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Atmospheric Measurement Techniques Discussions

Interactive comment on "Retrieval of tropospheric column-averaged CH_4 mole fraction by solar absorption FTIR-spectrometry using N_2O as a proxy" *by* Z. Wang et al.

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Response to Referee 2

We thank the reviewer, Geoff Toon, for carefully reading the manuscript and for providing many constructive comments. These comments are addressed below.

1)Section 3.1 (Strategy explicitly using tropopause pressure) should be removed. This section introduces an alternative strategy requiring knowledge of the tropopause pressure in addition to the gas column amounts. But this more complicated strategy seems to have little redeeming benefit. The authors find that explicit use of the tropopause

pressure only makes a difference at one site (out of 4). And at Spitsbergen the tropospheric CH4 derived from the "explicit" method (i.e. using the NCEP tropopause pressure) seem noisier than that derived using the "implicit" method. So understandably, the authors adopt the "implicit" method for all their final results and conclusions. So to me, the explicit method described in section 3.1 seems like a failed experiment: the authors tried something more complicated, but it didn't help. So why is it still in the paper?

We agree. This was presented to emphasise the fact that knowledge of the tropopause pressure doesn't help, and indeed is somewhat of a "failed experiement". We have now removed this section and the corresponding plots.

2) I found section 3.3 virtually impenetrable. It needs to be drastically shortened and simplified, or put into an appendix. Part of the problem is that the mathematical formalism seems designed to support the more complicated "explicit" strategy that makes use of the tropopause pressure. Consequently most of the equations contain P_t, the tropopause pressure, as do the terms alpha, beta, e1 and e2. If discussion of the explicit strategy (Section 3.1) were removed, then perhaps this would allow a simplification of Section 3.3 because there would no longer be a need for P_t in any of the equations. This would improve its comprehensibility and reduce its currently-excessive length.

We have indeed removed this section accordingly.

3) I feel that the Abstract and Conclusions somewhat over-state the advantages of the N2O method as compared with the HF method. If you look at the data points at the bottom of fig 10, below the 0.1% line, they are nearly all blue (HF). And if you look at the Spitsbergen results (vertical bars) the HF method produces smaller error bars than the N2O method for virtually all values of H2O. This implies that the HF method is slightly better for dry conditions. But for reasons not fully explained, Figure 11 shows the N2O method to be always better than the HF method at all sites, and it is these results (fig

11) that are summarized in the Conclusions, ignoring figure 10. In my opinion, a more objective and informative conclusion would be that for X_H2O below 0.002, the two methods are of comparable accuracy, but as X_H2O increases above 0.004 the HF method degrades rapidly. So for sites like Darwin with high H2O year-round, the N2O method is unquestionably better. But for colder, drier sites, the HF method is still very useful.

Yes, the conclusion and abstract contents concerning the comparison between the HF and N2O method are based on Figure 11. The Figure 10 is the relative uncertainty estimated based on Gaussian error propagation formula, its value depends on estimation and assumptions regarding the uncertainty of each component, namely, XCH4, XHF, XN2O, the tropospheric mole fraction of N2O and the slope b in Eq.1. But Fig. 11 is the calculated standard deviation during each day, which could be considered a more reasonable reflection of the actual repeatability arising from the data. Considering your comments, we have modified the relevant part in Abstract and Conclusion.

4) Is the HF-CH4 and N2O-CH4 correlation found in the column data consistent with those measured by ACE in fig.1? If not, this will bias the derived tropospheric CH4. You can't assume that the ACE and TCCON results are consistent, just because they use the same HF spectral line. Their different observation geometries and averaging kernels mean spectroscopic errors will affect ACE and TCCON differently.

The correlation plot in Fig. 1 is derived from ACE satellite data, and only the stratospheric part is used. The TCCON column data consist of all contribution in whole atmosphere. They can not be compared with each other.

5) The authors assume a linear relationship between CH4 and N2O. But figure 1 reveals that at high altitude both CH4 and N2O mole fractions tend towards zero, deviating from the fitted line. The authors should discuss this and explain why it doesn't make much difference.

The correlation presented by Fig.1 is for all global ACE-FTS data, however, the slope

used in this work is latitude dependent, with the ACE-FTS data separated into regions of 20 degrees. The nonlinearity mentioned only occurs for the region from -60 to -80 degrees and for mole fractions of N2O below 50 ppb. This corresponds to a pressure level of around 6 hpa, when the ch4 mole fraction is about 800 ppb. Assuming a surface pressure of 1000 hPa and an XCH4 of 1700 ppb, the fraction of CH4 above this level in the total column is 0.5*6*800/(1000*1700) = 0.0015. We have added a sentence to the manuscript highlighting the small influence of this deviation from the linear relationship that exists elsewhere.

6) Regarding the in situ measurement made from the Zeppelin Mountains. This site is at 470 m altitude. Do the authors assume that these measurements are representative of the entire troposphere? Might not the CH4 near the surface be biased high with respect to the free troposphere?

It is true that surface measurements will be biased high (in the case of a local source) relative to the free troposphere. In this case, given the relatively high altitude and the lack of strong local sources, combined with the lack of other available data, the measurements might be a good representation (or at least the best available) of the free troposphere above this site. We do not expect that these measurements are perfectly representative of the entire troposphere, and are presented only as an approximate reference. We have attempted to emphasise this in the manuscript.

A few minor technical issues:

Abstract, line 10: Change "of 20ppb around" to "of around 20ppb".

Page 1459, line 21. I don't think that it is correct to state that the HF method "is basedon the fact the HF is present solely in the stratosphere". This certainly simplifies the equations (a term becomes zero), but the HF method would still work with a non-zero tropospheric vmr, provided it had little variability.

Page 1459, lines 22-23: Delete "...with respect to changes in tropopause height". I

don’t know what this means. The implicit method doesn’t need a tropopause altitude.

Page 1460, line 1: Change "In stratosphere" to "In the stratosphere".

Page 1461, lines14-15: Delete sentence" The measured.....atmospheric gases".

Page 1462. line 14: Change "Learjet 35 aircraft" to "A Learjet 35 aircraft".

Page 1462, line 23: The authors use x_ch4(z) to denote the mole fraction of CH4 at a particular altitude, which is confusing because X_gas is commonly used to denote a *column-average* mole fraction. Suggest using a different symbol than x.

Page 1463, line 12: Change "derive" to "account for"

Page 1465, line 12: Change "overlooked" to "negligible".

Page 1467, line 7: Change "differs" to "differ"

Page 1467, lines 70-8: Don’t understand the sentence "The partial column reflected.....original value". I suggest deleting or re-writing.

Page 1468: line 20: Change "Integrating Eq (10)" to "Integrating Eq (9)".

Page 1474, line 14: Change "around 10ppb around." to "around 10ppb."

Page 1475, lines 9-10: states "every site has its characteristic tropopause pressure and HF column". This isn’t true. These depend on the origin of the airmass being observed. At mid-latitudes the tropopause altitude can vary from 7 km (polar airmass) to 16 km (sub-tropical airmass)

Page 1476, line 10: Change "the aircraft campaigns HIPPO and IMEC" to "the HIPPO and IMEC aircraft campaigns".

Figure 1 caption should mention that the plotted data are from ACE.

Figure 2: The symbols are too similar. Change shape or color.

Figure 6 caption: Change "...low aircraft flights" to "...low altitude aircraft flights".

Fig 11 caption should state "Same as fig 10" (not 9).

All of These have been done.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 1457, 2014.