

Response to interactive comment of anonymous Referee #1 on “Comparison of continuous atmospheric CH₄, CO₂ and N₂O measurements – results of InGOS travelling instrument campaign at Mace Head”

We wish to thank this reviewer for his/her helpful comments and suggestions for changes of our manuscript; our replies and respective changes are described below.

The submitted manuscript presents results from a two-month parallel on-site comparison of greenhouse gases observations with gas chromatography, cavity ringdown spectroscopy and Fourier Transform Infrared Spectrometry at Mace Head, Ireland. In particular, it reveals that novel spectrometric measurement techniques (such as the fourier transform infrared spectrometer used here) are suitable for quality control of continuous observations through short (in the order of a few weeks) side-by-side measurement campaigns in the field as the novel techniques are transportable, easily deployable, robust and are sufficiently precise to also detect small systematic differences in the range of the WMO/GAW compatibility goals.

The paper is scientifically sound, well written, and clearly structured and deserves to be published in ACP even if I agree with reviewer #2 that it is a conceptual paper that would have also fit (maybe even better there) in AMT. Moreover, I have a few comments that should be addressed prior to publication in ACP.

It should be clearly stated that the present approach, analysis and structure of the manuscript is very similar to the work of Hammer et al., 2013 (Atmos. Meas. Tech., 6, 1201–1216, 2013), but that it is applied to another measurement site, longer time series and also to N₂O in addition to CO₂ and CH₄.

Response

This is correct and we will make a respective statement now explicitly at the beginning of chapter 2.

The authors should carefully check the manuscript with respect to the use of acronyms. Some are most likely unnecessary, some are introduced several times (e.g. ILC twice in the abstract), some aren't at all (e.g. AGAGE, ICOS, HATS, CCGG) or at least not at first use (e.g. GC-MD). InGOS is introduced first in chapter 3.1 even if this acronym is even part of the title. I suggest to reduce the use of acronyms in particular in the abstract to a minimum and to introduce all acronyms at first use in the main text body even if it was already introduced in the abstract (e.g. ILC, page 10431, line 17).

Response

We will remove unhelpful acronyms in the abstract and introduce all acronyms at first use (and if occurring in the abstract, again in the main text body).

Is it of importance in the title that it is an InGOS travelling analyser? I suggest removing InGOS in the title.

Response

Yes, indeed this is not important and we will remove InGOS in the title (and abstract).

Page 10430, line 25: Remove citation (WMO, 2009) since it isn't common to use references in the abstract. The same statement with the same reference is once more made in the introduction.

Response

We will remove this reference in the abstract.

Page 10431, lines 20-21: add Rella et al. (Atmos. Meas. Tech., 6, 837–860, 2013) that also shows side-by-side results of a travelling instrument.

Response

We will add this reference.

Chapter 2.1: why don't you mention the manufacturer of the FTIR? what is the sample and recording rate of the FTIR?

Response

We will include the manufacturer (i.e. University Wollongong) in the Introduction and add the recording rate of the FTIR in Sec. 2.1.

Page 10433, line 2: "medium-size city", be more precise.

Response

We will add the number of inhabitants (about 150,000).

Page 10443, line 16: add Dlugokencky et al. (JGR, VOL. 110, D18306, doi:10.1029/2005JD006035, 2005) as reference.

Response

All scales are cited explicitly in Sec 2.1 of the manuscript, such as the above mentioned. Nevertheless, Dlugogencky et al., (2005) have also found small differences between both scales. So we agree that we should cite this paper also here.

Page 10435, line 3: The water correction parameters from Chen et al. (2010) were used for both Picarro analysers. It was shown that the parameters can slightly vary from instrument to instrument (see e.g. Rella et al. (Atmos. Meas. Tech., 6, 837– 860, 2013)). Have the parameters used been experimentally verified? See also the comment to paragraph 4.3 below.

Response

As noted in our reply to referee 3, we indeed used the correction described in detail by Chen et al. (2010) for both CRDS instruments. The water dependency of the wet CRDS

system (G1301) was tested at LSCE prior to installation at Mace Head station and was found to be described properly by the water correction provided by Chen et al. (2010). To confirm the correctness of the water correction we have now again compared the CO₂ differences of the wet (G1301) and dry (G2301) CRDS systems from March 2013 until July 2013. Both CRDS instruments showed a mean CO₂ difference of about $0.02 \pm 0.10 \mu\text{mol mol}^{-1}$. The difference is weakly correlated to absolute humidity (slope: $0.13 \mu\text{mol mol}^{-1} \text{CO}_2 / \% \text{ absolute humidity}$). This can be seen in Fig. D1 for CO₂ (D1a) and CH₄ (D1b) in the reply to referee 3. Since the CRDS G2301 was always measuring dried air, the absolute humidity dependency is most likely due to the incomplete water correction of the G1301 instrument. The absolute humidity during the comparison period varied between 0.55 and 0.8% (see gray shaded area in Fig. D1). Thus, the incomplete water correction would explain about 0.01-0.04 $\mu\text{mol mol}^{-1}$ higher CO₂ values of the G1301. However, this explains only less than 30% of the difference of $0.14 \mu\text{mol mol}^{-1}$, which was found during the comparison period between the TCI and the CRDC G1301.

Chapter 2.3: what are the sample and recording rates of the Picarro analysers? Which aggregates were used for the intercomparisons?

Response

The measurement frequency of the Picarro CRDSs is 0.2Hz (concentration updated every 5s in the data files) and the sample flow rate is about 0.3 slpm at about 1 bar absolute pressure. For the comparison we used hourly aggregates since the data is computed and stored like this in the common database. This information will be added in the manuscript.

Page 10437, lines 8 ff: is there any idea what could have happened?

Response

Unfortunately we don't have any idea. It cannot be due to a calibration issue since both cylinders use the same working standards for calibration and stable values have been reached during the standard gas measurements for both instruments.

Paragraph 3.3, Fig. 3: mention explicitly that different cylinders with different mole

Response

We will mention this in a revised manuscript.

Paragraph 3.4.2: no buffer volumes were used at Mace Head?

Response

This is correct and will be mentioned in a revised manuscript.

Page 10442, line 1: do I understand it correctly that the FTIR records 3-min averages and CRDS data are stored as 1-min means?

Response

This is correct, but we compare here hourly averages of both instruments.

Paragraph 4.3: As mentioned above, the water correction parameters published by Chen et al. are based on tests with one single G1301 analyser. Is it possible that the observed differences of the CO₂ (dry air) mole fractions determined with the two Picarro analysers can be caused by some improper humidity corrections? Standard and target gases are dry, thus, this could also explain why no systematic difference was observed for the measurements of the cylinders.

Response

With the information about the water correction given above, we would argue that the effect of the incomplete water correction does not exceed 0.04 $\mu\text{mol mol}^{-1}$ CO₂ during the comparison period. It definitely does not explain the majority of the difference between the G1301 and the TCI instrument. We will make a comment on the effect of a possibly incomplete humidity correction in the revised manuscript.

Page 10447, first paragraph: the authors could also consult the paper by Corazza et al. (Atmos. Chem. Phys., 11, 2381–2398, 2011) that retrieved bias corrections (in comparison to NOAA flasks) from different European stations based on inverse modelling.

Response

Ambient air differences in 2006 (as given in the paper by Corazza et al., 2011) have been significantly different to those we observed in 2013. Nevertheless, Corazza et al., 2011 indicate how important the assessment of biases between stations/networks is.

Page 10447, lines 9 and 10: correct the typo “flaks” (twice).

Response

Will be done in a revised manuscript.

Chapter 5: To my mind, this chapter doesn't really fit into this paper. Either skip it or elaborate.

Response

Principally, the referee is correct with this statement. However, in this section we wanted to emphasise that the precision of our measurements is excellent with this new instrumentation, but as long as we are not able to overcome biases in calibration and achieve a high compatibility between instruments, we will not be able to fully use this excellent instrumentation. We agree that Sec. 5 seemingly does not fit to the rest of the paper, which is more technical. In order not to disturb the clear structure of the paper, but nevertheless emphasize our statement, we will move this section into an Appendix.

E.g. Page 10448, line 24: add reference for CH₄ emissions from the ocean.

Response

Supersaturation of CH₄ in the ocean mixed layer potentially leading to a CH₄ flux from the ocean to the atmosphere has been observed, but the mechanisms are not fully understood yet (Bakker et al., 2014). We will also include this reference in the manuscript.

Add references to flux observations at Mace Head (if available). If flux strengths are available, do the observed gradients correspond to what is expected? Can you calculate fluxes out of the gradients?

Response

Principally, yes, however, it is not within the scope of this paper to present a flux estimate. For ²²²Rn-based flux estimates we would need to make assumptions about the temporal and spatial homogeneity of ²²²Rn fluxes and secondly we have not measured ²²²Rn at both heights. Nevertheless, we want to show that it principally is feasible to detect very small CO₂ or CH₄ gradients with the new optical instruments.

Page 10450, line 24: one week seems to be definitely on the short side.

Response

The gradient was measured over the course of two months. During this time the FTIR was measuring at 10 m a.g.l. and the CRDS system at 25 m a.g.l. Both instruments were measuring at the same height for one week.

Page 10450, lines 27 ff.: skip the part with the bureaucratic regulations. Such issues shouldn't be a determining factor.

Response

We will skip this part.

One thing that wasn't mentioned at all and that I suppose that can be crucial for a best possible comparison result is the perfect match of the time series taking exactly into account the residence times in the individual inlet lines etc. This is only possible if you compare two high time resolution analysers that allow shifting the time series even only by a few seconds before calculating higher aggregates. Table 1 and Figs. 4 to 7: which aggregates were compared and shown?

Response

It was stated before (p.10442 line 1) that one minutely CRDS data was compared with three-minutely TCI data, but these were not shown here. No asynchrony was detected in this comparison. The results in Table 1 and Fig. 4-7 are the comparison of hourly data from the CRDS and the FTIR.

Figure captions Fig. 4 and 6: figure captions says "concentration" while the y-axis labels says "mole fraction". Mole fraction is right. Please correct.

Response

This will be corrected in the revised manuscript.

Do you really need Fig. 5?

Response

We want to show how well both instruments agreed in Heidelberg and how the distribution of differences looked like. We think the Heidelberg comparison campaign is just as important as the Mace Head campaign, as this gives best confidence into the TCI setup as a whole.

References

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