

## Reply to anonymous referee#1

We thank referee#1 for the careful examination of the submitted manuscript, the useful remarks made with respect to content, and moreover would like to express our gratitude for the careful compilation of so many technical corrections. We do not list all technical corrections in this reply. We omitted minor corrections from this reply if we strictly followed the referee's request (still, all changes suggested by referee#1 are highlighted in red in the revised version of the manuscript with tracking of changes). Below, we refer only to items which deserve discussion in detail.

Reply to general comments:

***• Throughout there are many subjective and “colourful” terms used that are not strictly scientific. I have attempted to note these in the technical comments, but these should be tidied up.***

We have removed the terms under questions (also see reply to technical comments section).

***• There are likewise still a few language issues throughout that would make the article easier to read. In addition, the article features many long paragraphs, and would benefit with respect to clarity if these were broken up.***

We have changed these issues along the lines proposed by the referee. We would like to thank the reviewer for performing such a careful examination, we believe that the presentation quality significantly benefits from these improvements.

***• The comparisons between the standard measurements and those made using the prototype are often fairly rudimentary. In my opinion, the plots should also show the differences or ratios, and some more rigorous reporting of comparison statistics is necessary to understand the performance and limitations of the modified instrument. This should be done for all the comparisons reported.***

The suggestion of showing and discussing pair-by-pair ratios in general is not possible, because the measurements of the prototype and the other instruments were not synchronized (especially, a TCCON measurement takes a much longer scan time than required for a low-resolution spectrum). Only in case of Fig. 9, a pair-by-pair representation would be feasible, because here two different CH<sub>4</sub> products both derived from the prototype are shown. In order to present all results in a similar manner, we decided to add to each existing plot a correlation plot of the daily mean values (Figures 8, 9, and 11). Adding this kind of extension enables us to include some “more rigorous reporting statistics” as requested by the referee, as the intraday scatter of the results provides us with a useful measure for the width of the distribution underlying the daily mean. We also have added the standard deviation of the calibration characteristics derived from the observations.

***• Similarly, the description of the airmass correction is poorly described. The relative corrected and uncorrected values should be plotted against solar zenith angle, and the residuals with respect to the fitted function shown. The function itself (co-efficients) should be described, and if it fails at zenith angles greater than 75 this should also be shown (maybe greyed out). From Figure 10, there also appears to be evidence that the correction might fail at higher elevation angles, though presumably this is due to the variation on that particular day. Further work is obviously necessary to quantify this, particularly if the instrument is to be deployed to instruments in different latitude bands.***

The purpose of our paper is to describe the extension of the standard EM27/SUN with a second channel for XCO measurements and in addition to provide a sufficiently strong set of empirical evidence supporting our claim that the instrumental approach is actually working. The arguments are twofold: firstly, we demonstrate that the spectra of the extended channel are of adequate quality (signal-to-noise ratio, level of channeling artefacts, level of out-of-band artefacts), secondly we perform an explicit comparison with an independent source of XCO data (official TCCON data derived from our collocated high-resolution FTIR-spectrometer).

In our feeling, the demonstration that the XCO measuring capability of the spectrometer proves even at polar latitudes is beyond the scope of this paper. Increased uncertainties of column-averaged trace gas abundances derived from high airmass spectral observations is a general problem of remote sensing observations and not specific of the novel portable device introduced in this work. The problem results mainly from our limited capability of correctly modelling the absorption spectra at these high airmasses, they are not related to instrumental issues. We restricted the range of solar zenith angles (SZA) to 75 degrees because including high-airmass data would probably not enhance the significance of the comparison with the TCCON. Although TCCON reports values at larger SZA, the XCO error bars on the airmass-dependent correction increases towards large SZA. In case of XCO, the airmass dependent correction factor (ADCF) used by TCCON is larger than for any other TCCON species and moreover the uncertainty of this correction is larger than the correction itself (reference: corrections and calibrations in the TCCON wiki at [https://tccon-wiki.caltech.edu/Network\\_Policy/Data\\_Use\\_Policy/Data\\_Description](https://tccon-wiki.caltech.edu/Network_Policy/Data_Use_Policy/Data_Description)).

In this publication, we target at a verification of the prototype spectrometer's instrumental performance, which is most convincingly demonstrated in a comparison which avoids large SZAs, because then the TCCON data are a reliable reference. We have included additional information on the airmass correction we used for the prototype spectrometer: in addition to a best-estimate of the airmass-dependent correction, we have investigated the impact of an over-and undercorrection (now also shown in Fig. 10) and we were able to show that for the selected range of SZAs the details of the correction do not significantly affect our conclusions.

**• *From Figure 11, it seems like there is some serious day-to-day variability in the agreement with TCCON. Please quantify and discuss this further than what is already mentioned.***

Unfortunately, there was a technical error in the original analysis of the low-resolution spectra. The 2% systematic discrepancy was mainly evoked by a systematic smoothing error (as the vertical sensitivities of the spectrally high-resolution TCCON and the low-resolution prototype spectrometer differ). In contrary to the description in the text, an independent set of a-priori profiles (the set used by the NDACC FTIR network), resided in our analysis. We therefore repeated the complete analysis work for the low-resolution spectra, this time correctly adopting the a-priori profiles from TCCON (for H<sub>2</sub>O, HDO, CO<sub>2</sub>, CH<sub>4</sub>, and CO). This improved the level of agreement to a point where the XCO calibration factor of the prototype spectrometer becomes statistically indistinguishable from the TCCON (the 1  $\sigma$  uncertainty of the calibration slope is in the order of 0.5%). We have added the information “The demonstrated performance of the prototype indicates that the design is well suited for source attribution (Wunch et al., 2009, detected intraday XCO enhancements of up to 30 % in the Los Angeles Basin) and satellite validation (the TROPOMI accuracy and precision targets are 15% and 10%, respectively).” We also have extended the paragraph, adding references concerning vertical sensitivities and the impact of the smoothing error. In addition to this, we have included mid-infrared results for XCO, which support our interpretation that the remaining discrepancies between the TCCON and prototype observations are due to a mismatch of vertical sensitivities.

Moreover, we have added to the study a further day of measurements in mid of March 2016 during which the prototype also performed measurements in collocation with the unmodified EM27/SUN spectrometer used as a reference and with the TCCON spectrometer operated in Karlsruhe. The XCO value of this day was significantly higher than the values measured in autumn and this increase is well captured by the prototype.

Reply to technical comments:

***I would also suggest trying to reduce the number of words in the title, at presents it is quite cumbersome***

We have changed the title into a more compact form. It now reads “Addition of a channel for XCO observations to a portable FTIR spectrometer for greenhouse gas measurements”.

***p1, l18-19: you mention additional species here, but the article focussed on CO. I suggest removing “additional species, especially”***

Although we exclusively deal with CO here, which from the viewpoint of source attribution is the species of interest, other species become accessible via the second channel also, as HDO and N<sub>2</sub>O. The capability of also retrieving N<sub>2</sub>O from low-resolution spectra in this spectral region has been explicitly demonstrated by Hedelius et al., 2016. We feel that the information that other species also become observable by this technical modification might be useful for the reader and therefore would like to stick to the current phrase.

***... Also some better references for satellite validation would be good here. The Lindqvist reference in particular doesn't really fit. Seminal references for GOSAT, OCO-2 and/or SCIAMACHY validation using TCCON would be better, such as Reuter et al, 2011, Wunch et al, 2011, Butz et al, 2011, or Morino et al, 2011.***

We agree and have removed the Lindqvist reference, and we added the suggested references (with exception of the Morino et al., 2011 reference – here we instead refer to the latest study of this kind by Inoue et al., 2016, which includes more recent GOSAT results and additional TCCON sites).

***p3, l27-29: the sentence spanning these lines need rephrasing for clarity.***

The sentence in the original version reads: “In addition, a wider spectral response implies that different kinds of out of band artefacts, as nonlinearity and double passing are superimposing with the wanted spectral signal.”.

We have replaced this by a more extensive explanation: “However, as the FTIR technique reconstructs the irradiated spectrum by performing a Fourier transformation of the measured interferogram, it is susceptible for characteristic interfering influences which degrade the recorded interferogram. Periodic sampling errors, nonlinear detector response, or radiation which is reflected back into the interferometer and modulated twice before detection (double passing), can all generate parasitic signals in the spectral domain. If the spectrum is confined to a sufficiently narrow region, the spectral perturbations can be detected (and corrected, if necessary) by the characteristic out-of-band artefacts they create, or they can be tolerated, if the parasitic spectral signal does not overlap with the real spectral signal. Increasing the spectral bandwidth does not only significantly increase the noise level

of the spectrum, but is – more seriously – possibly harmful due to an insufficient level of control of the interfering influences mentioned above.”.

***p3, l29-31: I’m not 100% clear what you are trying to say here. Is there a dependence of XCO<sub>2</sub> and XCH<sub>4</sub> on the signal level?***

Yes, exactly, this is the problem the simple approach of using a detector with extended spectral coverage runs into. Very likely this is due to a nonlinear response of the detector element. We hope, that the new, more detailed explanation which now precedes this sentence helps to clarify. Moreover, we have added the likely diagnosis in the sentence under question, which now reads: “The application of an extended InGaAs diode has been investigated (J. Hedelius et al., 2016), but resulted in a significant dependence of XCO<sub>2</sub> and XCH<sub>4</sub> on the overall signal level of the interferogram, probably due to a nonlinear detector response.”.

***p4, l6: perhaps replace ‘feeding’ with ‘illuminating’***

We would prefer to keep “feeding”, as it is a widely used technical term in this context.

***p4, l11: replace ‘nasty’ with something more scientific***

Ok, right, we replaced “very nasty” by “highly undesirable”.

***p4, l17-23: this seems like an unnecessarily nepotistic example. Most TCCON sites use a dichroic to measure simultaneously on InGaAs and Si detectors.***

It is true that most TCCON sites use a dichroic to measure the InGaAs and Si in parallel. However, the Karlsruhe TCCON site is a deliberate deviation from the standard TCCON approach, as it splits the most important spectral domain for TCCON into two narrower spectral subsections. In our feeling, this is a substantial advantage, not only with respect to achievable spectral signal-to-noise ratio, but especially with respect to the diagnosis of other kinds of artefacts (nonlinearity, sampling ghosts, double-passing, etc.). Please note that the position of the spectral cut and the resulting spectral channels of the instrument proposed here are nearly identical to that of the TCCON spectrometer in Karlsruhe and that the underlying design considerations are the same, therefore we feel that the reference is adequate in the given context.

***p4, l30 - p5, l9: This section uses a lot of words to not say a lot. I would suggest shortening it.***

In our feeling, this paragraph presents two important items: it motivates a fundamental design decision in the development of the spectrometer (why the dichroic solution of the high-resolution spectrometer has not been copied), and explains why the chosen design can be added to an existing spectrometer with minimal effect on the instrumental characteristics (the empirical proof of this claim is given in section 5 of the paper). We tried, but did not succeed in condensing the paragraph without loss of relevant detail. Therefore we would prefer to keep the paragraph in its current shape.

***p6, l19: what do you mean by ‘definition’ in this context? I assume you are referring to the two spectral bandpasses as being well separated and independent***

Yes, correct. We changed the wording accordingly. The sentence now reads: “Both the primary and secondary channels are essentially free from channeling (we estimate the upper limit for the peak-to-peak amplitude in the primary channel to be 0.0005, in the secondary channel to be 0.0002) and the desired separated spectral bandpass for each channel is achieved: ...”.

***p6, l23-26: given that you have just said that the entrance window limits at high wavenumbers, why would this need to be replaced to extend further to lower wavenumbers?***

We added the following information: "...would in addition to a suited detector element require a replacement of the entrance window (because the glass-based window becomes intransparent at a wavelength of about 3  $\mu\text{m}$ )".

***p6, l31-32: how does this figure of 0.015% compare to the offset on the standard instrument?***

This value is similar to the level of out-of-band artefacts found for the standard instrument. (From the perspective of the primary channel performance, there is no real difference between the standard version of the spectrometer and the modified prototype presented in this work.)

***p7, l25-27: as mentioned in the general comments, it would be good to see evidence for discarding the higher zenith angle spectra. E.g. TCCON includes up to 82 degrees. Limiting the EM27/SUN to 75 degrees would limit the application at higher latitudes.***

We cover this topic in the general discussion. We do not think that including spectra observed at higher solar zenith angle would further improve the verification of the novel instrument design.

***p9, l26: what exactly is the polynomial that you use. Include the co-efficients.***

We added a more detailed description of the empirical airmass correction used for XCO in the figure caption of Figure 10. The figure caption now contains the information "For the airmass correction, we applied a second order polynomial fit, choosing 25 degree as the neutral point. For establishing the correction, spectra taken between 15 degree and 33 degree SEA have been taken into account. This choice results in the functional form  $(1 + 0.0027 * (\text{SEA} - 25^\circ) - 0.00007 * (\text{SEA} - 25^\circ)^2)$ , wherein SEA denotes the solar elevation angle." The fit parameters have slightly changed in the revised manuscript, (1) due to the reanalysis of spectra we performed with a-priori profiles compatible with TCCON, (2) due to including an additional measurement day, and (3) due to restricting the fit interval to below 33° SEA (at higher SEA, the airmass-dependent correction flattens out more and more so the intraday scatter becomes dominant).

***p10, l4: Please edit the start of the sentence here to make it objective.***

We rephrased and extended the statement: "The demonstrated performance of the prototype indicates that the design is well suited for source attribution (Wunch et al., 2009, detected intraday XCO enhancements of up to 30 % in the Los Angeles Basin) and satellite validation (the TROPOMI accuracy and precision targets are 15% and 10%, respectively)."

***Tables 1, 2: these could be consolidated into one table with a clear break at modification time***

Done.

***Figure 8: the numbers currently presented here give the impression that the ratios are different after the modification. I suggest including a measure of the uncertainty during each period, and an appropriate number of significant figures.***

We have added a second panel to the figure with statistical information. There is no statistical significant change of the calibration factor due to the modification.

## References

- J. K. Hedelius, C. Viatte, D. Wunch, C. Roehl, G. C. Toon, J. Chen, T. Jones, S. C. Wofsy, J. E. Franklin, H. Parker, M. K. Dubey, and P. O. Wennberg, “Assessment of errors and biases in retrievals of XCO<sub>2</sub>, XCH<sub>4</sub>, XCO, and XN<sub>2</sub>O from a 0.5 cm<sup>-1</sup> resolution solar viewing spectrometer”, AMTD, 2016.
- M. Inoue, I. Morino, O. Uchino, T. Nakatsuru, Y. Yoshida, T. Yokota, D. Wunch, P. O. Wennberg, C. M. Roehl, D. W. T. Griffith, V. A. Velazco, N. M. Deutscher, T. Warneke, J. Notholt, J. Robinson, V. Sherlock, F. Hase, T. Blumenstock, M. Rettinger, R. Sussmann, E. Kyrö, R. Kivi, K. Shiomi, S. Kawakami, M. De Mazière, S. G. Arnold, D. G. Feist, E. A. Barrow, J. Barney, M. Dubey, M. Schneider, L. Iraci, J. R. Podolske, P. Hillyard, T. Machida, Y. Sawa, K. Tsuboi, H. Matsueda, C. Sweeney, P. P. Tans, A. E. Andrews, S. C. Biraud, Y. Fukuyama, J. V. Pittman, E. A. Kort, and T. Tanaka: Bias corrections of GOSAT SWIR XCO<sub>2</sub> and XCH<sub>4</sub> with TCCON data and their evaluation using aircraft measurement data, Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2015-366, in review, 2016.