**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Calibration of column-averaged CH$_4$ over European TCCON FTS sites with airborne in-situ measurements" *by* M. C. Geibel et al.

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General comments

The authors describe the comparison of column average dry air mole fractions of methane retrieved from solar spectra acquired at six European TCCON stations with coincident aircraft measurements. These data are used to derive and refine the calibration of TCCON measurements on the WMO trace gas measurement scale, following the calibration formalism of Wunch et al (2010). Results from the European aircraft calibration campaign make (or will make) an important contribution to the demonstration of site-to-site consistency and network-wide calibration of the Total Carbon Column

Observing Network (TCCON).

Unfortunately the text is often clumsily or poorly worded, and I believe several of the equations are not correct as written. In particular, the iterative calibration approach (which is nominally the author's novel contribution to the calibration analysis) needs to be much better explained: in Figure 11b we have the apparently non-sensical result that one can make a reasonable estimate of the calibration factor even when there is virtually no aircraft data used in the calibration. The manuscript could also be better structured.

Consequently I believe a significant revision of the manuscript will be required before it can be deemed to provide an clear and accurate account of the calibration for the European TCCON sites.

I did also wonder whether the scope and content of the paper was not better suited to AMT, but assume this was considered in the quick review?

Specific comments

Manuscript structure

The introduction should provide a clear and succinct up-front description of the remote sensing measurement process, the reasons why the calibration to the in situ trace gas measurement scales is needed, and how this calibration can be achieved using aircraft data. Introductory material from sections 4.1 and 5 should be moved here and reworked. The column-average dry air mole fraction should also be defined here or early in the 'FTS data processing' section.

Section 2: IMECC campaign

There is insufficient detail on the the aircraft in situ data: What is the continuous in situ instrumentation? How is it calibrated? What are the statistics of the differences

between flask and continuous analyser measurements? The scale used (NOAA04?) should also be noted explicitly.

A revised Section 3 could become a subsection of section 2, with a similar subsection discussing the aircraft instrumentation and data processing.

Section 3: FTS data processing

The text here is weak. Would it not suffice to say all data were processed in accordance with the TCCON data protocol, using the same version of the standard TCCON processing algorithms to transform the interferograms to spectra (including correction for variations in solar intensity) and perform the trace gas retrievals.

It is important to note how the retrieval a prioris were adjusted (non-standard TCCON procedure). I would suggest the information on the GFIT a prioris (last paragraph of Section 4.1) and the adjustment procedure (section 4.2) be moved to section 3. Figure 6 does not provide enough information to be a useful illustration of the method, and should be revised or cut.

Regarding the discussion of the SIV correction, this is part of the standard TCCON data processing, so one wonders why non-standard and suboptimal procedures are discussed in some length here (a more useful discussion might have been the contrast between measurement precision (for retrievals using SIV correction) in clear and partially cloudy conditions i.e. how did sky conditions impact the calibration uncertainty?). The associated figures also raise questions: Why is the spectrum with DC-correction in Figure 4 twice the intensity of the uncorrected spectrum? Why are there not 'DC-corrected retrievals' for every 'uncorrected spectrum' retrieval in Figure 5? I suspect the material here is insufficient for someone unfamiliar with TCCON processing to appreciate the significance of the SIV correction (nor arguably is this the place to do so). On the other hand, we lack information on whether pre- or post-processing QC based on solar intensity variations was applied? Information on the the observation time windows

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which were applied when defining the FTS-aircraft coincidences and post-retrieval QC which are given in the results section (5.1) would usefully be moved here.

If for retrieval biases due to laser sampling errors have been empirically corrected, this should be noted/described here.

Section 4.1

The definitions of the variables in the second term of the right hand side of the equation 1 are insufficient to determine how this term can dimensionally be a dry air mole fraction.

It would be much clearer for the presentation of the methodology if the authors defined the 'smoothed tracer profile' which should be integrated to estimate the tracer vertical column (and then divided by the dry air total column to infer the column-average dry air mole fraction). An equation defining the calibration scale factor would also be useful (more useful say than paragraph beginning line 5 of page 1524). Similarly for a brief description of the calibration regression methodology (in a separate subsection if needed).

The wording of this section is very poor. The presentation would be simpler if the theory for the calculation of the FTS observation equivalent given a perfectly known tracer profile x_{true}, was described first, and issues arising with the approximation to x_{true} given by the aircraft data (x_h) were discussed in what is currently section 4.3 (but will become 4.2 assuming the GFIT a priori discussion is moved to the present section 3). This would also help in the presentation of results in section 6 (see below).

Section 5

The material in the first paragraph of this section should be in the introduction.

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Section 6

I do not follow how the statement 'the aircraft column has to be extrapolated with a calibration-factor-corrected GFIT a priori profile' and the figure 13 can be reconciled with the formulation of equation 3 (which additionally repeats the error noted for equation 1). Denoting the surface pressure P_s and the minimum pressure of the aircraft profile P_{min}, there must now be an explicit summation from P_s to P_{min} (where x_h is given by the aircraft data) and from P_{min} to zero, where x_h is iteratively approximated by $\gamma x_a/\psi_n$. I assume this is a notational error, and will continue to discuss the results presented assuming x_h is iteratively approximated by $\gamma x_a/\psi_n$ from P_{min} to 0 hPa.

The authors go on to present the counter-intuitive result that the iterative calibration converges to a solution which is essentially independent of the pressure range over which aircraft data is available (Fig 11b). To try and understand this behaviour, I extended the simple model presented by the authors to derive the expression for ψ_n:

$$\psi_n = \frac{\hat{c}}{(1-f)\chi_t + f\frac{\hat{\chi}}{\psi_{n-1}}} \qquad (1)$$

where $f = P_{min}/P_s \in [0:1]$, $\hat{c}$ is the FTS retrieval DMF, $\hat{\chi}$ is the mixing ratio inferred from the FTS retrieval and χ_t is the true mixing ratio, but which is only measured in the pressure range P_s to P_{min} (recall in this example $\hat{\chi}$ and χ_t are independent of pressure). Substituting recursively for the ψ_{n-1} gives:

$$\psi_n = \frac{\hat{c}}{(1 + f + f^2 + \ldots + f^n)(1-f)\chi_t + f^{n+1}\hat{\chi}} \qquad (2)$$

Thus in the case where $f = 1$ (no aircraft data) we have $\psi = 1$ for all n. $1 + f + f^2 + \ldots + f^n$ is a geometric series which converges to $\frac{1}{1-f}$ for $f < 1$ i.e. $\psi_n \to \frac{\hat{c}}{\chi_t}$ as $n \to \infty$ for all $f < 1$. For the current example $\hat{c} = \hat{\chi}$ and the method appears to converge to the correct solution for ψ, but only because we have assumed χ_t is a constant independent of pressure.

If we now consider the case where the true tracer mixing ratio is χ_t for $P \in [P_s : P^*]$ and χ'_t for $P < P^*$. In the case of a perfect column measurement A=I and the retrieved FTS DMF

$$\hat{c} = \psi_{true}((1 - \frac{P^*}{P_s})\chi_t + \frac{P^*}{P_s}\chi'_t). \tag{3}$$

In the case where P^* is less than P_{min} the iterative solution as $n \to \infty$ is unchanged ($\frac{\hat{c}}{\chi_t}$), and substituting for $\hat{c}$

$$\psi_\infty = \psi_{true}(1 + \frac{P^*}{P_s}(\frac{\chi'_t}{\chi_t} - 1)). \tag{4}$$

i.e. the solution converges, but not to the true value of the calibration coefficient (unless $\chi_t = \chi'_t$).

In the case where P^* is greater than P_{min}, I believe the results above hold, but with the term $(1 - f)\chi_t$ replaced by $(1 - f)\chi^*$ in Equation 1 and subsequent working, where

$$\chi^* = \frac{(1 - \frac{P^*}{P_s})\chi_t + (\frac{P^*}{P_s} - f)\chi'_t}{1 - f}. \tag{5}$$

and

$$\psi_\infty \sim \psi_{true}(1 + f(\frac{\chi'_t}{c_{true}} - 1)) \tag{6}$$

where c_{true} is the true column DMF, $(1 - \frac{P^*}{P_s})\chi_t + \frac{P^*}{P_s}\chi'_t$.

In these simple case studies the iterative solution converges to a solution for the aircraft estimate of the total column average dry air mole fraction that is equal to the partial column dry air mole fraction in the pressure range from P_s to P_{min}. This may explain the behaviour in figure 11b: the true tracer mixer ratio is fairly uniformly mixed over the pressure range observed by the aircraft so that the partial column dry air mole fractions from P_s to P_{min} when P_{min} is varied from $\sim P_s$–200 hPa are actually very similar.

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There is no doubt the iterative approach presented by the authors addresses a small inconsistency in the calibration methodology of Wunch et al (2010). However the approach and its convergence properties need to be much more clearly explained and a more detailed discussion of the calibration bias (including the difference between the Wunch et al ψ_1 and ψ_∞ solutions) should be given in this paper. This may need to be for a few suitably selected idealised cases, but these are none the less informative. The abstract and conclusions should be revised accordingly.

Additional technical corrections

Abstract

- reword first sentence: 'In September and October 2009 six European ground-based . . . (TCCON) were calibrated for the first time using aircraft measurements.'

- line 6: this sentence should be reworded for readability

- line 11: move 'using similar methods' from the start to the end of the sentence.

Introduction

- I believe the satellite community would say TCCON is only a validation data stream (calibration is reserved for the onboard and vicarious radiometric calibration activities).

- p 1519 line 5: column average dry air mole fractions not 'column integrals . . . in the form of volume mixing ratios'

- I am assuming this section will be rewritten and will not suggest further detailed edits

Section 2

- p 1520 l13: reword as 'Eight flights took place over four days and 20 flig ht hours.'

- p 1520 l22: reword as '... IFS125HR spectrometers operating in accordance with the TCCON data protocol (weblink) with one exception: The Karlsruhe FTS ... '

- p 1520 l27–28: clarify/distinguish continuous in situ data (in contrast to flasks; suggest rewording 'taken on board the aircraft' – it gives me an image of sampling the cabin air!

- p 1521 l 4-5: move 'The overall distance ...' to paragraph starting line 9 of p 1520 or cut

Section 3

- p 1521 l 15: the GFIT residuals have both random and systematic contributi ons so the bracketed text after statistical errors is not correct and should be cut.

Section 4

- p 1523 ' 8-12: These sentences need to be revised when drafting the description of the FTS measurement.

- p 1524 l16-18: cut the sentence 'Within the GFIT analysis ...' or reword a more appropriate description of the retrieval procedure in the 'FTS data proce ssing' section.

- section 4.2: There is a strange mix of discussion and citiation in this se ction. Briefly cite and describe the methodology of Wunch et al (2010). Provide any additional details that you needed to perform the correction and could not infer from their paper if needed.

- p 1525 l19: 'multiplied by' may be clearer than 'weighted by the retrieval scaling factor'. The following sentence is unclear (interjecting information on applying the smoothing in the midst of the discussion on the aircraft profile extension?)

- In general, the continual references to weighting x_a by the retrieval scaling factor are cumbersome. What about stating '$\gamma.x_a$ is the tracer mixing ratio profile corresponding to the FTS column retrieval, and will be referred to as the 'retrieved profile' hereafter.'

Section 5

- p 1526 l4: correct spelling of 'ultimately'

Figures

- The use of 'above' in the x-axis label in Figure 11 could be confusing. If I understand correctly, the pressure shown is the minimum pressure (P_{min}) and the aircraft data are limited to data acquired at pressures greater than P_{min}.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 1517, 2012.