Reply to Reviewers

Mapping spectroscopic uncertainties into prospective methane retrieval errors
from Sentinel-5 and its precursor

***Atmos. Meas. Tech. Discuss.**, 8, 1333-1363, 2015*

We thank the comments and remarks of the referees. We have considered for the new manuscript additional sensitivity studies on one side, and new FTS measurements on the other side. Because of the new FTS retrievals all the results shown on the new manuscript are based on new sensitivity studies. Details of the new calculations and our replies to the referees questions are included on this document

> ## Comment from Ilse Aben
> www.atmos-meas-tech-discuss.net/8/C429/2015/
>
> I think this is a very useful study. But I have a few questions/suggestions
>
> 1. In Galli et al., AMT, 2012 we did not find a dependence of the CH4 error on the amount of H2O (see fig. 8 top). This study however concludes that there is a significant correlation in the CH4 error with the amount of H2O. It would be good if some words are spent on the possible reasons for this apparent contradiction.
> 2. One of the possible reasons for the differences observed between Galli et al. and this manuscript lies in the use of the H2O spectroscopic database. It would be very useful to know if and to what extend switching to the Scheepmaker et al, AMT, 2013 database affects the errors on CH4 as presented here.
> 3. This study only uses two FTS measurements, one under dry conditions and one under wet conditions, to establish the effect of the amount of H2O on the error in CH4 column (In Galli et al. we used 40 measurements with varying H2O). Only using two measurements is very tricky as we have no idea of how accurate the established relation is. I would advice to use (much) more measurements to make the results (more) reliable

Reply:

In the revised version of the manuscript, we address all three points raised by Prof. Aben by replacing the 2 FTS measurements (at Karlsruhe) and HITRAN-2012 spectroscopy by the 50 FTS measurements (at Darwin) and the (Scheepmaker et al., 2013), spectroscopy previously used in (Galli et al., 2012). Therefore, A. Galli and V. Velazco are new co-authors. Using the (Galli et al., 2012), setup in general results in smaller residual CH_4 errors than the setup used for the initial submission which implies that the (Scheepmaker et al., 2013), spectroscopy is better suited for our retrievals than HITRAN-2012.

Our initial AMTD study mapped the spectroscopic uncertainties provided by 2 extreme dry and wet FTS retrievals to the whole Earth. In this first study the differences to the (Galli et al., 2012), analysis might have been due to one (or several) of the following factors:

1. Are there differences between the FTS retrieval methods used in (Galli et al., 2012), and for our TCCON-Karlsruhe approach: software, spectroscopy, a priori profiles, average kernels, etc.
2. Are differences due to the fact that our 10000 satellite scenes comprise a broader set of solar zenith angles, surface albedo and meteorological conditions than the 50 retrievals based on the Darwin-TCCON dataset covered by (Galli et al., 2012). Are the 50 FTS retrievals entirely representative of global conditions.

To exclude the first point, we have re-evaluated all our previous results with the same measurements and methods described in (Galli et al., 2012). The figures S1, S2, S3 show the results. We included the SW1 (SWIR1 only) and SW3 (SWIR3 only) retrievals in this reply, in the revised manuscript we have included also SW1+3 (SWIR1 and SWIR3 simultaneously):

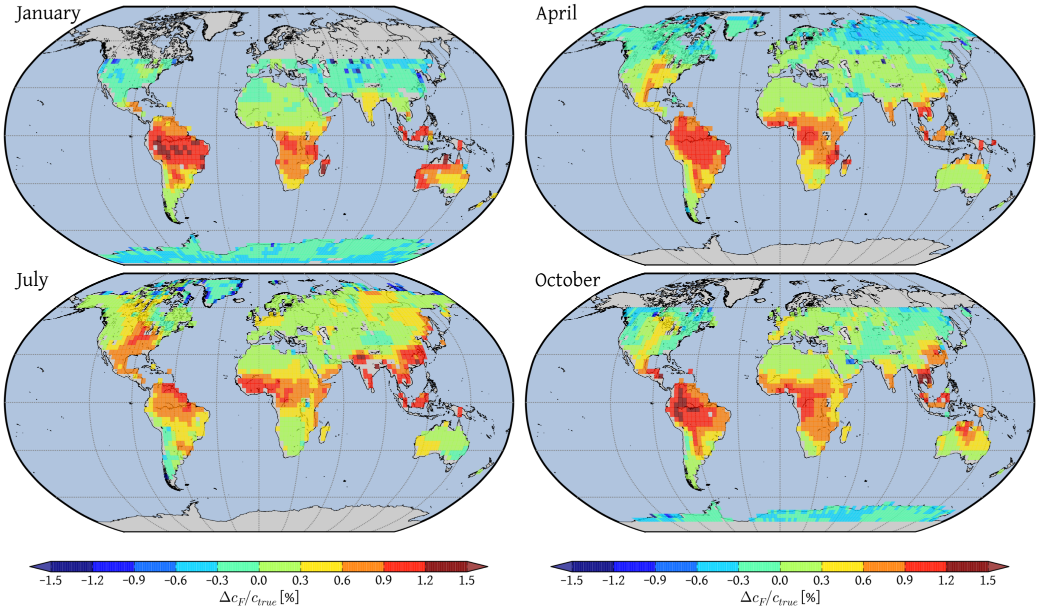

Figure S1: XCH$_4$ retrieval error $\Delta c_F/c_{true}[\%]$ for retrieval concept SW1. Given the (Galli et al., 2012), setup.

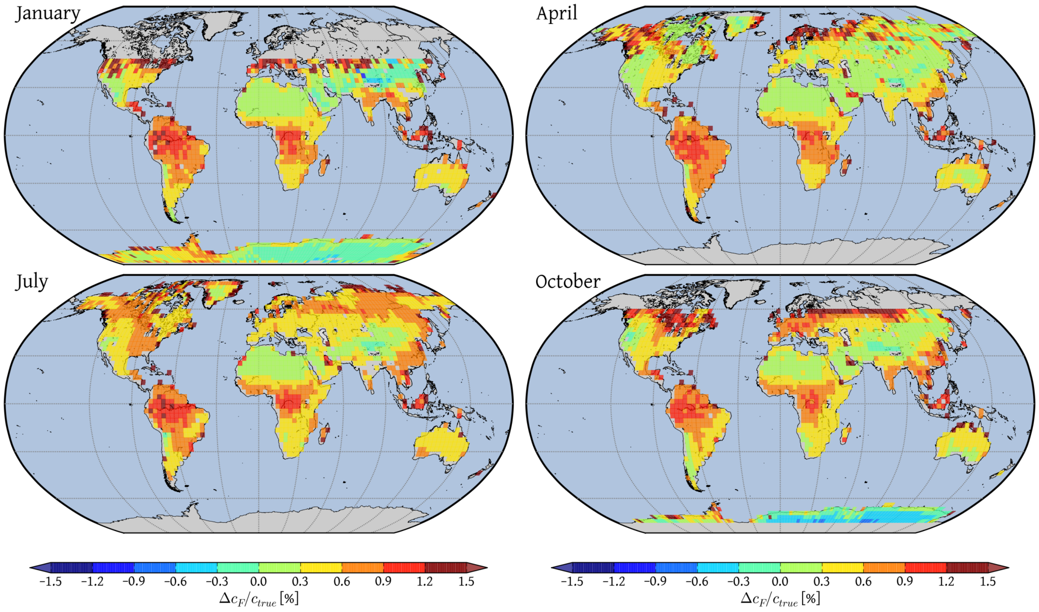

Figure S2: XCH$_4$ retrieval error $\Delta c_F/c_{true}[\%]$ for retrieval concept SW3. Given the (Galli et al., 2012), setup.

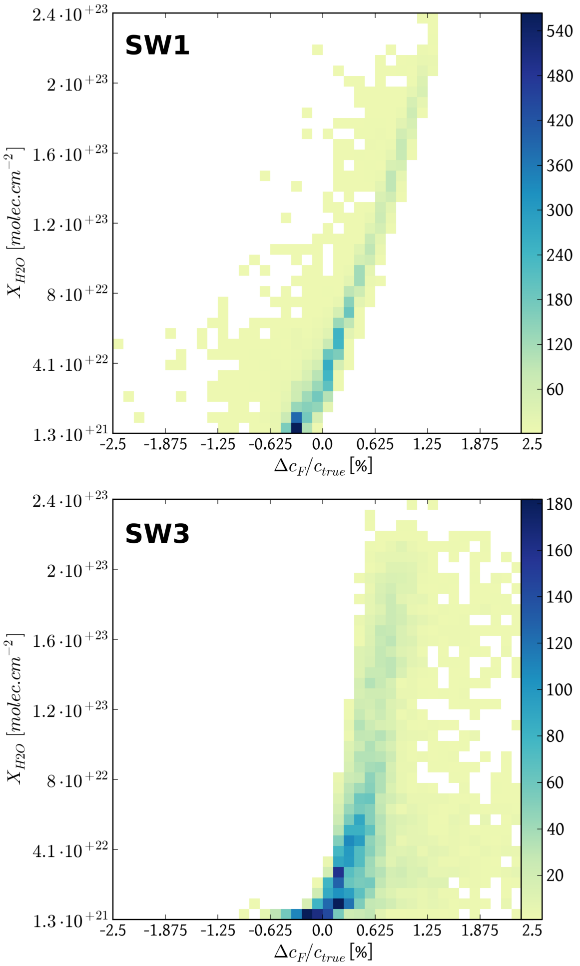

Figure S3: Bidimensional histograms of methane retrieval errors $(\%)$ with respect to XH_2O total concentration values. Analysis of SW1 and SW3 concepts. Given the (Galli et al., 2012), setup

The figures show that even with the same underlying FTS and spectroscopy information we observe correlations between water vapor and residual CH$_4$retrieval errors. A categorical (definitive) answer or calculation to discard the second explanation is not possible in the framework of our hypothesis and datasets

Besides addressing CH$_4$, Figure 8 in (Galli et al., 2012), shows two different CO retrieval analyses based on two slightly different SW3 retrieval windows, i.e. for a wide window and a CO optimized retrieval window. In the case of CO, the choice of retrieval window controls the strength of correlations between CO retrieval errors and total column water vapor. For this reason we have conducted additional, conceptually similar sensitivity studies and we detected differences in the correlation between CH$_4$ retrieval errors and total column water vapor for different choices of the CH$_4$ retrieval window. (Galli et al., 2012), used the $[4190, 4340]cm^{-1}$ window to retrieve CH$_4$. The Figure S4 compares the previous SW3 window: $[4200, 4325]cm^{-1}$ with another one covering the range $[4231, 4303]cm^{-1}$. This smaller window implies smaller dispersion with a larger bias for dry scences.

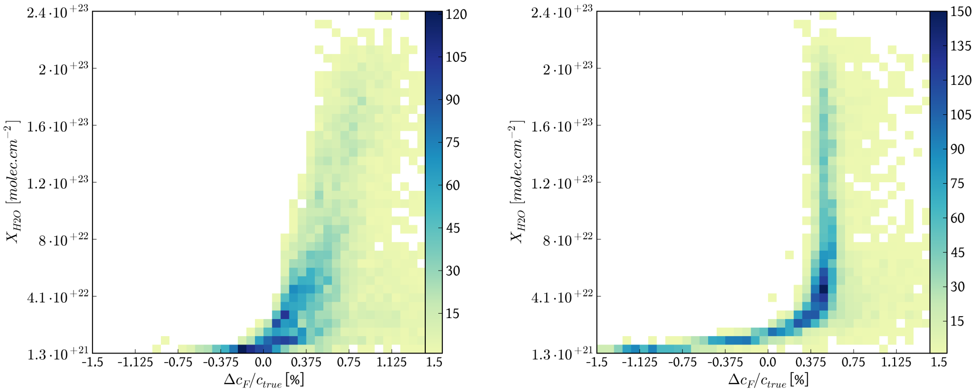

Figure S4: Bidimensional histograms of methane retrieval error $(\%)$ with respect to XH_2O total concentration values. S5P (TROPOMI) concept: SW3 band. Based on the dataset with 50 FTS measurements. Left panel SW3 window: $[4200, 4325]cm^{-1}$. Right panel SW3 window: $[4231, 4303]cm^{-1}$

Concerning the question about the goodness of new mapping based on the 50 FTS study we provide a test robustness. Since a robust mapping should not depend significantly on any specific selection of the FTS measurements, we compare results when selecting subsets of FTS measurements. If the results are similar we expect that our mapping is not contingent on the exact selection of ground-based residuals. We restrict our test to the SW1 configuration. The figures S5 and S6 show that the result for 30 FTS measurements has identical behavior to the standard run using 50 FTS measurements. An alternative approach would be a hierarchical classification of the FTS residuals according to specific properties of the water vapor profiles (for example using divergence measures from reference profiles). Such a classification could be used to refine the mapping of spectroscopic errors. This methodology will produce an adaptive mapping of the spectroscopic errors to representative profiles. However, since our subsampling test does not reveal any substantial difference to the standard run, we do not pursue refined mapping methods here.

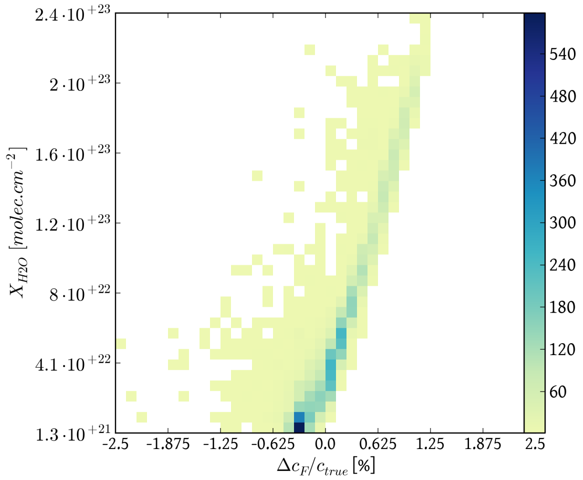

Figure S5: Bidimensional histograms of methane retrieval error $(\%)$ with respect to XH_2O total concentration values. Analysis of SW1 band concept. **Based on a sub-dataset with 30 FTS measurements**.

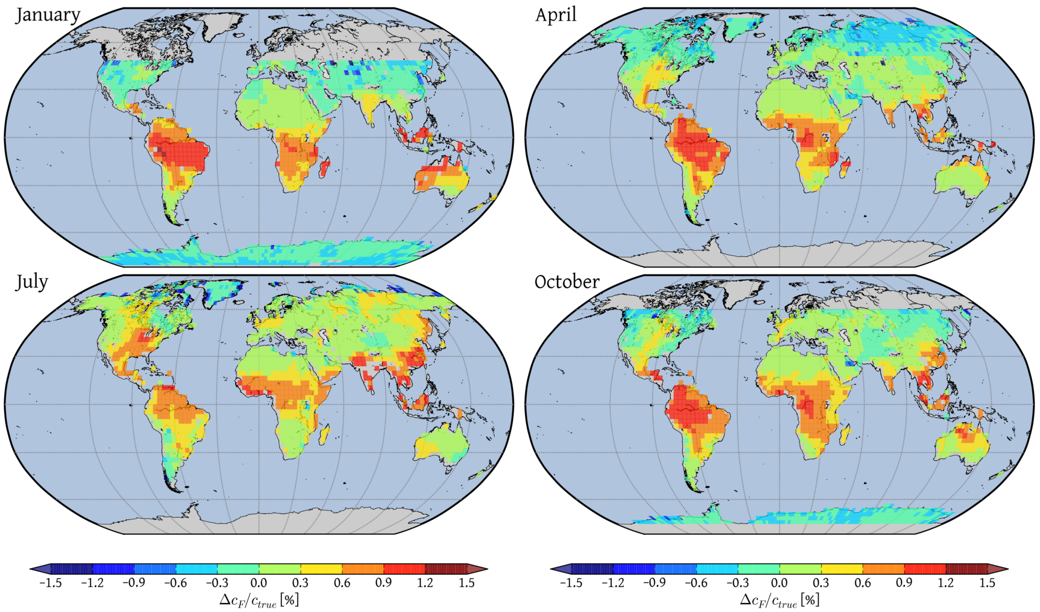

Figure S6: XCH$_4$ retrieval error $\Delta c_F/c_{true}[\%]$ for retrieval concept SW1. **Based on a sub-dataset with 30 FTS measurements.**

Comment from C.Frankenberg

http://www.atmos-meas-tech-discuss.net/8/C552/2015/amtd-8-C552-2015.pdf

The paper by Checa-Garcia et al deals with a very important topic, namely the impact of spectroscopic uncertainties in H2O for global space-borne methane retrievals in the short-wave infrared. Given the upcoming TROPOMI launch, this manuscript is timely, relevant and well suited for AMT. I do have some general remarks that should be considered before publication in AMT though, as outlined below and followed by a short list of small issues:

1. Major remarks: How was the FTS fit performed? I can see that the CH4 profile was fitted but was the same done for H2O? Given the wide range of lower state energies for H2O transitions, its T-dependence can be very large and a total column scaling fit may produce overly pessimistic spectral residuals (i.e. flaw 2). Please explain how H2O was fitted, both in the FTS but also in the satellite case, it may make a big difference as the profile assumption is a hard constrained, forcing the fit to push deviations from the profile into other state vector variables, presumably CH4.
2. The other question to me is the following: How can we potentially mitigate this problem? Yes, more spectroscopic studies are needed but experience shows that these are time-consuming and most likely won't have resolved the problem before the TROPOMI launch. Are there are partial solutions that could reduce this problem? What if you exclude the regions with the highest deltaT values in the spectral fit (or de-weight these regions)? How would results look like and can this be done without harming the CH4 retrieval itself? Right now, I have the impression that more could be done to mitigate the problem at hand and it would strengthen this paper by including such an additional analysis.
3. Also: You propagate a delta-T from 2 FTS stations to the globe, only taking the transmission into account. As I mentioned earlier, the H2O line intensities are very temperature dependent, hence transmission errors at similar H2O columns can actually vary substantially. The more "freedom" you provide the spectral fit to fit these features (e.g. fitting a H2O profile without stringent shape constraints), the less the cross-talk into CH4 will be. Ideally, I would like to see this analysis be repeated with a full H2O profile fit, both for the FTS but also the satellite retrievals. All atmospheric layers for the profile fit could be fitted independently, which can introduce jack-knifing but may help minimize the H2O impact.
4. Figure 1: It covers a wider wavelength range than the final fits using surrogate satellite data. How do spectroscopic errors look like if you shorten the window? The residuals appear rather large, » than 1%. These seem substantially larger than results in Frankenberg et al, where Jenouvrier et al H2O spectroscopy was used. This needs to be double-checked. Also, as shown in Frankenberg et al, the Q-branch was most sensitive; it would be very worthwhile to check the sensitivity after exclusion of the Q-branch in the spectral fit.
5. Review, some language checks needed.
    - Page 1336, line 19: "strives" –> describes
    - Page 1336, line 26: "chained" into the state. Is this a new "technical term" denoting a state vector element with really stringent prior constraints?
    - Page 1337, line 19: does not dispose –> does not include the SWIR
    - Page 1338, lines 5ff: The parameters are not extrapolated, the shape of the far wing is not well constrained so that the theoretical line-shape is not entirely certain. What does "only a small part of the lines was" mean in that context here? Not clear, part of a line (near the center) or not all transitions of a band? Please be more accurate

Reply:

1. The approach for the FTS retrieval used on the AMTD submitted draft its similar to the TCCON science team retrieval using the GFIT non-linear spectral fitting algorithm. Basically it scales an *a priori* profile of the specific interfering absorbers including H_2O. By scaling the different absorbers profiles it estimates the spectrum that best fits the measurements in a least-squares sense. For the target absorber (CH_4 in our study) a vertical profile is retrieved. So, we do not retrieve information on the shape of H_2O profile, i.e. the shape is hard constrained to the *a priori* profile. The Darwin TCCON FTS retrievals included on the revised manuscript is conceptually similar and specific details were given on (Galli et al., 2012).
   We agree that interferences between H_2O and CH_4 absorption lines could be minimized by retrieving an H_2O profile instead of a total column. Our experience indicates that inconsistencies on the spectroscopy between the retrieval algorithm and the measurements would produce non-physical oscillations on the retrieved profile, this is a reason to constrain the shape of the retrieved CH4 profile to mimic the *a priori*. In the case of the vertical profile of H_2O we would expect also an unrealistic profile. But given that our target is CH_4 this will not be critical if the additional degrees of freedom gained in the retrieval could potentially deal with the uncertainty on the spectroscopy mitigating interferences with our target species. Retrieving the H_2O vertical profile - allowing for unrealistic oscillations - implies quite substantial modifications to our inverse method. Therefore, we decided to keep the standard approach as used for TCCON FTS retrievals on one side, and previous satellite performance evaluations for Sentinel 5 Precursor (TROPOMI) [e.g.(Butz et al., 2012)] on the other side. We postpone changes of the inversion method to future analysis, but the conclusions of the revised manuscript mention the mitigation strategy suggested by the reviewer.

2. An optimization of the retrieval window is a possible mitigation meanwhile the spectroscopy information is improved. As commented on the reply to the first reviewer, we have explored possible differences between two SWIR3 windows finding that there is an impact on the retrieval errors with the window implemented on the retrieval. Additionally to answer the question 4 we have evaluated, as well, SW1 window retrievals skipping the Q-Branch. Figure xy shows the resulting XCH_4 errors. Excluding the Q-branch results in negatively biased CH_4 retrievals. The range of errors, however, is only slightly smaller than the standard setup ($\sim$0.5-1%) and the regional correlations persist. Therefore our approach for this paper is to maintain the original retrieval windows (also to be consistent with other sensitivity studies for S5P). The conclusions of the revised manuscript point to the mitigation option to select specific retrieval windows.

3. In general our retrieval was chosen to be consistent with the previous sensitivity studies focused on particle scattering related forward model errors for S5 and S5P and with the standard TCCON approach for ground-based measurements. As commented before, enhancing our inverse method to retrieve the H_2O vertical profile (with more than 1-2 degrees-of-signal in the vertical) is technically challenging and we cannot address it in the framework of this paper. To enhance the representativeness of our FTS dataset, we replaced the 2 FTS measurements from Karlsruhe by the 50 measurements from Darwin used previously by (Galli et al., 2012). We also use identical spectroscopy as in (Galli et al., 2012).

4. Concerning SW1 band we have done an additional sensitivity study excluding the Q-branch. The result is shown on the two figures S7 and S8. In the case of SW1 we have a negative bias with similar magnitude than the previous (in general) positive one. At least on the case of SW1 we cannot recommend exclude the Q-Branch, while for SW1+3 without Q-Branch the general negative XCH_4 errors for SW1 and positive XCH_4 errors for SW3 lead to a better performance which, however, is due to a cancellation of errors rather than due to an improved fit in the individual windows. For the SWIR3 window, we changed the H_2O spectroscopic database from HITRAN-2012 to (Scheepmaker et al., 2013), to be consistent with (Galli et al., 2012). This change indeed reduces spectroscopic errors and thus, the spectroscopy induced CH_4 error. Still the errors found are significant with respect to magnitude and regional correlation patterns.

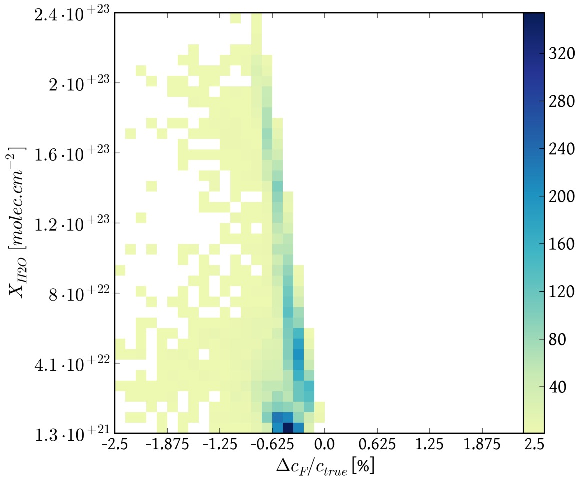

Figure S7: Bidimensional histograms of methane retrieval error $(\%)$ with respect to XH_2O total concentration values. S5P (TROPOMI) concept: SW3 band. Based on the dataset with 50 FTS measurements. **SW1 window excluding Q-Branch**.

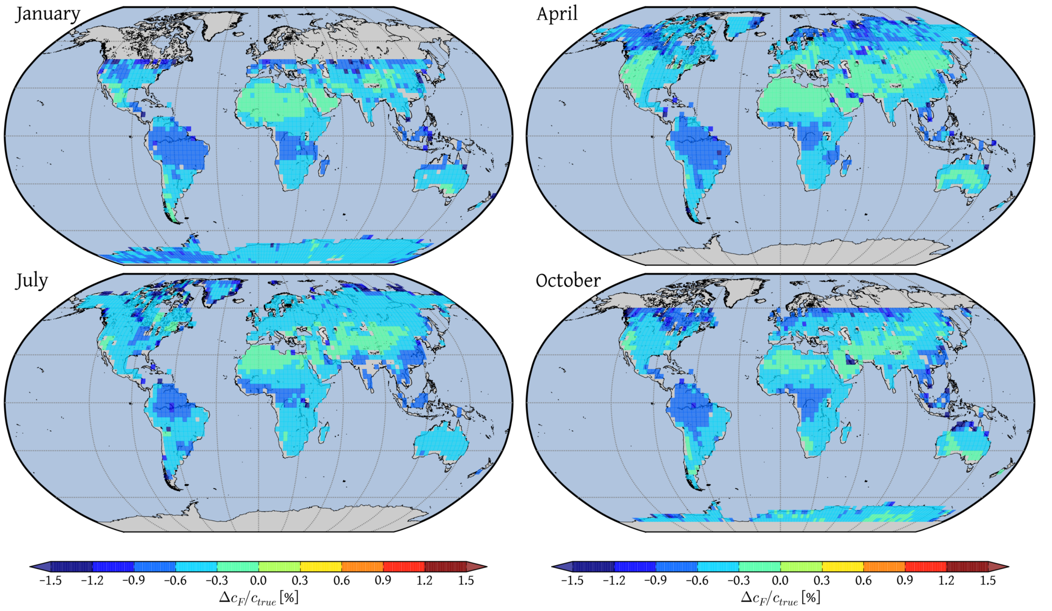

Figure S8: XCH$_4$ retrieval error $\Delta c_F/c_{true}[\%]$ for retrieval concept SW1. ased on the dataset with 50 FTS measurements. **SW1 window excluding Q-Branch**.

5. Reply to minor comments: All the typographical comments are incorporated. Thanks. Concerning the last point it is a short description of the CH$_4$ and H$_2$O spectroscopy line-lists on HITRAN 2008/2012, we will improve these lines on the manuscript.

Comment from Third Anonymous Reviewer

www.atmos-meas-tech-discuss.net/8/C624/2015/amtd-8-C624-2015.pdf

The study of Checa-Garcia et al., which estimates the propagation of spectroscopic errors on the XCH4 retrieval errors obtained from S5 and S5P-like spectra, is very relevant for AMT. The paper is very well structured and clear. Therefore, I recommend the publication of this paper in AMT, after a few remarks have been addressed.

1. If the goal of S5 and S5P is a total accuracy of better than 2% (or 30 ppb) for XCH4, then I would say that the -20 / + 20 ppb XCH4 errors obtained for SW3, even if not perfect, is good enough to achieve the goal, considering that the other main systematic error source has been estimated to less than 1% (Butz et al., 2012). Can the authors explain why they conclude differently ? Are there other expected error sources which, added to the spectroscopic ones, would make the 2% aimed accuracy not possible ?
2. In agreement with the referee C. Frankenberg, I suggest to investigate the role of the H2O profile on the residuals. Only a scaling factor is applied to the H2O *a priori* profile used in the retrievals. The H2O *a priori* profile is taken from a model ECHAM5-HAM, but it is not said if this *a priori* profile is calculated daily or hourly or interpolated to the time of measurement ? Do it improve the fit to, either make a preliminary retrieval of the H2O profile and use it as *a priori* in the CH4 retrieval, either make a simultaneous profile fit of H2O ? Even if only a scaling of H2O will also be used in the “real” S5/S5P retrievals, this test on ground-based data will help to determine if only the spectroscopy is responsible for the bad residuals.
3. The conclusions are different than the study of Galli et al. (2012). This study used 6-hourly H2O profiles from ECMWF as *a priori*. Could this be one of the reasons for the weakest dependence of CH4 retrievals on H2O found in this study (which also only scales the H2O *a priori* profile) ? What else could explain the different conclusions ?
4. The regularization for the CH4 profile retrievals is chosen to obtain 1 Degree-of-freedom (p.1339). Is it the expected maximum DOFS that can be obtained (i.e. no profile information can be derived; a “real” *a priori* covariance matrix would also lead to 1 DOF) ? If not, can a less strict constraint improve the residuals and the XCH4 errors?
5. Fig.8: can the authors provide some explanation for the different correlations observed at different humidity regimes, which is very pronounced in the case of SW3 (especially, for the decreasing interference for very humid cases) ? Is this behavior true at a regional / local scale (to remove the influence of very specific surface properties, such as the albedo, e.g Amazonia or the height, e.g Himalaya) ?
6. Minor or technical comments:
    - p.1335, l.22-26: You could add the information that the 20% overestimation of the tropical CH4 source was due to an overestimation of CH4 in the tropics up to 60 ppb, just to give to the reader the order of magnitude of the spectroscopic error found in this previous study compared to the present one.
    - p.1341, l.18: remove “approximately”.
    - p.1344 (Sect.4): I would prefer a more quantitative assessment of the correlation: e.g. it would be good to provide the correlation coefficients between the XCH4 errors and the AMF for each window configuration, and a similar figure than Fig. 8 for AMF. And for Fig. 8, the correlation coefficients could be provided.
    - p.1345, l.3: “mid-latitudes”
    - p.1355: Explain alpha_gb in the legend of the Table.

> - P1356-1357: Figs 1 and 2 could be plotted in the same figure (a and b) in AMT. I would use the same scale for the residuals in dry and humid cases.
> - p. 1363: Fig. 8: Enhance the font size, please.

Reply:

1. The threshold requirement of S5 and S5P is a total accuracy better than 2% for XCH_4 total column. The goal (defined by the best convenient level of requirement) is 1% for XCH4 total column. This value includes all possible error sources, therefore the total error budget has to deal with all possible forward model, noise and instrumental errors. The full physics retrieval approach described by (Butz et al., 2012) was used to evaluate the forward model error due to the particle scattering. Because currently state-of-the-art of XCH_4 and XCO_2 retrievals can only retrieve "effective parameters" describing particle properties in the atmosphere, they evaluated forward model errors due to this "incomplete" description. That study did not included other error sources, for instance, it assumed a perfect knowledge of the IRSF and spectroscopy. For this reason an additional forward model due to our imperfect spectroscopy knowledge between 0.5% to 1% compromise the total accuracy for XCH4. Beyond the problem of the total error budget we have to deal with regional correlated errors which is problematic for inverse modeling applications. There are several studies on this topic: (Meirink et al., 2006, 2008) concluded that biases around 1% have a role on the derived CH4 emissions, this is because systematic regional bias of 0.5% may imply an overestimation of regional emissions by multiples of the original bias. Our results show systematically correlated errors in several regions of the planet on this order of magnitude. Our conclusion to work on improving spectroscopy is a recommendation.

2. An answer to this question is included in the reply to the 2nd referee. Basically, we indeed retrieve a scaling factor for the H_2O profile but implementing a full profile retrieval (with more than 1-2 DFS resulting in profiles oscillations) is beyond the current assessment. However, the H_2O a priori for the FTS retrievals do not stem from ECHAM5-HAM but from actual meteorological data (ECMWF ERA interim as for (Galli et al., 2012)). The ECHAM5-HAM profiles are only used for the satellite simulations. The revised version refers to the idea of H_2O profile fitting in the conclusions.

3. An answer to this question in included in the reply to the 1st referee. In the end, we cannot track the difference to a specific cause but our answer to referee 1 suggests that the detailed selection of the retrieval windows controls interferences between CH_4 and H_2O.

4. In theory, the CH_4 absorption line shape as well as the large range of absorption optical depth within a retrieval window allows for retrieving CH_4 profile information. In practice, unknown or erroneous forward model parameters such as aerosol scattering properties or spectroscopic parameters blur profile information and hinder profile retrieval. Therefore, artificially enhancing DFS for CH_4 will result in oscillating profiles. It is not obvious, whether this actually dampens error propagation into total column CH_4 or not. For our retrievals from real GOSAT measurements e.g. (Butz et al., 2011; Guerlet et al., 2013), we find a substantial reduction of the XCH_4 (and XCO_2) scatter when allowing for 1.-1.5 DFS instead of 2-3.

5. We have shown on the reply of previous questions on this manuscript that this correlation is depending on the specific retrieval window definitions. In particular for the SW3 case, the retrieval window is relevant for the correlations on very dry scenes. For the SW1 case, differences on the retrieval window change the correlation on very humid situations from positive (+0.65%) to negative values (-0.6%). The information provided by FTS measurements to perform the mapping is not depending on surface properties (for instance albedo). For a given retrieval window configuration and relatively homogeneous surface properties but important differences on total column water vapor concentration we expect the behavior shown on the manuscript.

6. Reply to minor comments:
   - p.1335, l.22-26: *You could add the information that the 20% overestimation of the tropical CH4 source was due to an overestimation of CH4 in the tropics up to 60 ppb, just to give to the reader the order of magnitude of the spectroscopic error found in this previous study compared to the present one.* Thanks for the comment we improved the text.
   - p.1341, l.18: *remove "approximately".* Yes.

- p.1344 (Sect.4): *I would prefer a more quantitative assessment of the correlation: e.g. it would be good to provide the correlation coefficients between the XCH4 errors and the AMF for each window configuration, and a similar figure than Fig. 8 for AMF. And for Fig. 8, the correlation coefficients could be provided.* Concerning quantitative assessment on correlation (i.e. XCH4 errors vs H2O) we have not included such information for two reasons. First due to its dependence with the retrieval window. Second, because Pearson's correlation coefficient rely on several hypothesis of the sampling distribution that we cannot ascertain for the correlation plots analyzed. Depending on the window the relationship can be approximately linear or explicitly not linear. Other statistic parameters describing correlation, like Spearman's rank correlation coeficient, should be more robust given the family of behaviors we found but these analysis are beyond the initial objectives of our study. We are considering a more detailed statistical analysis for future projects, then additional comparisons like q-q plots or mutual information could be informative. Regarding AMF vs XCH_4 error we have included scatter-plots on this manuscript: Figures S9, S10, S11.

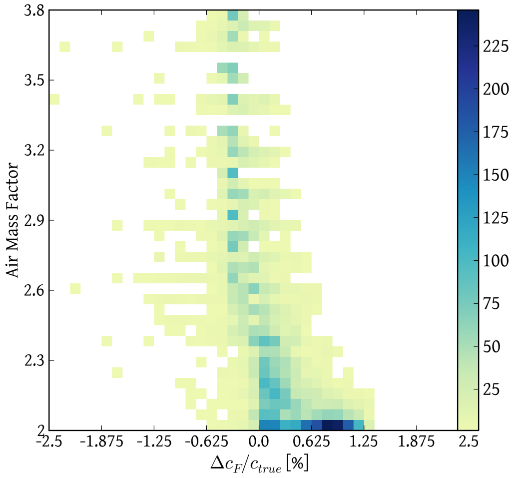

Figure S9: Bidimensional histograms of methane retrieval error $(\%)$ with respect to AMF. SW1 band. Based on the dataset with 50 FTS measurements.

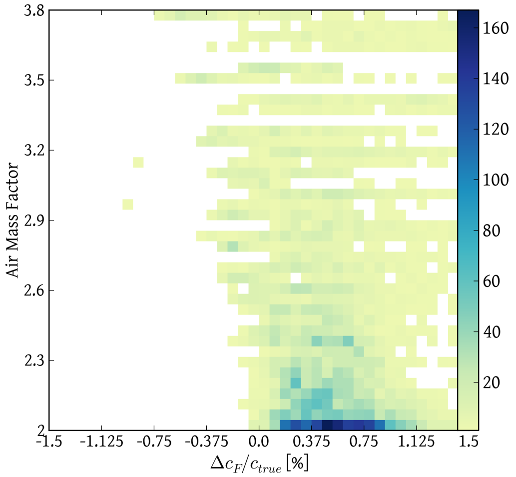

Figure S10: Bidimensional histograms of methane retrieval error $(\%)$ with respect to AMF. S5P (TROPOMI) concept: SW3 band. Based on the dataset with 50 FTS measurements.

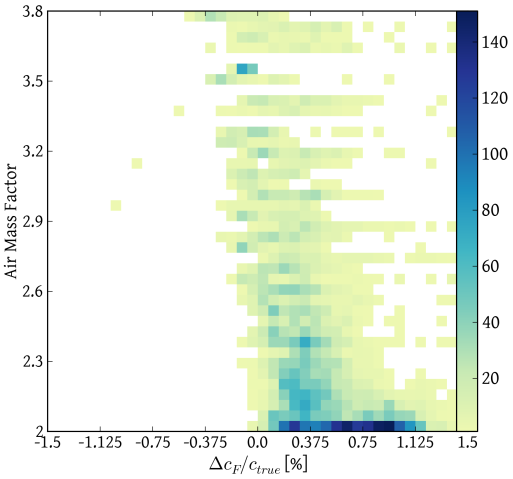

Figure S11: Bidimensional histograms of methane retrieval error $(\%)$ with respect to AMF. S5 concept: SW13 band. Based on the dataset with 50 FTS measurements.

- *p.1345, l.3: "mid-latitudes"*: Thanks.
- *p.1355: Explain alpha_gb in the legend of the Table*: This table is skipped on the new AMT manuscript.
- *P1356-1357: Figs 1 and 2 could be plotted in the same figure (a and b) in AMT. I would use the same scale for the residuals in dry and humid cases*: These figures are different on the new AMT manuscript. The comment has been incorporated on new plots.
- *Fig. 8: Enhance the font size, please*: This figure will expand along a full column on the AMT paper.

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