Authors' answer to the interactive comments of anonymous referee #1 on paper Frey et al., Atmos. Meas. Tech. Discuss., 8, 2735-2766, 2015

First of all, we would like to thank the anonymous referee #1 for the help in further improving the current presentment by a thorough assessment with regards of content and the careful technical proofreading resulting in the identification of several imprecisions and typos.

Referee: *"This paper and its companion (hereafter: P2), present the method and data from a field campaign taking place in and around Berlin, where a network (namely 5) of portable FTIR spectrometers are deployed to measure greenhouse gas columns and detect emissions from within the city. The papers (together) are within the scope of AMT, and relevant as a potentially important means of quantifying/validating city/urban scale greenhouse gas emissions, which future satellite missions such as CarbonSat hope to measure. The instrument(s) that are used to measure greenhouse gas columns have been introduced previously by the lead group at KIT. Their field application is somewhat novel. This paper is reasonably well written and structured. The contents, however, do not seem sufficiently novel to merit publication on their own. Indeed, it seems that the work within this part of the paper would suffice to create one section within P2, and that the two papers would have better impact as a combined paper with less padded out detail. I would therefore recommend simplification, and publication with P2 as one single paper."*

Authors: Concerning the suggested aggregation of both papers, we decidedly disagree. Especially, we reject the claim that "the contents … do not seem sufficiently novel to merit publication on their own". The level of consistency between the spectrometers demonstrated in this work is well beyond what has been demonstrated hitherto with ground-based low-resolution solar absorption measurements. Concerning the instrumental line shape (ILS) calibration procedure described in our work, we have already been contacted by several other working groups operating EM27/SUN spectrometers for further guidance. Therefore, it seems evident that the calibration procedures and consistency checks described in in our work are a vital prerequisite for any successful detection of XCO2 enhancements on the ppm level and will serve as a reference for defining a good practice for this kind of application in a much wider context than the subsequent demonstration concerning the observation of Berlin as a source of CO2, as treated in the Hase et al. submission. However, we are inclined to agree that the current titles might abet the erroneous assumption that the two publications are the result of an artificial separation. We therefore perform an adjustment of titles, for this work, we choose "Calibration and instrumental line shape characterisation of a set of portable FTIR spectrometers for detecting greenhouse gas emissions", and the Hase et al. publication becomes "Application of portable FTIR spectrometers for detecting greenhouse gas emissions of the megacity Berlin". In addition, we expanded the section concerning the ILS calibration procedure, considering some additional practical aspects of the procedure as raised in the discussions with other EM27/SUN users mentioned above. Furthermore we moved chapter 3.2 and 6 to the Hase et al. publication and added a chapter (5.2) in this work comparing barometric records and ground pressure derived from solar observations in Karlsruhe to avoid overlap between this publication and the Hase et al. work.

Referee: *"p2737, l5: within what? (or one another). or would 'smaller than' be more appropriate here?"*

Authors: We will rephrase as "The drifts … are smaller than …"

Referee: *"p2737, l11: these values are loosely tied to the WMO scale. TCCON is a secondary calibration at best. Given that different spectroscopy, different apriori assumptions and different spectral retrieval software are used, there are potentially other sources of further difficulty in tracing to the WMO scale. It is perhaps too strong to say that the measurements are 'compatible' to the WMO in situ scale. Traceable might pass."*

Authors: We will change the statement to "the records are traceable to WMO in-situ scale". In our feeling, the classification of TCCON being "a secondary calibration at best" might be too harsh. TCCON has been extensively been calibrated against in-situ aircraft measurements: Geibel et al., AMT, 2012 estimated the calibration error for methane of being accurate within 1 to 2 per mil and showed that results from the European IMECC campaign are consistent with earlier investigations by Wunch et al., 2010. For carbon dioxide, Wunch et al. estimate the calibration error to be within 2 per mil. For a remote sensing network as TCCON, these margins seems quite satisfactory, given that the claimed station-to-station consistency of the network is on the same level while the spectroscopic line strength errors typically range in the per cent range and above. Moreover, we do not see any reasons which raise doubt concerning the possibility of a transfer of the TCCON calibration to the low-resolution spectrometers within the sub-percent range: the systematic differences between TCCON and the low-resolution spectrometers results are dominated by the loss of spectral detail, which triggers significant changes in the retrieval results even when the same retrieval code and identical linelists are used. This behavior has been demonstrated for the GFIT code used by TCCON in the work of Petri et al, 2012. However, these effects, as other minor contributions due to different linelists and retrieval codes, are mostly systematic and so are absorbed into the calibration factor.

Finally, it should be noted that the aim of properly quantifying enclosed sources or sinks as pursued with an array of mobile spectrometers is not significantly compromised by a calibration error in the sub-percent range. We expect that even a state-of-the-art model setup will impose errors on the retrieved source strength of an order of magnitude higher (several percent), due to residual uncertainties in the wind field, assumptions on source distribution and vertical exchange.

Referee: *"p2738, l6: as far as I am aware, a TCCON instrument can be moved (see e.g. Geibel et al, 2010, and the system now at Manaus). They are, however, definitely not easily portable."*

Authors: Certainly TCCON measurements can be ceased at one site and can be started at another site using the same or another spectrometer, but the spectrometers are definitely not easily portable and – much more importantly – they fail to preserve the instrumental characteristics during transportation. Following our experience of shipping various high-resolution spectrometers of the kind used by TCCON and NDACC, an optical realignment is required after transport for achieving similar instrumental characteristics as before. On this occasion, we would like to

mention that in early 2014 we had to readjust the position of the TCCON spectrometer container operated in Karlsruhe by mere 20 meters. This task was carefully performed with a heavy-duty crane under our supervision, even this mild intervention required a realignment. In conclusion, we do not feel that the high-resolution spectrometer setup used by TCCON deserves the attribution of being a mobile device.

Referee: *"p2738, l6: comma after 'developed'"*

Authors: We will adopt this change.

Referee: *"p2738, l10: leightweight –> lightweight"*

Authors: We will correct the typo.

Referee: *"p2738, l11: platforms SUCH as ships?"*

Authors: We will adopt this change.

Referee: *"p2738, l11: is it appropriate to have a manuscript in preparation in the official list of references?"*

Authors: The manuscript by Klappenbach et al. has been submitted to AMTD by now.

Referee*: "p2738, l16: 'since long' does not make sense. 'for a long time'?"*

Authors: We will adopt this change.

Referee: *"p2738, l16: spectroscopy –> spectroscopic"*

Authors: Ok

Referee: *"p2738, l18-19: commas before 'which' and after 'vicinity'"*

Authors: Ok

Referee: *"p2738, l21: spectrometer –> spectrometers"*

Authors: Ok

Referee: *"p2738, l22: 'conurbation' is not a word that most would be familiar with. Consider replacing with 'metropolis' or something similar."*

Authors: We will use "metropolitan area"

Referee: *"p2740, l10: This sentence 'Bruker recently...' sounds like a sales pitch."*

Authors: We will rephrase "The manufacturer has recently released .."

Referee: *"p2740, l17: as –> because"*

Authors: Ok

Referee: *"p2741, l9: the 'O' in H2O should not be subscripted"*

Authors: Ok

Referee: *"p2741, l10: needed –> necessary"*

Authors: Ok

Referee: *"p2741, l12 'the MHB-382SD data logger'. This has not been introduced anywhere. Replace 'the' with 'a'. Does it have a brand name?"*

Authors: We will rephrase to "a Lutron MHB-382SD data logger"

Referee: *"p2741, l19-20: Suggest removing 'One can see'"*

Authors: We will omit this

Referee: *"p2741, l23: how did you arrive at the figure of 0.04% for XCO2?"*

Authors: We carried out a sensitivity test regarding the effect of an ILS change for the XCO2. Therefore we ran PROFFIT retrievals for one hour of measurements during noon assuming different ILS values with otherwise unchanged parameters. A change of 1 % in the modulation efficiency led to a change of 0.15 % in XCO2.

Referee:" *p2742, l3: a instrument –> an instrument"*

Authors: Ok

Referee: *"p2742, l7: include the year. It might be obvious now, but won't be some time in the Future"*

Authors: Ok

Referee: *"p2742, l16: what do you mean by 'considerably reduced'?"*

Authors: We will rephrase to "reduced … about 10 °C."

Referee: *"p2742, l20: insert a comma after 'Additionally'"*

Authors: Ok

Referee: *"p2742, l21: do you have a reference or details for the tall tower?"*

Authors: We will use "…are available from tall tower measurements (http://imkbemu.physik.uni-karlsruhe.de/~fzkmast/)."

Referee: *"p2743, l1: remove 'really'"*

Authors: Ok. Section 3.2 has been moved to the Hase et al. publication. So we will omit this chapter.

Referee: *"p2743, l3: insert the year again"*

Authors: Ok

Referee: *"p2743, l10: what constitutes 'a long time series'?"*

Authors: We will use "two weeks"

Referee: *"p2743, l11: data was –> data were"*

Authors: Ok

Referee: *"p2743, l11: how were these data used to calculate the altitudes? Why was this not obtained directly?"*

Authors: The site altitudes were taken from repeated GPS measurements and the results checked using a digital topographic map. Because all spectrometers were operated within an area of about 30 km extent, we decided to use a common pressure and temperature profile for the analysis at each site. Therefore, the individual records of the ground pressure collected at each site have been reduced to a common altitude and averaged. In the next step, the ground pressure value for each site has been recalculated from the averaged value according to the individual station altitude. This procedure has been chosen to avoid that a possible drift of one of the station pressure records induces a station bias.

Referee: *"p2743, l12: the concept of a model atmosphere has not been introduced."*

Authors: We will rephrase "…creation of the model atmosphere used by the radiative forward calculation".

Referee: *"p2744, l9: ascent –> increase ?? Otherwise this needs clarification"*

Authors: Ok, we will use "increase"

Referee: *"p2744, l14: stations –> stations'"*

Authors: Ok

Referee: *"p2744, l17: consider replacing 'proper' with a more appropriate word."*

Authors: We will rephrase: "… depends on the choice of linelists for …"

Referee: *"p2744, l22: parenthesis location around the reference needs to be fixed"*

Authors: Ok

Referee: *“p2745, l5: why do you expect large linelist errors because of the variability? Is it not rather because of the difficulty in measuring H2O line parameters in the first place?”*

Authors: You are absolutely right, we will omit this sentence.

Referee: *“p2745, l8: Section title needs rewording. Maybe "Results of calibration measurements” or "Calibration measurement results"”*

Authors: Ok, we will use “Calibration measurement results”

Referee: *“p2745, l22: consider a different date format”*

Authors: Ok

Referee: *“p2746, l3: ’for the largest part’ –> rephrase. Maybe ’can be mostly attributed...’”*

Authors: Ok

Referee: *“p2746, l11: minimize the residuum w.r.t. what? The mean? And how? By scaling each site? All sites???”*

Authors: Inside each bin all available values are averaged. This quantity is used as a reference value. For each individual station, the difference between each value and its bin reference is calculated. The squared sum of all these residuals sets the cost function contribution of the station. The total cost function is given by the sum of all station contributions. In an iterative procedure, the scaling factors for all stations are adjusted for minimizing the total cost function while the side constraint of an invariant average value of all scaling factors is respected. (We neglect the fact that the number of values per bin is slightly variable - in a more rigorous approach, an individual statistical weight of each bin could be taken into account when the associated contributions to the cost function is calculated.)

Referee: *“p2747, l1: perfect is too strong a word”*

Authors: Ok, we will use “excellent”

Referee: *“p2747, l4: with –> at”*

Authors: Ok

Referee: *“p2747, l9: date format”*

Authors: Ok

Referee: *“p2747, l13: But the SZA variation is higher during summer because the sun can get to lower zenith angles. So shouldn’t this be easier to see?”*

Authors: Yes, this is right – however, we restricted our measurements to smaller SZAs. The effect would be more evident if measurements are extended to larger

SZAs, as the airmass changes become more prominent. However, due to obstacles as trees or buildings, these were unaccessible.

Referee: *"p2747, l18: parenthesis location with reference"*

Authors: Ok

Referee: *"p2748, l2: This –> These"*

Authors: Ok

Referee: *"p2748, l3: shows –> show (the word 'data' is a plural)"*

Authors: Ok

Referee: *"p2748, l3: Consider rephrasing the sentence starting 'It turned out...'"*

Authors: Ok

Referee: *"p2748, l4: How do the SZA-dependences compare to those derived for TCCON? From what I remember, it is O2 that shows the airmass dependence within TCCON data, in contrast to what you see here."*

Authors: The airmass-dependency in O2 found with PROFFIT is significantly smaller compared to TCCON. For a detailed analysis of the airmass-dependent effects we refer to the Klappenbach et al. publication because there they are discussed in detail.

Referee: *"p2748, l17: how many significant figures are appropriate here? 5 seems to be too many. This is also true in other places (e.g. Table 3)"*

Authors: We changed the significant number of figures to 4, also in Table 3. In Table 4 however we think that it is appropriate to give 5 significant figures, given the very high stability and low drifts between the days.

Referee: *"Section 6: The point of doing this is not very clear. What do you mean by the 'slope' when referring to Figure 9? Also in Figure 9, you seem to ignore the fact that the instruments see larger variability at, and just after, the time of the maximum surface pressure."*

Authors: We will rephrase: "For each site, the record of the dry ground pressure and the retrieved water vapour column is in excellent agreement with the retrieved molecular oxygen column." This is a very sensitive test of the instrumental stability, because for the oxygen column, there is no (partial) compensation of instrumental problems as changes of ILS, presence of nonlinearity effects, as occurs in the ratioed XCO2 and XCH4 values.
This chapter has been moved to the Hase et al. publication but the statement also holds true for the added chapter 5.2.

Referee: *"Multiple locations: When referring to the higher spectral resolution instrument, the use of the term 'TCCON instrument' is not entirely accurate. The instrument is independent of TCCON. TCCON refers to more than the instrument -*

the measurement parameters, retrieval setup etc. If you have, as I suspect, not applied the standard TCCON analysis procedure to the retrievals from this instrument, then these are not TCCON measurements, nor should it be referred to as a 'TCCON instrument'"

Authors: The high-resolution spectrometer in Karlsruhe is part of the TCCON network and has the full (not provisional) TCCON status. The TCCON results shown have been generated with the current version of the TCCON processor, so these are TCCON measurements and the spectrometer is a TCCON instrument.

Referee: *"p2750, l1: How do you reach the conclusion of applicability to source/sink measurements? To me, that is too strong of a jump from what is presented in this paper, which deals with the stability and inter-instrument consistency."*

Authors: The level of stability of the spectrometers demonstrated in this work (drifts below 0.005% for CO2 and 0.035% for CH4) allows the unambiguous detection of XCO2 enhancements in the sub-ppm range for CO2 and sub-ppb range for CH4, which allows the detection of localized sources and sinks of various kinds.

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