Reply to the comments of Reviewer #1 on the manuscript "Greenhouse gas measurements from a UK network of tall towers: technical description and first results"

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We thank the reviewer #1 for their time and effort in evaluating this manuscript and for their suggestions for improvements. All points made by the reviewer are addressed on the following pages.

The manuscript presents a comprehensive overview of the recently set-up network of greenhouse gas measurements at tall towers in the UK. The paper is clearly structured and well written. It provides much useful information for readers that also aim at establishing monitoring capabilities at tall towers. Therefore, the manuscript merits publication in Atmospheric Measurement Techniques. However, the paper summarizes several things already published in the literature. Thus, I feel that some paragraphs of the manuscript could be shortened by minimizing repetition of already published technical description.

Response: It is understood that the paper summarises a number of things already published in the literature and that some paragraphs could be shortened.

Modification: A number of sections have been modified, as outlined below:

- Page 7, line 4: "This tubing is made from high-density polyethylene bonded to overlapped aluminium tape and has a total wall thickness of 1.57 mm (Andrews et al., 2014). The outer polyethylene coating makes it resistant to water (H_2O) vapour condensation on the inner aluminium core tube." was removed.
- Page 7, line 13: "For each inlet at RGL and TAC, an inverted stainless steel cup covers the inlet, acting as a shield to prevent H_2O entering the line. A monel mesh screen is inserted within the cup to help prevent large particles from entering the inlet lines." was changed to "For each inlet at RGL and TAC, an inverted stainless steel cup with a monel mesh screen inserted within the cup covers the inlet, acting as a shield to prevent H_2O entering the line."
- Page 7 line 23-24: "The filters are present to protect instruments and pumps from particles (Andrews et al., 2014)" was removed.
- Page 10, line 3-10: "H_2O can damage system components and interfere with measurements of GHGs, even at low levels (Andrews et al., 2014). H_2O influences

mole fractions of GHGs measured via CRDS through a dilution effect, whereby a difference of 100 µmol mol^{-1} (or parts per million; ppm) H_2O can cause a "dilution offset" of 0.04 µmol mol^{-1} in CO_2 (Andrews et al., 2014). Also, H_2O vapour differences between calibration gases and air samples can cause spectral issues within optical instruments. H_2O causes these spectral artefacts through line broadening effects on the spectroscopic line shapes, specifically Lorentzian broadening and Dicke line narrowing (Chen et al., 2010; Rella et al., 2013). The extent of these effects is dependent on the atmospheric mixing ratio of H_2O (Chen et al., 2010)" changed to "H_2O can damage system components and interfere with measurements of GHGs, even at low levels, through a dilution effect (Andrews et al., 2014) and pressure broadening effects (Chen et al., 2010; Rella et al., 2013)".

- Page 10, line 13: "The effectiveness of permeation Nafion dryers is dependent on the H_2O partial pressure gradient between the sample and counter purge flows (Andrews et al., 2014)" was removed.
- Page 11, line 7 – page 12, line 10: two paragraphs changed to "The SF_6 and N_2O analysis method used at RGL and TAC was similar to that described in detail in Ganesan et al. (2013), except P-5 carrier gas (a mixture of 5 % CH_4 in 95 % Ar; Air Products, UK) is used. Briefly, calibration gas and air samples are flushed through an 8 mL sample loop at 40 mL min^{-1} for 60 seconds at a fixed exhaust pressure (~ 20 psi; Fig. 3 Backflush) before decaying down to ambient pressure. Flow through the loop is controlled by a 'RED-y' smart series mass flow controller (GSC-A4TA-BB22, Voeglin Instruments AG, Switzerland) and pressure in the loop is measured using an 'All Sensor' pressure sensor (100PSI-A-DO, All Sensors, BS-Rep GmbH, Germany). Once equilibrated to ambient pressure, samples are then injected through an 8-port, 2-position valve (V3 in Fig. 2; EUDAC8UWEPH, VICI Valco AG International, Switzerland) onto a pre- (1.0 m Porapak Q, 80/100 mesh, 3/16" O.D.) and main column (2.0 m Porapak Q, 80/100 mesh, 3/16" O.D.) held at 90 °C, where N_2O and SF_6 are separated from air. Oxygen is "heart-cut" to vent (V3, Fig. 3) after it has eluted from the two columns, whilst the pre-column is back flushed with P5. The remaining, O_2 minimised, sample flows through the post-column (0.9 m of 1/8" O.D. stainless steel packed with molecular sieve 5Å, 45/60 mesh), housed in a thermostatically controlled heated inlet port of the GC at 180 °C. The post column reverses the elution order of SF_6 and N_2O to prevent the larger N_2O peak from tailing into the small SF_6 peak, improving sample reproducibility and precision. Detection occurs in the ECD which is held at 350 °C. The ECDs at TAC and RGL measure at a rate of 10 and 20 Hz respectively. Samples are dried using a Nafion Dryer (MD-050-72S-1, Perma Pure, USA) with a dry zero air counter purge (as outlined in Sect. 3.3)".
- Page 12, line 12: "The use of P5 carrier gas enables the omission of CO_2 doping" was removed.
- Page 12, line 13: "N_2O co-elutes with CO_2 on the column combination used within the UK DECC network, saturating the MS 5Å and providing a constant doping effect, thus care is taken to make sure the N_2O response if not affected by this" was changed to "N_2O co-elutes with CO_2 on the post-column, saturating the MS 5Å and providing a constant doping effect and reducing precision".
- Page 12, line 23 – page 13, line 11: paragraph changed to "CO and H_2 are measured at two sites, MHD and TAC, using a RGA (RGA3 (MHD) and Peak Performer 1 (TAC), Trace Analytical Inc., USA). Table 4 outlines RGA instrumental setup at TAC and MHD. The MHD RGA setup is different to TAC and is outlined in (Prinn et al., 2000). The TAC sample selection system is integrated within the GC-ECD

> system (Sect. 3.4) (Grant et al., 2010a; Grant et al., 2010b). The GC-ECD has a 10-port, 2-position valve (VICI Valco AG International, Switzerland) for V2 (Fig. 2), instead of an 8-port 2-position valve, as at RGL. This allows for a 1 mL RGA sample loop to be put in sequence before the ECD sample loop (Fig. 3 TAC). After samples have been dried using the Nafion Dryer (MD-050-72S-1, Perma Pure, USA), passed through the sample loops and decayed to ambient pressure, they are injected onto two isothermal packed columns held at 105 °C: a 0.768 m pre-column (1/8" O.D. stainless steel packed with 60/80 mesh Unibeads 1S) and a 0.768 m main column (1/8" O.D. stainless steel packed with MS 5Å, 60/80 mesh). After separation, gases are injected into the RGA for analysis using zero air plus (Air Products, UK) carrier gas, where the samples pass over a heated bed of mercuric oxide before being quantitatively determined using UV photometry (Grant et al., 2010a; Grant et al., 2010b)".

At the same time, the paper somehow lacks novelty or the author missed to emphasize the new approaches applied here. I suggest to highlight the new approaches and, in addition, to provide more information likely of interest for the reader. Issues to be addressed in this respect are (a) the required maintenance of the measurements (e.g. how did the regular maintenance look like, how many maintenance (regular as well as "emergency") visits were needed over the years); (b) how did the troubleshooting look like; (c) which major technical problems were the authors facing; and (d) add a paragraph on lessons learnt in the conclusions

Response: Two extra sections have been added to include information on maintenance (section 3.8; Table 5) and troubleshooting data issues (section 5.4; Table 8) as suggested.

How does this exercise refer to European efforts like ICOS, in terms of instrumentation, quality control and data processing?

Response: Instrumentation used within the UK DECC network is very similar to those used for CO_2 and CH_4 measurements in ICOS (Yver Kwok et al., 2015; Hazan et al., 2016) and other non-European measurement programmes, such as the Los Angeles Mega City Project (Verhulst et al., 2017). However, the data processing and quality control is different within ICOS to the UK DECC network. The ICOS method calibrates data by linear interpolation between a linear fit of a suite of calibration gases (n = 4) spanning above and below ambient concentrations (Hazan et al., 2016), whereas the method used in the UK DECC network uses a daily standard to calibrate out daily instrumental drift and then suite of calibration cylinders to correct for effects above and below ambient mole fractions.

Specific comments:

Page 1, line 17: I don't agree with the wording "automated custom-built instrumentation" here as a large part of the instruments is commercially available.

Response: "custom-built" removed.

Page 2, lines 11-13: ". . .independent emission estimates for comparison with the UK national inventory . . .": if this is mentioned in the abstract, I expected to see some related results in the manuscript but I couldn't find it.

Response: This sentence has been removed as these modelling results are outside of the scope of this paper.

Page 4, line 6: "...except MHD which samples every 20 minutes ...": this statement is in contradiction to Table 1 where a Picarro G2301 is listed for MHD.

Response: "Except MHD which samples every 20 minutes" has been removed.

Page 5, line 19 – 20: Here for RGL and also further below for TAC: what is the rationale for measuring at several heights?

Response: CO_2 and CH_4 are measured at several heights on the tall towers to try and asses for boundary layer stratification. Measurements at several inlet heights with instruments that are able to measure at high-frequency has the added benefit of aiding troubleshooting of data.

Page 7, lines 3 – 10: why Synflex tubing is used at RGL and TAC while stainless steel tubing is used at MHD. What are the pros and cons for using one of them? What is the difference between Synflex 1300 and Synflex 3000?

Response: Historically, stainless steel (SS) has been used at MHD when it was set up as a sampling site for AGAGE in 1994, as well as for the predecessor research programmes, as SS does not outgas any of the halocarbons measured in the research programme. The relatively short run of tubing from the inlet to the line pump at MHD meant that ¼" i.d. SS tubing gave the desired flow rates with the line pumps used. However, for the tall towers, a wider i.d. tubing (½" i.d.) was necessary to get the necessary flow rates to flush the tubing on the tall towers well enough. The weight of ½" i.d. stainless steel tubing on the tall towers, as well as the impracticality and financial cost of installing a continuous length of ½" i.d. SS tubing, Synflex tubing was used instead. Double bonded aluminium core tubing, such as Synflex, is used routinely on tall tower sites (it is recommended in the ICOS specifications: http://icos-atc.lsce.ipsl.fr/filebrowser/download/27251) due to its flexibility, light weight and cost.

The difference between Synflex 1300 and Synflex 3000 is the material used for the inner core of the tubing. The former has a polyethylene inner core, whilst the latter has a nylon inner core. Both plastics outgas; hence, the flow rates at the sites were kept high (~ 20 L min^{-1}) to ensure that no build-up of trace gases being measured at the sites occurred.

Page 7, line 17: I suppose that only the bowl is made out of Perspex. What is the material used for the other wetted parts like the body, the seals and the gaskets?

Response: Correct, only the bowl is made of Perspex on the H_2O decanting bowl. The other wetted parts of the system are made of the following: the housing is aluminium, the filter is sintered polypropylene and the seals are either nitrile or neoprene. The sentence on page 7, line 17, has been reworded to make it clearer what the wetted parts of the system are.

Page 9, line 22 to page 10, line 1: If the temperature is maintained at 318 +/- 0.004 K, line 1 must read a few / thousandth of a K . . ., correct?

Response: Correct, changed to thousandths.

Page 10, line 11: 0.25 % volume ratio of water: which dew point is that?

Response: 0.25 % H_2O is -10.3˚C dew point. This has been added to the sentence in parentheses.

Page 10, lines 12 – 14: did you test for potential CO2 losses in the Nafion?

Response: No tests were undertaken by the authors themselves but we had spoken to collaborators at AGAGE and Scripps Institute of Oceanography about the use of Nafion dryers and their tests into cross membrane leakage of CO_2 and CH_4; work presented in Welp et al. (2013). CO_2 and CH_4 bias from cross-membrane permeation was observed by the authors; however, the procedure of passing both the sample air and calibration gases through the Nafion cancels out the bias (Welp et al., 2013).

Page 10, lines 17 – 21: Correction for water vapour interferences: are the correction coefficients listed in Rella (2010) the ones that are implemented in the Picarro software? If so, the direct Picarro output internally corrected for H2O cross-talk can be used right away, correct? Did you also use the Rella (2010) factors for TTA when running the measurements w/o dryer? If so, why didn't you use individually determined correction factors as suggested by Rella et al. (2013) (https://www.atmos-meastech.net/6/837/2013/).

Response: The correction coefficients listed in (Rella, 2010) are implemented in the Picarro software to derive CO_2 and CH_4 dry mole fractions and were used at TTA to correct data to dry mole fractions. No individually determined correction factor were assessed or implemented at TTA due to time constraints and site access issues.

Page 12, lines 11 – 12: It reads like it was a new idea of the authors to use 5% CH4 in Ar (i.e. P5) instead of CO2-doped N2 as carrier gas. However, this is already done by many groups for many years. You may refer to Schmidt et al. (2001) (JGR; http://onlinelibrary.wiley.com/doi/10.1029/2000JD900701/epdf).

Response: The sentence was reworded as part of the first comment on minimising repetition of previously published work. The above reference has also been added to the sentence so that it now reads: "The SF_6 and N_2O analysis method used at RGL and TAC was similar to that described in detail in Ganesan et al. (2013), except P-5 carrier gas (a mixture of 5 % CH_4 in 95 % Ar; Air Products, UK) is used (Schmidt et al., 2001)."

Page 16, line 13: what is a "significant change"? Was the change detected automatically or manually (visually)? Which criterion was applied?

Response: Any changes in the coefficients from the curve of the nonlinearity correction would be classed as a significant change. The word significant was removed from the sentence to make the sentence clearer. Changes were detected manually when comparing the previous coefficients with the new coefficients. The sentence was altered to make this clearer.

Page 16, lines 14 – 15: "A second order non-linear curve is fit to the data . . .": Does that mean that the Picarro has a non-linear response? If so, please state it clearly and elaborate on it and quantify the effect when erroneously neglecting the non-linear response.

Response: For all of the UK DECC network sites there is a small non-linear response on the CRDSs. The non-linear response is not thought to be due to calibration gases used within the network as the non-linear effect has been seen when NOAA standards have been used to calibrate instruments that use GCWerks. An extra figure has been added to shown an example of the non-linear fits used within the network (Figure 4) and histograms to show the offsets between data with and without the non-linear corrections (Figure 5). Additional information has been put in the paragraph to exemplify what the effect would be on the data if the non-linear correction wasn't applied. There was a small difference between the two steps in data correction, with a median value of < -0.01 µmol mol^{-1} CO_2 and < -0.002 nmol mol^{-1} CH_4 (linear corrected - non-linear corrected data).

Page 17, lines 9 – 11: If I understand correctly, only a one point calibration approach is applied. This only works when assuming no detector signal at zero concentration, right. Was that tested and how was the one point calibration approach applied when a GC-MS signal > 0 was detected for species-free air?

Response: Yes, the one point calibration approach does assume that no detector signal at zero concentration for the Medusa GC-MS and was tested when the Medusa system was being developed at Scripps Institute of Oceanography (Miller et al., 2008; Arnold et al., 2012). Weekly, a system blank is analysed on the Medusa GC-MS, whereby He is passed through the pre-concentration system instead of a sample gas to the GC-MS. The system blank allows the detection of signals >0 in species-free gas and GCWerks can then integrate the contamination peak. A blank correction can then be implemented to cancel out the contamination.

Page 17, lines 25 – 26: how did you make sure that there were no traces of N2O and SF6 in the zero air? The zero air, was it real air having N2O and SF6 trapped or was it a N2/O2 mixture? How about the Argon content in the zero air?

Response: The zero air used for the non-linearity tests was analysed for traces of N_2O and SF_6 during the non-linearity test sequence. Any cylinders with detectable N_2O or SF_6 contamination were not used. A sentence has been added to clarify this. The zero air used for the tests was a synthetic blend of N_2/O_2, with Ar contents < 0.01 µmol mol^{-1}.

Page 18, 8: troubleshooting is mentioned here but it remains too vague if and how and how often troubleshooting was required. See one of my general comments above.

Response: Created section 5.4 to discuss troubleshooting of data within the network.

Page 18, line 18: I suggest skipping the explanation of the naming convention of the stripchart files. This is irrelevant.

Response: Removed.

Page 18, line 23: remove "with time stamps corresponding to the beginning of the measurement, and stored" It isn't of importance here.

Response: Removed

Page 19, line 5: add a table with the thresholds for the maximum allowed standard deviations?

Response: Added in Table 7 with parameter thresholds for all CRDS filters.

Page 19, line 24: remove explanation of the naming convention.

Response: Removed

Page 20, line 1: is it important that there was a 4:1 compression ratio?

Response: Removed

Page 21, lines 3 – 5: how often did it happen that the processing routines filtered false negatives? Is that a time consuming (and important) task to review the automatically filtered data?

Response: When the data filters were introduced to the software and the parameterisation of the filters had not been finely tuned, there were a number of occasions when the air filter was not set high enough and real pollution events were filtered out. Through reviewing the stripcharts, this was spotted and the standard deviation filter was increased to 10. It is important to check all of the data, including filtered data, to ensure that no instrumental problems are missed due to the filtering of data. Within GCWerks, all data is shown within the stripcharts, so the viewing of the stripcharts is not that time consuming (a day's worth of data can usually be checked in about 5 minutes).

Page 21, lines 9 – 10: Is the flagging of spurious data a manual process? If so, I suggest clarifying it by saying "Spurious data are manually flagged and a justification . . . can be added and logged."

Response: "manually" added.

Page 21, line 11: what is "GCcompare"?

Response: GCcompare is data visualisation software, where data within a tab delimited format can be imported to the visualisation software and the time series data can be overlain with other time series data from other sites for the same compound to look and investigate errors.

Page 22, lines 18 – 19: where are the 21 nmol mol-1 coming from? Add a reference.

Response: The amplitude data is from Mace Head. This has been clarified in the sentence.

Page 23, lines 2 -3: Add a statement that the trend of 0.8 nmol mol-1 per year is lower than the global trend. You may refer to the latest WMO GHG bulletin #12 (https://library.wmo.int/opac/doc_num.php?explnum_id=3084). What do you mean by seasonal trend, isn't it the annual growth rate?

Response: There is a 0.8 nmol mol^{-1} amplitude in the seasonal trend at MHD, not the annual growth rate. The sentence has been altered to clarify this.

Page 23, chapter 6.2: this is all largely text-book knowledge and can be considerably shortened.

Response: The section has been shortened.

Page 25, Summary and conclusions: It only summarizes what was said before. I would like to read some kind of outlook and some recommendations that go beyond a simple description of the setup as given above. Topics to be potentially addressed could be: are there any modifications planned (based on some lessons –learnt); are there any major flaws in the setup which cannot be easily changed anymore; with the experience gained during the few years of operation, would the setup again look the same when you may be able to once more start from scratch?

Response: A new recommendations section has been added (section 7) to discuss recommendations based on experiences within the network. Additionally, future work planned in the network has been added the conclusion.

Page 26, lines 11 – 13: The reference given here to underline the benefit for such measurements for GHG inventory verification was published in 2011, i.e. before the presented measurements were implemented. Either remove the reference to the emission verification on page 26 (and in the abstract) or elaborate on the benefit of additional observations for the GHG inventory assessment based on tall-tower measurements and inverse modelling.

Response: Removed

Page 31, footnote a to Table 1: I cannot find the data on the ICOS carbon portal as indicated.

Response: When accessed on 12/01/18, data was found to be available in the ICOS data portal search (https://data.icos-cp.eu/portal/#search?station=%5B%22Mace%20Head%20%22%5D).

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