

Interactive comment on “Retrieval of atmospheric CH₄ vertical information from TCCON FTIR spectra” by M. Zhou et al.

Anonymous Referee #1

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The manuscript "Retrieval of atmospheric CH₄ vertical information from TCCON FTIR spectra" by Zhou et al. uses FTIR spectra recorded at six different TCCON sites and applies an alternative data processing: instead of column-averaged dry air mole fractions of CH₄, altitude profiles (or rather partial columns) of tropospheric and stratospheric CH are retrieved. The results are compared to several other data sets for validation:

- standard TCCON CH₄ column data product
- stratospheric profiles from ACE-FTS
- a proxy used in TCCON to separate tropospheric and stratospheric partial columns
- tropospheric and stratospheric in-situ CH₄ profiles at one site

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In my opinion, the manuscript addresses an important issue: a profile data product from TCCON spectra has been requested by several TCCON data users in the past. So, in principle, this study could be very useful to evaluate the possibilities and limitations of such a data product.

Unfortunately, the manuscript fails at this attempt. While the retrieval itself looks fine, the validation of the method is flawed. The most obvious way to validate the method would have been comparing the retrieved profiles to in-situ aircraft and balloon (aircore) profiles. Dozens of such profiles have been measured at various TCCON stations during the last 10-15 years and are available for the TCCON community. However, the authors decided to select six sites of which only one actually had in-situ profiles during the - also arbitrarily chosen - time period. Why did they not use TCCON sites that had aircraft profiles in the past and processed the spectra from those days? It would have been easy to apply the profile retrieval to other time periods. I understand that they did not want to process many more years of TCCON data. But I also do not see a need for using the same time period for all stations in the first place.

As a result, they struggle with the validation of their retrieved stratospheric profiles. They resort to using satellite CH₄ profiles from ACE-FTS where the precision and accuracy is more than an order of magnitude worse than the TCCON standards for column retrievals. Instead, they could have done an extensive comparison with data from the Sodankylä site but for an extended time period. And there are more TCCON sites that have been covered by aircore measurements than just Sodankylä. One is even part of this study: Orleans!

In the troposphere, it is even more puzzling why the bulk of available in-situ aircraft profile data was completely ignored. Even during the same time period, several TCCON sites were overflown by the NASA ATom campaign. And several of the sites that have been used in this study had aircraft profiles taken during the IMECC campaign in 2009. Why did they not process spectra from that time period for these sites instead? There is no obvious reason why they limited themselves to 2016-17.

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Given these issues, I cannot recommend publication without major revisions. The authors should try their profile retrieval on stations and time periods with corresponding aircraft or aircore in-situ CH₄ profiles. If they do so, the whole section 3.3 (ACE-FTS comparison) could be dropped. The section 3.4 (TCCON proxy) would still be useful to evaluate if the proxy method or a profile retrieval provides better results.

General comments:

Why were these sites selected? Among those, Sodankylä is the only one where in-situ profiles were used to validate the retrieval. Other sites do have profiles but they were ignored. To my knowledge, two sites never had any kind of in-situ profiling but were included nonetheless.

Aircraft/aircore profiles at the selected sites:

- Sodankylä: many during several campaigns (but only a limited set used here)
- Bialystok, Bremen, Orléans: aircraft profiles taken 2009 but not used here. Data published in Geibel et al.: Calibration of column-averaged CH₄ over European TCCON FTS sites with airborne in-situ measurements, Atmos. Chem. Phys., 12, 8763-8775, <https://doi.org/10.5194/acp-12-8763-2012>, 2012.
- Ny-Alesund: never
- St Denis: never

ATom visited several TCCON sites in 2016/17, the data is publicly available. Why was this data set completely ignored?

Why was the time frame 2016-17 selected? There is no special reason given and I cannot see one that requires to try the SFIT4TCCON on the same time period for all sites. It would make much more sense to use it on periods with available in situ profiles for each site.

Why was St. Denis used as the prototype? It is probably the wettest site of all and not

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necessarily characteristic for the others.

Specific comments:

The data product is called SFIT4TCCON but - unlike the name suggests - it is not an official TCCON data product. Was the use of the name "TCCON" approved by the TCCON PIs? I am a TCCON PI myself but I am not aware of this.

Section 2.2.1:

- p. 4, l. 11: Diurnal variations above St Denis are assumed to be small. What about the biomass burning season, would this still be true? At least there should not have been much biomass burning activity during the chosen time frame. It would have been easy to check other sites TCCON sites for these criteria. Also, I do not understand why the band with the highest correlation between CH₄ and dry air pressure is best choice.
- p. 6, l. 8-10: the " $Sa^{-1} = \dots$ " should be a numbered equation. The quantity "L1" is not described at all, "T" only very briefly. Provide a reference linking Tikhonov 1963 to the Sa matrix. In the cited reference, Tikhonov certainly did not refer to Rodger's original work which came years later (1976).

Section 2.2.4:

- How was the SFIT4 column AVK calculated?

Section 2.2.5:

- 5% uncertainty might be fine for St Denis sources but may underestimate the variability closer to CH₄ sources. Check available aircraft profiles (e.g. from ATom, HIPPO etc.) as well as aircore to get a better grip on the expected variability. The model data might be too smooth.

Eq. 7: Why did you use pressure and not the O₂ column for the calculation dry air column? There are good reasons why TCCON abandoned surface pressure as a proxy for airmass years ago.

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Section 3.2

- It is doubtful that surface in-situ measurements from 20 km away are representative for St Denis. If you think otherwise, you should provide some reasonable arguments.

Section 3.3

- I understand the need to find something to compare the stratospheric profiles with. However, given the very limited precision and accuracy listed here for the 10-year old ACE-FTS data (25%), I wonder how useful this comparison can be. One might be better off comparing to zonal means with better statistics. That might also provide profiles for the stations where reasonable co-locations could not be found.
- If you decide to make the major revisions that I recommend, this whole section could be dropped.

Section 3.4

- Due to the H₂O interference in the HF microwindow, I would trust the N₂O method more for a tropical site like St. Denis.
- p. 14, l. 10 to p. 15, l. 4: it should not be too difficult to find out which of the raised possibilities 1) to 3) are true. The source code for the TCCON retrieval as well as the handling of the a priori profiles is available.

Section 3.5

- I believe that the Aircore measurements are the most trustworthy validation source for the tropospheric and stratospheric profiles. There are other TCCON sites which had simultaneous aircore profiles taken. Why were these not used? This would also improve the statistics in Fig. 11, which suffers from the fact that a very limited range of XCH₄ was compared.

References:

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- All the TCCON dataset citations are wrong: they still refer to Oak Ridge National Laboratory from which the TCCON data archive moved away already in October 2017! Strange how this can happen when several TCCON PIs are listed as coauthors.
- Acknowledgments: the authors should check if the acknowledgments are in line with what is required by the TCCON Data Use Policy.

Minor comments:

- p. 3, l. 15: I would not call that "noise".
- p. 3, l. 21: Please explain the ATM acronym at least once.
- Fig. 2: poor color choice for dry air pressure as green is already used for band 3.
- p. 6, l. 5: should be "constrain" instead of "constraint"
- p. 7, l. 5: should be "panel" instead of "penal"
- p. 8, l. 17: "The other retrieved parameters have do not contribute ..."
- p. 8, l. 28: "retrieved CH 4 total column are 3.2 and 0.5%" units missing
- Table 5: Why not just write "<0.1%" instead of "-"?"
- p. 9, l. 2: "applied to" instead of "applied for"
- p. 11, l. 1: Are there really no authors and no better reference for the EDGAR database?
- Eq 6: TC is probably total column but should be explained
- p. 14, l. 3: "systematically" instead of "systematic"
- p. 14, l. 4: drop "relatively". It is also absolutely higher.
- p. 14, l. 5-6: the sentence started in line 5 should end with ", respectively."
- p. 18, l. 23: The URL points to the TCCON Data Archive, not to the TCCON wiki.

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