Atmos. Meas. Tech. Discuss., 7, C1605–C1613, 2014
www.atmos-meas-tech-discuss.net/7/C1605/2014/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Derivation of tropospheric methane from TCCON CH₄ and HF total column observations" *by* K. M. Saad et al.

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Received and published: 8 July 2014

We thank Anonymous Referee #2 for his/her comments.

Pg 3473, Ln 11 (and Pg 3475, Ln 15): why this restriction to a subset of TCCON sites, because of observing limitations, available manpower? Please specify.

HF is not measured at all TCCON sites because the spectral ranges of several of the instruments do not include the band where HF is retrieved ($4038cm^{-1}$). The subset of

TCCON sites that do measure HF was chosen based on length of dataset and zonal representation.

*Pg 3475, Ln 8: I would write: "these retrievals are *also* sensitive to error in the instrument ..."*

This sentence was reworded.

Pg 3478, Ln 7: is this threshold very strict? In other words, do you lose many measurements when applying it?

The number of measurements with uncertainties above 1% varies across sites, but about 98% of the data over all sites and years fall within a 1% error (about 950,000 out of 970,000 individual measurements included in this analysis). The figures only include daily medians for days with more than 5 measurements with less than 1% uncertainty in order to illustrate how the total and tropospheric DMF standard deviations compare over time; however, measurements with errors higher than 1% are not filtered out of the dataset. Because the figure caption clearly states how this filter was applied, the sentence on p. 3478, line 7 has been removed to avoid confusion. Additionally, we added a section in which the uncertainty is explained more explicitly so that end-users can choose an error threshold appropriate for their purposes.

Pg 3478, Ln 15: how are the ACE-FTS errors evaluated? We would like to know more about the quality, reliability, error estimations and filtering of the ACE products. How do they compare to HALOE data, used previously? A brief description and/or appropriate references are needed (also true for H2O, your sentence Pg 3480, Ln 1-2).

ACE-FTS DMF errors are statistical errors from the retrieval fitting; this clarification has been added. We have also included Waymark et al. (2014) as a reference for the

updated version 3.0 data, which describes the data quality, especially with respect to earlier versions of ACE-FTS profiles. We also reworded the description of the post-processing filters applied specifically for this analysis.

For both CH_4 and HF, ACE-FTS profile shapes agree with those of HALOE, and ACE-FTS CH_4 tends to be slightly larger within error. Additionally, altitudes at which the variability is high, also show similar increases in variances comparing ACE-FTS HF and H_2O with HALOE (De Mazière et al., 2008; Mahieu et al., 2008). CH_4 standard deviations for the two instruments are also similar between 0-70 km (De Mazière et al., 2008), while the standard deviations for HF above 30 km tend to be higher for ACE-FTS measurements (Mahieu et al., 2008). These references are now cited in the paper.

ACE-FTS H_2O DMFs are about 10% larger than those of HALOE but are consistent with newer instruments, such as MIPAS, SAGE III and Aura-MLS (Hegglin et al., 2013). The impact of using the H_2O vapor profile to convert "wet" profiles to column-averaged DMFs is small, typically changing CH_4 by less than a ppb. Moreover, ACE-FTS H_2O is only included in the method validation of the derivation and not part of the TCCON tropospheric CH_4 calculation.

Pg 3478, Ln 21: why is Fig. 2 introduced before Fig. 1? Or do you mean Fig. 2 of Washenfelder et al. (2003)?

Figure 1 is now introduced before Figure 2.

Pg 3478, Ln 26: I am wondering if the sparser data from 0-30S and 0-30N could not have been merged, to get more meaningful or robust statistics for the tropical region.

Although the β values in the tropics tend to be within the standard error, the northern and southern tropics do differ in some years. Applying a combined tropical β in the method validation tends to cause greater residuals from the one-to-one line, so we

chose to keep the seperate values with larger standard errors.

Pg 3479, Ln 2: "NH slopes are more steep than their zonal counterparts": is this still true when accounting for the statistical uncertainties affecting the various slopes (e.g. at 2-sigma)?

For most years, the NH slopes are more steep than their zonal counterparts in the SH, even taking into account the 2σ uncertainties, as illustrated in the inset of Fig. 2, which is now referenced in this sentence.

Pg 3479, Ln 8: "For 2013": but you indicated before (pg 3478) that the ACE-FTS dataset was limited to 02/2004-12/2012?

ACE-FTS data exist through March 2013, but these data were excluded from the calculation of β so that the seasonal variations that occur over these three months do not bias the annual slope. We removed "for which data past March are unavailable," to prevent confusion on this point.

Section 2.2, Pg 3479: I have several concerns here: if I am not wrong, the ACE-FTS occultation measurements go at best down to 6 km *altitude. How did you select the ACE-FTS measurements, was the lowest available tangent altitude a criterion for selection? You mention that when ACE information was missing, you used the TCCON priors. Is this the best approach? What about an extrapolation down to the surface level using a mean value from the available ACE-FTS tropospheric profile? A sensitivity study and its brief description would be helpful.*

ACE-FTS profiles included in the method validation had minimum and median minimum retrieval altitudes of 5.5 km and 9.5 km, respectively. The largest tangent angles (at 30 km) included for this were ± 63 degrees, although removing values with tangent

angles of $> \pm 55$ did not change the slope or r^2 values of the best fit lines. Because this experiment tested the internal consistency of the methodology and is not compared to coincident TCCON measurements, a reasonable extrapolated profile should not impact the results of this validation significantly. However, the assumption that $\gamma_{CH_4} = 1$, which we must make because we are not including TCCON retrievals, does impact the results. Larger values for γ_{CH_4} in the Southern Hemisphere compared to the Northern Hemisphere, which reflect a small bias in the a priori profiles between the Northern and Southern Hemispheres, seem to contribute to the underestimation of tropospheric CH_4 in the Northern Hemisphere and overestimation in the Southern Hemisphere in the method validation. For TCCON measurements, γ_{CH_4} is generally within 1% of 1, but a difference of 0.01 between hemispheres can shift the residuals from the one-to-one line by 4 ppb, or about one quarter of the offset.

Pg 3480, Ln 25: it is unfortunate that results from the very high-latitude site of Ny Alesund are not included. What is the reason for this?

While the results for only a subset of TCCON sites are included, this methodology can be applied to all sites that measure HF. Ny Alesund provides an extremely valuable dataset for examining zonal trends of tropospheric methane, but because of the seasonal data gaps during polar night and within the polar vortex, the time series is less illustrative of the methodology.

Pg 3480, discussion of Figure 3: could you characterize and provide a measure of the quality of the intercomparisons, i.e. slopes of the linear fittings, R factors...?

Linear regression lines and r^2 values have been added to each of the plots in Figure 3. The values have also been copied below for reference.

Latitude	Slope	Slope Error	r^2
90	0.99	± 0.0003	0.75
60	0.99	± 0.0005	0.70
30	1.00	± 0.0005	0.96
-30	1.00	± 0.0003	0.99
-60	1.00	± 0.0005	0.68
-90	1.00	± 0.0002	0.63

Additionally, for the method validation, occultations for which the lowest HF values are more than 10 km above the tropopause are no longer included, as these profiles contained mostly interpolated values and therefore were less indicative of actual stratospheric conditions.

Section 3.1, Pg 3481: does your correction generally results in lower intra-day variability for tropospheric methane, when compared to Washenfelder (see error bars on Fig. 8)? If yes, can you quote this improvement, e.g. by providing typical relative standard deviations for both methods? And would this be verified at other TCCON sites?

We have included box plots illustrating the distribution of the differences between daily tropospheric CH_4 standard deviations using the Washenfelder et al. (2003) and updated methods for all sites used in this analysis. The biggest impact of the updated method is the inclusion of the CH_4 averaging kernel, which adjusts the airmass-dependence, and thus the seasonality, of the tropospheric CH_4 calculation. This improvement is now stated more explicitly in the comparison.

Pg 3498, Figure 8 caption: I would identify the blue dots in the caption (e.g. "Daily mean median total (blue) and tropospheric..."

Total column CH_4 daily medians were removed from the figure upon recommendation

of referee comments, so those points are no longer referenced in the caption.

Pg 3501, Figure 11: do you need to make a distinction between off and on shore winds data? This is not discussed in the text. If yes, I would use symbols allowing for a better identification of the two subsets (i.e. two different colors).

Baring Head flask measurements taken in onshore wind conditions are less likely to be comparable to the FTS measurements at Lauder, and these conditions tend to be when the largest deviations occur. This section mentions "changing wind directions impacting the covariance between the two sites" as a possible explanation for larger Lauder tropospheric DMFs in the late summer and early fall. Because verifying this point would require further analysis, however, we have removed the distinction of wind direction in the figure.

Additional Changes

The TCCON data was updated as follows:
- The errors for CH_4 DMFs included measurement precisions that were underestimated. In the new section on tropospheric CH_4 errors, the updated precisions are explained. Because of this update, fewer measurements are filtered out by the 1% threshold, and figures now show daily medians for days in which at least 10 tropospheric CH_4 measurements exist.
- Laser sampling errors for Darwin, Wollongong, Sodankylä and Izaña are now better characterized, and the CH_4 DMFs have now been corrected.
- A small number of points in the Park Falls dataset were not processed consistently, and these points have been corrected.

Minor grammatical errors and wording choices were changed.

The color schemes for several of the figures were changed to make the plots more clear.

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Interactive comment on Atmos. Meas. Tech. Discuss., 7, 3471, 2014.