Interactive comment on "First ground-based FTIR-observations of methane in the tropics" by A. K. Petersen et al.

Title changed to "First ground-based FTIR observations of methane in the inner tropics over several years"

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AUTHOR COMMENT:
We would like to thank the referee for reviewing the manuscript and for the helpful comments.

The paper discusses measurements of methane total column amounts from groundbased FTIR measurements in the near-infrared at the station of Paramaribo in Suriname. The ground-based data are compared with SCIAMACHY data from 2 algorithms, namely WFM-DOAS and IMAP-DOAS, with TM5-4DVAR model data, and with data from local surface samplings. In the present version of TM5, surface data from the NOAA ESRL air sampling network, at marine and continental background stations, have been assimilated.

General Comments
The purpose of the paper is to present the first validation of SCIAMACHY retrievals in the tropics. However, the discussion of the validation results is done in a very qualitative way. The only validation results are the figures 1 and 2. One can see a 'reasonable' agreement between the FTIR data and the SCIAMACHY data, during the short campaign periods where FTIR data are available. One cannot say anything about the seasonal variation, as confirmed in the paper, because there are no FTIR data covering all seasons. Moreover, part of the years 2004 and 2005 have to be discarded from the validation because the FTIR data were affected by biomass burning pollution.
This makes the finally available datasets for validation very limited. This is a serious drawback of this validation work. Of course in the tropics, FTIR measurements are very difficult, beacuse they need dry weather conditions which are not available allyear- round. Still, the paper requires more quantitative and more in-depth discussion of the validation results, taking into account also the large scatter on the SCIAMACHY data. At present, the paper does not really provide a clear answer to what one can really conclude from this validation exercise. Also the SCIAMACHY data from the 2 algorithms look very different in Fig. 1. Apart from saying why they are so different (pg. 2309), this is completely neglected in the discussion of the validation results.
The aim of the paper is the comparison of the only available FTIR measurements in the inner tropics with satellite and model data.
We found a good agreement between the FTIR observations and satellite retrievals and model data apart from biomass burning pollution.
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 CH4/CO2. Methane enhancements due to biomass burning are hidden in the CH4/CO2 ratio as both species are enhanced in a similar way
given typical emission factors. The good agreement of the CH4/CO2 ratio of FTIR and satellite, and the differences between FTIR and satellite XVMR(CH4) shows that the influence of biomass burning can hardly be detected by the satellite with this retrieval method. The consistency of the FITR and the satellite observations of the CH4/CO2 ratio also confirms biomass burning is the cause for the differences observed between the FTIR and SCIAMACHY XVMR(CH4).

The comparison with SCIAMACHY retrievals has been complemented by adding a table and a plot with yearly means and campaign-means of the days with FTIR observations.
Since we use FTIR and in situ data to compare with the TM5 model, we can use the TM5 model to make conclusions on the seasonal cycle of the SCIAMACHY data.

We wanted to make clear, that the validation of the satellite retrievals is difficult, because
a.) of the large scatter of the satellite retrievals. So conclusions about the agreement can only be taken within the limitations of the data quality. Averages over time or space should be used when satellite data should be compared.
b.) it highly depends on the satellite retrieval: As can be seen from the different satellite retrievals, the difference in seasonality depends strongly on the a apriori used for the retrieval.
Also should people be aware that the way the satellite retrieval is done, there is always an uncertainties of what is real and what comes from the retrieval (e.g. the use of a model for the CO2 volume mixing ratio; or what is masked out by dividing column(CH4)/column(CO2)).

The validation focused mostly on the IMAP-Doas retrieval, since the WFM-Doas is only available for 2004 and 2005.

Another point to be better explained in the paper is the validation approach. In Fig. 1, one compares XVMR values from SCIAMACHY and ground-based FTIR that are derived in 2 different ways: for the FTIR according to Eq. 2, for SCIA according to Eq. 3. Is it not feasible to compare SCIA with ground-based FTIR XVMR values that are derived identically according to Eq. (3) ? As far as I know, the FTIR measures CO2 simultaneously with CH4 ? And as far as I understand, the data in Fig. 2 are derived using the simultaneously measured CO2 column? And what is the exact usefulness of Fig. 2 in the whole validation approach?

We also looked at the FTIR data derived similar to Eq.3 (by dividing the measured columns of CH4 and CO2 and multiply with a CO2 vmr-model or the FTIR-measured volume mixing ratio of CO2 (than derived similar to Eq. 2 using the pressure). To our opinion we do not want to bring additional information into the retrieval by the use of a model. That is why we used the pressure to derive the total column of air. In order to get an idea of the influence of the used CO2-model and the retrieval, we compare the FTIR and the satellite "directly" in Figure 2:
In Figure 2 the FTIR data is used similar to the satellite product: column(CH4)/column(CO2).

Usefulness of the validation approach related to Figure 2:

- *The comparison of the directly measured ratio of CH4/CO2 allows the validation of the satellite retrieval without further model assumptions*
- *The influence of the model is taken out of account for the validation and its influence can be tested.*
- *One has to keep in mind, that the CO2-model is influencing the retrieval of XCH4 and can also bring information in (e.g. if there are biomass burning signatures in the CO2 model, they will be in the XCH4 as well).*
- *The retrieval method of SCIAMACHY does not allow the detection of biomass burning, because the methane emissions due to biomass burning are hidden in the CH4/CO2 ratio as both species are enhanced in a similar way. The good agreement of the CH4/CO2 ratio of FTIR and satellite and the differences between the FTIR and satellite XCH4 shows that the influence of biomass burning for methane can hardly be detected by the satellite (with this retrieval method) and that it is important to know how the retrieval method is done and to be careful with conclusions from the satellite observations. The SCIAMACHY retrievals are so far the best for a global picture of methane concentrations but have their limitations!*

The consistency of the FTIR and the satellite observations of the CH4/CO2 ratio also confirms biomass burning is the cause for the differences observed between the FTIR and the SCIAMACHY XVMR(CH4).

Another - to my opinion - weak point of the paper is the statement that the data confirm the recent findings by Rigby et al and Duglokencky et al. concerning the enhanced methane levels in 2007 compared to earlier years. The figures shown in the paper are not at all convincing me that this enhancement was also seen in the FTIR ground-based data or in the model or in situ data.
The new figures and tables make this point hopefully more clear. Especially the middle panel of Figure 1 (without the error bars) shows clearly the enhancement of the TM5 model in 2007. Since the TM5 model is assimilated with surface observations showing the 2007 anomaly, the TM5 model shows the 2007 enhancement. We only want to state a good agreement between the TM5 model with the observed FTIR and in situ data; the conclusion is that the FTIR data and the in situ data are not in contradiction with the recent findings, but confirm the methane anomaly.

Specific comments

- Title: The title is not correct as it is. With 'the tropics' one usually refers to the latitude region included between the Tropic of Cancer and the Tropic of Capricorn, ie between 23.5_ N and S. Senten et al. (ACP, Vol. 8, 3483-3508, 2008) already reported CH4 measurements at 21_S (hence the tropics) from ground-based FTIR observations. The Senten et al. measurements were performed in the mid infrared, whereas the actual work reports measurements in the near-infrared. As far as I know- the authors are right that these are the first reported ground-based measurements of CH4 in the tropics in the near-infrared spectral region. Or the authors wanted to point out that they are measuring in the equatorial region ? In any case, the title should be corrected.

We changed the title to "First ground-based FTIR observations of methane in the inner-tropics over several years".
We want to point out, that the measurements are the first FTIR observations in the inner-tropics, so close to the equator. Hawaii, as well as La Reunion and Darwin are at the outer part of the tropics (around 20 N or 20 S). Especially for Hawaii it is documented, that the probed air masses are mostly influenced by the Northern Hemisphere, especially North America and Eurasia (Buermann et al., The changing carbon cycle at Mauna Loa Observatory, PNAS, 2006). Another point is, that the measurements in Paramaribo are the only ones over several years. Even if they only take place during the dry seasons (), interannual variations and especially differences between the northern and southern hemisphere can be investigated. The measurements on Hawaii of Rinsland et al. report only on 4 day average for methane and have not been compared with model data or satellite data. Senten et al. present measurements of two measurements campaign between 2002 and 2004 and do not investigate the seasonal or interannual variation of methane because of the limited amount of data.
We added a small section in the introduction addressing the FTIR measurements performed in Hawaii and La Reunion.

* Introduction, lines 20-24: 'The first space-borne measurements for CH4....'. SCIAMACHY is not the first satellite experiment to provide data for methane. There are many other satellite sensors before SCIAMACHY that have provided CH4 profiles in the upper troposphere - stratosphere, like ATMOS, HALOE, ACE-FTS, ... SCIAMACHY is probably the first one that provides good data for total column CH4. Although one should not forget that IMG has provided limited data sets for total column CH4 → see the following paper: 'Latitudinal distribution of methane as observed by IMG sensor aboard ADEOS satellite' (Proceedings

Paper) Author(s): Ryoichi Imasu; Toshihiro Ogawa; Haruhisa Shimoda in SPIE Proceedings Vol. 3501, Optical Remote Sensing of the Atmosphere and Clouds, Jinxue Wang; Beiying Wu; Toshihiro Ogawa; Zheng-hua Guan, Editors, pp.84-91 Date: 18 August 1998.
So the authors should formulate their statement more correctly.
Changed to "Space-borne measurements for total column CH4 have become…".

* Section 2 - pg. 2306, line 2: 'with respect of instrumental influences' -> correct to ' taking into account instrumental influences' *Done.*
- pg. 2306, line 29: ' a too restricted retrieval algorithm due to profile retrieval' : I think the authors wanted to say ' due to profile scaling only retrievals' . In any way, as stated actually, this is very misleading. *Done.*
- pg.2306, line 26: it is said that the CH4 total column results strongly depend on the a priori choice. It is known that this is also the case (even if it may be less) when you make a real SFIT2 profile retrieval. This sensitivity is not discussed in the paper. Moreover, the paper does not provide any information about the uncertainties (error budget) associated with the CH4 total column data that are presented and used for the validation of SCIAMACHY. The authors should provide this information and discuss how it is taken into account in the validation exercise.
We use the diurnal variation as a measure for the precision: part of the diurnal variation will be caused by real variations in CH4 over the day, therefore this measure gives an upper limit for the precision (Warneke et al., 2006).
The lower most panel shows the daily mean FTIR observations with the standard deviations, this gives an idea of the precision of the observations.
- pg. 2307, line 5: 'as commonly applied for the retrievals of trace gas profiles in the MIR': what exactly out of the previous characteristics of your SFIT2 approach are you comparing to the common approach in the MIR? The sentence is not clear.
Commonly, only single absorption lines are fitted and not a whole absorption band. In the MIR region, the retrieval is commonly done with single absorption lines, while in the NIR absorption bands are used. The use of a whole absorption line needs a lot of computing time, when the optimal estimation retrieval SFIT2 is used.
We changed the sentence to make it more clear to:
"…, we used the SFIT2 algorithm for profile retrieval based on optimal estimation. Volume mixing ratios and total column amounts of methane have been derived from NIR spectra with SFIT2 by 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)."
- pg. 2307, lines 10 to 15: it is not clear whether the a priori choices listed here are the same ones as the ones used initially in the profile scaling approach ? If so, then all this information about the a priori choices should be moved forward in the text. If not, then it should be made clear how the a priori was changed.
For the profile scaling approach, we tested several a priori, to check the influence on the results, also theses ones.
The a priori profiles used within this publication are the most common ones, which are used within the TCCON network.
The information about the a priori choices belong to the retrieval method used in this publication. The parts about the retrieval description are now all in one paragraph.
- pg. 2307, Equation (2) In this equation, one needs the total column of H2O: does it come from the retrieval or from NCEP? What is its uncertainty? So how does this uncertainty affect the uncertainty on XVMR(CH4)?
The column of H2O can be taken from NCEP or from the retrieval. We used the H2O column retrieved from the same NIR spectra as the CH4 retrieval.
The uncertainty of the H2O column on the XVMR is small.

- pg. 2308, line 5: How to interpret the sentence 'The potential errors in the FTIR observations ....compared to the diurnal variations' ? This is not clear...

The diurnal variation can be used as a measure for the precision of the observations. Part of the diurnal variation will be caused by real variations in CH4 over the day, therefore this measure gives an upper limit for the precision (Warneke et al., 2006).
The lower most panel of Figure 1 shows the daily mean FTIR observations with the standard deviations, this gives an idea of the precision of the observations.
We derived the CH4 column by using the measured surface pressure. We estimate for the pressure measurements an uncertainty of 2hPa (which is a quite large estimate), this would result in an uncertainty of 0.2% of the CH4 XVMR. Since we use the diurnal variation as a measure of the total precision of our observations and the diurnal variation is larger than the 0.2% of the pressure uncertainty, the uncertainty in the pressure can be neglected.

- pg. 2310, lines 6-7: 'we expect 0.3 to 1.7 x 10E17 molec/cm2': I don't see where the value of 1.7x10E17 comes from?
CO is enhanced by 12% relative to background levels. That means, during the events of CO up to 3.5x10^18molec cm-2 (background in LDS 2005: 2.0x10^18molec cm-2), there are 1.5x10^18 molec cm-2 in addition to the background coming from biomass burning (see Petersen et al., 2008). With the Efs of CH4 and CO, we get 0.114 x 1.5x10^18 molec cm-2 = 1.71x10^17 molec cm-2.

We included this calculations in more detail in the electronic supplement, together with the calculations for CH4/CO2. Please see the electronic supplement for a detailed answer to this comment!

- pg. 2312, lines 4-5: 'indicating the strong influence of local and regional sources'. Can the authors give more information aboiut these sources ? Do the authors have some 'proof' of the impact of these sources on the data ?
The in situ measurements are high precision measurements. The strong variation of the surface data indicates the influence of local sources. We took air samples at different locations in Suriname (results not shown) to get an idea of these sources (e.g. the influence of tropical rainforest on methane emissions), but couldn't yet draw any conclusion on these sources. We couldn't find strong correlations with precipitation or humidity or other meteorological conditions.
We assume that the sources are due to urban pollution of the city of Paramaribo e.g. gas used for cocking purposes and others.
The in situ data is very sensitive to local sources, because they only probe the air at the time of the measurement at one single location. Total column measurements are probing the whole atmospheric column, so they show less sensitivity to local influences. In addition, the in situ samples have been performed only once or sometimes twice per day, so they don't contain averaging.
We assume the influence of these sources to be small on the total column measurements.

- pg. 2313, line 4: 'which we account to biomass burning' should be 'which we assign to biomass burning'. *Done.*