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First ground-based FTIR-observations of methane in the tropics

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Abstract

Total column concentrations and volume mixing ratio profiles of methane have been retrieved from ground-based solar absorption FTIR spectra in the near-infrared recorded in Paramaribo (Suriname). The methane FTIR observations are compared with TM5 model simulations and satellite observations from SCIAMACHY, and represent the first validation of SCIAMACHY retrievals in the tropics using ground-based remote sensing techniques. Apart from local biomass burning features, our methane FTIR observations agree well with the SCIAMACHY retrievals and TM5 model simulations.

1 Introduction

Methane (CH_4) is the second most important anthropogenic greenhouse gas in the atmosphere and is one of the target gases of the Kyoto protocol. Its global atmospheric concentration has more than doubled since preindustrial times and reached 1774 ppb in 2005 (IPCC: Forster et al., 2007). Very recently, surface measurements from global monitoring networks revealed a renewed growth of atmospheric methane in 2007 and 2008 on a global scale (Rigby et al., 2008; Dlugokencky et al., 2009). This change in the growth rate is yet not understood and reveals the importance of measurements and research on the global methane budget. Sources and sinks of atmospheric methane are not yet well quantified. In particular, in the tropics a large uncertainty in the methane budget exists due to the scarcity of measurements (e.g. Meirink et al., 2008b). The first space-borne measurements for CH_4 have become available from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument onboard ENVISAT, with an estimated relative accuracy on the order of 1–2% (Frankenberg et al., 2005, 2006; Buchwitz et al., 2006; Schneising et al., 2009). However, due to the complexity of the retrieval algorithms and the limited availability of independent measurements for validation, the quantification of potential systematic retrieval errors remains very difficult. Recently, Franken-

berg et al. (2008a) reported a major revision of their CH_4 retrievals, mainly a consequence of the identification of systematic errors in the spectroscopic parameters of CH_4 (Frankenberg et al., 2008b) and H_2O vapor (Frankenberg et al., 2008a). This revision resulted in significantly lower column-averaged CH_4 mixing ratios, especially in the tropics. The systematic retrieval errors were caused by spectroscopic errors and interference between H_2O and CH_4, and were most pronounced in the tropical regions, where H_2O vapour is most abundant. Inversions based on the revised SCIAMACHY retrievals using the TM5-four-dimensional variational (TM5-4DVAR) inverse modeling system (Meirink et al., 2008a) yield ∼20% lower tropical emissions compared to the previous retrievals (Frankenberg et al., 2008a; Bergamaschi et al., 2009). In this paper, we present the first tropical ground-based FTIR measurements for validation of the SCIAMACHY methane retrievals. For this purpose, we compare our ground-based observations with the two existing SCIAMACHY methane retrievals, the WFM-DOAS (Weighting Function Modified DOAS; Buchwitz et al., 2006) and the IMAP-DOAS (Iterative Maximum A Posteori-DOAS; Frankenberg et al., 2005) retrieval. Furthermore, we compare the simulations of the TM5-4DVAR model with our ground-based FTIR observations as well as with surface in situ measurements of CH_4.

2 Site description and retrieval

The FTIR measurements were performed at the Meteorological Service in Paramaribo, Suriname (5.8° N, 55.2° W) during seven consecutive dry seasons between September 2004 and November 2007. Campaigns during short dry seasons (SDS, February to March) are denoted as SDS campaigns, while campaigns during long dry seasons (LDS, August to November) as LDS campaigns. The Intertropical Convergence Zone migrates twice a year over the measurement site. During the SDS, Paramaribo belongs to the meteorological Northern Hemisphere, and during the LDS, it belongs to the meteorological Southern Hemisphere (Fortuin et al., 2007). The retrieval of trace gas concentrations from absorption spectra is based on the comparison of measured

with simulated spectra. For an apriori state of the atmosphere (e.g. pressure and temperature profiles) and with respect of instrumental influences, a transmission spectrum of the atmosphere is calculated. Through variation of certain parameters (e.g. trace gas concentrations) in the simulation, the calculated spectrum is fitted to the measured spectrum. Total column trace gas concentrations can be retrieved by scaling the trace gas concentration profiles during the fit. In addition to the retrieval of total column amount, for some trace gases it is possible to retrieve information about the vertical distribution from the analysis of the shape of absorption lines, e.g. by optimal estimation. The spectral line shape of an absorption line is influenced by the natural line width, Doppler broadening and pressure broadening. Since pressure decreases with height, pressure broadening dominates in the troposphere whereas in the stratosphere, Doppler broadening is dominant in the infrared. Information about the volume mixing ratio profiles can be gained as high up as pressure broadening is dominant. Solar absorption spectra, recorded with a Bruker 120M FTIR spectrometer were analysed using the SFIT2 vs2.92 algorithm (Spectral Least Square Fitting Program) developed at NASA Langley Research Center and the National Institute for Water and Atmospheric Research in New Zealand based on optimal estimation (for further details see Rinsland et al., 1998) to retrieve volume mixing ratios and total column amounts of methane. The FTIR solar absorption spectra were analysed in the same spectroscopic regions and with the same spectroscopic linelists as used for the CH_4 IMAP-DOAS retrieval from low-resolution spectra from SCIAMACHY (Frankenberg et al., 2008a). The retrieval of methane total column amounts in the near-infrared (NIR) spectral region by profile scaling has been used successfully in a number of cases (Warneke et al., 2006; Washenfelder et al., 2003), showing good agreement with model simulations and surface observations. In the tropics, the methane total column retrieval by profile scaling showed strong sensitivity to the a priori information as well as to different microwindows used for the retrieval, which we ascribe to too unrealistic a priori information for the tropical trace gases, water interference, a too restricted retrieval algorithm due to profile retrieval (no freedom to adjust the shape

of the profiles) or other unknown effects. In order to solve the problem of reduced freedom due to profile scaling, we used the SFIT2 algorithm for profile retrieval based on optimal estimation and derived volume mixing ratios and total column amounts of methane in the NIR, fitting a whole transition band instead of single absorption lines, as commonly applied for the retrieval of trace gas profiles in the mid-infrared (MIR). The spectral window used is one order of magnitude larger than commonly used for the profile retrieval. Since a whole transition band (consisting of several absorption lines with different temperature sensitivities) is used for the retrieval, inconsistencies of spectroscopic parameters for certain absorption lines and assumed a priori information for the pressure-temperature profiles have less impact on the results. The a priori profiles used for the FTIR retrievals are based on the a priori profiles used for tropical sites within the TCCON network (http://www.tccon.caltech.edu/). Profiles of pressure, temperature and relative humidity are obtained from the NOAA Climate Diagnostics Center (CDC) based on NCEP (National Centers for Environmental Prediction) Reanalysis data (http://www.cdc.noaa.gov/data/gridded/data.ncep.reanalysis.html). Methane total columns are retrieved from FTIR spectra in the spectral window from 5995 to 6115 cm^{-1}. The interfering species CO_2 and H_2O are also fitted in the same microwindow. Total columns of carbon dioxide are retrieved from the same NIR spectra as methane (microwindow: 6180–6260 cm^{-1}) by profile scaling. The retrieved total column amounts are converted to column averaged volume mixing ratios (XVMR) by dividing the retrieved vertical column by the total dry air mass:

$$\mathrm{XVMR}(CH_4) = \frac{\mathrm{column}(CH_4)}{\text{total dry column}}, \tag{1}$$

where the total dry column is

$$\text{total dry column} = \frac{P_{\mathrm{obs}}}{m_{\mathrm{dry}} g(\varphi)} - \mathrm{column}(H_2O)\left(\frac{m_{H_2O}}{m_{\mathrm{dry}}}\right). \tag{2}$$

P_{obs} denotes the observed surface pressure, m_{dry} the mean molecular mass of dry air (0.028964 kg mol^{-1}), m_{H_2O} the mean molecular mass of water vapour

(0.01802 kg mol^{-1}) and $g(\varphi)$ is the latitude dependent surface acceleration due to gravity. We estimate the error on the CH_4 XVMR to be less than 0.2% for a pressure uncertainty of 2 hPa, which we assume to be the maximal error for the pressure measurements during all campaigns. The potential errors in the FTIR observations due to errors in the pressure measurements are negligible compared to the diurnal variations.

3 Comparison with space-borne measurements

Methane retrievals from SCIAMACHY are performed in a microwindow in channel 6, ranging from 5983 to 6138 cm^{-1} (1629–1671 nm). The SCIAMACHY retrievals used within this work are described in detail by Frankenberg et al. (2008a) for the IMAP-DOAS v50, and by Schneising et al. (2009) for the WFM-DOAS v1.0 retrieval algorithm. The SCIAMACHY products represents the measured total column of CH_4 normalized to the measured total column of CO_2. The measured columns of CH_4 and CO_2 are derived from neighboring spectral regions, ensuring very similar light path distributions for both species. The column averaged CH_4 mixing ratio XVMR(CH_4) is obtained by

$$\mathrm{XVMR}(CH_4) = \frac{\text{meas. column}(CH_4)}{\text{meas. column}(CO_2)} \cdot \mathrm{XVMR}_{\mathrm{model}}(CO_2) \tag{3}$$

using modeled column averaged mixing ratios of CO_2 ($\mathrm{XVMR}_{\mathrm{model}}(CO_2)$). Global spatio-temporal CO_2 concentration fields have been calculated by the atmospheric tracer transport model TM3 driven with re-analysed meteorological data (NCEP) (Rödenbeck, 2005). Surface CO_2 fluxes supplied to the model comprise detailed representations of fossil fuel emissions, land biosphere exchange, and oceanic exchange, as well as an large-scale inversely calculated correction flux ensuring consistency with measured atmospheric CO_2 concentrations at many sites around the globe. Details about the model are described by (Rödenbeck, 2005), and data are available online (http://www.bgc-jena.mpg.de/~christian.roedenbeck/download-CO2-3D/). For the

IMAP-DOAS v50 retrieval, prior methane fields are applied using the TM5-4DVAR model fields at 180° W at the same days and latitudes; for the WFM-DOAS v1.0, a single (constant) vertical methane profile is used. Due to the relatively high single measurement noise of satellite retrievals, we use a ±15 days running average of the daily means of the satellite observations over 4.0–8.0° N, 54.0–57.0° W containing both land and ocean pixels. The 15 days running average of the daily means (IMAP-DOAS: blue line, WFM-DOAS: red line) is shown in Fig. 1 together with the standard deviations of the daily means.

The FTIR observations of methane at Paramaribo are in general in good agreement with the satellite observations retrieved with different algorithms (see Fig. 1). The WFM-DOAS v1.0 retrievals, available only for 2003 to 2005, use a single (constant) methane a priori and show less temporal variation at this location than the IMAP-DOAS v50, using prior methane fields of the TM5-4DVAR model fields, which seems to be the major reason for this temporal variation. During the LDS 2004 and 2005, the FTIR observations are higher than the ±15-day running average of SCIAMACHY. From FTIR observations of carbon monoxide (CO) and other biomass burning related trace gases, model simulations and trajectory analysis it is known that Paramaribo experienced air masses polluted by biomass burning during this time (Petersen et al., 2008). During the whole LDS 2005 campaign, CO levels are enhanced by 12±2.5% compared to other campaigns. We observe CO levels of 2.3×10^{18} molec/cm^2 with peaks of up to 3.5×10^{18} molec/cm^2. Using typical emission factors of CO and CH_4 from biomass burning of tropical forest or savanna/grassland (see Table 1), we can estimate the enhancement of CH_4 relative to background levels emitted from biomass burning.

$$
\begin{aligned}
(\mathrm{CH_4})_{BB} &= \frac{EF(\mathrm{CH_4})}{EF(\mathrm{CO})}\cdot(\mathrm{CO})_{BB} \\
&= \frac{6.8}{104}\frac{[\mathrm{g\ CH_4}]}{[\mathrm{g\ CO}]}\cdot(\mathrm{CO})_{BB(\text{Tropical Forest})} \\
&= \frac{6.8}{104}\left(\frac{16.04\mathrm{g\ mol^{-1}}}{28.01\mathrm{g\ mol^{-1}}}\right)\cdot(\mathrm{CO})_{BB(\text{Tropical Forest})} \\
&= 0.114\cdot(\mathrm{CO})_{BB(\text{Tropical Forest})}
\end{aligned}
$$

From the observed CO levels during the LDS 2005, we expect 0.3 to 1.7×10^{17} molec/cm^2 of additional methane from biomass burning assuming tropical forest emission factors. This is equivalent of around 0.1 to 0.5% (2 to 9 ppb).

Using the observed CO as a tracer for biomass burning, we can calculate the amount of methane coming from biomass burning by using typical emission factors for tropical biomass burning. Our calculations can only confirm part of the observed methane enhancement. The reason can be deviations of the used emission factors from the true ones, or the much shorter lifetime of CO compared to methane: From back trajectory analysis we know, that the observed CO was transported to Suriname from the South American continent, mostly from Brasil (Petersen et al., 2008). Due to a shorter lifetime of CO compared to CH_4, it is possible, that we observe less CO but still higher methane from the biomass burning source.

The enhanced CH_4 observed by the FTIR during the LDS 2005 cannot be seen in the SCIAMACHY XVMR observations because of the large footprint of the SCIAMACHY retrievals and the retrieval method itself. Column averaged volume mixing ratios are derived from the measured ratio CH_4/CO_2. Methane enhancements due to biomass burning are hidden in the CH_4/CO_2 ratio as both species are enhanced in a similar way given typical emission factors by Andreae and Merlet (2001). Furthermore, model assumptions may introduce biases when unrealistic CO_2 model data are used.

The comparison of the directly measured ratio of CH_4/CO_2 from both instruments allows the ground-based validation of the satellite retrieval without further model assumption. The directly measured CH_4/CO_2 ratios retrieved from FTIR spectra are in good agreement with the SCIAMACHY observations (Fig. 2).

4 Comparison with model simulations

In the following we compare our observations with 3D model fields assimilated with the TM5 four-dimensional variational (4D-VAR) data assimilation system. Essential parts of the TM5-4DVAR system are described in detail in (Meirink et al., 2008b) and subsequent further improvements in (Bergamaschi et al., 2009). We use here global coarse resolution ($6^\circ \times 4^\circ$) simulations, interpolated to the coordinates of the Paramaribo site. Surface observations of methane, taken from the NOAA ESRL global cooperative air sampling network (Dlugokencky et al., 1994, 2003), have been used to optimize the two-dimensional distribution of surface emissions. Only measurements from marine and continental background sites have been used for the inversion.

During the first part of the LDS 2004 and during the LDS 2005, when Paramaribo experienced air masses polluted by biomass burning, the FTIR observations exceed the model simulations by far. Using CO as a tracer for biomass burning, we identify biomass burning as a source for the enhanced CH_4 levels (see Sect. 3). Apart from these biomass burning events, the FTIR CH_4 retrievals are in general in good agreement with the model output. The FTIR data shows higher variation than the model representing methane background levels. As reported recently by Rigby et al. (2008) and Dlugokencky et al. (2009), global surface in situ measurements show enhanced methane levels in 2007 compared to earlier years. The TM5 model based on assimilation of NOAA surface observations show this anomaly being ~10 ppb higher in 2007 than in the years before. The ground-based methane total column FTIR observations are consistent with the observations of Rigby et al. (2008) and Dlugokencky et al.

(2009) and are the first CH_4 total column measurements showing this anomaly.

Surface samples were taken in Paramaribo and analysed by the MPI Jena by gas chromatography (Fig. 3). The results of the flask measurements in Paramaribo show in general a very high variation, indicating the strong influence of local and regional sources. Assuming that the lower values are representative for air with small local pollution, it can be concluded that there is a good agreement of the "clean air" flasks with the TM5 model. We assume that the high methane pollution events are due to urban pollution from the city Paramaribo or local and regional biomass burning. The good agreement also in 2007 of the TM5 model with the "clean air" surface in situ measurements as well as with our FTIR observations is consistent with the observations reported by Rigby et al. (2008) and Dlugokencky et al. (2009).

5 Conclusions

From FTIR CO observations in Paramaribo it is known that during the first part of the LDS 2004 and during the whole LDS 2005 air masses above Paramaribo were highly influenced by regional biomass burning pollution (Petersen et al., 2008). During these biomass burning periods, we observe enhanced methane levels with the ground-based FTIR solar absorption measurements. Apart from these biomass burning events, the TM5 model simulations reproduce the FTIR observations. The FTIR measurements, being more sensitive to local influences, show higher variations than the low resolution model. Surface in situ observations performed in Paramaribo show in general a very high variation, indicating the high influence of local and regional sources. However, the lower values of the surface observations, representing the "clean air" observations, agree well with the TM5 model simulations for all campaigns. The TM5 model based on assimilation of NOAA surface observations shows the methane anomaly of 2007, first reported by Rigby et al. (2008). The agreement of the TM5 model with the in situ and the ground-based total column FTIR observations of methane are consistent with the findings of Rigby et al. (2008) and Dlugokencky et al. (2009). The FTIR observations of

methane at Paramaribo agree with the SCIAMACHY observations retrieved with different algorithms (IMAP-DOAS and WFM-DOAS) within the standard deviation of the daily means apart from the enhancements observed by the FTIR during the LDS in 2004 and 2005 which we account to biomass burning. The biomass burning enhancements cannot be seen in the SCIAMACHY XVMR observations because of the large footprint of the SCIAMACHY retrievals and the retrieval method itself: Column averaged volume mixing ratios are derived from the measured ratio CH_4/CO_2. Methane enhancements due to biomass burning are hidden in the CH_4/CO_2 ratio as both species are enhanced in a similar way given typical emission factors. Since the FTIR measurements are restricted to the dry seasons, no firm conclusions can be drawn regarding the differences in the seasonality between the two different SCIAMACHY products. The directly measured CH_4/CO_2 ratios retrieved from FTIR spectra are in good agreement with the SCIAMACHY observations. These FTIR measurements represent the first ground-based validation of the methane retrievals from SCIAMACHY spectra in the tropics, showing good agreement.

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Table 1. Emission factors (EF) for biomass burning of savanna/grassland and tropical forest (Andreae and Merlet, 2001). EFs are given in gram species per kilogram dry matter burned.

	Savanna/Grassland	Tropical Forest
CH_4	2.3±0.9	6.8±2.0
CO_2	1613±95	1580±90
CO	65±20	104±20

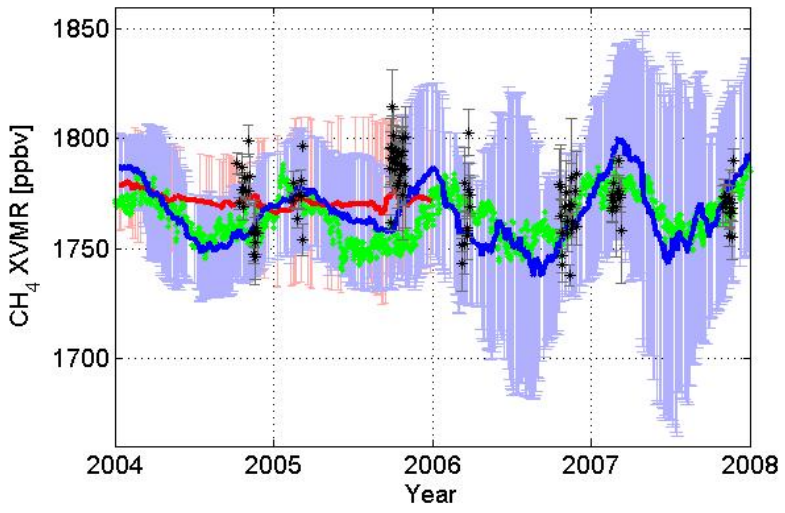

Fig. 1. Retrieved XVMR(CH_4) from FTIR (black stars) and SCIAMACHY spectra (IMAP-DOAS in blue, WFM-DOAS in red) averaged over 4.0–8.0° N, 54.0–57.0° W compared with TM5 model simulations assimilated with NOAA surface observations (green). Shown is the 15-days running average over daily means from SCIAMACHY retrievals (lines) and the standard deviations of the daily means (errorbars). The IMAP-DOAS retrieval is scaled with 1.01. Due to the degradations of chanal 6 of the SCIAMACHY instrument, larger standard deviations are observed in 2006 and 2007. The FTIR observations are daily means (black stars) with standard deviations. TM3 model data for CO_2 is used to derive the XVMR(CH_4) in the SCIAMACHY retrievals.

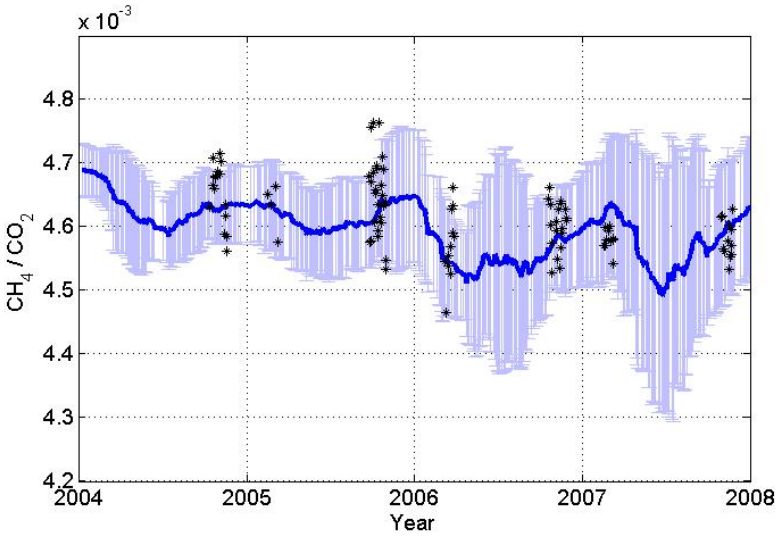

Fig. 2. CH_4/CO_2 ratios retrieved from FTIR and SCIAMACHY (IMAP-DOAS) spectra averaged over 4.0–8.0° N, 54.0–57.0° W. Shown is the 15-days running average over daily means from SCIAMACHY IMAP-DOAS retrieval (blue line) and the standard deviations. Due to the degradations of chanal 6 of the SCIAMACHY instrument, larger standard deviations are observed in 2006 and 2007.

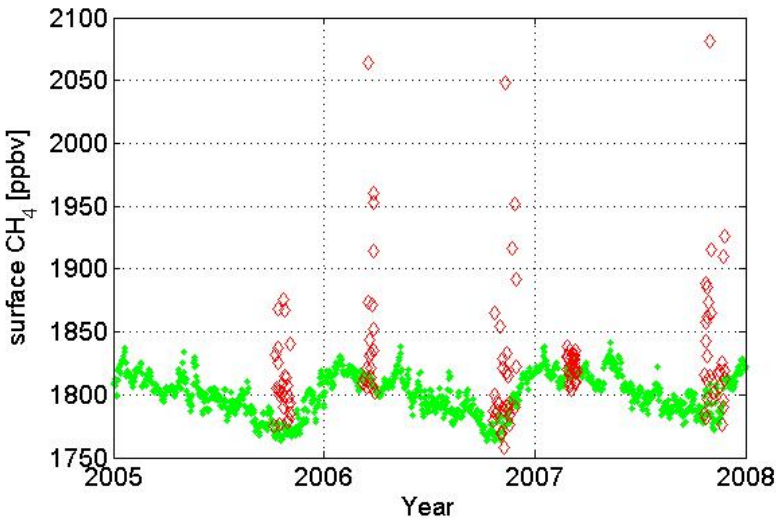

Fig. 3. Methane surface volume mixing ratios from TM5 model simulations, assimilated with NOAA surface observations (green) compared with in situ air samples taken in Paramaribo (red diamonds).