

Interactive comment on “On the consistency of methane isotopologue retrievals using TCCON and multiple spectroscopic databases” by Edward Malina et al.

Anonymous Referee #2

Received and published: 20 July 2020

The paper by Malina et al. reports on the consistency and quality of four spectroscopic databases for methane (CH₄) absorption parameters in the shortwave infrared spectral range. The evaluation tool is measurements of atmospheric absorption spectra collected in direct-sun geometry at high spectral resolution across a range of atmospheric path lengths and meteorological conditions. Topic-wise, the study is of interest to the readers of AMT since the respective spectroscopic parameters are relevant for ground-based as well as current and future satellite remote sensing experiments which aim at CH₄ column concentrations with sub-percent accuracy.

The study is based on state-of-the-art, high quality data. But, the paper is not written

Printer-friendly version

Discussion paper



well. As I already indicated in my technical review for the discussion phase, the paper provides too much specific information without guidance which piece is important and, on the other hand, the paper is too superficial in some of the important aspects. If to be published, the paper needs to be made more concise and to-the-point having in mind the interests of the atmospheric sciences community.

In that regard, I recommend dropping the entire 13CH₄ discussion since it is essentially impossible (for the thinkable future) to perform a remote sensing experiment that yields errors low enough to make it useful for scientific purposes.

But I highly recommend discussing in more depth the following aspects. The key challenge of methane remote sensing is avoiding retrieval biases that correlate with geophysical conditions in a way that the errors mimic source (or sink) patterns. Reported and notorious candidates for such spurious errors are correlations between CH₄ and H₂O absorption (e.g. inducing tropical biases), correlations between CH₄ and the lengths of the lightpath (e.g. inducing seasonal or high-latitude biases), and wrong CH₄ profile shapes in the stratosphere. TCCON is known for requiring an empirical lightpath correction for low sun observations which, presumably, originates from deficient spectroscopic parameters. A meaningful evaluation of spectroscopic databases and retrieval windows requires addressing and quantifying these dependencies. The study here does not really include these aspects although it uses a range of spectra from two TCCON sites which could serve the purpose (e.g. plotting retrieval differences against H₂O content or against path length). The study resorts to a sensitivity assessment with perturbed a priori and meteorological parameters which is not indicative for real atmospheric performance in particular since the perturbations are mere educated guesses. The most important sensitivity, the one to the stratospheric profile shape, is deliberately left out.

The required changes to the manuscript are at the edge of what seems possible under a major revision.

[Printer-friendly version](#)[Discussion paper](#)

Specific comments:

P1 L18: What are “pressure cross sections”? Do you refer to the pressure dependence of molecular absorption cross sections?

P3 L72: “an average of three retrieved values from three TCCON fit windows” Why is CH₄ separately retrieved from different windows and averaged a posteriori? If this is a simple arithmetic averaging, it tends to compromise the entire idea of using a maximum likelihood or an optimal estimation technique.

P3 L76: The discussion of the 13CH₄ retrievals lacks the point that atmospheric gradients will be even smaller in column-average concentrations than in in-situ observations.

P4 L107: “least-squares optimal estimation” To the best of my knowledge, it is either least-squares (cost function without side-constraint, maximum likelihood principle) or optimal estimation (a priori departure as side-constraint, Bayesian statistics principle), not both together. From the list of retrieval parameters, it looks like the inversion problem is well suited for a pure least-squares technique. Optimal estimation would be required if the problem was ill-posed e.g. when retrieving concentration vertical profiles instead of column scaling factors.

P5 Table 2. Windows 1 and 5, 4 and 6 are only different because of technical details of the particular software used (apparently there can only be one target molecule per window). This is not of interest to the general reader and the discussion could be simplified.

P7 L175: The metrics should include the definition of the column-average mole fraction and its “X” notation. Later (section 3.3), the X-notation comes as a surprise.

P8 L210: Using CO absorption as a proxy for 13CH₄ is inadequate, since precision and accuracy requirements for 13CH₄ are factors higher.

P9 L256: I think the term “a priori errors” is misleading since not all parameters that are perturbed are retrieval parameters (for which an a priori exists).

The assumed perturbations for the parameters are not based on actual real-world problems reported previously. For example, simply assuming a 2% bias in the methane total column will not yield any significant error sensitivity because the retrieval of the total column is unconstrained. The a priori assumption that causes problems for the CH₄ retrieval is the shape of the a priori profile e.g. assuming a mid-latitude stratospheric profile where there is in reality a polar vortex profile. Likewise a perturbation of the water vapor column by 10% cannot assess the actual real-world problem that spectroscopic interferences between CH₄ and H₂O might induce spurious gradients between the mid-latitudes and the tropics since the H₂O content differs by factors, not just 10%. I would recommend replacing the entire perturbation study by a correlation analysis how CH₄ retrievals from TCCON spectra (from Tuckaba, Ascension Island, and possibly other stations to increase the parameter range) correlate with H₂O content, CH₄ stratospheric profile conditions, and path length dependencies and how these correlations are better or worse for the various spectroscopic databases.

P11 Fig.1: What is the spurious error (as a function of path length, season, latitude) that the difference of 12CH₄ and 13CH₄ averaging kernels could imply for delta-13CH₄?

P12 Fig.2: The figure is much too small and contains unnecessary information such as the CO and HDO transmittances. The four spectroscopic databases are indistinguishable. I would recommend dropping all panels except for the residuals – if a residual is clearly correlated with a molecular absorber you can put a symbol on the residual peak.

P16 Table 4: The table is tough to understand. It would be helpful if the table indicated which window is 12CH₄ and which one is 13CH₄ since the definitions are far away. I recommend adding an extensive caption.

P21 Fig.7 and 8: Together with a very short and superficial discussion on page 22 the figures are largely useless (too small, point clouds masking each other). Drop for the sake of conciseness.

[Printer-friendly version](#)[Discussion paper](#)

P23 Why is the section called “a priori” error (similar question as for section title 2.4.2)? The section reports on the perturbation of the imposed temperature profile. Since temperature is not a retrieval parameter but a forward model parameter, there is no corresponding a priori. Probably, the title should rather be “parameter error”.

P23 Fig 9: Units missing. Wouldn't it be the purpose of the sensitivity study to distinguish between the performance of the four databases with respect to temperature errors propagating into the retrieved CH₄ columns? From Fig.9 I cannot find any conclusive hint in that regard. Fig. 10 is more useful.

P24 Fig 10: “perturbed pressure column retrievals” Isn't it temperature perturbation?

I do not comment on many editorial issues: acronyms need to be defined at first usage, jargon of the spectroscopic or TCCON community should be avoided, inconsistent use of parentheses when referencing, extensive use of the empty word “value”.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2020-86, 2020.

[Printer-friendly version](#)[Discussion paper](#)