# Response to Reviewer 1

Jia Chen

Manuscript Title:
"Differential Column Measurements Using Compact Solar-Tracking Spectrometers"

We would like to thank reviewer 1 for carefully reading the paper and giving helpful comments.

Below, the reviewers' original text is included in typewriter font. The answers are highlighted in blue, sans-serif fonts.

## General Comments:

In section 2 there is too little description of the basic solar FTS measurement - the reader is assumed to be familiar with solar FTS remote sensing and previous work. A summary of the measurement technique, referring back to TCCON and the previous papers on this instrument, would be useful for all readers, as it is it is directed only to those who are already involved in these measurements.

Thanks for this comment. We added one subsection (2.1), providing background information about the solar-tracking FTS techniques and discussing existing networks. References to TCCON, i.e., [Toon et al. 2009], [Wunch et al. 2010], [Wunch et al. 2011] are added. For EM27/SUN, [Gisi et al. 2011], [Gisi et al. 2012], [Frey et al. 2015], [Hedelius et al. 2016] are cited.
The added section is:

### Column Measurement and Existing FTS Network

Solar-tracking FTSs can be used to measure the gas column number densities, i.e. the number of gas molecules per unit area in the atmospheric column (column$_{\mathrm{G}}$, unit: molec. m$^{-2}$). The sun is used as light source and the FTS is located on the ground for measuring the solar radiation transmitted through the atmosphere. The recorded sun radiation spectrum is broadband and covers the absorption fingerprints of diverse gas species including $CO_2$, $CH_4$, $H_2O$ and $O_2$. The attenuation of the solar intensity at specific frequencies provides a measure for the column number density for various gases. For further details of the modeling of the atmsopheric transmittance spectrum, please see Wunch et al. (2011) and Hase et al. (2004), for the working principles of FTS please refer to Davis et al. (2001), and Griffiths and De Haseth (2007).
The existing FTS networks include NDACC (Hannigan (2011)), i.e. Network for the Detection of Atmospheric Composition Change, and TCCON (Toon et al. (2009), Wunch et al. (2010), Wunch et al. (2011)). NDACC measures at mid-infrared wavelengths and detects atmospheric $O_3$, $HNO_3$, HCl, HF, CO, $N_2O$, $CH_4$, HCN, $C_2H_6$, and $ClONO_2$, chosen to help understand the physical and chemical state of the upper troposphere and the stratosphere. The TCCON network focuses on column measurements of greenhouse gases, mainly $CO_2$, $CH_4$, $N_2O$ and CO, at near-infrared wavelengths. It uses the Bruker IFS 125HR spectrometer that is large in dimension (room size) and heavyweight ($>$500 kg, Bruker (2006)). The spectra in the TCCON network are recorded with a spectral resolution of approx. 0.02 cm$^{-1}$.

Section 3 is about precision and accuracy of the technique, but the authors use the term "precision" and other terms in incorrect ways. I recommend a reading of the IUPAC publication commonly known as GUM: "Evaluation of measurement data Guide to the expression of uncertainty in measurement" by the Joint Committee for Guides in Metrology, JCGM 100-2008 (available from the BIPM website). "Precision" is a general term, which is not defined for quantitative uncertainty assessment - quantities such as repeatability and reproducibility have specific, quantitative meanings and should be used in quantitative assessments of random uncertainty. Similarly there are more specific terms for "accuracy". In particular, in most cases in this paper, "precision" is used very loosely and mostly means "repeatability". Since the focus of section 2 is quantification of uncertainty, I recommend using correct terminology.

Thank you very much for this comment and providing the references. We looked at ”JCGM 200:2008 International vocabulary of metrology  Basic and general concepts and associated terms (VIM)”, which defines ”measurement precision” as the closeness of agreement between indications or measured quantity values obtained by replicate measurements on the same or similar objects under specified conditions, and explicitly says that ”Measurement precision is used to define measurement repeatability, intermediate measurement precision, and measurement reproducibility.”

In the Wikipedia article Accuracy and precision, ”precision” is described as: ”The precision of a measurement system, related to reproducibility and repeatability, is the degree to which repeated measurements under unchanged conditions show the same results”. According to ISO 5725-1, precision is the closeness of agreement among a set of results.

Therefore, we would like to keep the terminology ”precision”, which is also used in the field of laser spectroscopy for quantifying the measurement repeatability.

The authors use "gradient" throughout when they really mean "difference". A gradient is difference per unit length, eg 2 ppm km-1, not 2 ppm as used here. I found this confusing when reading, and recommend that all instances of "gradient" be sought out and replaced with "difference" where appropriate.

Thank you for pointing out this problem. We fully agree and changed the document accordingly. Note, that there are some occurrences of ”gradient” in the doc, where we actually mean difference per unit length, and thus kept the expression.

Units: please quote units correctly, eg m s-1 not m/s, molec m-2 s-1, not molec/(m2s) as in Table 1. See the IUPAC "green book" (Cohen, et al. (2007)). Quantities, Units and Symbols in Physical Chemistry. Cambridge, IUPAC, RSC Publishing) for authority on units.

We changed the units written as the product of units, without any multiplication sign and left one space between the unit symbols.

Use of "%" and "‰"- I found it very confusing to mix these two quantities, it is too easy not to notice "‰" and read it as %. I recommend using % throughout.

Thanks for this note, we changed all the ”‰” to ”%”.

## Specific and technical comments:

Section 3.1. It is acceptable to use "precision" when speaking generally, but in cases when a value is assigned to an uncertainty, "precision" should be replaced by the appropriate specific quantity "repeatability" or "reproducibility". There are many instances, please search and replace P4 L10: "underestimate the true precision" really means repeatability and is ambiguous since high precision is a small number. Perhaps replace with "overestimate the true random uncertainty of the measurement."

Thank you very much for this comment. As for our answer to the use of ”precision”, please see our reply to your general comment.
We changed ”underestimate the true precision” to ”overestimate the true random uncertainty of the measurement.”

P4 L21: Allan Deviation, not standard deviation.
P4 L28: "Allan standard deviation" is incorrect - replace with "Allan deviation", which is the square root of the Allan Variance; they are not calculated in the same way as variance and standard deviation and should not be confused. There are many examples of "Allan standard deviation" throughout which should be replaced by "Allan deviation" = please search and replace (including figures, eg axis labels in Fig 1).

Thank you for pointing this out. We replaced ”Allan standard deviation” with ”Allan deviation” throughout the document.

P5 L11 "System Robustness" "Robust" has a specific meaning in statistics, and since this is a statistical section, I would recommend System stability as a better title.

We changed ”robustness” to ”stability”, thanks!

P11 L12: "Column gradient observations" - first of many examples where "difference" or "differential" should replace "gradient"

We changed ”column gradient observations” to ”differential column observations”.

P6 L5: Here "precision" is used when "accuracy" is meant - this sentence describes a systematic error.

Thanks. The previous sentence was misleading, we changed the sentence to ”Retrievals for ha have been scaled with RG for the Allan analysis (Sec. 3.1) ”

P7 L23: The usual Reynolds notation is to use u' for the turbulent wind speed component, rather than uturb, so that u(t) = u(bar) + u'(t)

Thanks for pointing this out. We would prefer to use $u_{\text{turb}}$, because u’(t) could be mis-understood as the first derivative of the signal u(t). Hence, $u_{\text{turb}}$ might be less prone to misunderstanding, especially, for readers from other disciplines.

P10 L19: Please explain the transient peak around solar noon - what is it due to, and justification for its removal.

We added section 4.3.2 ”Transient Peak at Chino” to discuss the possible cause of the transient peak. The transient peaks are removed for the calculating the column difference, since they are not associated with the local dairy farms.

### Transient Peak at Chino

Not only for side-by-side measurements, but also in the field measurement, short term peaks are observed, as mentioned in Sec. 4.1. Transient peaks are moving from the upwind to the downwind site: they are observable at upwind site $hb$ between 0.1 and 0.7 hours after solar noon, and at $ha$ between 0.5 and 1.1 hours after solar noon (Fig. 1). They are not observable at $pl$ site, probably because the plume is very narrow. Compared to the upwind peaks, the downwind peaks have a time shift, and are weaker and broader due to air dispersion. The peaks travelling from upwind to downwind site along the trajectory provide a proof that the same air mass is sampled.

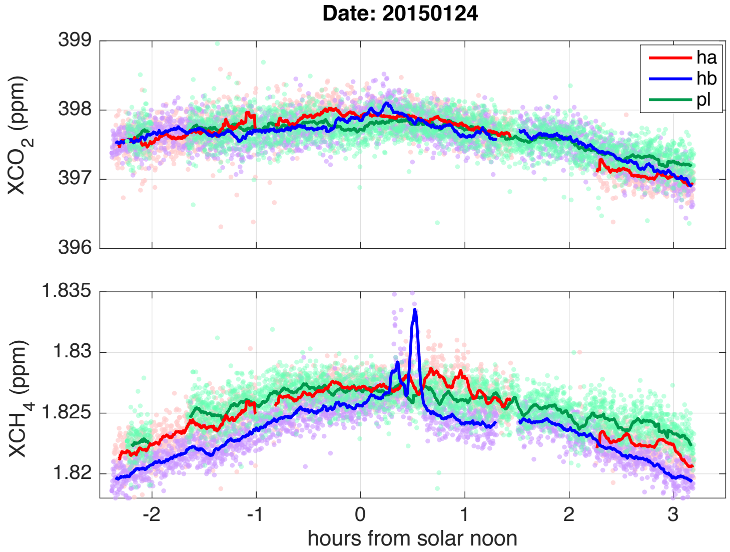

Figure 1: Observed column differences $\Delta X_{\mathrm{CO_2}}$ (upper panel) and $\Delta X_{\mathrm{CH_4}}$ (bottom panel) on 24 Jan. 2015, the transient peaks in $X_{\mathrm{CH_4}}$ are not observed for $X_{\mathrm{CO_2}}$.

The transient peaks are not observed in $X_{\mathrm{CO}_2}$, indicating they are not caused by passing clouds, or from a powerplant. They may come from natural gas leaks from the pipelines in the Chino area, with some evidents being reported by environmental defense fund (EDF (2016)). The transient peaks are removed from the column difference study (Sec. 4.1), because they are not associated with the local dairy farms.

`P11 L14: The meaning of this heading is quite unclear. I suggest replacing "gradient" with "difference" as commented earlier, and bring the second paragraph beginning Pasadena ..." ahead of the first, so it comes first after the heading. Most of the confusion lies in the incorrect use of "gradient".`

Thanks! We changed the title, and changed ”gradient” to ”difference”. Also we changed the order of the first two paragraphs and hopefully the content is more clear now.

`P12 Fig 4: The lowest panel would be very much improved if both the XCO2 and XCH4 axis had a common zero line.`

Thanks much for this suggestion! Now the two y-axis share a common zero line in Fig. 4.

`P12 Fig 5: In the plot, cut off the negative axes at (-1,-5) to better utilize the space and avoid large empty area of the plot. Use the (0,0) axes rather than L and right axes, so the origin is clear.`

Thanks. We cut off the negative axes, and we use ”+” to illustrate (0,0).

`P13 Fig 6: Same comment as Fig 5. The origin crosses cant be seen. The time periods are quite unclear - what are [-1 h : 0 h] etc, relative to what time? More detail in caption required.`

Thank you for the question and the suggestion. We changed the caption as ”Left figure shows the time period between 1 hour before solar noon and 2 hours after solar noon, and right plot shows the time period 2 to 4 hours after solar soon.”

`P13 L1: please rephrase as " a lower DXCH4/DXCO2 ratio" since this is the way the plots are presented (with CH4 on the Y axis).`

Done.

`Appendix D: The plots of the O2 column are not particularly informative, since they reflect mostly pressure not spectroscopic retrieval. I suggest to replace these plots with the O2 column : pressure ratio (corrected to dry air) - this should be a constant with a known value related to 0.2095 mole fraction of O2 in air.`

Thanks for this suggestion. Here the $\mathrm{O}_2$ column are shown for determining column dry air and its uncertainty. We added one paragraph in Appendix D to clarify the purpose:
”According to Fig. 11, the oxygen column number density over the dairy area is $4.493 \cdot 10^{28}$ molec. $\mathrm{m}^{-2} \pm 1\%$ (Sec. 4.1), which is used for calculating $\mathrm{column}_{\mathrm{dryair}}$, the column number density of dry air. This calculation should be equivalent to determining the dry air column using the water column number density and surface pressure.”

## References

Bruker: IFS 125HR User Manual, BRUKER OPTIK GmbH, 1st edn., 2006.

Davis, S. P., Abrams, M. C., and Brault, J. W.: Fourier transform spectrometry, Academic Press, 2001.

EDF: Natural gas: Local leaks impact global climate, https://www.edf.org/climate/methanemaps, last visited 22 May 2016, 2016.

Griffiths, P. R. and De Haseth, J. A.: Fourier transform infrared spectrometry, vol. 171, John Wiley & Sons, 2007.

Hannigan, J. W.: NDACC IRWG: Evolution of Ground-Based Global Trace Gas Infrared Remote Sensing, in: Fourier Transform Spectroscopy, p. FMC1, Optical Society of America, 2011.

Hase, F., Hannigan, J., Coffey, M., Goldman, A., Höpfner, M., Jones, N., Rinsland, C., and Wood, S.: Intercomparison of retrieval codes used for the analysis of high-resolution, ground-based FTIR measurements, Journal of Quantitative Spectroscopy and Radiative Transfer, 87, 25–52, 2004.

Toon, G., Blavier, J.-F., Washenfelder, R., Wunch, D., Keppel-Aleks, G., Wennberg, P., Connor, B., Sherlock, V., Griffith, D., Deutscher, N., et al.: Total column carbon observing network (TCCON), in: Fourier Transform Spectroscopy, p. JMA3, Optical Society of America, 2009.

Wunch, D., Toon, G. C., Wennberg, P. O., Wofsy, S. C., Stephens, B. B., Fischer, M. L., Uchino, O., Abshire, J. B., Bernath, P., Biraud, S. C., et al.: Calibration of the Total Carbon Column Observing Network using aircraft profile data, Atmospheric Measurement Techniques, 3, 1351–1362, 2010.

Wunch, D., Toon, G. C., Blavier, J. F., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W., Sherlock, V., and Wennberg, P. O.: The total carbon column observing network, Philos Trans A Math Phys Eng Sci, 369, 2087–112, 2011.