

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.

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Sepulveda et al. describe the retrieval of tropospheric CH₄ in the mid-IR region using the PROFFIT algorithm. Two approaches are evaluated for removing the effects of stratospheric variability: (i) profile retrieval which uses the shape of the CH₄ lines to infer the vertical distribution, (ii) use the column HF to estimate the stratospheric CH₄ column which, together with the retrieved total CH₄ column, is used to infer tropospheric CH₄. The results are compared with in situ measurements of tropospheric CH₄. The profile retrieval technique (i) appears to give better results. For the remainder of this review, I will refer to method (ii) as the "HF procedure".

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This is basically a good paper with interesting scientific methods and results that fall within the scope of AMT. I have no problems with the part of the paper that deals with the profile retrieval. The authors are experts in this field. My main difficulty with this paper is that the author's HF procedure leaves artifacts in the tropospheric CH₄, which were not seen in the Washenfelder [2003] analysis of NIR Kitt Peak spectra. Specifically, the authors' HF procedure seems to result in a substantial low bias of tropospheric CH₄ (3% in fig 6), whereas in Washenfelder's work the bias was < 1%. Given this discrepancy, it is not surprising that the authors' HF procedure seems to only partially remove the CH₄ variation arising from the stratosphere, whereas Washenfelder's HF procedure was much more effective. This calls into question the HF post-correction procedure being used by the authors.

Closer scrutiny of the authors' HF procedure reveals several differences from that used by Washenfelder: 1) the authors fit a periodic function to their HF column abundances "in order to reduce the influence of the chemical variability of HF in the calculations." But the variability should represent real changes in the column of HF, which would correlate with stratospheric CH₄. So removing the variability in HF will impair the correction process. This is why Washenfelder used the directly measured HF columns. 2) An explicitly-stated assumption in the Washenfelder paper was that a linear relationship exist between HF and CH₄ vmrs. Stratospheric measurements support this. But isn't clear from the authors' paper whether this holds for their WACCM profiles since they present no plots of their HF-CH₄ vmr correlation. Nor do the authors validate their HF and CH₄ WCCAM profiles by comparing them (or their correlation) to measurements. What value do they determine for the slope, b, and how does that compare to Washenfelder et al? 3) The author's retrieval appears not to involve the use of column O₂ retrievals, which Washenfelder's did. 4) The authors use CH₄ lines in the 2600-2900 cm⁻¹ region, whereas Washenfelder used 5900-6000 cm⁻¹. The profile retrieval will work better in the MIR than in the NIR because the doppler widths are narrower, spectroscopy is better, and instrumental spectral resolution and ILS are likely better. So comparing profile retrievals in the mid-IR to a stratospheric CH₄-HF

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correction procedure intended only for near-IR spectra is unfair, and does not address the more important issue of which technique is best for analyzing NIR spectra, such as those measured by TCCON.

So the authors' HF procedure seems to have serious deficiencies that were not apparent in Washenfelder's work. And there are substantive differences of implementation. So it isn't clear whether the poor reported performance of the HF procedure is truly a fundamental drawback, or whether it has been exacerbated by differences of implementation. Until these deficiencies are remedied, this comparison cannot be considered objective. And by describing their HF procedure as being similar to Washenfelder's, the authors taint Washenfelder's work.

I'm perfectly willing to accept that the profile retrieval technique, applied to lines in the MIR where Doppler widths are narrow and the spectroscopy/ILS are in good shape, has significant advantages over the HF procedure. And this may also be true in the NIR. But I think that this paper in its current form exaggerates the weaknesses of the HF procedure.

Washenfelder, R. A., P. O. Wennberg, and G. C. Toon. 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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