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"

Anonymous Referee #1
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AUTHOR COMMENT:
We would like to thank the referee for reviewing the manuscript and for the helpful comments.

General comments

The authors present a comparison of ground-based FTIR observations of methane total columns in Suriname with satellite observations and chemistry-transport model simulations. The FTIR observations are interesting and unique for this part of the world, and they deserve to be published. On the other hand, they also deserve a more thorough analysis. Here are some directions for such further analysis.

• The comparisons are very general and can hardly be called validation. Fig. 1 shows that ground- and space-based observations are approximately in the same 50-ppb range but there appears to be little correlation. In fact, during some episodes (LDS 2005 and SDS 2007) there is clear disagreement. Can the authors somehow convince the reader that the FTIR observations are really in line with the satellite observations and model results?

The satellite observations show a large scatter, when not averaged over long time periods (it is very common to look at mean concentrations of one year or at least of each 3-month season for satellite data). The problem is to compare these satellite observations with observations at one station.
We changed Figure 1 and added one panel without the error bars (standard deviations) of the observations, and one panel showing the yearly means and the means for each measurement campaign, which makes it easier to compare the different data sets.

During the LDS 2005, the FTIR observations are highly influenced by biomass burning events (for details see Petersen et al., 2008). This cannot clearly be seen in the satellite observations due to the large footprint of the satellite and the retrieval method itself.
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 (see electronic supplement, which is now added). 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).
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).
Unfortunately, due to the detector degradation, the quality of the satellite retrieval gets worse after 2005. This might be one reason, why the satellite retrieval in 2007 is much higher than the TM5 model and the FTIR observations.

We used the TM5 model to compare with the FTIR observations. The model is used to interpretate the FTIR observations and analysed the differences between model and FTIR observations. The use of a global model allows to see the observations in Paramaribo in a global context.

The model allows also to compare indirectly the FTIR total column measurements with surface in situ measurements: the model gives us the simulated surface CH4 concentrations. They are in good agreement with the "clean air" surface observations sampled in Paramaribo.

(It should be stated that also the satellite needs clear sky conditions to measure methane concentrations. The FTIR observations take place only during the dry seasons when we have the highest chance of clear sky.)

• The biomass burning events need more attention. Perhaps the authors could zoom into one of the episodes (e.g., LDS 2005), make comparisons with measured CO concentrations, present back trajectories, perform model simulations at higher resolution and with realistic biomass burning emissions, etc.
Paper Petersen et al., 2008 is mentioned as a reference. In this paper the CO concentrations observed by the FTIR are compared with satellite observations and model simulations. The electronic supplement of this paper provides also back trajectories.

We included now more details from the paper Petersen et al. 2008 in this manuscript:
"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 (for details see Petersen et al. (2008)). During the whole LDS 2005 campaign, CO levels were enhanced by 12± 2.5% compared to other campaigns. We observed CO levels of 2.3× 1018 molec/cm2 with peaks of up to 3.5× 1018 molec/cm2 . These enhancements are clearly caused by emissions from fires on the South American continent confirmed by back-trajectory analysis and fire counts from satellite measurements (see electronic supplement of Petersen et al. (2008)). Statistics on the fire counts during the LDS 2004 and 2005 give no evidence for enhanced biomass burning during the LDS 2005, neither in Africa nor in South America. However, back-trajectories indicate that during the LDS of 2005, a higher percentage of air masses are coming from South America compared to the same season in the years 2004 and 2006, so that greater levels of regional biomass burning pollution are experienced (Petersen et al., 2008)."

• The effect of the CO lifetime should be quantified in order to assess whether it could be significant .
We included in the manuscript:
" The significance of the lifetime and transport can be estimated: assuming a lifetime of 1month for CO, after 5 days, only 85% of the CO is left. Assuming a lifetime of 10 years for CH4, less than 1% is gone in the 5 days."
The back trajectories take about 3 to 5 days from the fires in Brazil.
This is only a rough estimate, but sufficient for the conclusions of this paper. A more quantitative assessment would exceed the limitations of this manuscript.

• The 'renewed growth' of methane concentrations in 2007 is mentioned but not visible in the figures. A table should be added with annual mean concentrations for observations and model to clarify this feature.
T*he renewed growth of methane is most clearly seen in the surface model simulations, shown in Figure 3. The methane anomaly of 2007 is visible in the model simulations, because the TM5 model is assimilated with surface in situ measurements from the NOAA network, showing the enhanced methane observations. The surface model simulations of 2007 are around 10ppb higher than those from 2004-2006. We added a table with annual means, as well as with means for each measurement campaign. In addition, we changed Figure 1 and provide three panel, so more details can be seen in the figure.*

• What can be concluded from the relatively good agreement between groundbased and space-based CH4/CO2 ratios?

- *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 (see electronic supplement, which is now added). 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).

We included now some discussion about this in the manuscript and added a electronic supplement about the CH4/CO2 ratio.

Specific comments

P2305, L7-10: This is the other way round: Meirink et al. (2008a) used 'unrevised' retrievals and found higher tropical emissions.

Yes, but this paper (Meirink et al, 2008a) is the citation for the TM5-4DVAR inverse modeling system. The 20% lower tropical emissions are in the publications cited at the end of the sentence (Frankenberg et al., 2008a, Bergamaschi et al., 2009). We removed the citation of Meirink et al, 2008a, to avoid confusions.

Section 2: The retrieval description is not completely clear. What is actually used? The profile scaling technique or optimal estimation? Can the authors give an indication of the differences (in ppb)? Warneke et al. (2006) presented ship-based measurements including the tropics. Since they used a profile scaling technique, can we conclude that their retrievals are erroneous?

We used the optimal estimation technique for the retrieval, but used a large spectral region in the NIR. Our retrievals showed a strong sensitivity to the a priori, when we used profile scaling. In Paramaribo, due to the movement of the ITCZ, we measure Northern Hemispheric air during the SDS, and Southern Hemispheric air during the LDS. For methane, with a strong gradient between the Northern and Southern Hemisphere, the profile scaling gives no robust results using a single a priori profile (it shows different results for different a priori profiles). The use of two different a priori profiles for the different season may introduce a unrealistic seasonality in the results.

We assume that the ship-based measurements in the tropics (Warneke et al. 2006) are correct because they do not have a sharp gradient, but a smooth transition between northern and southern hemisphere. In addition, the a priori profiles used for the study by Warneke et al., 2006 contained vertical profiles of water vapor from regularly launched sondes (up to 3 times daily). Water vapor in the tropics can be highly variable. The profile-scaling only retrieval might not be able to account for this variability, while the optimal estimation profile retrieval method allows more freedom to retrieve water vapor (methane and water vapor are fitted at the same time in the retrievals).

The sondes-based water vapor profiles from the ship are well suited for the apriori, while in Paramaribo we do not have sondes-data several times per day.

We added an explanation about the a priori sensitivity to the manuscript to make this point more clear.
Instead of "In the tropics…" we say:
"At the tropical site Paramaribo, …"
And:
"Due to its specific location within the migration zone of the ITCZ, at the Paramaribo site air masses belonging to the Northern (SDS) as well as to the Southern Hemisphere (LDS) are observed by the FTIR. The use of one single a priori profile for the profile scaling only retrieval may be too restrictive for a trace gas like methane having very different concentrations and profiles in both hemispheres. On the other hand, the use of two different a priori profiles for the LDS campaigns and SDS campaigns respectively may introduce unreal seasonality by the a priori."

We changed the section about the retrieval to make it more clear:
" In order to solve the problem of reduced freedom due to profile scaling only, 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 singe 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. "

P2309, L9: Can you really claim a 'good' agreement? As stated above, there appears to be little or no correlation between space- and ground-based observations.
We added "within the limitations of the satellite data quality". The modified Figure 1 and the additional tables with the annual means and the means for each measurement campaign give a more qualitative conclusion.

P2309, L14-15: During the last part of LDS 2004, the FTIR observations are lower than SCIAMACHY.
Changed to "during the first part of the LDS 2004 and during the whole LDS 2005…"

P2309-2310: How does the CH4 time series (e.g. for the LDS 2004) compare with CO? Is it likely that enhancements have been caused by the same source?
For details, see Petersen et al. 2008, in the electronic supplement the back-trajectories are shown. Due to the very strong signal of CO and also other trace gases related to biomass burning (not shown), we know the influence of biomass burning during the first part of the LDS 2004 and the whole LDS 2005. The back-trajectories shown in the electronic supplement of Petersen et al. 2008 show the origin of the air masses.

P2310, L6-7: Can the authors clarify where these numbers come from? Specifically, what value for the CO background column is used?
See P2309, L19-21. In the publication Petersen et al. 2008, we present the mentioned CO observations.
More details about this publication have now been included in this manuscript.

P2310, equations: The CH4 column is denoted 'column(CH4)' on the previous page, but here '(CH4)'. This is not consistent. Actually, these equations are not really needed.
I suggest adding the EF(CH4)/EF(CO) ratios in Table 1, in kg/kg as well as in mol/mol. At the same time, the savanna/grassland values may be removed because they are not used or

discussed in the paper. Otherwise, it should be noted that if part of the burning was from savanna fires, the resulting CH4 enhancement would be even smaller.
The CH4 column is denoted as column(CH4). In the equation on P2310, the (CH4)_BB is the enhancement of CH4 relative to background levels emitted from biomass burning, as mentioned just before the equation. This is not the column, but the part of the column coming from biomass burning, therefore it is named (CH4)_BB.
We removed the values for savanna and grassland, because it is not necessary for this publication.
See the electronic supplement for more details of the calculations.

P2310, L9-11: This was already done in the previous paragraph. Sentence can be removed.
Done.

P2310, L15-17: Based on estimates of the transport time and the CO lifetime it should be possible to assess the impact of chemistry on CO concentrations, and to account for this in the estimate of CH4 enhancements.
We included in the manuscript:
"The significance of the lifetime and transport can be estimated: assuming a lifetime of 1month for CO, after 5 days, only 80% of the CO is left. Assuming a lifetime of 10 years for CH4, less than 1% is gone in the 5 days."
The back trajectories take about 3 to 5 days from the fires in Brazil.
This is only a rough estimate, but sufficient for the conclusions of this paper. A more quantitative assessment would exceed the limitations of this manuscript.

P2310, L21-23: For savanna/grassland fires the ratio of CH4 to CO2 emission factors is 0.004 mol/mol, which is similar to the CH4/CO2 mixing ratio in the atmosphere. However, tropical forest burning, which is assumed to be relevant for this study, is characterized by a three times larger CH4/CO2 emission ratio. Hence, these emissions should to a large extent be visible in the observed CH4/CO2 column ratio.
Methane enhancements due to tropical biomass burning cannot be detected by the satellite, because they are hidden in the CH4/CO2 ratio as both species are enhanced in a similar way.
We included now an electronic supplement with details of the calculations. Please see the electronic supplement to answer these comments!

P2311, L3-4: What can be concluded from the fact that ground-based and space-based CH4/CO2 ratios are relatively consistent?
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 (see the electronic supplement, we added now). 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). The consistency of the FTIR and satellite observations of CH4/CO2 also confirms biomass burning is the cause for the observed differences between the FTIR observations and the satellite observations of XCH4 and the TM5 model.

P2311, L21-22: This statement is only valid if the model data are daily means (instead of running averages), which I could not find in the text.
The model data is not a running average, but daily averages. We included this in the text, also in the Figure-subscription:
"Daily mean values from the TM5 model are compared with the daily averaged FTIR observations (Figure 1)."

P2311, L21-22: The model is not only representing background levels, but also the effect of nearby emissions, be it at a rather coarse resolution.
As mentioned on P2311, L14-15, surface observations of the NOAA network have been used to optimize the distribution of surface emissions. Only measurements from marine and continental background sites have been used for the inversion.
We included, that the model is on a coarse resolution.

P2311, L22 – P2312, L1: From Fig. 1 it is for none of the datasets (FTIR, SCIAMACHY, TM5) clear that methane concentrations are on the order of 10 ppb higher than in previous years. Please add a table showing annual mean (or dry-season-mean) concentrations for all years to clarify this point. Otherwise, the last sentence of this paragraph is not justified.
A table and a plot has been added to the manuscript in order to clarify this point.

P2311, L25: I'm not sure 'anomaly' is the right word here. An anomaly implies that there is a reference. What is the reference in this case?
Anomaly was the word used in this discussion so far (e.g. Dlugokencky et al., 2009). So the reference are the years before with stagnating methane concentrations since 2001, while since 2007 the methane concentrations are higher than expected.
This renewed growth of methane is yet not understood, this is expressed by the word anomaly.

P2312, L2-5: Were the flask measurements all taken during the day? Otherwise, high concentrations may be the result of CH4 build-up in the stable nocturnal boundary layer.
The flask measurements were mainly taken during the day. But we couldn't find any relation between high methane values and time.
We excluded measurements before 8am and after 6pm (not shown), but there was no reduction in the variability of the data.

Fig. 1: The FTIR observations are daily means, whereas the SCIAMACHY data are at around 10h local time. Does diurnal variability influence the comparison?
The FTIR observations are daily means, but the take only place during day time (since they need direct sunlight). The diurnal variability of the observations is shown in the errorbars (standard deviation of the daily means) and is rather small, especially compared to the variability of the satellite observations.

Fig. 2: For completeness add that the unit on the vertical axis is mol/mol, and that these are column ratios.
The unit of the vertical axis (molec/cm2) / (molec/cm2), as commonly used for total column amounts. We added the units to the vertical axis and included in the figure subscript, that it is the ratio of the total columns.

Technical comments
Title: Remove hyphen between FTIR and observations *Done.*
P2304, L16: yet not -> not yet *Done.*
P2305, L8: Remove hyphen between TM5 and four *Done.*
P2305, L15: Posteori -> Posteriori *Done .*
P2306, L1: apriori -> a priori *Done .*
P2306, L2: with respect of -> taking into account (?) *Done .*
P2305, L26: Maybe start new paragraph after (Fortuin et al. 2007) *Done .*
P2308, L21: an -> a *Done .*
P2309, L13: Remove second 'fields' *Done .*
P2309, L19: are -> were *Done .*
P2309, L20: observe -> observed *Done .*
P2311, L21: shows -> show *Done.*
Caption Fig. 1, L5: errorbars -> error bars *Done.*

Caption Fig. 1, L6: chanal -> channel *Done.*
Caption Fig. 2, L4: chanal -> channel *Done.*

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2303, 2010.