,b). We use PMD1 which corresponds to the spectral region 320–380 nm located in the UV part of the spectrum. Strictly speaking, the algorithm detects enhanced backscatter in the UV. Enhanced UV backscatter mainly results from clouds but might also be due to high aerosol loading or high surface UV spectral reflectance. As a result, ice or snow covered surfaces may be wrongly classified as cloud contaminated. This needs to be improved in future versions of our retrieval method.

25 The quality of the WFM-DOAS fits in the near-infrared is poor (i.e., the fit residuals are large) when applying WFM-DOAS to the operational Level 1 data products. In

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order to improve the quality of the fits and thereby the quality of our data products we pre-process the operational Level 1 data products mainly with respect to a better dark signal calibration (see [Buchwitz et al., 2004a,b](#)). In addition, there are indications that the in-orbit slit function of SCIAMACHY is different from the one measured on-ground (Hans Schrijver (SRON), personal communication) due to the “ice issue” (see Sect. 2). 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 (see [Buchwitz et al., 2004a,b](#)).

#### 4. WFM-DOAS data products: time coverage

The WFM-DOAS trace gas column data products have been derived by processing all consolidated SCIAMACHY Level 1 operational product files (i.e., the calibrated and geolocated spectra) of the year 2003 that have been made available by ESA/DLR (up to mid-2004). Figure 1 gives an overview about the number of orbits per day that have been processed. The maximum number of orbits per day is about fourteen. As can be seen (blue lines), all 14 orbits were available for only a small number of days. For many days no data were available. Many of the large data gaps are due to decontamination phases (see Sect. 2) which are indicated by red lines). For November and December 2003 no consolidated (i.e., full product) orbit files have been made available (for ground processing related reasons).

#### 5. Carbon monoxide (CO)

For CO column retrieval a small spectral fitting window (2359–2370 nm) located in SCIAMACHY channel 8 has been selected which covers four CO absorption lines. The retrieval is complicated by strong overlapping absorption features of methane and water vapor. The first results of CO from SCIAMACHY have been presented in [Buchwitz](#)



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et al. (2004a) focusing on a detailed analysis of three days of data of the year 2003. For details concerning pre-processing of the spectra (for improving the calibration), WFM-DOAS v0.4 retrieval, vertical column averaging kernels, quality of the spectral fits, and a quantitative comparison with MOPITT Version 3 CO columns (Deeter et al., 2003; Emmons et al., 2004) we refer to Buchwitz et al. (2004a). An initial error analysis using simulated measurements can be found in Buchwitz and Burrows (2004) where it is shown that the errors are expected to be less than about 20%. In Buchwitz et al. (2004a) it has been shown that strong plumes of CO can be detected with single overpass data which are in good qualitative agreement with MOPITT. Globally, for measurements over land, the standard deviation of the difference with respect to MOPITT was shown to be in the range  $0.4\text{--}0.6 \times 10^{18}$  molecules/cm<sup>2</sup> and the linear correlation coefficient between 0.4 and 0.7. The differences depend on time and location but are typically within 30% for most latitudes. Perfect agreement with MOPITT is, however, not to be expected for a number of reasons (differences in overpass time, spatial resolution, etc.). In this context it is important to point out that the sensitivity of SCIAMACHY measurements is nearly independent of altitude whereas the sensitivity of MOPITT to boundary layer CO is low. On the other hand, retrieval of CO from SCIAMACHY is not unproblematic. For example, the WFM-DOAS v0.4 CO column are scaled with a constant factor of 0.5 to compensate for an obvious overestimation. This overestimation is most probably closely related to the difficulty of accurately fitting the weak CO lines. The fit residuals, which are on the order of the CO lines, are not (yet) signal-to-noise limited but dominated by (not yet understood) rather stable spectral artifacts.

Figure 2 shows a comparison of monthly averaged WFM-DOAS version 0.4 CO columns with CO from MOPITT (version 3). Because of the low surface reflectivity of water the NIR nadir measurements are noisy over the ocean (outside sun-glint conditions). Therefore, we focus on SCIAMACHY measurements over land. Only these measurements are shown in Fig. 2. The same land mask as used for SCIAMACHY has also been used for MOPITT to ease the comparison. SCIAMACHY data have only been averaged if the CO fit error is less than 60% and when the pixels have been

flagged cloud free. As can be seen from Fig. 1, there are large SCIAMACHY data gaps for April and August. Therefore, the SCIAMACHY data shown in Fig. 2 are not monthly averages obtained from a bias free sampling. The August data, for example, are strongly biased towards the beginning of August.

5 Only for September were nearly all orbits available. As a result, nearly all land areas are covered by measurements during this month. But even for this month there are quite large gaps. There are nearly no data over Greenland because the cloud detection algorithm can not discriminate between clouds and snow/ice covered surfaces. There are also large gaps in the tropics due to persistent cloud coverage. When comparing the September data from SCIAMACHY and MOPITT one can see that overall  
10 the agreement is good. Both sensors show that the columns are typically between 1.7 and  $2.5 \times 10^{18}$  molecules/cm<sup>2</sup> (shown in green). Both sensors show large regions of enhanced CO in the northern part of South America and over the central/southern part of Africa. It is well known that a lot of biomass burning is going on in these regions  
15 in September. In the northern hemisphere the overall agreement is reasonable but there are also substantial differences. For example, SCIAMACHY sees much higher CO over the eastern part of the United States compared to MOPITT. If this is due to the higher sensitivity of the SCIAMACHY measurements for boundary layer CO or for other reasons is currently unclear.

20 The April data of SCIAMACHY show large gaps in time and space. The agreement with MOPITT is not so good as for September. The main difference with respect to MOPITT is the significantly weaker inter-hemispheric difference as seen by SCIAMACHY. Both sensors give (on average) quite similar values over the northern hemisphere but over the southern hemisphere there are significant differences. Here the MOPITT data are mostly in the range  $1.2\text{--}1.7 \times 10^{18}$  molecules/cm<sup>2</sup> whereas the SCIAMACHY data are systematically higher (around  $2 \times 10^{18}$  molecules/cm<sup>2</sup>). The SCIAMACHY data over  
25 South America are sparse. Nevertheless, outside the Andes mountains, there are clear differences concerning the observed CO plumes. Whereas MOPITT only detects strongly elevated levels of CO in the most northern part of South America, SCIA-

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MACHY shows elevated CO also at various other places. The northern hemisphere is difficult to compare because of the large gaps in the SCIAMACHY data. Both sensors observe elevated CO over a large part of western central Africa north of the equator, where significant biomass burning is typically going on during this time of the year.

5 For August 2003 there are also large gaps in the SCIAMACHY data. In general, the agreement is fair but there are large differences over several parts of the world. For example, SCIAMACHY sees high CO over a large part of northern Africa not seen by MOPITT. The reason for this difference is not clear. The MOPITT data are in better agreement with typical biomass burning areas for August. There are also large regions  
10 over the northern hemisphere where SCIAMACHY sees higher columns than MOPITT. For example, SCIAMACHY sees high CO over Portugal (but not over Spain) which is not observed by MOPITT. This coincides with large fires which were going on in August 2003 in Portugal (but not in Spain).

In summary, the agreement is reasonable but there are significant differences at  
15 certain locations during certain times of the year. More investigations are needed to explain the observed differences taking into account the different altitude sensitivities of both sensors.

## 6. Methane (CH<sub>4</sub>)

The methane columns have been retrieved from a small spectral fitting window (2265–  
20 2280 nm) located in SCIAMACHY channel 8 which covers several absorption lines of CH<sub>4</sub> and several but much weaker absorption lines of nitrous oxide (N<sub>2</sub>O) and water vapor (H<sub>2</sub>O). The main scientific application of the methane measurements of SCIAMACHY is to obtain information on the surface sources of methane. The modulation of methane columns due to methane sources is only on the order of about one percent.

25 This is much weaker than the variation of the methane column due to changes of surface pressure (because methane is well-mixed the methane column is highly correlated with the total air mass and, therefore, with surface pressure). To filter out these much

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larger disturbing modulations, the methane columns need to be normalized by the observed air mass to obtain a so called dry air column averaged mixing ratio of methane (denoted XCH<sub>4</sub>). To accomplish this, oxygen (O<sub>2</sub>) columns have been retrieved in addition to the methane columns. From the oxygen columns the air mass can be calculated using its constant mixing ratio of 0.2095. The O<sub>2</sub> columns used to compute the WFM-DOAS v0.4 XCH<sub>4</sub> data product have been retrieved from the SCIAMACHY measurements in the O<sub>2</sub> A band spectral region (around 760 nm; channel 4).

First CH<sub>4</sub> results from SCIAMACHY have been presented in [Buchwitz et al. \(2004b\)](#) focusing on a detailed analysis of four days of data of the year 2003. For details concerning pre-processing of the spectra (to improve the calibration), WFM-DOAS (v0.4) processing, averaging kernels, quality of the spectral fits, and a quantitative comparison with global models we refer to that study. An initial error analysis using simulated measurements is given in [Buchwitz and Burrows \(2004\)](#). According to this error analysis errors of a few percent due to undetected cirrus clouds, aerosols, surface reflectivity, temperature and pressure profiles, etc., are to be expected. It has been shown in [Buchwitz et al. \(2004b\)](#) that the WFM-DOAS Version 0.4 methane columns have a time dependent (nearly globally uniform) bias of up to –15% (low bias of SCIAMACHY) for one of the four days that have been analyzed. The bias is correlated with the time after the last decontamination performed to get rid of the ice layers on the detectors. Therefore, [Buchwitz et al. \(2004b\)](#) concluded that the bias might be due to the “ice-issue” (see Sect. 2). This is consistent with the finding of H. Schrijver (SRON) (see Sect. 2) that the ice build-up on the detectors results in a broadening of the instrument slit function (the wider the slit function compared to the assumed slit function, the larger the underestimation of the retrieved methane column).

Because the methane source signal is weak it typically cannot be detected with single overpass or single day SCIAMACHY data. To detect methane sources averages have to be computed. Because of the time dependent bias of the WFM-DOAS v0.4 methane columns, however, this is not directly possible.

In the following we describe how we have processed the SCIAMACHY data to get

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an improved version of our methane data product (Version 0.41) by applying a bias correction to the v0.4 methane columns. We assume that the data can be sufficiently corrected by dividing the columns by a globally constant scaling factor which only depends on time (on the day of the measurement). The correction factor has been determined as follows: For each day all cloud free v0.4 XCH<sub>4</sub> measurements over the Sahara have been averaged. The ratio of these daily average mixing ratios to a constant reference value (chosen to be 1750 ppbv) is approximately the methane bias (because methane is not constant this is not exactly the methane bias and a certain systematic error is introduced by this assumption). This time dependent methane bias is shown in Fig. 3 (black diamonds). This bias shows a similar time dependence as the independently measured channel 8 transmission loss also shown in Fig. 3 (red diamonds). The transmission has been determined by averaging the signal of the channel 8 solar measurements normalized to a reference measurement at the beginning of the mission. The varying transmission is a consequence of the varying ice layer on the detectors. Figure 3 shows a third curve, the (daily) correction factor (magenta diamonds). The correction factor curve has been obtained by linearly transforming the transmission curve. The coefficients of the linear transformation have been selected such that a good match is obtained with the methane bias curve. In order to correct the WFM-DOAS v0.4 methane columns for the systematic errors introduced by the ice layer the correction factors are applied as follows: All v0.4 methane columns of a given day have been divided by the correction factor for this day. The corrected WFM-DOAS v0.4 methane columns are the new WFM-DOAS v0.41 (absolute) methane columns.

In order to generate the new WFM-DOAS v0.41 XCH<sub>4</sub> product a second modification has been applied: Instead of normalizing the methane columns by the oxygen column retrieved from the 760 nm O<sub>2</sub> A band (as done for v0.4 XCH<sub>4</sub>), they have been normalized by the CO<sub>2</sub> columns retrieved from the 1580 nm region (for details on CO<sub>2</sub> retrieval see Sect. 7). Using CO<sub>2</sub> rather than O<sub>2</sub> for normalizing the methane columns has been first proposed by C. Frankenberg et al. (presented at ENVISAT Symposium, Salzburg, Austria, 6–10 September 2004). The reason why normalizing methane by

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CO<sub>2</sub> rather than O<sub>2</sub> is expected to give better results for XCH<sub>4</sub> is because errors of CO<sub>2</sub> and CH<sub>4</sub> are similar and cancel much better when the ratio is computed. Errors due to aerosols, residual cloud contamination, surface reflection, etc., are expected to be the more similar, the more similar the radiative transfer is. In general, this requires that the two spectral intervals from which the two columns are retrieved are located as close as possible (in wavelength). As the CO<sub>2</sub> fitting window (at 1580 nm), is much closer to the CH<sub>4</sub> fitting window (2270 nm) than the O<sub>2</sub> window (760 nm) canceling of errors will be better using CO<sub>2</sub>. In addition, when the column retrieval from the two spectral regions suffers from similar instrumental/calibration errors, also these errors cancel to a certain extent.

The drawback of this approach is that CO<sub>2</sub> is not as constant as O<sub>2</sub> mainly because of the surface sources and sinks of CO<sub>2</sub>. This approach requires that the variability of the column averaged mixing ratio of CO<sub>2</sub> is small (ideally negligible) compared to the variability of the column averaged mixing ratio of CH<sub>4</sub>. According to global model simulation this is a reasonable assumption. The model simulations shown in Buchwitz et al. (2004a) indicate that the variability of the methane column is about 6% ( $\pm 100$  ppbv) and the variability of the CO<sub>2</sub> column is about 1.5% ( $\pm 5$  ppmv), i.e., a factor of four smaller than for methane. This means that the error introduced by essentially assuming that CO<sub>2</sub> is constant is less than about 1.5%. This is comparable to the estimated error on the WFM-DOAS v0.4 CO<sub>2</sub> columns as reported in Buchwitz and Burrows (2004).

Figure 4 shows a comparison of WFM-DOAS v0.41 XCH<sub>4</sub> with TM5 model simulations. The TM5 model is a two-way nested atmospheric zoom model (Krol et al., 2004). It allows to define zoom regions (e.g. over Europe) which are run at higher spatial resolution ( $1 \times 1^\circ$ ), embedded into the global domain, run at a resolution of  $6 \times 4^\circ$ . We employ the tropospheric standard version of TM5 with 25 vertical layers. TM5 is an off-line model and uses analyzed meteorological fields from the ECMWF weather forecast model to describe advection and vertical mixing by cumulus convection and turbulent diffusion. CH<sub>4</sub> (a priori) emissions are as described by Bergamaschi et al. (2004). Chemical destruction of CH<sub>4</sub> by OH radicals is simulated using pre-calculated

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OH fields based on CBM-4 chemistry and optimized with methyl chloroform, for the stratosphere also the reaction of CH<sub>4</sub> with Cl and O(<sup>1</sup>D) radicals are considered. The comparison is limited to observations over land as the SCIAMACHY observations over ocean are less precise because of the low ocean reflectivity in the near-infrared. For SCIAMACHY all measurements from cloud free pixels have been averaged where the CH<sub>4</sub> column fit error is less than 10%. The bi-monthly averages have been computed from daily gridded data. For the model simulations only those grid boxes where measurements were available have been used to compute the averages.

The comparison with the model simulations shows similarities but also differences. Very interestingly and in qualitative agreement with the model simulations, the measurements show high CH<sub>4</sub> mixing ratios in the September to October 2003 average over India, southeast Asia, and over the western part of central Africa, which are absent or significantly lower in the March to April average. In the model the high columns in these regions are a result of methane emissions mainly from rice fields, wetlands, ruminants, and waste handling. The good agreement with the model simulations indicates that SCIAMACHY can detect these emission signals. However, there are also differences compared to the model simulations. For example, in the March-April average the SCIAMACHY data are few percent lower over large parts of South America but over large parts of the northern hemisphere the SCIAMACHY data are a few percent lower. More investigations are needed to find out what the reasons for these discrepancies are.

Figure 5 shows a quantitative comparison of the daily data with the TM5 model, the correlation coefficient and the bias for the two versions of SCIAMACHY XCH<sub>4</sub> data products, namely v0.4 and v0.41. As can be seen, the bias is significantly smaller for the v0.41 data, although not zero. There still appears to be a systematic bias due to the ice issue indicating that the bias correction applied to generate the v0.41 data is not perfect. Also the correlation with the model results is better for the version 0.41 data, especially in the middle of 2003, where the correlation coefficient can be as high as 0.9.



## 7. Carbon dioxide (CO<sub>2</sub>)

The CO<sub>2</sub> columns have been retrieved using a small spectral fitting window (1558–1594 nm) located in SCIAMACHY channel 6 (which is not affected by an ice layer). This spectral region covers one absorption band of CO<sub>2</sub> and weak absorption features of water vapor. As for methane v0.4 (air or) O<sub>2</sub>-normalized CO<sub>2</sub> columns have been derived, the dry air column averaged mixing ratios XCO<sub>2</sub>. First results of CO<sub>2</sub> from SCIAMACHY have been presented in [Buchwitz et al. \(2004b\)](#). For details concerning pre-processing of the spectra (for improving the calibration), WFM-DOAS (v0.4) processing, vertical column averaging kernels, quality of the spectral fits, and a quantitative comparison with global model simulations we refer to [Buchwitz et al. \(2004b\)](#). An initial error analysis using simulated measurements is given in [Buchwitz and Burrows \(2004\)](#). According to this error analysis errors of a few percent due to undetected cirrus clouds, aerosols, surface reflectivity, temperature and pressure profiles, etc., are to be expected. In [Buchwitz et al. \(2004b\)](#) it has been shown that the WFM-DOAS v0.4 CO<sub>2</sub> columns agree with model columns within a few percent. To compensate for a not yet understood systematic underestimation the WFM-DOAS v0.4 CO<sub>2</sub> columns have been scaled with a constant factor of 1.27 (see [Buchwitz et al., 2004b](#), for details). It has been shown that the spatial and temporal pattern of the retrieved column averaged mixing ratio is in reasonable agreement with the model data except for the amplitude of the variability. The measured variability is about a factor of four higher than the variability of the model data (about 6% compared to about 1.5% for the model data).

In the following we restrict the discussion to the comparison of three months of data with TM3 model simulations as shown in Fig. 6. More results can be found in [Buchwitz et al. \(2004b\)](#). TM3 3.8 ([Heimann and Körner, 2003](#)) is a three-dimensional global atmospheric transport model for an arbitrary number of active or passive tracers. It uses re-analyzed meteorological fields from the National Center for Environmental Prediction (NCEP) or from the ECMWF re-analysis. The modeled processes comprise tracer advection, vertical transport due to convective clouds and turbulent vertical transport

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by diffusion. Available horizontal resolutions range from  $8\times 10^\circ$  to  $1.1\times 1.1^\circ$ . In this case, TM3 was run with a resolution of  $1.8\times 1.8^\circ$  and 28 layers, and the meteorology fields were derived from the NCEP/DOE AMIP-II reanalysis. CO<sub>2</sub> source/sink fields for the ocean originate from Takahaschi et al. (2002), for anthropogenic sources from the EDGAR 3.2 database and for the biosphere from the BIOME-BGC model. For SCIAMACHY the averages have been computed using only the cloud free pixels with a CO<sub>2</sub> retrieval error of less than 10%. Shown in Fig. 6 are only the data over land because of the problems with measuring over the ocean in the near-infrared (see Sect. 5).

For SCIAMACHY Fig. 6 shows absolute column averaged mixing ratios of CO<sub>2</sub> in the range 335–385 ppmv. For TM3 “uncalibrated” XCO<sub>2</sub>-offsets are shown which are in the range 0–13.7 ppmv. These offsets do not include the (current) background concentration of CO<sub>2</sub>. Therefore, not the absolute values but only the variability in space and time should be compared. The model simulations show low columns (compared to the mean column) over the northern hemisphere in July 2003 compared to higher values in May and September. This is mainly due to uptake of CO<sub>2</sub> by the biosphere which results in minimum columns around July. Qualitatively the SCIAMACHY data show a similar time dependence with also lower columns in July compared to May and September. This indicates that SCIAMACHY is able to detect the uptake of CO<sub>2</sub> by the biosphere over the northern hemisphere when the vegetation is in its main growing season. The measured variability ( $\pm 25$  ppmv) is about a factor of 3–4 higher than the variability of the model data ( $\pm 7$  ppmv).

Over large parts of the (mostly western) Sahara SCIAMACHY sees “plumes” of relatively high CO<sub>2</sub> (red colored areas) not present in the model simulations. These probably a few percent too high CO<sub>2</sub> mixing ratios may result from the high surface reflectivity over the Sahara (probably in combination with aerosol variability). Currently, only a constant surface albedo of 0.1 is assumed for WFM-DOAS (and only one aerosol scenario). According to the error analysis presented in Buchwitz and Burrows (2004) the error on the CO<sub>2</sub> column is +1.4% if the albedo is 0.3 instead of 0.1 (for a solar zenith angle of  $50^\circ$ ). The corresponding error for O<sub>2</sub> is –3.0%. The XCO<sub>2</sub> error is to

a good approximation the difference of these errors, i.e., +4.4% or 16 ppmv. This indicates that the high values seen by SCIAMACHY over the Sahara may be explained by retrieval algorithm limitations as the current version does not take albedo (and aerosol) variations fully into account.

## 8. Conclusions

Nearly one year (2003) of SCIAMACHY nadir measurements have been processed with the WFM-DOAS retrieval algorithm (v0.4, for methane also v0.41) to generate a number of data products: vertical columns of CO, CH<sub>4</sub>, and CO<sub>2</sub>. In addition, O<sub>2</sub> columns have been retrieved to compute dry air column averaged mixing ratios for the relatively well-mixed greenhouse gases CH<sub>4</sub> and CO<sub>2</sub>, denoted XCH<sub>4</sub> and XCO<sub>2</sub>, respectively. The data products have been compared with independent measurements (CO from MOPITT) and model simulations (for CH<sub>4</sub> and CO<sub>2</sub>).

For the CO columns the agreement with MOPITT is mostly within 30%. SCIAMACHY detects enhanced concentrations of CO due to biomass burning similar as MOPITT. SCIAMACHY seems to systematically overestimate the CO columns over large parts of the southern hemisphere at least for certain months where MOPITT sees systematically lower columns in the southern hemisphere compared to the northern hemisphere. This discrepancy is most probably related to the difficulty of accurately fitting the weak CO lines covered by SCIAMACHY. Investigations are ongoing on how to improve the precision and the accuracy of the CO retrieval. This includes using a larger fitting window to cover more CO lines.

The WFM-DOAS Version 0.4 methane columns have a time dependent bias of up to about -15% related to ice build-up on the channel 8 detector. Using a simple bias correction an improved methane data product (v0.41) has been generated. The comparison with model simulations shows agreement within a few percent. The comparison indicates that SCIAMACHY can detect enhanced columns due to emissions from surface sources such as rice fields and wetlands over India, southeast Asia and central

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The WFM-DOAS Version 0.4 CO<sub>2</sub> columns show agreement with model simulations within a few percent. The comparison indicates that SCIAMACHY is able to detect low columns of CO<sub>2</sub> resulting from uptake of CO<sub>2</sub> over the northern hemisphere when the vegetation is in its main growing season. Over highly reflecting surfaces such as over the Sahara SCIAMACHY seems to systematically overestimate the column averaged mixing ratio of CO<sub>2</sub> by a few percent most probably because of limitations of the current version of the retrieval algorithm.

A summary of our findings from the comparisons with independent data shown here and elsewhere (Buchwitz et al., 2004a,b; Gloudemans et al., 2004; de Maziere et al., 2004; Susmann and Buchwitz, 2005; Warneke et al., 2005) is given in Table 1 which shows our current best estimates of precision and accuracy of our data products.

Our future work will focus on identifying the reasons for the observed biases and to improve the accuracy of the data products. This will also comprise the use of larger spectral fitting windows, especially for CO, to improve the precision. So far only a small subset of the large spectral region covered by SCIAMACHY has been analyzed.

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**Table 1.** Current estimates of precision and accuracy of the SCIAMACHY/WFM-DOAS vertical column data products.

Gas	Horizontal resolution [km <sup>2</sup> ]	Estimated precision (scatter) [%]	Estimated accuracy (bias) [%]
CO (v0.4)	30×120	10–20	10–30 (mostly positive)
CH <sub>4</sub> (v0.4)	30×120	1–6	2–15 (mostly negative)
CH <sub>4</sub> (v0.41)	30×120	1–4	2–5
CO <sub>2</sub> (v0.4)	30×60	1–4	2–5

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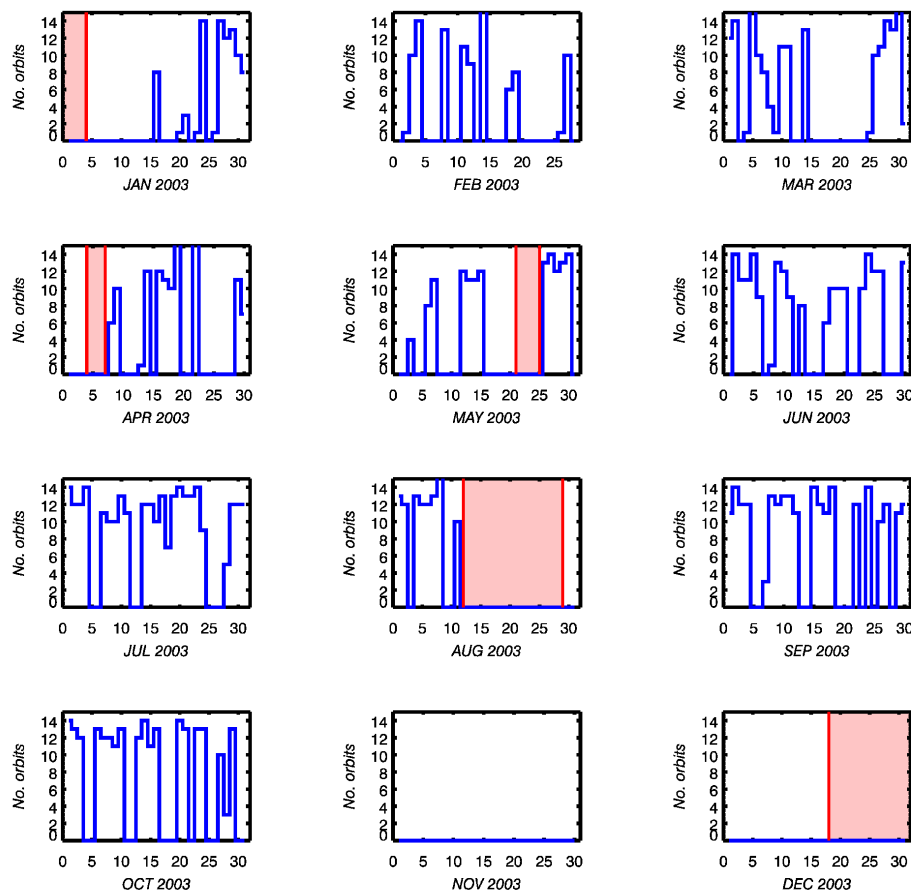
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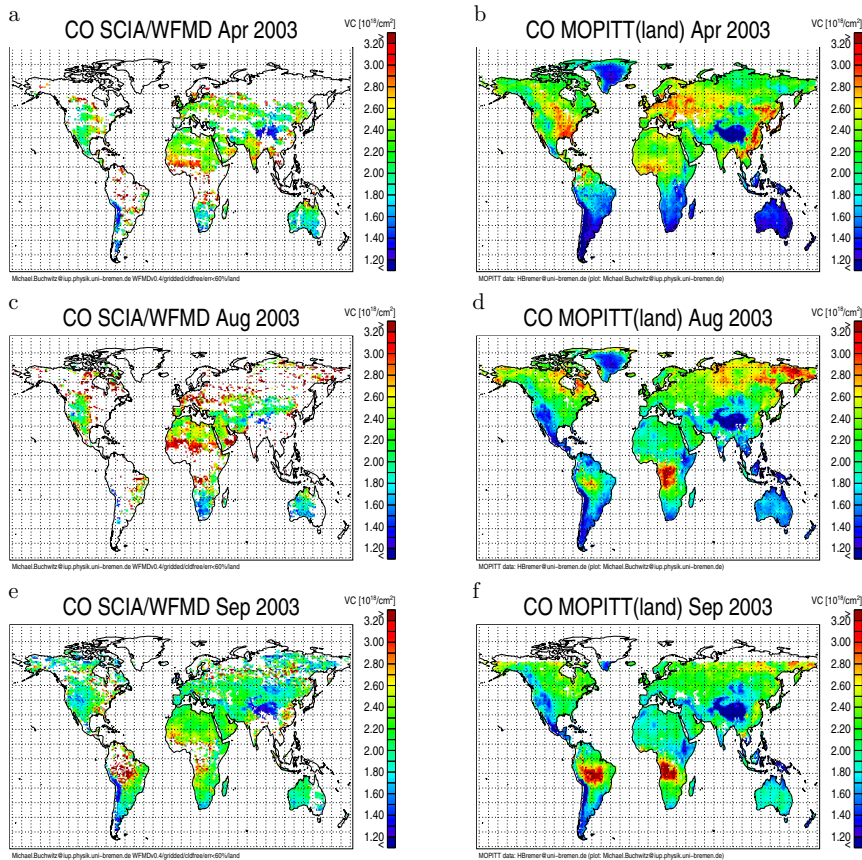
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**Fig. 1.** Number of orbits per day of the year 2003 processed by WFM-DOAS (blue lines). The maximum number of orbits per day is about 14 (~100%). The red shaded areas indicate the decontamination phases performed to get rid of the ice layer that grows on the near-infrared detectors of channels 7 and 8. Data gaps are due to decontamination but also due to other (mostly ground processing related) reasons.

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**Fig. 2.** Monthly mean CO columns over land from SCIAMACHY/ENVISAT (left) and MOPITT/EOS-Terra (right). Only the columns over land are shown because the quality of the SCIAMACHY CO columns over water is low due to the low reflectivity of water in the near-infrared. For SCIAMACHY only data have been averaged where the CO fit error is less than 60% and where the PMD1 cloud identification algorithm indicates a cloud free pixel. For MOPITT cloudy pixels are also not included.

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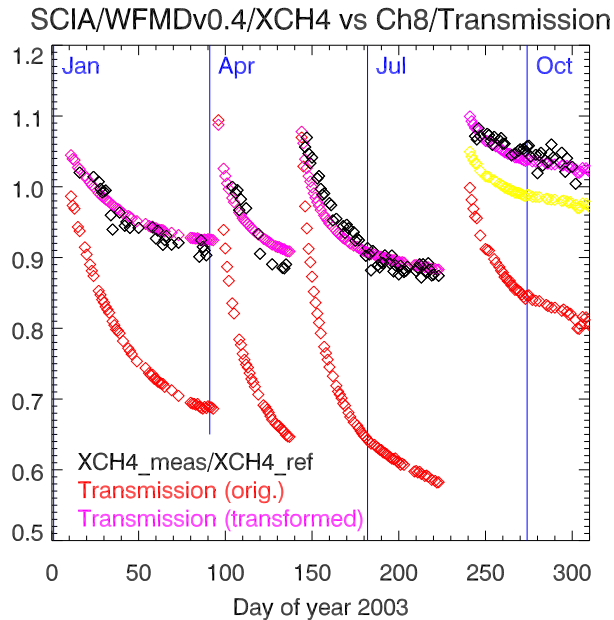
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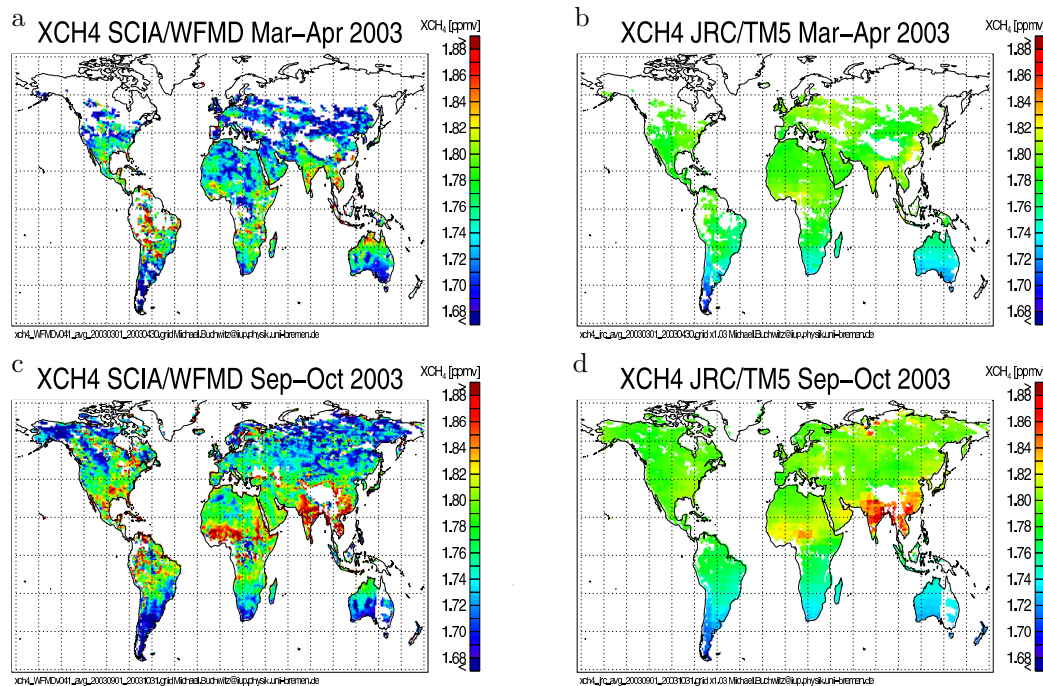


**Fig. 3.** The black diamonds show XCH<sub>4</sub> (v0.4) as measured by SCIAMACHY over the Sahara normalized by a constant reference value of 1750 ppbv as a function of day of the year 2003 (“methane bias curve”). The red diamonds (“Transmission (orig.)”) show independent measurements, namely the SCIAMACHY channel 8 transmission which changes due to the varying ice layer on the detector. This curve has been determined by averaging the signal of the solar measurements and normalizing it. The magenta (*m*) “Transmission (transformed)” curve has been obtained by a linear transformation of the red (*r*) curve, i.e., by computing  $m=A+Br$ . Applying the same coefficients *A* and *B* as used for the data before day 230 also to the data after day 230 results in the yellow curve which still shows an offset with respect to the black methane bias curve. To get a better match, two sets of coefficients have been used, one for the data before day 230 and another set for the data after day 230. The reason why different coefficients are needed is most probably because the spatial distribution of the ice layer on the detector changes with time.

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## CO, CH<sub>4</sub>, and CO<sub>2</sub> columns from SCIAMACHY

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**Fig. 4.** Column averaged mixing ratios of methane (XCH<sub>4</sub>) as measured by SCIAMACHY over land using WFM-DOAS v0.41 (left) compared to JRC/TM5 model data (right). For SCIAMACHY only the measurements from cloud free pixels have been averaged where the methane error is less than 10%. The model data have been averaged taking into account the daily sampling of the SCIAMACHY data. The model data have been scaled by a constant factor of 1.03 to compensate for the bias between the two data sets (see Fig. 5).

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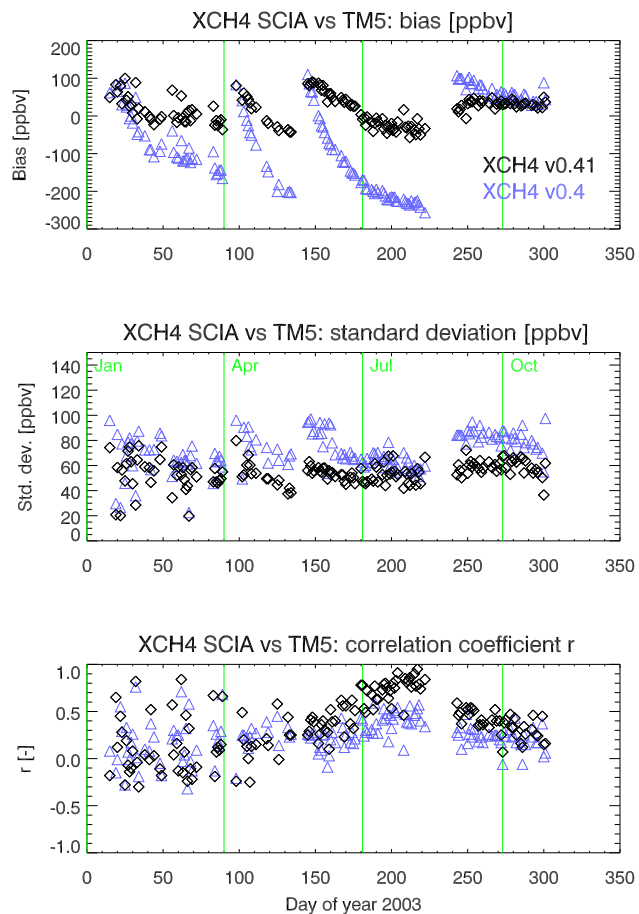
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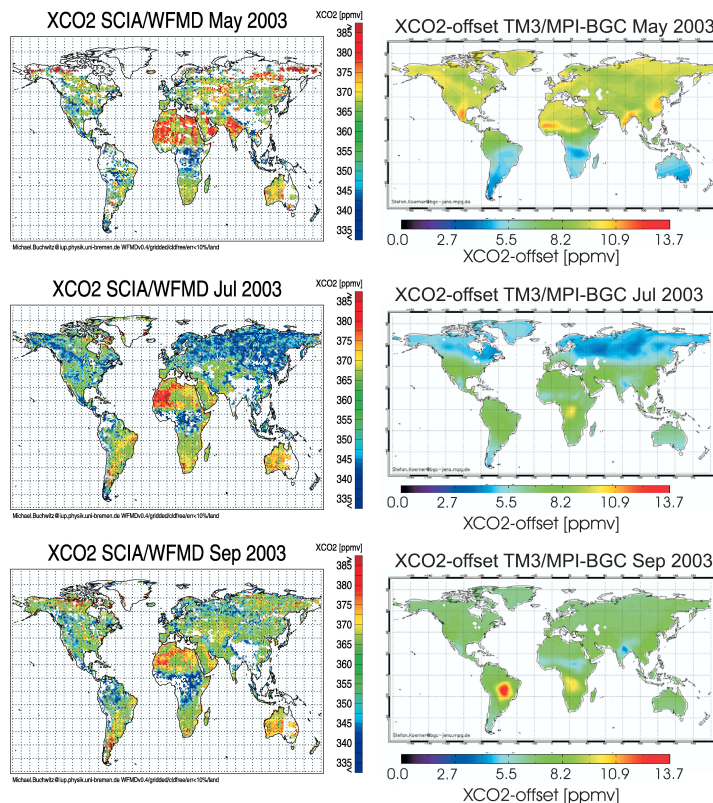


**Fig. 5.** Comparison of daily SCIAMACHY XCH<sub>4</sub> measurements (versions 0.4, blue triangles, and 0.41, black diamonds) with TM5 model simulations. The top panel shows the bias (SCIAMACHY-model), the middle panel the standard deviation of the difference, and the bottom panel Pearson's linear correlation coefficient  $r$ .

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## CO, CH<sub>4</sub>, and CO<sub>2</sub> columns from SCIAMACHY

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**Fig. 6.** Comparison of SCIAMACHY XCO<sub>2</sub> (left) with TM3 model simulations (right). For SCIAMACHY all cloud free measurements over land have been averaged where the CO<sub>2</sub> column fit error is less than 10%. For TM3 monthly averaged XCO<sub>2</sub>-offsets are shown. These offsets do not include the background concentration of CO<sub>2</sub>. Therefore, only the spatial and temporal variability should be compared. Two different scales have been used, one for SCIAMACHY ( $\pm 25$  ppmv) and one for the model simulations ( $\pm 6.85$  ppmv), to consider the 3–4 times higher variability of the SCIAMACHY data compared to the model data.

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