Reply to anonymous reviewer #1

First of all, we would like to thank the reviewer for helpful suggestions and moreover for the careful detection of language errors. We have adopted most of the technical corrections, however, we feel uncomfortable with respect to the suggestion of mixing the pair of submitted papers into one blend – we will detail our opinion on this point below.

We erroneously missed to add the ceilometer data to the electronic supplement, which would be useful for the users of the dataset. The electronic supplement has been expanded accordingly. We also added the following information in the text: "In addition to the FTIR data, the electronic supplement contains the results derived from the ceilometer observations in both tabulated and graphical form."

This is part 2 of 2 papers in which five portable solar FT spectrometers were deployed around Berlin to measure total column CO2 and CH4 in the atmosphere over a 3 week campaign in summer 2014. In part 1 the intercalibration of the spectrometers before and after the campaign is described in detail. Part 2 describes the campaign measurements and their interpretation through a relatively simple Lagrangian dispersion model to estimate city-wide emissions of the target gases. This is an valuable piece of research and the papers are suited to publication in AMT. The work is thoroughly presented and I recommend publication, however I suggest a re-structuring of the work into a single, more concise paper. The splitting into two papers makes the publication longer than necessary and awkward to read either paper without referring to the other. Some material must be repeated in each paper, and some material in one paper better belongs in the other. While the need for tight intercalibration is essential for the purposes of the field campaign and is well justified, it does not justify a standalone paper as it does not have clear relevance without the context of the measurement campaign.

Concerning the suggested aggregation of both papers, we decidedly disagree. We do not think that the judgement *"while the need for tight intercalibration is essential for the purposes of the field campaign and is well justified, it does not justify a standalone paper as it does not have clear relevance without the context of the measurement campaign*" is correct. Firstly, it should be noted that the level of consistency between the spectrometers demonstrated in the work of Frey et al. is well beyond what has been demonstrated hitherto with ground-based solar absorption FTIR spectrometers, which in our opinion - given the orientation of AMT - merits a publication of these results in their own right. Secondly, concerning the proposed 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 by Frey et al. are from a general viewpoint useful requisite for any reliable detection of XCO2 enhancements on the ppm level using this kind of instrumentation. We expect that the work by Frey et al. will in future serve as a valuable reference for defining a good practice in a much wider context than the subsequent application described in the work by Hase et al.. However, we agree with the reviewer 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 part 1 we will use the title "calibration and instrumental line shape characterisation of a set of portable FTIR spectrometers for detecting greenhouse gas emissions", and part 2 becomes "application of portable FTIR spectrometers for detecting greenhouse gas emissions of the megacity Berlin". With regard to content, the section concerning the ILS calibration procedure in the work of Frey et al. has been expanded in the revised version, now treating in further depth practical aspects of the procedure as raised in the requests of other EM27/SUN users mentioned above, whereas sections overlapping with the description of the Berlin campaign itself have been moved to the Hase et al. manuscript.

Part 1 contains introductory material that better belongs to part 2 (e.g. 1. Introduction, and 3.2 describing the Berlin campaign). Part 2 relies heavily on part 1 to follow the measurements. I therefore recommend combining the two papers into one, merging the duplicated material in introductions campaign descriptions and conclusions, with a single introduction to the importance of the work, followed by (existing) sections on the instrumentation, (calibration, ILS determination, spectrum analysis and processing to total columns), Berlin campaign description, results and modelling. This is mostly a matter of reorganising existing sections rather than rewriting new material. The authors might consider placing some of the calibration material from part 1 into an appendix to the combined paper. It is important that this material be included, but for the less interested reader it can be summarised in the main text with details in the appendix to enhance readability.

We have revised the introductory part of the work by Frey et al. towards a clearer focus on quality issues of ground-based remote sensing. The contents of section 3.2 have been removed and now are addressed in the manuscript of Hase et al..

Technical corrections – part 2

P2768 L11: Remove "unique" – there are other studies – see below

In our feeling, the term is appropriate here. To our best knowledge there is no other comparable set of ground-based remote sensing data available: the application of a larger number of mobile spectrometers around a source region is novel and the precision and consistency of the resulting dataset has not been demonstrated with this kind of equipment before.

P2769 L10: The site locations and campaign details belong here in part 2 if the work is separated into 2 papers.

We have moved this into the Hase et al. manuscript.

L14: form => from

Corrected

P2770 L12: remove “applied”

Removed

L13: replace originally” with “initially”

Corrected

L16: measurements => measurement

Corrected

L24: nicely => clearly

Corrected

P2771 L 3: H2O varies considerably

Corrected

P2773 L15: inspired by informations => informed by information

We rephrased this statement and now use "… and contribution of each box have been adjusted according to informations …".

P2774 L 2: "molecules" Descriptions of Langrangian models usually refer to the more generic term particles, not in quotes, rather than "molecules". I suggest replacing "molecules" throughout with particles.

We have adopted this technical term as suggested by the reviewer.

P2775 L20 and figures 6-8: expand figure captions to include this information about background asumptions

We have included this information in the figure captions.

L27: the peaks are "well captured" but much broadened in time – is this local sources or model resolution, or dispersion?

This is correct: the peaks are much wider in the model prediction. The simple model does not incorporate any point sources, only the distributed source regions. We suspect that a more localized source in region 1 is responsible for the sharpness of the observed peak (as outlined in P2776, first section). Further diagnosis would require additional model runs with a more refined source setup.

P2777 L1. Define MACC

MACC is the EU-funded "monitoring atmospheric composition & climate" project. We have added this information to the first occurrence of the abbreviation and a link to the project page in the acknowledgement (project page: http://www.gmes-atmosphere.eu).

L13: quality-filtered XCH4 (add hyphen)

Done.

Figure 9: There are no scales or labels on X or Y axes

The coordinates are omitted in this map of central Europe, because the coastlines are shown in the plot for orientation. We have added this information in the figure description.