# IMPROVED CARBON DIOXIDE AND METHANE RETRIEVED FROM SCIAMACHY ONBOARD ENVISAT: VALIDATION AND APPLICATIONS

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## ABSTRACT

Improved global data sets of atmospheric carbon dioxide and methane column-averaged dry air mole fractions - which are the quantities needed for inverse modelling to get information on the sources and sinks - retrieved from SCIAMACHY nadir observations are presented upgrading pre-existing greenhouse gas information derived from European EO data. The multi-year data sets are validated with ground-based Fourier Transform Spectrometer (FTS) measurements at Total Carbon Column Observing Network (TCCON) sites providing realistic error estimates of the satellite data, which is a prerequisite to assess the suitability to be used in inverse modelling. The validated greenhouse gas data sets are then analysed in terms of geophysical applications. The presented applications include an analysis of the atmospheric greenhouse gas variability on a spatial and temporal basis caused by biogenic and anthropogenic processes.

## 1. INTRODUCTION

Carbon dioxide  $(CO_2)$  and methane  $(CH_4)$  are the two most important anthropogenic greenhouse gases contributing to global climate change. The atmospheric abundance of both gases has increased significantly since the start of the Industrial Revolution. While the carbon dioxide concentrations have risen steadily during the last decades, the atmospheric methane increase temporarily paused from 1999 to 2006 [10, 4] before a renewed growth was observed from surface measurements since 2007 [20, 11].

Despite their importance, there are still many gaps in our understanding of the sources and sinks of these greenhouse gases and their biogeochemical feedbacks and response in a changing climate (see, e.g., [23]). Theoretical studies have shown that satellite measurements combined with inverse modelling can significantly reduce surface flux uncertainties, if the satellite data are accurate and precise enough [19, 13, 17, 8]. The significant reduction of regional-scale flux uncertainties additionally requires high sensitivity to the lowest atmospheric layers where the variability is largest. Sensitivity to all altitude levels, including the boundary layer, can be achieved by using reflected solar radiation in the near-infrared/shortwaveinfrared (NIR/SWIR) spectral region. SCIAMACHY onboard ENVISAT (launched in 2002, end of mission declared in 2012) [7, 5] was the first and is with TANSO onboard GOSAT (launched in 2009) [25] one of only two satellite instruments yielding measurements of the relevant absorption bands of both gases in this spectral range. OCO-2 (originally scheduled to be launched in 2013 but temporarily put on hold due to re-evaluation of launch vehicle options) [9, 3] will be another satellite designed to observe atmospheric carbon dioxide in the same spectral region as SCIAMACHY and TANSO. CarbonSat [6], which is one of two candidate Earth Explorer Opportunity Missions (EE-8, to be launched in 2018), and the CarbonSat Constellation shall also measure XCO<sub>2</sub> and XCH<sub>4</sub> in this spectral range. Hence, SCIAMACHY is playing a pioneering role in the relatively new area of greenhouse gas NIR/SWIR retrievals from space and is essential to initiate consistent long-term time series of carbon dioxide and methane column-averaged dry air mole fractions retrieved from satellite measurements.

The WFM-DOAS (Weighting Function Modified Differential Optical Absorption Spectroscopy) XCO<sub>2</sub> and XCH<sub>4</sub> data sets, which have been described in the peerreviewed literature [21, 22], were further improved: The carbon dioxide data set was updated using an improved cloud filtering and correction method using the strong water vapour absorption at 1.4  $\mu$ m and the O<sub>2</sub> A-band [12], as well as a bias-correction based on multivariate linear regression. For methane a correction based on simultaneously retrieved water vapour (1.56  $\mu$ m) to account for spectroscopic interferences was added. These improved data sets are the basis of the analysis presented here.

## 2. RESULTS

All available SCIAMACHY Level 1b version 6 spectra for the years 2003–2009 have been processed using the improved retrieval algorithm WFM-DOAS. The respective carbon dioxide and methane results are validated and discussed separately in the following subsections.

	XCO <sub>2</sub> [ppm]	XCH <sub>4</sub> [ppb]
Global Offset	0.3	-10
<b>Regional Precision</b>	1.3	16
Relative Accuracy	0.8	12

Table 1. Summary of the WFM-DOAS validation results. The global offset is the average of the mean differences to TCCON over all sites, the regional precision is the mean standard deviation of the differences, and the relative accuracy is the standard deviation of the mean differences (for details, see [22]).



Figure 1. Overview of the long-term global carbon dioxide data set; shown are column-averaged dry air mole fractions as a function of latitude and time.

#### 2.1. Validation

The improved long-term global carbon dioxide and methane column-averaged dry air mole fraction data sets from SCIAMACHY derived using WFM-DOAS are validated with ground-based Fourier Transform Spectrometer (FTS) measurements from the Total Carbon Column Observing Network (TCCON) [24] recording direct solar spectra in the near-infrared/shortwave-infrared spectral region. The intercomparison is performed at the following 8 TCCON ground sites: Białystok (Poland), Bremen (Germany), Orléans (France), Garmisch (Germany), Park Falls (USA), Lamont (USA), Darwin (Australia), and Wollongong (Australia).

The updated validation results using the comparison method described in detail in [22] provide the realistic error estimates of the satellite data summarised in Tab. 1 demonstrating the potential value in regions where there is sparse sampling by surface flask measurements.

#### 2.2. Carbon dioxide

An overview of the long-term global carbon dioxide data set is shown in Fig. 1. In addition to the pronounced seasonal cycle due to growing and decaying vegetation, the



Figure 2. Comparison of the SCIAMACHY (black) and CarbonTracker (red)  $XCO_2$  for the Northern Hemisphere (top) and the Southern Hemisphere (bottom) based on monthly means (coloured circles). The saturated solid lines have been smoothed using a four-month Hann window (which has a similar frequency response to a twomonth boxcar filter but better attenuation of high frequencies). The pale solid lines represent the corresponding deseasonalised trends. Shown below are the derivatives of these deseasonalised curves. Noted are yearly mean values of the derivatives (in pale colours) as well as the mean values of the whole time period.

steady increase of atmospheric carbon dioxide primarily caused by the burning of fossil fuels can be clearly observed.

To examine the increase with time and the seasonal cycle more quantitatively the SCIAMACHY results are compared to the CarbonTracker release 2010 assimilation system [18] based on monthly data (see Fig. 2). The CarbonTracker XCO<sub>2</sub> fields as used for this study have been sampled in space and time as the SCIAMACHY satellite instrument measures. The SCIAMACHY altitude sensitivity has been taken into account by applying the SCIA-MACHY column averaging kernels to the CarbonTracker  $CO_2$  vertical profiles. The retrieved continuous increase with time is consistent with CarbonTracker. The analysis of global and hemispheric averages demonstrates that the annual mean increase agrees with the assimilation system within the error bars amounting to about 1.8 ppm/yr, respectively.



Figure 3. Comparison of SCIAMACHY (black) and CarbonTracker (red) XCO<sub>2</sub> seasonal cycle amplitudes and annual increases.

We also find good agreement of the retrieved phase of the carbon dioxide seasonal cycle with the model resulting in a pronounced correlation of the two data sets (r=0.98and r=0.99 for the two hemispheres, respectively). There is also no significant difference of the southern hemispheric seasonal cycle amplitudes. However, the amplitude of the SCIAMACHY northern hemispheric seasonal cycle is about 40% larger than for CarbonTracker. This is qualitatively consistent with the recent finding that a boreal NEE (Net Ecosystem Exchange) enhancement in the CASA (Carnegie-Ames Stanford Approach) biogeochemical model improves the comparison with TCCON [14, 16] indicating that the actual NEE is larger than predicted by the CASA model, which is also used in CarbonTracker. The global and hemispheric mean values of the seasonal cycle amplitude and annual increase, as well as the corresponding errors, are depicted in Fig. 3.

Another related aspect analysed is the boreal forest carbon uptake during the growing season and its local partitioning between North America and Eurasia. To this end, longitudinal gradients of atmospheric carbon dioxide are studied during May-August (the period between the maximum and minimum of the seasonal cycle), which are the basic signals to infer regional fluxes, using a region consisting of equally sized slices in North America and Eurasia covering the bulk of the boreal forest area of the planet. When an air parcel flows over the boreal forest, more and more carbon is steadily taken up by the growing vegetation leading to a gradient parallel to wind direction with smaller values at the endpoint compared to the starting point. Due to the fact that the prevailing wind direction in mid- to high-latitudes is from west to east, one would expect a negative west-to-east longitudinal gradient for the considered region because the air masses are mainly moving according to this wind direction over the uptake region.

The gradients are derived by calculating meridional averages of seasonally averaged (May–August) SCIA-MACHY and CarbonTracker  $XCO_2$  as a function of longitude (in 1° bins) and linear fitting the corresponding west-to-east gradient weighted according to the standard deviations of the meridional averages. The associated er-



Figure 4. Annual west-to-east longitudinal XCO<sub>2</sub> gradients from SCIAMACHY (black) and CarbonTracker (red) for boreal forests during the growing season. The examined boreal forest region is composed of two equally sized regions in North America and Eurasia. The gradients and associated errors are illustrated for the overall region and the North American and Eurasian slice separately.

ror is derived from the square root of the covariance of the linear fit parameter. This investigation of the boreal forest carbon uptake during the growing season shows good agreement between SCIAMACHY and CarbonTracker concerning the annual variations of the gradients (see Fig. 4). While there is also very good quantitative agreement of the gradients for the overall region, there are systematic differences if both slices are analysed separately suggesting stronger American and weaker Eurasian uptake. However, these differences are not significant because both data sets agree within their error bars.

Another analysis on the regional scale addresses the retrieved  $XCO_2$  enhancement over anthropogenic source regions. As can be seen in Fig. 5, the SCIAMACHY  $XCO_2$  correlates reasonably well with anthropogenic emissions from the Emission Database for Global Atmospheric Research (EDGAR), release version 4.2, with significantly elevated values, e.g., in the Rhine-Ruhr metropolitan region, the Benelux, the East Coast of the United States, the Greater Tokyo Area, the Bohai Economic Rim, and the Yangtze River Delta. However, an exact agreement cannot be expected due to atmospheric



Figure 5. SCIAMACHY XCO<sub>2</sub> over selected anthropogenic source regions during 2003–2009 compared to anthropogenic CO<sub>2</sub> emissions (EDGAR v4.2) on a longitude-latitude grid with  $0.5^{\circ} \times 0.5^{\circ}$  resolution.

transport and the long atmospheric lifetime of CO<sub>2</sub>. A rigorous analysis of anthropogenic emissions would require the usage of a transport model. Nevertheless, the locally elevated  $XCO_2$  indicates that regional anthropogenic carbon dioxide emissions can potentially be detected from space.

## 2.3. Methane

An overview of the long-term global methane data set is shown in Fig. 6. Clearly visible is the interhemispheric gradient with higher atmospheric methane abundance on the Northern Hemisphere and the global renewed methane growth in recent years following a period with rather stable atmospheric methane mole fractions. Besides the interhemispheric gradient, Fig. 7 additionally shows the global spatial methane pattern from which major methane source regions, like the Sichuan Basin in China, which is famous for rice cultivation and gas production, or Siberian wetlands, can be identified.

To examine the renewed methane growth in recent years more quantitatively, Fig. 8 shows the temporal evolution of retrieved SCIAMACHY methane based on monthly means as well as the corresponding deseasonalised trend and its derivative analogue to the figure for carbon dioxide shown before. Also shown is the TM5-4DVAR model [15, 1, 2] being sampled in space and time as SCIA-MACHY measures and optimised by assimilating highly accurate NOAA surface measurements. The anomaly



Figure 6. Overview of the long-term global methane data set; shown are column-averaged dry air mole fractions as a function of latitude and time.



Figure 7. Seven years mean (2003–2009) of retrieved SCIAMACHY methane. Clearly visible are major methane source regions like the Sichuan Basin in China and the interhemispheric gradient.

since 2007 is derived from the difference of the mean values of the derivative of the deseasonalised trend after and before middle of 2006. To avoid a potential distortion of the analysed curves caused by the alteration of the detector pixel mask in November 2005, which was necessary due to detector degradation in the spectral range used for the methane column retrieval, all values during the period of plus or minus 6 months from this date are not considered in the calculation of the mean value before 2007. The global anomalies of SCIAMACHY and TM5-4DVAR are in good agreement amounting to about 7 ppb/yr, respectively.

This demonstrates that the retrieved recent renewed methane increase is consistent with surface observations and corresponding model simulations. Possible drivers of this increase are positive anomalies of arctic temperatures and tropical precipitation [11]. These potential causes are consistent with the retrieved anomalies for different latitude bands (see Fig. 9) showing that the largest increase is retrieved for the tropics and northern mid- and highlatitudes.



Figure 8. As Figure 2 but for SCIAMACHY and TM5-4DVAR XCH<sub>4</sub> and global monthly averages. The anomaly (numbers on the right) is defined as the difference of the mean values of the derivative of the deseasonalised trend after and before middle of 2006. Values one year around the detector pixel mask alteration in November 2005 (due to detector degradation) are not considered because potential systematic offsets might be distorting the deseasonalised trend and its derivative.



Figure 9. The derivative of the deseasonalised trend and the corresponding anomaly as a function of latitude.

## 3. SUMMARY

The improved long-term (2003–2009) global data sets of atmospheric carbon dioxide and methane columnaveraged dry air mole fractions retrieved from the spectral near-infrared/shortwave-infrared nadir observations of the SCIAMACHY instrument onboard the European environmental satellite ENVISAT using the scientific retrieval algorithm WFM-DOAS were validated at 8 TCCON sites and analysed in terms of geophysical applications.

The analysis of the carbon dioxide data set has shown that the retrieved annual mean increases for different latitude bands and the southern hemispheric seasonal cycle amplitude are consistent with the assimilation system CarbonTracker. However, the retrieved amplitude of the northern hemispheric seasonal cycle is about 40% larger than for CarbonTracker supporting the indication that NEE is actually larger than predicted by the CASA biosphere model. The investigation of the longitudinal gradients for boreal forests during the growing season being valuable to infer regional fluxes shows good agreement between SCIAMACHY and CarbonTracker concerning the overall magnitude of the gradients and their interannual variations. The systematic differences to Carbon-Tracker if the North American and Eurasian slices are analysed separately suggesting stronger American and weaker Eurasian uptake are not significant because both data sets agree within their error bars. Elevated XCO<sub>2</sub> over several anthropogenic Source regions indicates that regional anthropogenic CO<sub>2</sub> emissions can potentially be detected from space.

The global methane data set clearly demonstrates the detection of major source regions. The observed recent renewed increase is consistent with surface observations and model simulations. The largest increase is retrieved for the tropics and northern mid- and highlatitudes, which is in line with the assumption that possible drivers of this increase are positive anomalies of arctic temperatures and tropical precipitation.

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