Referee Report: amt-2020-86

"On the consistency of methane retrievals using TCCON and multiple spectroscopic databases"

This paper details the impact of using 5 different spectroscopic databases (GGG2014, GGG2020, GEISA2020, HITRAN2016, SEOM-IAS) on the retrieval of 12CH4 DMFs in 4 different spectral windows from TCCON measurements. The retrieval is performed with the GFIT retrieval algorithm from the GGG2014 environment. Retrievals are carried out on approximately a year of data at 4 TCCON stations (Ascension Island, Darwin, Ny Alesund, Tsukuba), chosen to have an optimal coverage of different atmospheric and measurement conditions (temperature, water vapour, SZA). A second objective of the paper is also to assess the impact of the spectroscopy and fit windows on the retrieval of 13CH4 (and indirectly, the d13C) and check for consistency of the results.

The scientific work is extensive, with a lot of data processing performed to represent retrievals under various atmospheric and measuring conditions. The presentation and analysis of the data is sometimes a little laborious, since so many different parameters are considered (spectroscopic database, retrieval window, TCCON sites). Still, the paper mostly presents the results adequately and underlines the important results. The results are scientifically relevant and interesting, and are certainly within the scope of AMT.

However, the paper sometimes lacks in style and clarity. It is obvious that it has been through some re-writes and major changes, and it should be proof-read and improved in terms of flow and clarity. I would recommend publication of the paper in AMT pending some minor revisions of the manuscript, as detailed below. Note that I started to note down many typos or problematic sentences in the first half of the paper but was not as systematic in the second half and would recommend to proof-read this more thoroughly.

Comments

1/ The abstract should be improved.

a) The third paragraph is especially difficult to read. It would be helpful to be more quantitative in your description of the impact. The sentence "We also find strong evidence that different windows in different spectroscopic databases exhibit different levels of sensitivity to changing local conditions such as light path length and water vapour" is technically correct, but it is so general it becomes almost meaningless.
b) You should mention the work on the 13CH4 retrieval and d13C determination.

2/ The introduction should be re-worked to improve clarity and fluidity:

a) I find it strange that a paper studying the impact of the spectroscopy on the retrieval of methane does not directly discuss the spectroscopy aspect of the retrieval in its introduction. Maybe add 1-2 lines to describe how it is relevant?
b) L44-50 should be at the end of the introduction, detailing the content of the paper. Up until that point, methane isotopologues have not been mentioned at all, maybe add a line about this aspect of the work as well beforehand.
c) I am not certain of the necessity of Table 1. One could either have a table with more information (a graph of the respective instruments and their spectral coverage), or a sentence in the text? Since you discuss TCCON windows shortly thereafter, maybe it would be a good opportunity to describe the TCCON windows used for the CH4 retrieval there (instead of referring to Table 4)?
d) L62-66: Maybe I misunderstood the point being made, but it seems simplistic to suggest that the retrieval window differences between TCCON and TROPOMI may be the major source of biases in the validation of the methane product, especially when many other factors will affect the quality of the retrieval on both GB and satellite platforms. This is surely an interesting point to study, but this seems to inflate the importance of the retrieval window on the validation.
e) L71: "We can infer some of the potential spectroscopic related biases in satellite retrievals…": yes, but since the resolution of TCCON and satellite instruments are quite different, TCCON should be much more affected than satellite retrievals, maybe something to mention.
f) L75: I think that Eq. 2 should be moved here. [?]

3/ L119: "GFIT assumes a fixed profile shape for each trace gas, …": you should mention how the a priori is determined

4/ L198-201: Is there already an estimate of the errors due to these assumptions? It would be interesting to have some figures on this in the paper.

5/ Section 3

Before analysing the retrieval data, I would *suggest* to first perform a comparison of synthetic spectra using fixed state vectors with different spectroscopic databases to look at the expected impact in terms of transmissions in various windows. It might also be interesting to look at the contribution of H2O and CH4 to the total transmission, and hence have a better feeling for which windows should be more sensitive to the water content (would help with the analysis of section3.3?)

6/ Section 3.1

A slightly more exhaustive analysis of these results would be interesting. Could you associate some of the more prominent residuals with species? For instance, in window 3, HITRAN and GEISA have large discrepancies around 6001 cm-1 compared with GGG, why is that? Are there clear improvements due to the use of non-Voigt parameters?

7/ Section 3.3

In this part of the analysis, you sometimes seem to confuse correlation and sensitivity, which are quite different, or only consider one of these aspects. For instance, on L346 you state that "results from Ascension Island, which operates at lower SZAs than any of the other TCCON sites considered in this study also indicates large correlations, suggesting further complexity". The truth is that the sensitivity (slope) of the linear fit to SZA is close to 0, so despite a large correlation, very little effect is observed.

On L348-349: "For window 1, there is weak sensitivity to water vapour variations at the Ny-Ålesund and the Darwin sites," where the sensitivity is substantial for Ny Alesund in Window 1, but correlation is weak.

Section 3.3 could also gain from an indication of statistical significance in the results. It is mentioned on L385 that large p-values were found in some cases, I would suggest identifying clearly results that are not significant, and state at what level you consider these not statistically significant.

Typos and technical corrections

Comments with a "[?]" at the end are for your own consideration

L1: "methane retrieving satellites" -> "methane retrieving satellites instruments" [?]

L3: "is as important as when TCCON…" -> "is as important now as when TCCON…"

L6: the 'S' in "Spectroscopic databases" should not be capitalized

L7: Leave out spectroscopic databases in "TCCON GGG2014 and GGG2020 spectroscopic databases;", as it is redundant [?]

L14: consider changing "~x3" by "up to three times" [?]

L16: "vapour. Such" -> the period should be a comma, and the Such should be lowercase. But you should consider changing this sentence as it is very long and uses 5 times the word "different"

L24: "bottom up" -> "bottom-up"

L25: "top down" -> "top-down"

L28: "S5P" -> "Sentinel 5-Precursor (S5P)" , "TROPOMI" -> "TROPOspheric Ozone Monitoring Instrument (TROPOMI)" . These acronyms have been defined in the abstract, but consider adding a proper acronym description in the full paper as well. [?]

L40: "will therefore be relying" -> "therefore rely"

L43: "from the 6000 cm-1 spectral range": a range has two values. Either define the window precisely, or reformulate

L44: "make" -> "perform"

L48: "…quantify the variations in retrieval abundances when using five separate spectroscopic databases,…" : this seems like the main aspect of this paper, it should probably be mentioned first.

L50: "are studies" -> "are studieD"

L50: consider a reformulation of this sentence

L52: (an weighted …" -> "(a weighted…)

L81: Either state "98.9% and 1.1%", or "99% and 1%", but it should add up to 100%. Considering the "roughly" in the sentence, I would go with 99% and 1%.

L86: "structured as follows," -> change the comma to a colon

L90: "…conclusions are shown…" -> "conclusions are drawn" [?]

L100: Add a comma after "In this study"

L100-101: "which is summarised briefly here." -> "briefly summarised here."

L101: "A forward" -> lowercase "a"

L104: "fit" -> "fitted" [?]

L105: after "In the case of GFIT", add a comma

L106-115: I am not convinced that these are essential to the papers and may be omitted. Maybe reconsider listing these, and put a reference instead [?]

L116: confusing wording: "Note that not all of the above are not routinely…" I imagine that you should omit the second "not".

L116: Start a new sentence with "For example" [?]

L124: "the 7885 cm-1 spectral range multiplied" -> a spectral range needs 2 numbers. Maybe use a formulation such as "in the vicinity of 7885 cm-1…" [?]

L127: Not sure if it's necessary to state the resolution again, which was already mentioned on L57

L131-132: not sure if this sentence needs commas after "window" and "study" or if they are optional [?]

Table 4 has a beginning o f a sixth row

L135: is there a reference for the "standardised process"?

L137: this sentence should not be its own paragraph

L158: "Further to exploring the…" -> maybe "Beyond exploring …" [?]

L167: "line mixing" -> "line-mixing"

L169: "They find a 1.1% difference in total methane…" -> is it a positive or negative bias? Maybe just add a "+" to emphasize [?]

L184: "standard methane window" -> either "standard methane windows" (there are 3 of them, right?) or, better yet "standard TCCON CH4 product"

L185-189: These are technically correct sentences but are a bit long and wordy. Maybe consider adding equations ? [?]

L194: This sentence should not start a new paragraph, but simply continue the previous one.

L196: there should not be a period before the "2)". Maybe a semi-colon?

L197-198: "apriori" or "a priori" -> be consistent, I have seen both in the paper

L202: “cross sections” -> “cross-sections”

L204-207: consider re-writing this sentence, it’s a bit convoluted. Maybe with more than one sentence?

L208-209: you probably should not hyphenate an acronym

L213: leave out “metric”

L214: you repeat “varies in the total”

L216: “<<1‰ in…”: are these French quotation marks (<<) or is it the symbol “much smaller than”? If it is the latter, it should be bigger, if it’s the former, the close quotation mark is missing. Also, is this figure for a single measurement? monthly mean?

L221-223: “Nevertheless precision errors will be low due to the nature of TCCON, and through the fact that TCCON sites are situated in a fixed position, allowing for long term averaging to reach a required precision target.” -> This sentence is unclear (especially the second part: “through the fact…”). Also, it should be “long-term”, not “long term”.

Figure 1: Would it be possible to plot or mention the instrumental uncertainty along the residuals?

L238: “shown in Fig 2,” -> “presented in Fig 2,” [?]

L239: “Fig 2 is the fit” -> “Fig 2 is that the fit”

L247: Maybe start a new paragraph with “There are differences in the…” as you focus more on the TCCON sites now, not on the windows?

L256: “Likely reasons” -> for what? If you refer to the last paragraph, you should stay in that paragraph, and state more clearly what you mean.

Figure 2: Maybe indicate in one plot (window4?) the number of spectra used for the statistics? I know this information is available in Table 2, but this is always nice to have this information on hand.

Figure 2 caption: maybe indicate the colour of each statistics in parenthesis: “Each subplot shows the RMSE (black) and chi2 values (blue) …”

L263+264: “fit” -> “fitted” [?]

L263: “there no specific” -> “there are no specific”

L267: “GESIA” -> “GEISA”

L270: “the the” -> “that the”

L270: “retrievals each database” -> “retrievals for each database”

L274-275: This is not a complete sentence, there seems to be missing something (maybe Firstly we present the results?)

L289: “from in” -> delete “in”

L290: “the standard in” -> “the standard retrieval in”

L304: “sides” -> “sites”

L327-330: This sentence encapsulates nicely the differing atmospheric conditions of the various TCCON stations. Maybe it should be moved to the introduction to better explain this part of the work [?]

Figure 8: This figure is not very easy to read, but I am not quite sure how to improve it. It does give a good overview of the results. Maybe instead it could be a pcolor-type graph for the slope of the linear fit and the intercept for each station and parameter sensitivity (each row of the graph could be a different spectroscopic database, each column a different window?). Just a suggestion

Figure 8 legend: "GEISA", not "GESIA"

L334: The description of Figure 8 is really not clear. Could you be more precise in what is being shown in the figure instead of "the qualitative distributions"?

L342: "indicate the retrieval" -> "indicate that the retrieval"

L345: "creep" -> maybe find a better word [?]

L349: "opposite databases" -> what are opposite databases?!

L362: "vapour. Thus" -> should be in the same sentence "vapour, thus"

L396: "in the calculation" -> its more in the analysis of the results than the actual calculation of the value…

Table 5: The table should be slightly improved in such a way that there is less text. Maybe use sub-rows for each station, one for each database (basically like it's done now, but just mention the database name once, not in each cell…). Also, would it be possible to either add the uncertainty on the d13C value, or mention an order of magnitude in the text? This would add important information to the discussion of L398-401 about the uncertainty.

L398: "that we calculate the mean uncertainty" -> "that the mean uncertainty…is between…"

L404: "show results" -> "are"

L407: "combination" -> "combinations" [?]

L420: "Further to this" -> "Furthermore" [?]

L447: "question that is of some interest to the community, "can we calculate realistic and constant 13C values from TCCON"." -> this formulation is a bit strange to me, maybe use colon after community (instead of a comma), or change the formulation to something like "question that is of some interest to the community, namely whether it is possible to calculate realistic and constant 13C values from TCCON."

L453: "calculate" -> "calculated"

L459: "this is an assumption based" -> "it is based"