GEOPHYSICAL RESEARCH LETTERS, VOL. 35, L15811, doi:10.1029/2008GL034300, 2008

Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT

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Received 18 April 2008; revised 22 May 2008; accepted 26 June 2008; published 12 August 2008.

[1] Methane retrievals from near-infrared spectra recorded by the SCIAMACHY instrument onboard ENVISAT hitherto suggested unexpectedly large tropical emissions. Even though recent studies confirm substantial tropical emissions, there were indications for an unresolved error in the satellite retrievals. Here we identify a retrieval error related to inaccuracies in water vapor spectroscopic parameters, causing a substantial overestimation of methane correlated with high water vapor abundances. We report on the overall implications of an update in water spectroscopy on methane retrievals with special focus on the tropics where the impact is largest. The new retrievals are applied in a four-dimensional variational (4D-VAR) data assimilation system to derive a first estimate of the impact on tropical CH_4 sources. Compared to inversions based on previous SCIAMACHY retrievals, annual tropical emission estimates are reduced from 260 to about 201 Tg CH_4 but still remain higher than previously anticipated. **Citation:** Frankenberg, C., P. Bergamaschi, A. Butz, S. Houweling, J. F. Meirink, J. Notholt, A. K. Petersen, H. Schrijver, T. Warneke, and I. Aben (2008), Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT, *Geophys. Res. Lett.*, *35*, L15811, doi:10.1029/2008GL034300.

1. Introduction

[2] Methane (CH_4) is, after carbon dioxide, the second most important anthropogenic greenhouse gas, directly contributing 0.48 W/m^2 to the total anthropogenic radiative forcing of 2.63 W/m^2 by well-mixed greenhouse gases [*Intergovernmental Panel on Climate Change*, 2007].

[3] According to established knowledge, methane is, apart from fossil fuel and biomass burning, primarily produced by strictly anaerobic methanogen microbes occurring in wetlands, rice paddies, landfills and the digestive tracts of ruminants. *Frankenberg et al.* [2005a] found significantly enhanced tropical methane abundances by analyzing near infrared spectra recorded by the SCIAMACHY instrument onboard ENVISAT. *Buchwitz et al.* [2006] confirmed this finding using a different retrieval algorithm. Source inversion studies based on SCIAMACHY retrievals [*Frankenberg et al.*, 2006] indicate significantly larger tropical emissions than estimated by inversions using surface observations only [*Bergamaschi et al.*, 2007; *Meirink et al.*, 2007b], even though model fields of the latter (i.e. based on the surface observations only) were found to be consistent with ship-borne CH_4 measurements over the tropical Atlantic ocean [*Warneke et al.*, 2006]. However, also recent ground-based and airborne measurements point to substantial tropical emissions [*Miller et al.*, 2007].

[4] In addition to these observational surprises, a recent study by *Keppler et al.* [2006] challenged the textbook knowledge on methane sources by reporting emissions from terrestrial plants under aerobic conditions, supposedly largest in the tropics. This new source type is heavily debated, especially with respect to reported global emission estimates (62–236 Tg/yr), which were considered too high by several studies [*Houweling et al.*, 2006; *Kirschbaum et al.*, 2006; *Ferretti et al.*, 2007]. The work was also heavily debated from a laboratory point of view [*Dueck et al.*, 2007] but confirmed by later studies [*Vigano et al.*, 2008; *Keppler et al.*, 2008] while its importance in the global methane budget remains highly uncertain.

[5] Here, we identify previously unaccounted spectroscopic interferences with water vapor as a source of error in SCIAMACHY methane retrievals and report on its effect of overestimating tropical sources.

2. SCIAMACHY Instrument: Retrieval Methods and Changes in Water Spectroscopy

[6] SCIAMACHY onboard the European Space Agencies environmental research satellite ENVISAT is an 8 channel grating spectrometer measuring in the ultraviolet, visible and near infrared wavelength region (240–2380 nm) [*Bovensmann et al.*, 1997].

[7] Methane is retrieved from nadir spectra in a micro-window of channel 6, ranging from 1630 to 1670 nm. A recent reanalysis of methane spectroscopic parameters in this spectral range [*Frankenberg et al.*, 2008] already resolved a potential retrieval bias due to erroneous pressure-broadening coefficients.

[8] Details about the retrieval method can be found in *Frankenberg et al.* [2005b, 2005a, 2006]. While methane is the strongest absorber in the retrieval window, minor absorptions by carbon dioxide and water vapor exist. For CO_2, we use a spectroscopic database by *Toth et al.* [2007]. H_2O parameters have so far been taken from an updated version (2006) of the 2004 HITRAN spectroscopic database [*Rothman et al.*, 2005]. *Jenouvrier et al.* [2007], however,

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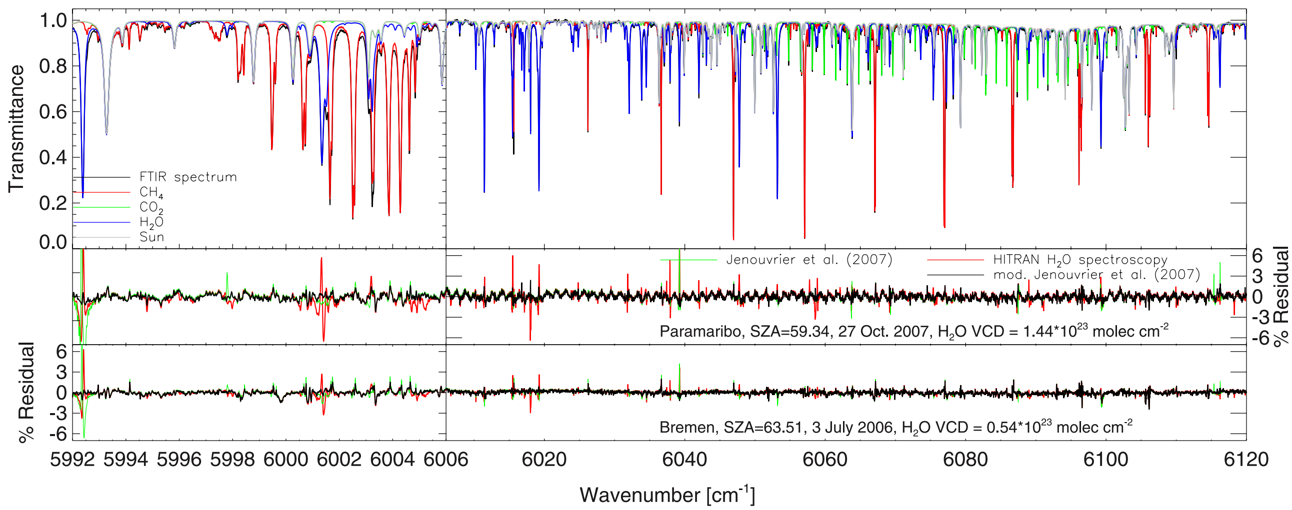

Figure 1. (top and middle) Spectral fit of a Paramaribo spectrum and corresponding residuals. Contributions from individual gases (multiplied by the sun reference) are shown in color. The methane $2\nu_3$ Q branch (from 5998–6006 cm^{-1}) to which SCIAMACHY is most sensitive is shown in more detail. (bottom) Residuals of a fit using a Bremen spectrum.

recently determined new line parameters (denoted as Bxl-Reims database) using high-resolution water vapor absorption spectra in the 4200–6600 cm^{-1} range, covering the SCIAMACHY microwindow in which about 200 new weak water lines could be identified and further systematic differences compared to HITRAN exist.

3. Atmospheric Observations

[9] Here, we use ground-based high-resolution Fourier transform (FTIR) solar absorption spectra to evaluate systematic differences between the H_2O spectroscopic data sets before reporting the impact on low-resolution SCIAMACHY spectra.

3.1. High-Resolution FTIR Retrievals

[10] We analyzed two solar absorption Fourier transform spectra covering the SCIAMACHY retrieval window in the near infrared. The FTIR measurements were performed at the Meteorological Service (MDS) in Paramaribo, Suriname (5.8°N, 55.2°W) and at the University of Bremen (53.1°N, 8.9°E) with a Bruker 120M and a Bruker 125HR spectrometer, respectively. Measurements were performed with a resolution of 0.055 cm^{-1} and 0.022 cm^{-1}, respectively.

[11] Figure 1 clearly shows that at a tropical location such as Paramaribo, water vapor exhibits substantial absorptions, partially overlapping with strong methane lines. Using HITRAN spectroscopic parameters for water vapor, we find systematic residuals of up to 6% which mostly vanish using a modified Bxl-Reims database. Similar findings hold for the Bremen fit, even though discrepancies are smaller due to lower water vapor abundances. For a few strong transitions in this spectral range, we modified the Bxl-Reims database as pressure-shift or air-broadening parameters were not provided or resulted in systematic residuals, as indicated by the green residuals in Figure 1 (see auxiliary material[1] for a list of changed intensities, broadening coefficients and pressure shifts). Overall, it can be concluded that systematic errors in spectroscopic parameters provided by HITRAN are substantially reduced in the Bxl-Reims database. Considering the FTIR-fits, this has a substantial impact on residuals but the fitted methane column remains unchanged within 0.15% as absorption lines are well resolved, minimizing interference of CH_4 and H_2O absorptions.

3.2. Impact on SCIAMACHY Retrievals

[12] In contrast to high-resolution FTIR spectra, SCIAMACHY does not resolve individual absorption lines, hence being more susceptible to potential spectroscopic interferences. We retrieved methane for the entire year 2004 from SCIAMACHY spectra with two different retrieval versions, using HITRAN and the modified Bxl-Reims database for H_2O, respectively. In contrast to previous retrieval versions [*Frankenberg et al.*, 2005a, 2006], we use ECMWF pressure, temperature and water vapor profiles as prior information and NOAA CarbonTracker CO_2 fields [*Peters et al.*, 2007] to convert retrieved CH_4/CO_2 ratios to column-averaged mixing ratios [*Frankenberg et al.*, 2005a, 2006]. In addition, we apply prior methane fields that are optimized with ground-based measurements from the NOAA ESRL global air sampling network using a 4D-variational data assimilation system [*Meirink et al.*, 2007b]. In the following, these fields are denoted as TM5-4DVAR model fields.

[13] To discern a retrieval bias depending on water vapor from a latitudinal bias or a potentially true correlation of methane emissions with specific humidity, we analyzed correlations over a Saharan region from January through July 2004, as illustrated in Figure 2. The use of the HITRAN database clearly results in a positive correlation of retrieved methane (expressed as the ratio of the retrieval versus TM5-4DVAR model columns) with the total water vapor column, taken from ECMWF. Given that tropical water columns can be $1 \cdot 10^{23}$ molec/cm^2 higher than mid-latitudinal values, the overestimation can be on the order of 3%. Using the modified Bxl-Reims database in SCIAMACHY retrievals virtually eliminates this bias which is crucial due to the high variability of water vapor in the atmosphere and the potentially largest effects in humid

[1] Auxiliary materials are available in the HTML. doi:10.1029/2008GL034300.

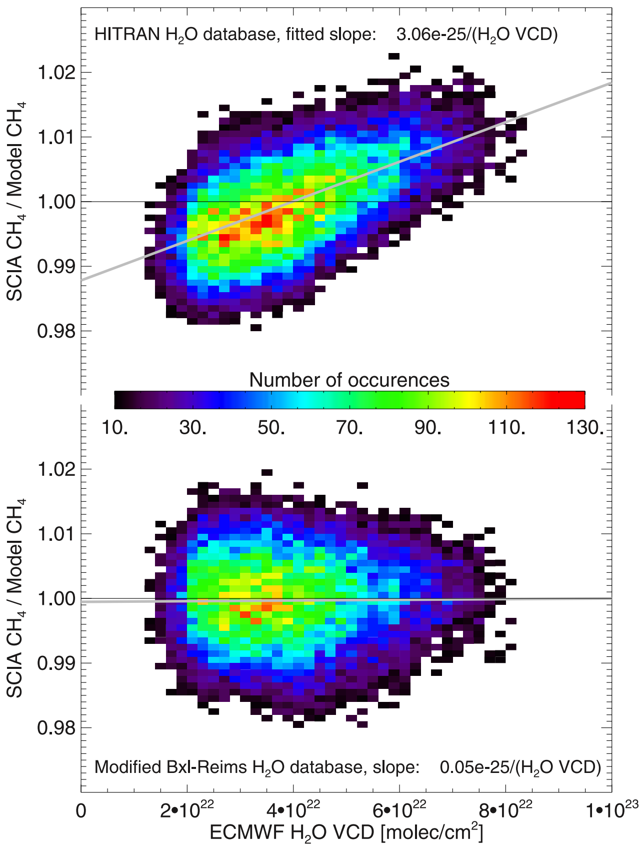

Figure 2. Frequency distribution of the ratio of SCIAMACHY methane retrievals and TM5 model columns over the Sahara (25°N–30°N, 0°E–40°E) in 2004 as a function of water column (only data from January through July are taken to avoid transport from rice paddy emissions in Asia). The top shows retrievals using the HITRAN database for water spectroscopic parameters while the bottom uses a modified version of the Bxl-Reims database [*Jenouvrier et al.*, 2007]. A linear fit to the data is shown as gray line. SCIAMACHY retrievals haven been scaled with a factor of 1.008 in order to result in a best possible agreement with the model.

tropical regions. The reason for the spectroscopic interference is complex and outlined in the auxiliary material where we also show that the usage of the original Bxl-Reims database would yield very similar results.

[14] Figure 3 shows a yearly average of SCIAMACHY (using the modified Bxl-Reims database) methane retrievals (upper panel) and the differences between the retrieval versions differing only in water spectroscopic parameters (i.e. Bxl-Reims - HITRAN, lower panel). Over tropical regions, methane abundances are reduced by up to 60 ppb and the patterns are similar to the enhancements observed by *Frankenberg et al.* [2005a]. Seasonal changes in water abundances also caused parts of the seasonal bias over Australia that was reported by *Frankenberg et al.* [2006]. As shown by *Frankenberg et al.* [2008], there is now a very good agreement between measurements and TM5-4DVAR model fields over Australia.

[15] Despite these systematic changes, some tropical enhancements still exist. However, they should not be confused with near-surface mixing ratios as strong mixing and a high tropopause contribute substantially to elevated column averaged mixing ratios. It should be noted that latitudinal gradients in methane column-averaged mixing ratios are modified by variations in tropopause height and stratospheric depletions. Hence, high-latitude column averaged mixing ratios are often lower than in the tropics even though surface concentrations are higher.

4. Impact on Tropical Emission Estimates

[16] We use a 4DVAR inverse modeling system based on the TM5 model [*Meirink et al.*, 2007a, 2007b] to obtain a first estimate of the impact on tropical emissions. As from *Bergamaschi et al.* [2007] and *Meirink et al.* [2007b], we use the satellite retrievals simultaneously with ground-based measurements from the NOAA/ESRL network as a high-accuracy reference. The reference data set for old SCIAMACHY retrievals is based on the work by *Frankenberg et al.* [2006].

[17] Table 1 shows results for different source-inversion scenarios and the prior emissions used by *Frankenberg et al.* [2005a] as a further reference. The scenario based on ground-based measurements only (S1) already yields higher tropical emissions than those prior values, particularly in South-America and despite the lack of nearby measure-

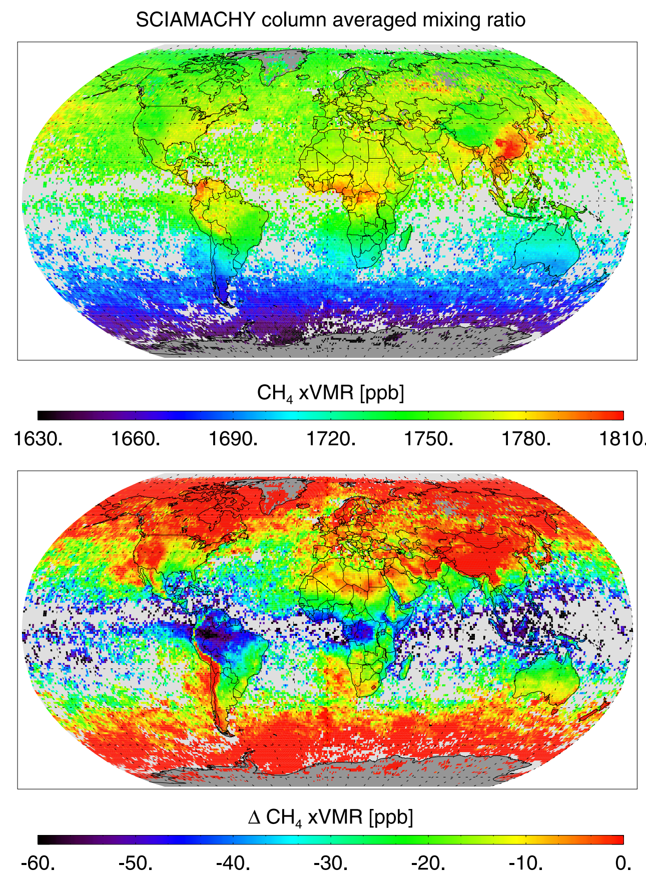

Figure 3. (top) SCIAMACHY column averaged mixing ratios (xVMR) of methane gridded on 1° by 1° in 2004 using the modified Bxl-Reims database. (bottom) Difference plot, subtracting a retrieval version using the HITRAN database for H_2O.

Table 1. Annual Tropical Methane Emissions in Tg CH_4 as Obtained From Different Source Inversions for the Year 2004[a]

	Prior[b]	S1[c]	S2[d]	S3[e]
Trop. S-Am	34.6	70.6 (±6.6)	102.5	79.0 (±3.8)
Trop. Africa	52.2	66.1 (±5.0)	98.6	71.2 (±1.3)
Indonesia	48.9	53.8 (±5.9)	58.8	50.7 (±4.4)
Tropics total	136	190.5	259.9	200.9

[a]In addition, the prior values as used in the model-SCIAMACHY comparison by *Frankenberg et al.* [2005a] are shown. Sources are given for tropical regions (15°S–15°N), viz. tropical South-America (180°W–30°W), tropical Africa (30°W–65°E) and Indonesia (90°E–180°E). Uncertainties of posterior emissions (given as 2-σ only for scenarios S1 and S3) are estimated based on the leading Eigenvectors of the posterior covariance matrix [*Meirink et al.*, 2007a], and have been aggregated to yearly numbers for the respective regions taking into account spatio-temporal error correlations. It should be noted that these estimates furthermore depend on the specific settings of the inversions (e.g. uncertainties and spatio-temporal correlations of prior emissions) and do not account for systematic model errors.

[b]Prior values used by *Frankenberg et al.* [2005a].

[c]Using only ground-based observations.

[d]Ground-based and *Frankenberg et al.* [2006] SCIAMACHY retrievals.

[e]Ground-based and new SCIAMACHY retrievals.

ments. The main reason is that the model needs to increase tropical emissions in order to reconcile measured and modeled interhemispheric gradients. The inclusion of old SCIAMACHY retrievals [*Frankenberg et al.*, 2006], however, results in a considerable further increase of tropical CH_4 emissions (S2). Applying the new SCIAMACHY retrieval (S3) largely reduces these fluxes, emission estimates being only slightly higher than scenario S1.

[18] Overall, tropical emissions are still substantially higher than the prior fluxes assumed by *Frankenberg et al.* [2005a], but the new inversion yields a more consistent picture with inversions based on ground-based stations only. The inclusion of SCIAMACHY data adds important constraints on spatial and temporal emission distributions. A detailed discussion of these regional distributions, however, is beyond the scope of this paper and will be presented elsewhere.

5. Conclusions

[19] We detected systematic errors in previous SCIAMACHY methane retrievals caused by an erroneous H_2O spectroscopic database. We have shown that these resulted in a positive correlation of retrieved methane with water vapor abundances and thereby led to a systematic overestimation of tropical methane abundances. An updated spectroscopic data set for water (modified Bxl-Reims database, *Jenouvrier et al.* [2007]) largely eliminated this dependence in a new retrieval version which has been applied in a 4-D variational data assimilation system to invert methane sources. Compared to inversions based on previous SCIAMACHY data, tropical emission estimates are reduced from 260 to about 201 Tg CH_4/yr, being more consistent with inversions based on ground-based measurements only.

[20] **Acknowledgments.** CF is supported by the Dutch science foundation (NWO) through a VENI grant. We acknowledge John Burrows, PI of the SCIAMACHY instrument, for having initiated and realized the SCIAMACHY project. The Netherlands SCIAMACHY Data Center and ESA is greatly acknowledged for providing data and R. van Hees for having written the versatile NADC tools software package. We thank Ed Dlugokencky for providing NOAA ESRL CH_4 data, Wouter Peters for providing CarbonTracker results and A. Segers, C. Schrijvers, and O. Tuinder for providing ECMWF data. We acknowledge the European Commission for supporting the 6th Framework Programme project HYMN (contract 037048), GEOMON (contract 036677) and GEMS-IP (contract SIP4-CT-2004-516099). We further acknowledge exchange of information within the EU 6th FP Network of Excellence ACCENT.

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