

**SCIAMACHY carbon
gas measurements:
CO, CH₄, and CO₂**

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Atmospheric carbon gases retrieved from SCIAMACHY by WFM-DOAS: improved global CO and CH₄ and initial verification of CO₂ over Park Falls (46° N, 90° W)

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The three carbon gases carbon monoxide (CO), carbon dioxide (CO₂), and methane (CH₄) are important atmospheric constituents affecting air quality and climate. The nadir spectra of reflected and scattered solar radiation in the near-infrared region, as observed by SCIAMACHY/ENVISAT, contain information on the vertical columns of these gases. A modified DOAS algorithm (WFM-DOAS) has been developed to retrieve this information. The main SCIAMACHY/WFM-DOAS data products are CO vertical columns and dry-air column averaged mixing ratios of methane and CO₂, denoted XCH₄ and XCO₂, respectively. For CO and methane we present new results obtained with an improved version of WFM-DOAS (v0.5). The SCIAMACHY data products have been compared with global reference data (MOPITT for CO, TM5 model simulations for XCH₄). The comparisons indicate that major problems of the previous version of WFM-DOAS (v0.4x) related to the varying ice-layer on the SCIAMACHY channel 8 detector have been solved. On average, the SCIAMACHY CO agrees within 10% (standard deviation 30%) with MOPITT but regionally, especially over northern South America, large differences have been found (up to about 80%). For methane we present global and regional maps which are compared to TM5 model simulations performed using standard emission inventories. Overall, there is good agreement but regionally there are substantial differences, e.g., due to limitations of current methane emission inventories. It still needs to be assessed by how much emission inventories can be improved by using the SCIAMACHY data. Concerning CO₂ we present a comparison of SCIAMACHY XCO₂ (WFM-DOAS v0.4) with TM3 model simulations over Park Falls, Wisconsin, USA. The peak-to-peak XCO₂ variability as measured by SCIAMACHY (seasonal cycle of year 2003–2005 data) is ~13 ppmv, in good agreement with preliminary analysis of ground-based Fourier Transform Spectrometer (FTS) measurements, which is a factor of 2.3 larger than the XCO₂ variability of TM3 model simulation for 2003. Park Falls is one of the few FTS ground stations which measure column averaged CO₂ and detailed comparison with these measurements (after data

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release) will help identifying the reason for the observed differences between SCIAMACHY and global (atmospheric) carbon models such as TM3 as reported here and in previous studies. For all three carbon gases we present regional results including seasonal variation focusing on China.

1. Introduction

Carbon monoxide (CO) is a major tropospheric air pollutant and carbon dioxide (CO₂) and methane (CH₄) are the two most important anthropogenic greenhouse gases. Knowledge about the global distribution of these carbon gases is therefore important. CO 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 the troposphere and, therefore, its self-cleansing efficiency and the concentration of greenhouse gases such as CH₄. CO also has large air quality impact as a pre-cursor to tropospheric ozone, a secondary pollutant associated with respiratory problems and decreased crop yields. Satellite measurements of CH₄, CO₂, and CO in combination with inverse modeling have the potential to help better constrain the uncertainty of the strength of the surface fluxes than currently possible with the highly precise but rather sparse data from the network of surface stations (see Houweling et al., 1999, 2004; Rayner and O'Brien, 2001, and references given therein). A better understanding of the sources and sinks of CH₄ and CO₂ 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.

High quality trace gas column retrieval from the SCIAMACHY near-infrared spectra is a challenging task for many reasons, e.g., due to calibration issues mainly related to high and variable dark signals and changing detector characteristics (Gloude-mans et al., 2005), because it is difficult to detect the weak CO lines which are close to the noise level and due to strong interference of water and methane lines, and because

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of the challenging accuracy and precision requirements for CO₂ (Rayner and O'Brien, 2001; Houweling et al., 2004) and CH₄ (Meirink et al., 2005). When developing a retrieval algorithm many decisions 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/precision of the data products. In this context it is important to point out that other groups are also working on this important topic using different approaches (see Gludemans et al., 2004, 2005; Frankenberg et al., 2005a,b,c; Houweling et al., 2005).

SCIAMACHY is not a dedicated CO₂ and/or CH₄ mission such as the planned future near-infrared/nadir missions OCO/USA (Crisp et al., 2004) and GOSAT/Japan (http://www.jaxa.jp/missions/projects/sat/eos/gosat/index_e.html) which have for example higher spectral and spatial resolution compared to SCIAMACHY in order to maximize the spectral information and to have a higher probability for cloud free ground pixels. Nevertheless, SCIAMACHY makes similar measurements as will be done by OCO and GOSAT and therefore can be considered the pre-decessor of these dedicated carbon missions. The SCIAMACHY spectral measurements are currently used for example by the OCO team to critically test the OCO retrieval algorithm and validation approach.

Initial carbon gas vertical column data products derived from SCIAMACHY nadir observations with the WFM-DOAS version 0.4x (0.4 and 0.41) retrieval algorithm have been presented and compared with global reference data in Buchwitz et al. (2005b). The version 0.4x data set has also been compared with independent ground based Fourier Transform Spectroscopy (FTS) measurements (de Maziere et al., 2004; Sussmann and Buchwitz, 2005; Warneke et al., 2005; Dils et al., 2005; Sussmann et al., 2005). A comparison of the new version 0.5 data products with ground-based FTS measurements is currently being performed. A detailed comparison with our main CO₂ data product, the dry-air column averaged mixing ratio XCO₂, has however not yet been done. In Dils et al. (2005) a comparison is shown for the absolute CO₂ columns

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(in molecules/cm²) using three NDSC stations (two located at non-ideal position for the validation of tropospheric gases: the high latitude station Ny Alesund, and the mountain station Jungfrauoch). In [Warneke et al. \(2005\)](#) SCIAMACHY XCO₂ over Africa is compared with (nearby) ship-based FTS XCO₂ over the atlantic ocean but only for a few days end of January to beginning of February 2003. In this context it is important to point out that the SCIAMACHY carbon gas measurements over water are of significantly lower quality compared to measurements over land due to the low albedo of water in the near-infrared (outside sun-glint conditions).

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. In Sect. 4 the new WFM-DOAS version 0.5 CO and methane data are presented and compared with global reference data and a comparison is shown between SCIAMACHY/WFM-DOAS version 0.4 XCO₂ over Park Falls with TM3 model simulations. In Sect. 5 regional results for all three carbon gases are presented focusing on China. Conclusions are given in Sect. 6.

2. The SCIAMACHY instrument

SCIAMACHY ([Burrows et al., 1995](#); [Bovensmann et al., 1999](#)) 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). On the Earth's day side SCIAMACHY mainly performs a sequence of alternating nadir and limb observations. The horizontal resolution of the nadir measurements depends on orbital position and spectral interval but is typically 60 km (e.g., for methane and CO₂) or 120 km (e.g., for CO) across track times 30 km along track. These measurements can be inverted to obtain a large number of (primarily) atmospheric data products ([Bovensmann et al., 1999](#)).

Overall, the in-flight optical performance of SCIAMACHY is as expected from the on-ground activities. One exception is a time dependent optical throughput variation

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in the SCIAMACHY near-infrared (NIR) channels 7 (the main CO₂ channel) and 8 (the only CO channel and main CH₄ channel) due to ice build-up on the detectors which adversely affects the trace gas retrieval (Buchwitz et al., 2005b; Gloudemans et al., 2005). This effect is minimized by regular heating of the instrument. The WFM-DOAS version 0.5 methane and version 0.4 XCO₂ results presented in this paper have been derived from channel 6 not affected by an ice-layer. The WFM-DOAS version 0.5 CO is retrieved from channel 8 using a correction procedure for ice-layer induced errors.

3. WFM-DOAS retrieval algorithm

The retrieval of a long-lived and therefore relatively well-mixed gas such as carbon dioxide and methane is extremely challenging as only small variations on top of a large background are of relevance in order to obtain information on their surface sources and sinks. Therefore, the retrieval algorithm has to be very accurate. In addition, the algorithm also has to be very fast to process huge amounts of data. We have developed the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval algorithm to accomplish this task.

WFM-DOAS (Buchwitz et al., 2000b, 2004, 2005a,b; Buchwitz and Burrows, 2004) 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. the observed sun-normalized radiance. The WFM-DOAS reference spectra are the logarithm of the sun-normalized radiance and its derivatives computed with a radiative transfer model (Buchwitz et al., 2000a). In order to avoid time-consuming on-line radiative transfer simulations, a fast look-up table scheme has been implemented.

There are strong indications that due to the varying ice-layers on the channel 7 and 8 detectors (see Sect. 2) the in-orbit spectrometer slit function of SCIAMACHY is different

from the one measured on-ground and changes with time (GlouDEMANS et al., 2005; Buchwitz et al., 2005b). The slit function currently used by WFM-DOAS has been determined by applying WFM-DOAS to the in-orbit nadir measurements. The one that resulted in best fits, i.e. the smallest fit residuum, has been selected.

5 Basically the same algorithm is used to retrieve CO, methane, and CO₂. There are however also trace gas specific differences which will be discussed in the following subsections.

3.1. CO specific aspects

Carbon monoxide is retrieved from a small spectral fitting window (2324–2335 nm) located in channel 8 covering several absorption lines of CO. Compared to our initial version 0.4 CO (Buchwitz et al., 2004, 2005b) the version 0.5 data product presented here has been significantly improved. The v0.5 CO column product is retrieved from an optimized spectral fitting window and generated without the application of a scaling factor (the v0.4 product was scaled with the factor 0.5). In addition, a correction has been applied to reduce time dependent biases caused by the ice-issue (e.g., slit function change), to improve the retrieval for partially cloud covered scenes and to make the retrieval less sensitive to aerosols and albedo variability. The correction is based on the retrieved methane column obtained from the same fitting window. A correction factor, defined as the ratio of an assumed (a-priori) methane column (see below) and the retrieved methane column, is applied to the retrieved CO column. The a-priori methane column is computed using a single (scene independent) profile of the methane mixing ratio, of pressure, and of temperature, and is computed taking into account the surface elevation of the corresponding ground pixel. The assumed methane column is 3.6×10^{19} molecules/cm² for a ground pixel with a surface elevation equal to sea level. This approach makes use of the fact that the variability of the methane column is low (few percent) compared to the variability of the CO column (up to or even larger than 100%). A quality flag is set for each pixel to indicate a (potentially) successful measurement. To decide if a measurement is successful, a number of criteria have been

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defined based on the value of the root-mean-square of the fit residuum, the CO fit error, and the methane correction factor. The methane correction factor, for example, has to be close to 1.0 (within 20%) for a measurement to be classified as successful. Otherwise the disturbances due to clouds, aerosols, surface reflectivity, calibration issues, etc., are considered too large to be corrected for. Note that because of the correction procedure the quality flag does not depend on the cloud mask (i.e., the quality flag is independent of the pixel being classified as cloud contaminated or not). This is also a major difference to version 0.4 CO. Because of this the number of useful pixels for version 0.5 is about ten times larger than for version 0.4.

3.2. Methane specific aspects

For methane we derive dry air column averaged mixing ratios (denoted XCH_4) by normalizing the retrieved methane column by the simultaneously observed air mass estimated by retrieving the column of a reference gas whose column is less variable than methane. Initially (for v0.4) we used oxygen (O_2) columns retrieved from the O_2 A band (760 nm) (Buchwitz et al., 2005a,b). Unfortunately, the sensitivity of the retrieved O_2 column with respect to, e.g. aerosol, is quite different compared to methane mainly because of the large spectral distance between the two fitting windows. For v0.41 XCH_4 (Buchwitz et al., 2005b) we use CO_2 retrieved from the 1.6 μm region (channel 6). The new v0.5 XCH_4 data product is derived in a similar way as the v0.41 data product, except that methane is retrieved from channel 6 (1629–1671 nm) instead of channel 8. There are two reasons why methane retrieval from channel 6 should give better results compared to channel 8: (i) channel 6 is not affected by an ice-layer, and (ii) the channel 6 methane absorption band is located close to the channel 6 CO_2 band thus improving the cancellation of errors when the ratio is computed (see also Frankenberg et al., 2005c, who are using a similar approach for the same reasons). For each ground pixel a quality flag is set using similar criteria as for CO. As for CO the quality flag is independent of the pixels being classified as cloud contaminated or not. There is however one difference compared to CO: Because of the high accuracy and precision

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requirements for methane and the reduced quality of measurements over water, only pixels over land are classified useful.

3.3. Carbon dioxide specific aspects

The SCIAMACHY/WFM-DOAS version 0.4 XCO₂ retrieval algorithm is described in detail in [Buchwitz et al. \(2005a\)](#) and [Buchwitz et al. \(2005b\)](#). The results which are shown here have been obtained with WFM-DOAS v0.4, i.e. we have not yet implemented an improved algorithm such as v0.5 for CO and methane. In short, the CO₂ columns are 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 CO₂ and weak absorption features of water vapor. Similar as for methane, dry air column normalized CO₂ columns are derived, the dry air column averaged mixing ratios XCO₂. For CO₂ the air column is obtained from the simultaneously measured oxygen (O₂) column retrieved from the O₂ A-band spectral region.

One of the problems of version 0.4 as applied to year 2003 SCIAMACHY Level 1 version 4.0x spectra is a systematically underestimation of the CO₂ columns and an overestimation of the O₂ columns ([Buchwitz et al., 2005a,b](#)). To compensate for this, the initially retrieved CO₂ and O₂ columns are scaled with constant factors (1.27 for CO₂ and 0.85 for O₂). The factors have been chosen to make sure that the CO₂ (O₂) column is near its expected value of about 8×10^{21} molecules/cm² (4.5×10^{24} molecules/cm²) for a cloud-free scene with a surface elevation close to sea level and moderate surface albedo. Because of this scaling we focused in [Buchwitz et al. \(2005a\)](#) and [Buchwitz et al. \(2005b\)](#) on spatial and temporal variability and not on absolute values when comparing the SCIAMACHY XCO₂ with reference data, e.g., by comparing XCO₂ anomalies. In this paper we present first results of WFM-DOAS version 0.4 applied to SCIAMACHY version 5.04 Level 1 spectra of the years 2004 and 2005 which have an improved calibration. The resulting year 2004/2005 CO₂ columns have the right order of magnitude without the application of a scaling factor. Therefore, the year 2005/2005 results shown

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in this paper have been obtained without scaling the CO₂ columns.

One of the most important problems with WFM-DOAS v0.4 XCO₂ results from the fact that the sensitivity of the spectral nadir measurements with respect to aerosols, (partial) clouds, and surface reflectivity is quite different for CO₂ and O₂. This is due to the large spectral distance between the CO₂ band (1580 nm) and the O₂ band (760 nm) which introduces (average light path length related) errors on the retrieved CO₂ and O₂ columns (by up to several percent) which do not perfectly cancel when the column ratio (i.e., XCO₂) is computed (Buchwitz et al., 2005a) (see also Houweling et al., 2005 for SCIAMACHY CO₂ retrieval and van Diedenhofen et al., 2005 for SCIAMACHY O₂ retrieval). We currently estimate the XCO₂ error to about a few percent (~1–6% depending on conditions) with both random and systematic components (Buchwitz et al., 2005a,b). Our future work will focus on the development of a retrieval algorithm that is less sensitive to these and other types of error sources.

Because of the high precision and accuracy requirements for XCO₂ the contamination by clouds needs to be minimized as much as possible. In order to identify cloud-contaminated ground pixels we currently use a simple threshold algorithm based on sub-pixel information provided by the SCIAMACHY Polarization Measurement Devices (PMDs) (details are given in Buchwitz et al., 2005a). In short, 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 (despite the low sensitivity of PMD1 for surface reflectivity changes compared to PMDs 2–7; note that PMD1 has been selected because of its low sensitivity to surface properties). As a result, ice or snow covered surfaces may wrongly be classified as cloud contaminated. This also needs to be improved in future versions of our retrieval method.

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4. Trace gas results

So far we have processed all SCIAMACHY spectra made available by ESA for the year 2003 by WFM–DOAS (details concerning the processed orbits are given in Buchwitz et al., 2005b). The WFM–DOAS version 0.5 2003 data set will be discussed in the following for CO (Sect. 4.1) and XCH₄ (Sect. 4.2). In addition, for XCO₂ we have processed all orbits of the years 2004 and 2005 over the area of Park Falls, Wisconsin, USA. These data will be presented in Sect. 4.3.

4.1. Carbon monoxide (CO)

To assess the quality of the new SCIAMACHY/WFM–DOAS version 0.5 CO column data product we have compared it with operational version 3 CO columns from MOPITT/EOS-Terra (Deeter et al., 2003). Figure 1 shows a comparison of year 2003 averages of both independent data sets. The figures have been produced by independently averaging all available data products of both sensors, i.e., without implementing any specific measures to consider or correct for their different spatial and temporal sampling, horizontal resolution, and differences in their altitude sensitivity. This needs to be done in the future but is outside of the scope of the present study. The comparison is restricted to measurements over land because of the reduced quality of the SCIAMACHY measurements over oceans caused by the low albedo of water in the near-infrared which results in low signals and low signal-to-noise ratios. Overall, both data sets agree quite well but there are also significant differences. On average, the SCIAMACHY data are higher compared to MOPITT by 0.20×10^{18} molecules/cm² (+10.5%) but regionally the differences can be much larger (up to 80%) as shown by the panels on the right hand side. The reason for these differences is currently unclear and more investigations are needed, e.g., to find out to what extent this is due to the higher sensitivity of SCIAMACHY for boundary layer CO. Globally, the standard deviation of the difference is 0.48×10^{18} molecules/cm² (27.9%)

Figure 2 enables a comparison between the two data sets for different seasons.

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Shown are tri-monthly averages excluding and including the SCIAMACHY measurements over water. Especially over the oceans the differences to MOPITT are large, e.g., because of the low signal-to-noise ratios of the SCIAMACHY measurements over water (see above). Despite the noisy measurements over water Fig. 2 suggests that large regions of elevated CO over the oceans are observed with SCIAMACHY but with a pattern significantly different from the one observed by MOPITT. We are currently not in the position to judge in detail the quality of the SCIAMACHY CO measurements over water. This aspect needs to be assessed by a detailed comparison including CO model simulations. We also performed a detailed comparison of monthly means (not shown here) using the measurements over land and water. We found that the monthly mean inter-hemispheric CO concentration difference as measured by both sensors is in good agreement (mean difference within 3%, correlation coefficient $r=0.9$).

Focusing on the measurements over land, where the signal-to-noise ratios are typically significantly higher, Fig. 2 indicates that at least qualitatively SCIAMACHY and MOPITT show a similar time dependence of the CO over Africa which (mostly) originates from biomass burning during the dry season. Qualitatively, similar remarks also hold for South America. Quantitatively, however, there is large disagreement, with SCIAMACHY seeing higher columns throughout the year. Also this aspect needs further study. In this context one has to remember that for a number of reasons (e.g., different altitude sensitivity, spatial resolution, and measurement time) perfect agreement is not to be expected.

4.2. Methane (CH₄)

Figure 3 shows a comparison of the new SCIAMACHY/WFM–DOAS version 0.5 XCH₄ data product derived from channel 6 with independent TM5 model simulations performed at the EC Joint Research Centre (JRC), Ispra, Italy (Bergamaschi et al., 2000, 2005a). The TM5 model is a two-way nested atmospheric zoom model (Krol et al., 2005). It allows to define zoom regions (e.g. over Europe) which are run at higher spatial resolution (1 × 1 degrees), embedded into the global domain, run at a resolution

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of 6×4 degrees. 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. Methane (a priori) emissions are as described by Bergamaschi et al. (2005a). Chemical destruction of CH₄ by OH radicals is simulated using pre-calculated OH fields based on CBM-4 chemistry and optimized with methyl chloroform. For the stratosphere also the reaction of CH₄ with Cl and O(¹D) radicals are considered. The TM5 simulations are currently being refined (Bergamaschi et al., 2005b). For this study we used the same TM5 data set as used for comparison with our previous methane v0.4x products (Buchwitz et al., 2005b).

Figure 3 shows the methane mixing ratios for two orbits over Europe on 2 August 2003, for a synoptic situation where methane enriched air masses are transported over eastern Europe resulting in a slight but significant west-to-east methane gradient over Europe. For TM5 XCH₄ two types of airmasses can be identified, one with relatively high XCH₄ (>1750 ppbv) and one with relatively low XCH₄ (<1750 ppbv). The mean and standard deviation of the “low methane” airmass for TM5 (the corresponding SCIAMACHY values for pixels classified by TM5 XCH₄ are given in brackets) is 1728 ppbv (1737 ppbv) and 7 ppbv (32 ppbv), respectively. The mean and standard deviation of the “high methane” airmass for TM5 (SCIAMACHY) is 1770 ppbv (1797 ppbv) and 9 ppbv (38 ppbv), respectively. From this one can conclude that the upper limit (because of the model errors) of the precision of the SCIAMACHY measurement is about 2% (35 ppbv of 1750 ppbv). The mean XCH₄ difference between the two air masses is 42 ppbv (=1770–1728) according to TM5 and 60 ppbv (=1797–1737) according to SCIAMACHY. From these values an upper limit (because of the model errors) of the (relative) accuracy of the SCIAMACHY measurements can be estimated which is less than 1% (18 ppbv). The good agreement of the SCIAMACHY measurements with the TM5 simulations indicates the high precision and accuracy of the new SCIAMACHY methane data product. Note that we performed a similar comparison with our previous v0.4x methane data products derived from channel 8 showing significantly larger

deviations when compared to TM5.

Figure 4 shows a comparison for bi-monthly averages. On average, the SCIAMACHY measurements are somewhat lower than the TM5 XCH₄, especially at high northern latitudes. For the tropical region Fig. 4 is consistent with the findings of Frankenberg et al. (2005c) who also analyzed differences of SCIAMACHY data and model simulation but for the time period August to November 2003 and found strong indications for a missing methane source or a significant underestimation of the strength of a known source over tropical rain forests. Frankenberg et al. (2005c) used a different retrieval algorithm (IMAP–DOAS) and compared with a different model (TM3 model of KNMI). Figure 4 confirms these important findings and shows that they neither depend on the details of the retrieval method nor on the model used for comparison.

Figure 4 shows that under certain conditions the observed XCH₄ is significantly lower than the model XCH₄: over Antarctica values less than 1560 ppbv are measured and north of 20° N values below 1670 ppbv are found, in particular during January to March and around October (this follows from an analysis of monthly means not shown here). Recent investigations including an initial comparison with ground based FTS measurements (Bart Dils, BIRA-IASB, private communications) indicate that the error primarily depends on the solar zenith angle (SZA). A possible cause is a small calibration offset. The SZA dependent bias is roughly given by $BIAS=0.9+0.15\cos(SZA)$, i.e., dividing the retrieved methane columns by BIAS appears to be a good first order correction. For this paper, however, no correction has been applied as this issue needs further study. Excluding the low values mentioned above from the SCIAMACHY – TM5 comparison yields a mean difference of the global daily data of less than 2% for nearly all days of the year 2003 (nearly constant low bias of SCIAMACHY of about 1%). The standard deviation of the difference is in the range 1.5–2.5% and the correlation coefficient is typically larger than 0.6. Figure 5 shows a comparison of SCIAMACHY with TM5 when the low values of SCIAMACHY are excluded.

In order to show the rich level of detail provided by the SCIAMACHY methane measurements Fig. 6 displays detailed results for four regions (Europe, northern America,

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southern America, and south-east Asia) including a comparison with TM5. The figures show major surface source regions of methane due to fossil fuel related activities, wetlands, waste handling, ruminants, rice cultivation, etc. The comparison with the TM5 XCH₄ shows in general good agreement but there are also significant regional differences which are, at least partially, due to shortcomings of current methane emission data bases. Similar as for CO, our future work on methane will focus on using the SCIAMACHY data to better constrain our knowledge on regional methane sources. To which extent this is possible with the version 0.5 data product presented here still needs to be assessed.

4.3. Carbon dioxide (CO₂)

Detailed results such as global maps and time series of the SCIAMACHY/WFM–DOAS version 0.4 XCO₂ data product have been presented in [Buchwitz et al. \(2005b\)](#) including a comparison with global model simulations using TM3 ([Heimann and Körner, 2003](#)). Here we present a comparison with TM3 at Park Falls, Wisconsin, USA, which is one of the few ground-based FTS stations that measure XCO₂. A detailed comparison with the Park Falls FTS data will be performed once the FTS data will be released. 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 by diffusion. Available horizontal resolutions range from 8×10 degrees to 1.1×1.1 degrees. In this case, TM3 was run with a resolution of 1.8×1.8 degrees and 28 layers, and the meteorology fields were derived from the NCEP/DOE AMIP-II reanalysis. CO₂ 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 ([Thornton et al., 2002](#)) including a simple parameterization of the diurnal cycle in photosynthesis and respiration.

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A detailed comparison of SCIAMACHY and TM3 for the year 2003 has been presented in [Buchwitz et al. \(2005b\)](#) showing reasonable to good agreement after temporal and spatial averaging of the SCIAMACHY data except for the amplitude of the XCO₂ variability, which was found to be typically about two to five times larger for the SCIAMACHY measurements compared to TM3. Here we show a comparison of the same data sets but restricted to the Park Falls area. In addition to the year 2003 data (which had large gaps) we have recently processed all available year 2004 and 2005 SCIAMACHY orbits covering the Park Falls area. Still there are large gaps in the SCIAMACHY data mainly for two reasons: (i) the instrument is heated regularly for one or two weeks to get rid of the ice-layers on the near-infrared detectors, (ii) not all orbits have yet been made available by ESA for (to be solved technically) ground processing related reasons.

The processing of the SCIAMACHY year 2004/2005 spectra has been performed as described in [Buchwitz et al. \(2005a\)](#) and [Buchwitz et al. \(2005b\)](#) for 2003 but with one exception. For the years 2004 and 2005 operational Level 1 data products (spectra) have been made available by ESA which are significantly improved with respect to their calibration. Therefore, it was not necessary for the year 2004/2005 spectra to improve the calibration by “patching” the operational data products (i.e., by replacing the dark signals in the binary Level 1 data files by improved dark signals) as done for the year 2003 data ([Buchwitz et al., 2005a,b](#)). We have not yet investigated if this difference in data processing results in any inconsistencies between the year 2003 and the years 2004/2005 time series. For the comparison shown here this is only of minor importance because we focus on XCO₂ variability (anomalies). For the year 2003 time series the by far most computer time consuming step was the extraction of the spectra including the patching of the binary data products. Processing of the years 2004/2005 spectra was much faster because the binary files have not been patched. The final and most important step, namely the processing of the extracted spectra by WFM–DOAS, is very fast and last typically less than one minute per fitting window and per orbit (consisting of several thousand measurements) on a single-processor standard PC. This means

that WFM–DOAS (Level 1 to Level 2) processing is two orders of magnitude faster than real time (one ENVISAT orbit lasts about 100 min).

In order to demonstrate the good quality of the SCIAMACHY spectral measurements over Park Falls, Fig. 7 shows the result of a spectral WFM–DOAS fit for a measurement on 15 July 2004. The fit error of the CO₂ column for the single measurement is 2.3% and the root-mean-square (RMS) difference between the SCIAMACHY measurement and the fitted WFM–DOAS linearized radiative transfer model is 0.24%. Both numbers are (not yet) limited by the signal-to-noise performance of the instrument but dominated by rather stable spectral artifacts (i.e., the fit residuals of other ground pixels are very similar to the one shown in Fig. 7) resulting from, e.g., residual calibration errors, uncertainties of the spectroscopic parameters (Rothmann et al., 2003), or for other reasons.

In contrast to our version 0.5 CO and methane data products we have not yet implemented an automatic computation of a standard quality flag for the version 0.4 XCO₂ data products. For the Park Falls comparison we have defined a number of quality criteria in order to determine if a measurement can be classified successful. When investigating the new year 2004/2005 data we found that the quality of the spectral fits was slightly better compared to 2003 probably due to the improved calibration (for 2003 we used Level 1 spectra version 4.0x, for 2004/2004 version 5.04). Therefore, we use a stricter quality criterium for the XCO₂ fiterror for 2004/2005 compared to 2003. Due to the improved calibration of the year 2004/2005 the application of a scaling factor for the CO₂ columns is not needed anymore, i.e., the year 2004/2005 columns are not scaled. To make a consistent comparison over Park Falls we have removed all scaling factors, i.e., the comparison is based on unscaled XCO₂. The quality criteria for the year 2003–2005 data set are: (i) only cloud-free pixel over land, (ii) only forward scan pixel, (iii) RMS of fit residuum: <0.7%, (iv) XCO₂ fiterror <8% for 2003 (<4% for 2004/2005), (v) O₂ fiterror <1.8%, (vi) (unscaled) CO₂ column (in units of 8×10²¹ molecules/cm²) within 0.85–1.1 (also for 2003 but divided by 1.27), (vii) (unscaled) O₂ column (in units of 4.5×10²⁴ molecules/cm²) within 1.0–1.13. In addition,

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only pixels have been selected whose center coordinates have a distance of less than 350 km from Park Falls.

Figure 8 shows the comparison of the SCIAMACHY and TM3 XCO₂ data sets. As can be seen, the peak-to-peak XCO₂ variability (seasonal cycle of the year 2003–2005) as measured by SCIAMACHY is about 13 ppmv. This is a factor of two to three larger than the year 2003 peak-to-peak XCO₂ variability of TM3. A similar disagreement between SCIAMACHY and TM3 has also been found in previous studies (Buchwitz et al., 2005a,b). Based on initial (preliminary) analysis of the Park Falls FTS data a peak-to-peak XCO₂ variability of 13 ppmv has been reported (Washenfelder et al., 2005). More studies are needed to understand the differences between the TM3 and the SCIAMACHY XCO₂ measurements. We hope that this issue can be clarified in the near future by a detailed comparison with XCO₂ ground-based FTS measurements at Park Falls (after official data release) and other ground stations.

5. Carbon gases over China

Air pollution resulting from large-scale fossil fuel combustion and fossil fuel related activities has become a problem with increasing importance, especially for countries with an increasing energy demand and inherent fuel consumption such as China. The quantification of concentrations near the sources and the subsequent transport of pollutants is important, for example, for monitoring and forecasting of air pollution.

The left panel of Fig. 9 shows yearly averaged vertical columns of CO over China. Clearly visible are large regions of elevated CO (shown in red) indicating CO source regions. Elevated CO is present over a large area south of Beijing, in the region around Chengdu/Chongqing, around Shenyang, and over Hainan Dao and Zhanjiang. The elevated CO detected with SCIAMACHY clearly correlates with major industrialized areas. The three panels on the right hand side of Fig. 9 show tri-monthly averaged CO columns for (from top to bottom) February–April, May–July, and August–October 2003. The largest CO columns are observed in the May to July 2003 average over

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large parts of eastern China and around Chongqing/Chengdu. High columns are also observed over these regions during other times of the year, especially around Beijing during February to April. Our future work will focus on a more detailed analysis of this interesting region with the goal to obtain as much quantitative information as possible on the strength of the surface sources of CO. If the quality of the WFM–DOAS version 0.5 CO data product is high enough for such a challenging application still needs to be assessed.

China emits significant amounts of methane, e.g., due rice cultivation, ruminants, and wetlands. We have processed all SCIAMACHY measurements available for the year 2003. The left panel of Fig. 10 shows the dry air column averaged mixing ratio of methane over China during 2003. Shown in Fig. 10 are only those measurements for which the quality flag determined by the retrieval algorithm indicates a (potentially) successful measurement. Using this approach the measurements over the Himalaya region are filtered out. Clearly visible are large regions of elevated methane (shown in red) indicating methane source regions over large parts of south-east China (as well as over India, south-east Asia, and Japan). The panels on the right hand side show tri-monthly averages of the year 2003 methane data set. During February to April 2003 (top panel) elevated concentrations are present around Chongqing/Chengdu (see annotation of Fig. 9, left panel) and along the Pacific ocean coast line around and south of Shanghai, over Hainan Dao and north of Hainan Dao. During May to July (middle panel) the methane concentration is significantly higher compared to February–April over large parts of south-east China. The highest mixing ratios occur over the Chongqing/Chengdu area. Even larger methane concentrations are visible in the August to October average (bottom panel) were highly elevated mixing ratios are observed over large parts of eastern China (as well as over India and south-east Asia) broadly in line with the seasonality of methane emissions from rice paddies.

Figure 11 shows WFM–DOAS version 0.4 XCO₂ over China including seasonal variation. The regional SCIAMACHY XCO₂ measurements are significantly more difficult to interpret compared to the SCIAMACHY CO and methane measurements because

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the estimated measurement error of a few percent is on the order of the expected XCO₂ variability. The year 2003 average (left panel) shows maximum values over north-east China and India. The tri-monthly averages (right hand side panels) show high values over north-east China during February to April and August to October but significantly lower values over this region in the May to July average. Over India XCO₂ is high during February to July but significantly lower during August to October. The overall seasonal dependence over this region, especially at the higher latitudes, is broadly as expected from the seasonality of carbon dioxide uptake and release by vegetation, i.e., higher values in winter/early spring and late autumn/winter and lowest values during the growing season late spring/summer.

6. Conclusions

We have presented an improved algorithm (WFM–DOAS version 0.5) for the retrieval of vertical columns of carbon monoxide (CO) and dry-air column averaged mixing ratios of methane (XCH₄) from the spectral near-infrared nadir observations of the SCIAMACHY instrument onboard the European environmental satellite ENVISAT (launch March 2002). An initial comparison with global reference data (MOPITT satellite data for CO, TM5 model simulations for XCH₄) is presented giving clear indications for a significant improvement of the CO and methane data products compared to earlier versions (0.4, 0.41). Concerning carbon dioxide we have presented a comparison of SCIAMACHY measured dry-air column averaged mixing ratios XCO₂ – obtained with WFM–DOAS version 0.4 – over Park Falls, Wisconsin, USA, with TM3 model simulations. The peak-to-peak XCO₂ variability as measured by SCIAMACHY is about 13 ppmv broadly in line with preliminary analysis of ground-based May 2004–June 2005 FTS XCO₂ measurements. This is a factor of two to three larger than the year 2003 peak-to-peak XCO₂ variability of TM3. Similar disagreement between SCIAMACHY and TM3 has also been found in previous studies. We are confident that this issue can be resolved in the near future by a detailed comparison with XCO₂

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ground-based FTS measurements at Park Falls (after official data release) and other ground stations. We presented measurements of all three carbon gases over China including seasonal variations. These measurements enable the identification of source regions of carbon monoxide which are correlated with major industrialized areas and source regions of methane due to, e.g., methane emissions from rice paddies and wetlands. More studies are needed to understand the remaining differences between the SCIAMACHY CO, XCH₄, and XCO₂ measurements and the reference data used for comparison in order to fully assess the information content and quality of the SCIAMACHY carbon gas data products. Our primary goal is to generate high quality data products that can provide qualitative and as much as possible quantitative information on the surface sources and sinks of these major carbon gases via inverse modeling.

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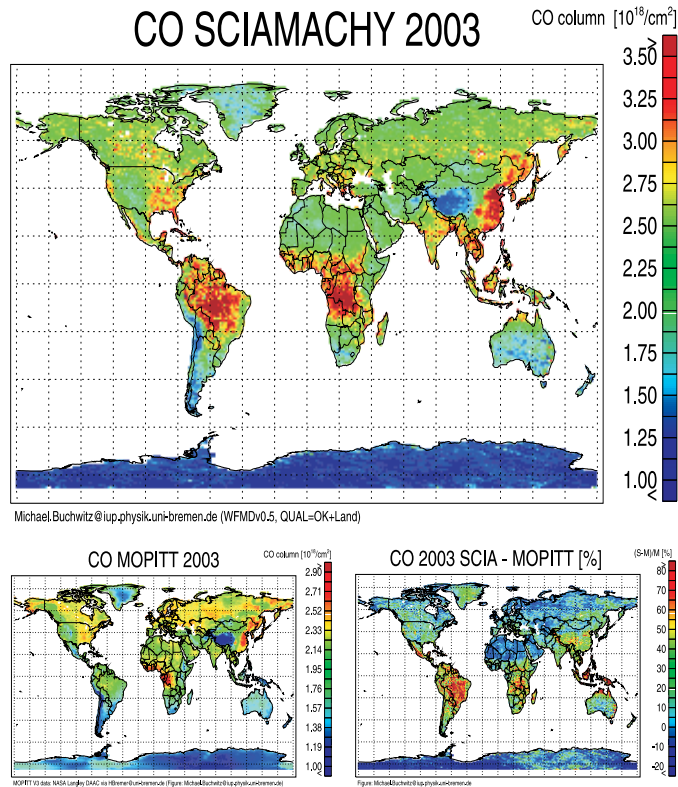


Fig. 1. Year 2003 average of SCIAMACHY/WFM–DOAS version 0.5 CO columns (top) compared to MOPITT operational CO columns (Lv2V3) over land (bottom left). Note that the scale is different for both figures because the SCIAMACHY CO columns are typically higher than the CO columns measured by MOPITT. The relative CO column differences (SCIAMACHY – MOPITT) are shown in the bottom right panel.

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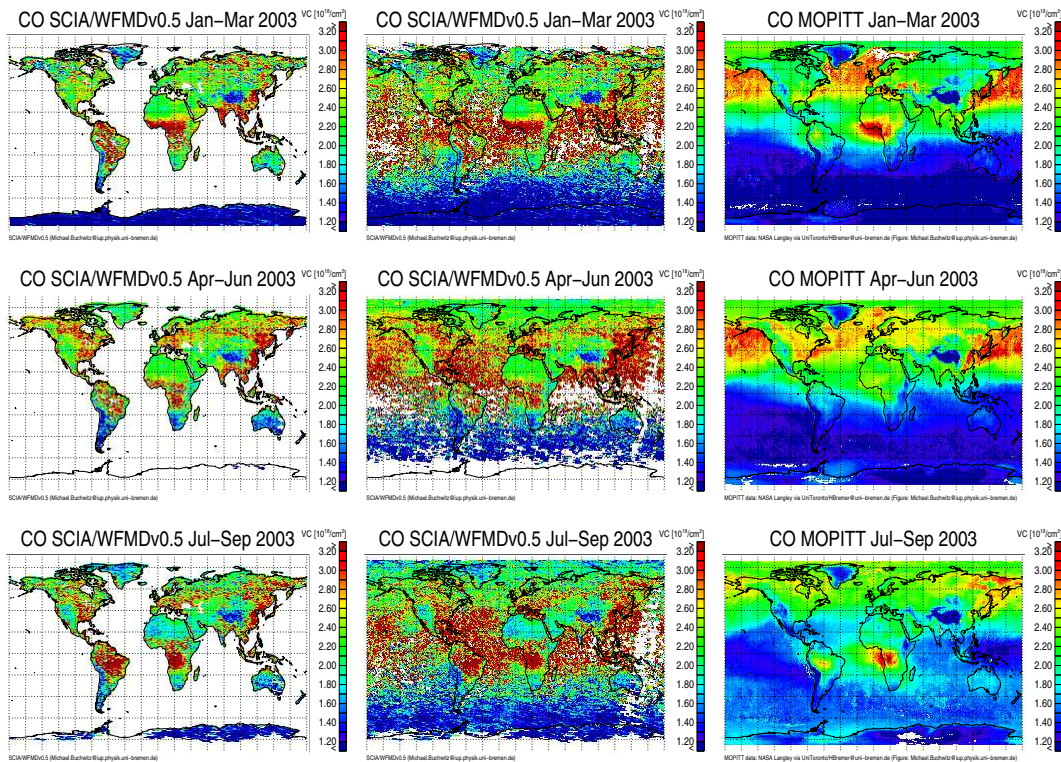


Fig. 2. Comparison of year 2003 tri-monthly averages of SCIAMACHY/WFM–DOAS version 0.5 CO (left: over land, middle: over land and water) with MOPITT (right).

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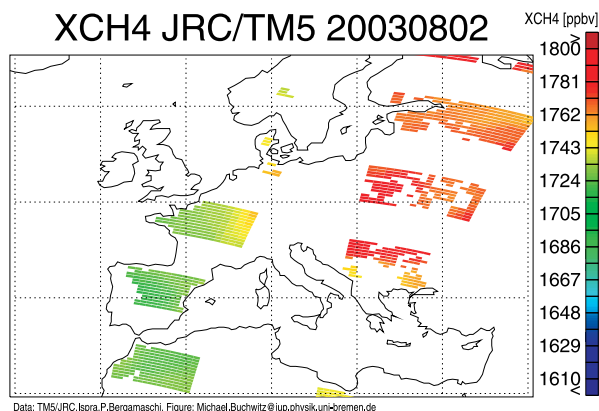
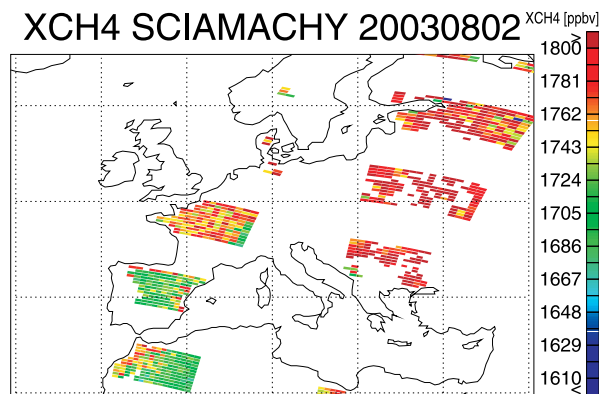


Fig. 3. Single orbit (non-averaged) SCIAMACHY/WFM–DOAS version 0.5 methane over Europe (top) compared with independent JRC/TM5 model simulations (bottom) sampled at the location of the SCIAMACHY ground pixels. The regular gaps in between the nadir measurements are due to the SCIAMACHY limb observations. Only those ground pixels are shown for which the WFM–DOAS v0.5 quality flag, which is part of the data product, indicates a potentially successful measurement.

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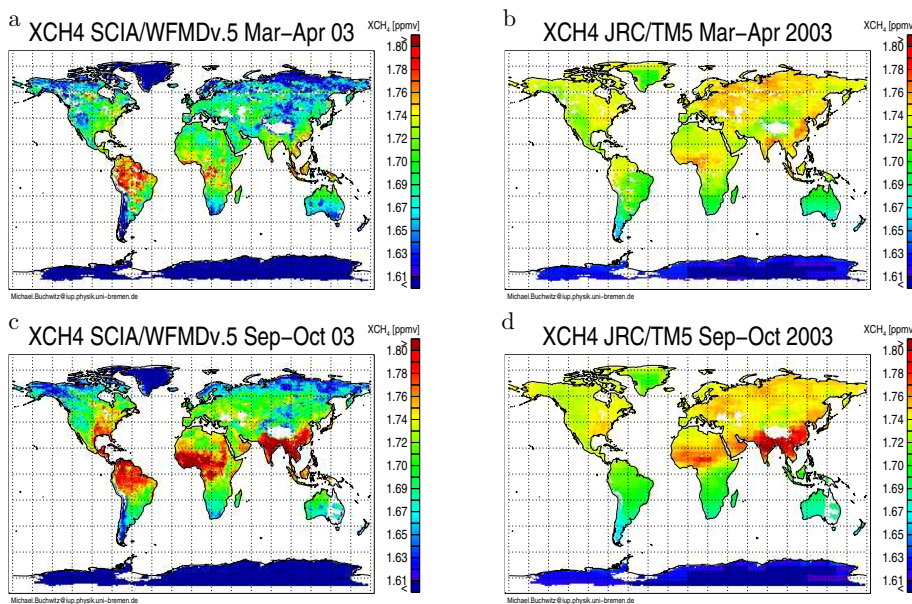


Fig. 4. Comparison of bi-monthly year 2003 averages of SCIAMACHY/WFM–DOAS version 0.5 methane (left) with JRC/TM5 model simulations (right).

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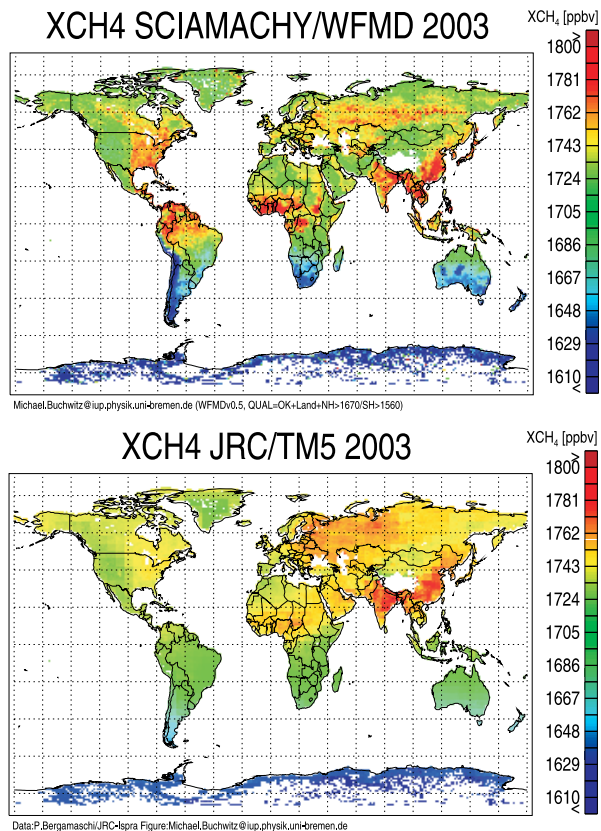


Fig. 5. Year 2003 average of SCIAMACHY/WFMD–DOAS version 0.5 methane (top) with JRC/TM5 model simulations (bottom). The low values of SCIAMACHY observed over Antarctica and high northern latitudes have been filtered out (see main text for details).

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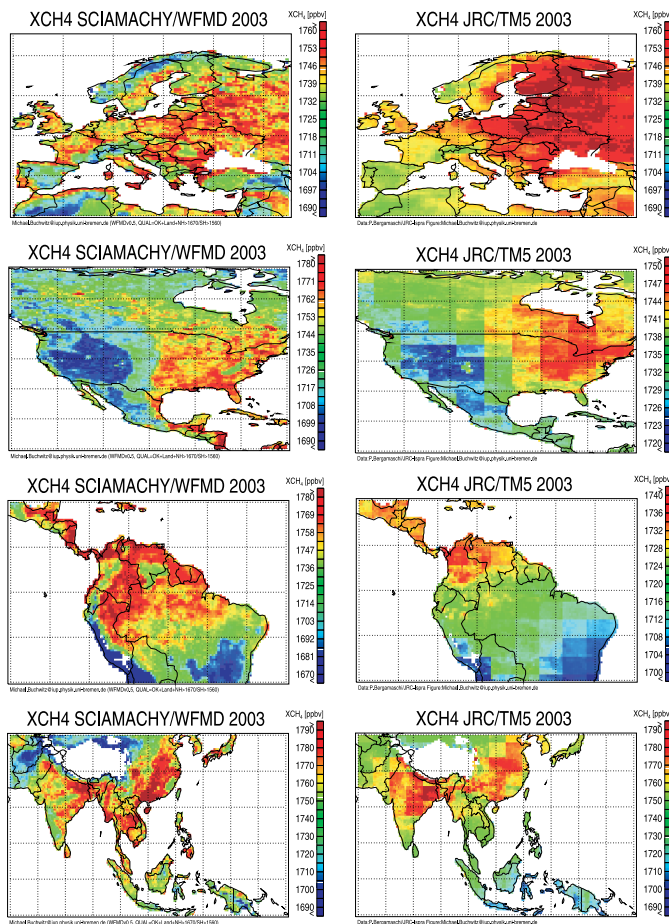


Fig. 6. Comparison of SCIAMACHY/WFM–DOAS version 0.5 XCH₄ (left) with JRC/TM5 model simulations (right) for various regions using the TM5 high resolution zoom mode over Europe and low resolution mode outside Europe.

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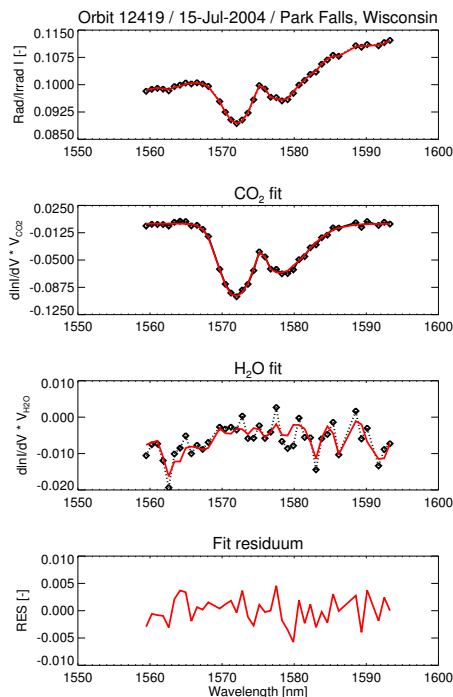


Fig. 7. SCIAMACHY/WFM-DOAS version 0.4 CO₂ spectral fit for Park Falls, Wisconsin, USA, 15 July 2004. The top panel shows the measured sun-normalized radiance (black diamonds) and the WFM-DOAS modeled radiance after the fit (red line). The second and third panels show separately the spectral CO₂ and H₂O fits (red: scaled weighting functions (trace gas derivatives), black symbols: as red curves but with fit residuum (i.e., difference between measurement and model after the fit) added). The bottom panel shows the fit residuum, i.e., the difference between the WFM-DOAS model and SCIAMACHY measurement after the fit. The (unscaled) WFM-DOAS version 0.4 retrieved CO₂ column is 6.65×10^{21} molecules/cm² $\pm 2.3\%$. The root-mean-square (RMS) of the fit residuum is 0.24%.

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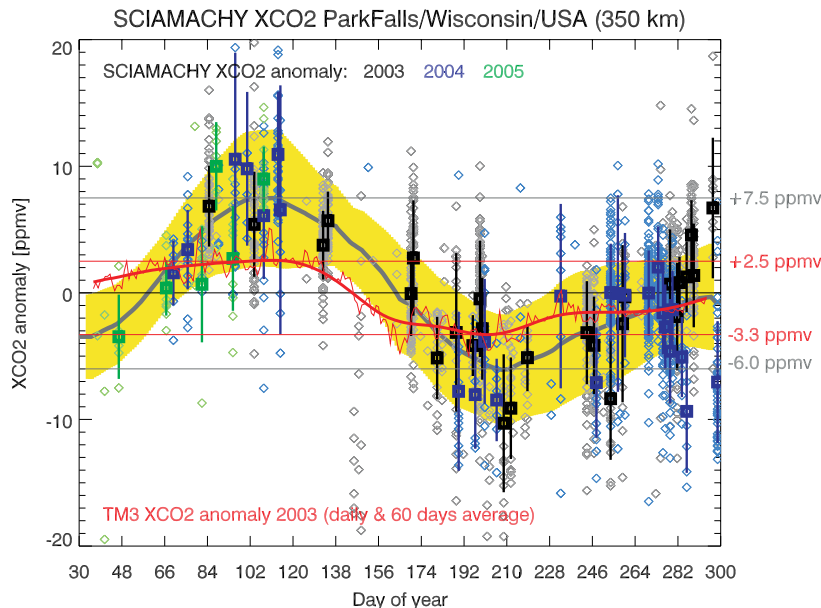


Fig. 8. Comparison of SCIAMACHY/WFM-DOAS XCO₂ time series anomalies with MPI-BGC/TM3 model simulations for Park Falls, Wisconsin, USA. The SCIAMACHY XCO₂ is shown for individual ground pixels as diamonds (year 2003: grey, 2004: blue, 2005: green). The mean values and standard deviations of the daily overpass data (single orbits) are shown as squares and vertical lines, respectively (only computed if 9 or more measurements per day were available within a distance of 350 km around Park Falls; 2003: black, 2004: blue, 2005: green). The SCIAMACHY XCO₂ 60 days running mean and standard deviation is shown as thick grey line and yellow shaded area, respectively. The red curves show the XCO₂ of the MPI-BGC/TM3 model (daily data (thin line) and 60 days average (thick line)). The grey horizontal lines mark the peak-to-peak variation of SCIAMACHY XCO₂ which is 13.5 ppmv. The red horizontal lines mark the peak-to-peak variation of TM3 XCO₂ which is 5.8 ppmv. Note that not all ENVISAT orbits were available for this study resulting in large data gaps, especially at the beginning and the end of each year and for April to June 2004/2005.

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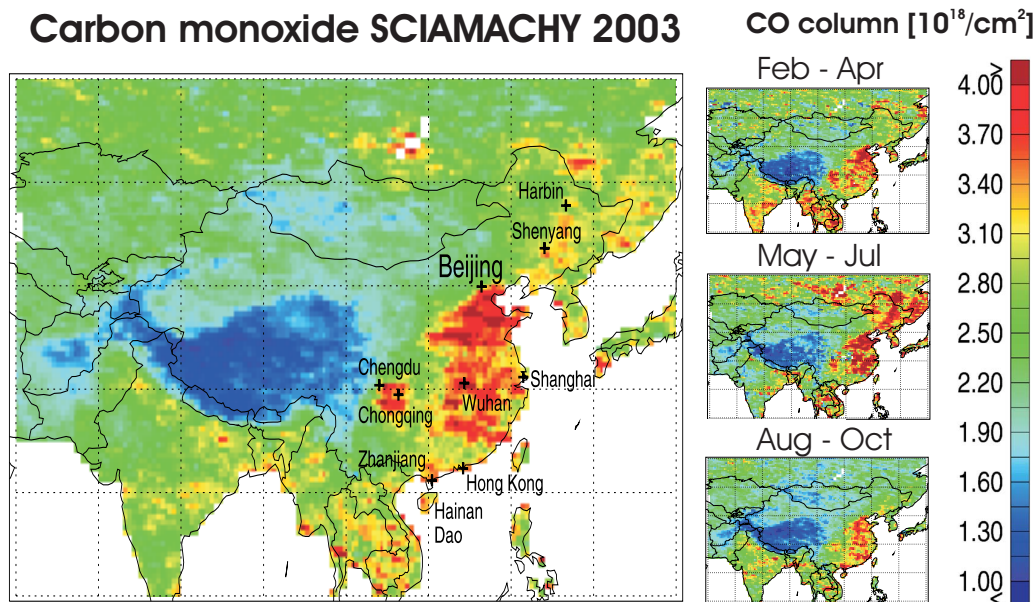


Fig. 9. Year 2003 SCIAMACHY/WFM-DOAS version 0.5 CO over China including seasonal variation (right). All CO measurements over land are shown for which the WFM-DOAS quality flag indicates a (potentially) successful measurement (see main text for details).

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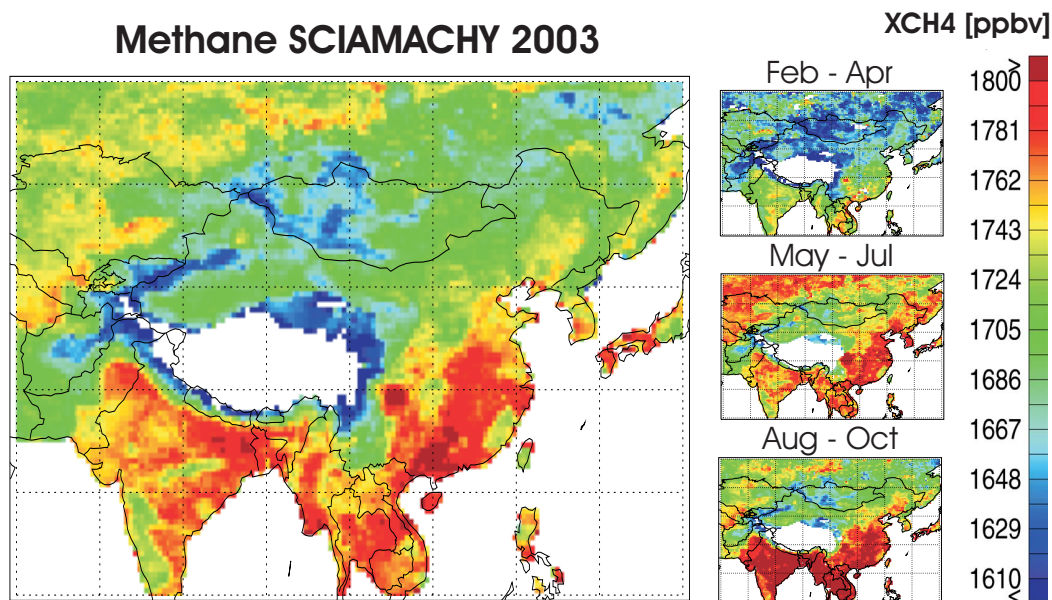


Fig. 10. As Fig. 9 but for SCIAMACHY/WFM–DOAS version 0.5 methane.

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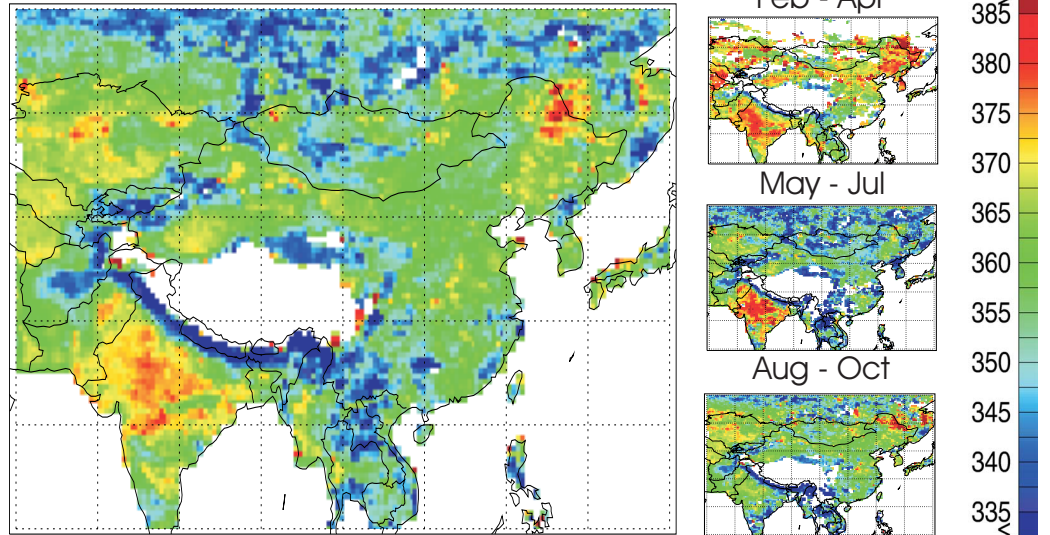


Fig. 11. As Fig. 9 but for SCIAMACHY/WFM–DOAS version 0.4 carbon dioxide.