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Interactive comment on “Long-term validation of total and tropospheric column-averaged CH₄ mole fractions obtained by mid-infrared ground-based FTIR spectrometry” by E. Sepúlveda et al.

Anonymous Referee #2

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Sepulveda et al. investigated two retrieval methods for CH₄: profile scaling and profile retrieval. They used 4 microwindows in the MIR region. They showed that the profile scaling procedure leads to large smoothing errors due to changes in the CH₄ profile shape. Furthermore, they showed that applying a posteriori correction using HF only removes part of the stratospheric CH₄ contribution and does not result in a good estimate of tropospheric XCH₄. On the other hand, they showed that profile retrieval allows for a good estimate of tropospheric XCH₄ amounts. They compared this with GAW CH₄ data, which resulted in a very good agreement.

A retrieval guideline for NDACC CH₄ products has been under scrutiny for quite some

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time because CH₄ measurements using FTIR spectrometry in the MIR region is a very challenging task. The objective of such a guideline is to achieve CH₄ measurements in the MIR that are comparable or close to the high-precision requirements of TCCON. Since many NDACC stations have MIR measurements that go as far back as 15 years before TCCON started, the NDACC CH₄ time series could be a valuable complement to the TCCON time series, once a working strategy for the NDACC CH₄ is established. The work of Sepulveda et al. is very significant in paving the way to achieving this goal.

I completely agree with the comments of reviewer 1 (Geoff Toon) regarding the whole profile scaling retrieval using the HF correction procedure introduced by the authors. I think this manuscript is publishable in AMT once this and a few minor issues that I'd like to point out have been resolved.

Minor issues:

1. I think there has been a reasonable overlap between the NDACC MIR and TCCON NIR data for Izana. So I think that it would be useful to show comparisons of retrievals of tropXCH₄ using the true method of Washenfelder et al., 2003 and the profile retrieval method shown here. I think this would complete the paper.
2. If I understood it correctly, the profile retrieval uses optimal estimation with a Tikhonov-Philips regularization, which means the retrieved profiles are constrained by an a-priori profile and its shape. This also means that the variability of the retrieved profiles are constrained by the variability of the a-priori (reflected in the Sa matrix). Therefore, one can achieve a very good precision in the retrieved values simply by adequately constraining the retrieval through the Sa matrix, or if the retrieved molecule exhibits small variability from the climatology used. Therefore the extent of variability in the retrieved data may not be the best measure of precision, because the Sa can be tweaked or "tuned" or influenced by climatology.
3. Page 1397, Lines 9-10: The authors mention: " Indeed, we observe a strong anti-correlation between the HF amounts and the CH₄ mixing ratio at 21 km (altitude that is

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well representative for the UT/LS region, see Sect. 3.4.2). This strong anti-correlation ($R = -0.81$, see Fig. 8) confirms the good quality of the CH₄ concentration retrieved for the UT/LS region." The kernel for 21 km is very wide and has a contribution of about -0.02 coming from around 5 km and below, where CH₄ VMRs are usually high. Will this not mean that high tropospheric CH₄ values will contribute to lower the CH₄ VMR at 21 km and vice versa? So in some way, this might explain the strong anti-correlation.

4. The authors used the retrieval set-up suggested by Sussmann et al. (2011) and compared the results with the results from their retrieval set-up. However, Sussmann et al. (2011) used a different retrieval code (SFIT2) and the authors used PROFFIT. Furthermore, the authors mention "The retrieval setup suggested by Sussmann et al. (2011) fails to improve the reconstruction of the annual cycle" (Page 1400, last sentence). Are the differences in the two retrieval codes (at least for CH₄ profile retrievals) negligible enough to arrive at this conclusion?

Some minor technical corrections.

Separate long sentences and clauses with commas, for example (but not limited to):

Abstract, Lines 1-2: "At the Izana Atmospheric Research Center(comma here)"

Lines 19-20: "Concerning totXCH₄ the agreement between the FTIR data product and the in-situ measurement is rather poor documenting that totXCH₄ is no valid free tropospheric CH₄ proxy(comma here)"

Page 1385 Line 25: Change "wood" to "woodland" in "(from a wood that surrounds the station....)"

Page 1389, Line 10: Change "WCCAM" to "WACCM"

References: Sussmann, R., Forster, F., Rettinger, M., and Jones, N.: Strategy for high-accuracy-and25 precision retrieval of atmospheric methane from the mid-infrared FTIR network, Atmos. Meas. Tech., 4, 1943–1964, doi:10.5194/amt-4-1943-2011, 2011.

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Washenfelder, R. A., Wennberg, P. O., and Toon, G. C.: Tropospheric methane retrieved from ground-based near-IR solar absorption spectra, *Geophys. Res. Lett.*, 30, L017969, doi:10.1029/2003GL017969, 2003.

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