

Reply to anonymous referee #1

We gratefully appreciate the time taken for the referee to read and evaluate our manuscript. We thank her/him for the helpful comments and suggestions to clarify issues and to improve the content, readability and presentation of the manuscript. Below we address each question, suggestion, correction or criticism individually. Referees' comments are shown in blue. Responses are in regular font. Quotes from the manuscript are in quotation marks, with altered manuscript wording given in bold type.

This paper describes the history and technical aspects of an in-situ analyser that has been running in Lauder, NZ since January 2007. The spectrometer measures a variety of trace gas species, but this paper focuses on methane, carbon monoxide, and nitrous oxide. This paper describes a high quality, unique dataset that promises to be very valuable to the scientific community.

General comments:

The paper is well written, although it is quite long and reads much like a technical report or detailed owner's manual. The most interesting parts of the paper (in my opinion) are sections 1, 5.12, 6, and 7.

Reduction of manuscript length was also recommended by referee 2. We are glad to hear the referee enjoyed sections 1, 5.12, 6 and 7. The content is technical and detailed by the nature and focus of the manuscript (and stated as such in the last paragraph of the introduction). We are looking at the long term technical performance of the FTIR analyser system (both good and bad points), which no one has done before. We would like to think that the technical details presented would assist a research group that was thinking of setting up a long-term network of multiple FTIR systems (i.e. performance, stability, reliability and logistics). Such information is not in any current peer reviewed research papers, technical manuals or the instrument manual, thus whilst detailed we are not repeating material (and hopefully seen as a welcome addition to FTIR analyser literature). This manuscript could be viewed as a continuation of FTIR analyser performance reports by Griffith et al., 2012 and Hammer et al., 2013. Long term performance was not in the scope these papers. We also believe such detailed investigation and reporting are within the AMT journal remit and scope. AMT papers such as Andrews et. al., 2014 and Winderlich et al., 2010 are similar in aim, manuscript length, technical/detail scope and evaluation techniques.

I was unable to determine why some sections of the paper were put into appendices whereas other sections were not.

Sorry that this was not clear. Appendices A and B are related to the MALT spectral retrieval analysis, common to all FTIR systems, not just to the Lauder prototype. These retrieval improvements have not been published (in peer viewed literature) before. Appendix C describes the custom-made calibration suite. Details of such are not required in the main body. Appendix D describes the method used to determine local baseline conditions. It was easier to put into the appendices as baseline is mentioned prior to its determination, hence referencing to an appendix is more efficient and logical than detailing it on first mention (in section 2, Location). Routine maintenance details were moved to Appendix E to reduce manuscript size and increase readability, but still relevant technical details for FTIR analyser operation.

I recommend that the authors consider putting more of section 5 into appendices, and in the main body state only information that is required to understand the time series analysis.

This is a good suggestion, but the focus of the paper is not just to provide support/interpretation for time series analysis, but to also understand instrument operation (performance, stability, reliability) as whole along with practicalities of logistical servicing.

Section 5 is abnormally large compared to other manuscript sections. To reduce the size, section 5.4 (routine maintenance) has been moved to the appendices (as appendix E). Details about intel port configuration (section 5.2.3) and the front-end pump (section 5.2.4) have been reduced. Additionally, details pertaining to the air sampling line maintenance in section 4 have been moved to appendix E.

We decided sections on interferometer performance, Data QC/QA filtering and concentration dependent bias (sections 5.4, 5.10 and 5.8.3 respectively) should stay in the manuscript as such FTIR analyser details have not been published before and pertinent in assessing instrument performance (and in interpretation of the dataset time series). The detailed section on instrument upgrades (section 5.2) is needed as the changes mentioned have a large impact on the dataset time series uncertainty estimates and provide a heuristic link between the operation of FTIR analysers prior and post cell pressure flow decoupling. There still are FTIR systems in use (worldwide) that have not been upgraded.

We would have liked to reduce the amount of detail in section 6 but like the FTIR this is the first time the Lauder flask sampling system and related time series has been published. We cannot simply reference already published material, hence the length of this section.

Changes to the manuscript:

Appendix E added.

In the last paragraph of section 5.3 we have added a sentence stating:

“Extended periods of automation are possible (such as at remote unmanned sites) with a different measurement schedule but given that the FTIR is located on-site and accessible, regular checks and intervention are not an issue. **Details on routine maintenance can be found in appendix E.**”

Section 5.2.3 and 5.2.4 has been shortened (reduction in technical detail).

Details pertaining to the air sampling line maintenance in section 4 have been moved to appendix E.

All figures in the manuscript from figure 7 onwards have been relabelled, as figures 5 and 6 are now figures E2 and E3. Sections 5.4 to 5.12 have been relabelled due to section 5.4 now appendix E.

Note: All further replies to comments are relate to the new section and figure numbering in the manuscript.

Specific comments:

P2L12: You may want to motivate your work by reminding the reader that there are few emissions in the SH, so these SH mid-latitude measurements are crucial for pinning down the true background values.

Thanks for this advice. There is an indirect referral (via the references) to this point in the introduction: “There is also a need for increased coverage in the southern hemisphere (Thompson et al., 2014; Wells et al., 2015), which is relatively data sparse compared to the northern hemisphere.” and a direct referral: “Such conditions also make it an ideal site for clean air trace gas observations.”. In section 2

we state: “All these conditions make Lauder an ideal site to take baseline measurements (baseline conditions are defined Appendix D).”

To strengthen and reiterate this point in the summary (section 9) the manuscript has been changed to read:

“Despite these misgivings, the current FTIR system employing single WS calibrations is sufficient to capture CH₄, CO and N₂O seasonal and annual trends in southern hemisphere **mid-latitude baseline** atmospheric composition within GAW reproducibly guidelines.”

Whilst we state Lauder is a good site for ‘clean air’ measurements we do not explicitly mention that southern hemisphere (*anthropogenic?*) emissions are less than the northern hemisphere.

Technical comments:

P14L17: Did you also assess the modulation efficiency of the FTIR, along with the phase and FOV?

For the fitting of the Lauder spectra the modulation efficiency is not retrieved. The FOV is fitted instead as this gives better fits due to more consistent and lower fit residuals. This does not mean the ILS is not monitored as the fitted FOV is effectively acting as a proxy ILS diagnostic. Since the input aperture is fixed (all measurements made at with the same aperture size, 1.5mm), and the focal length of the IRCube input optics is constant, the FOV is a static quantity. Fits of the FOV should be constant. The ILS is dependent on the FOV (see Griffiths, 2007), thus any changes in the fitted FOV are indicative of a probable change in the ILS, which in practice, means a change in the alignment.

The manuscript (section 5.4) has been changed to read:

“The field of view (FOV) and spectrum phase are fitted to monitor of linewidth and asymmetry. **The ILS modulation efficiency is not retrieved. The FOV is fitted instead, as this gives more consistent and lower fit residuals whilst effectively acting as an ILS diagnostic, i.e. changes in the fitted FOV are indicative of an ILS alignment, acquisition or analysis issue.** The fitted FOV and phase are displayed in Fig. 5c. There is a gradual decline in phase, but the overall phase is very small (< 0.01 rad) indicating a stable near symmetric ILS. The small step changes in phase are related to a change in the cell temperature sensor, laser replacement and operation of the FTIR with a different FOV. The theoretical FOV of the IRCube is unvarying at 21.73 mrad, (apart from brief testing period in mid-2011). ~~thus any deviations in the fitted FOV indicate an issue in spectra acquisition or analysis.~~ Prior to September 2011 the calculated FOV was lower than expected but still stable. This was because the background spectra acquisition aperture setting (3 mm) differed from the sample spectra acquisition aperture setting (1.5 mm). The background aperture size was set to 1.5 mm in September 2011. After this change the fitted FOV agrees well with the physical FOV.”

P15L25: I would have liked to see more of these linear regression curves. You show one in the appendix, so either refer to that figure or plot a few more here.

On P16L4, we state that only Pressure RCS corrections are used. An example of an N₂O RCS_p linear regression curve is given in Fig. A1b. This figure is now referred to in the manuscript.

Section 5.6.1 in the manuscript has been changed to read (this change is part of more an extensive manuscript change to answer another question by referee 1):

“**The linear regression includes errors in the measured pressure and dry mole fraction measurement spread. For example, Fig. A1b displays the retrieved N₂O dry mole fraction as a function of cell**

pressure from tests conducted in December 2013, the resulting RCS_p is 0.005 ± 0.0008 ppb hPa⁻¹ (from table 1).”

P16L8: You have not yet defined QC/QA.

Thanks for spotting this.

The manuscript has been changed to read:

“This is not uncommon, both H13 and Lebeque et al. (2013) also found such experiments challenging. With strict **data quality assurance and quality control (QC/QA)**, based on cell temperature and retrieved water absolute amounts along with the relative difference between sample and calibration amounts, the associated RCS corrections are minimised (**QC/QA filtering detailed in section 5.10**).”

Also altered in the manuscript the section header:

“**5.10 Data quality assurance and quality control** ~~(QC/QA)~~”

P16L23: How do you determine RCS_p ?

Good point, we state that “Experimental determination of RCS_p is easily done” but then neglect to tell the reader how we do it. Sentences have been added to the manuscript to explain how data for RCS_p derivation is obtained, how RCS_p is calculated along with an example.

The manuscript has been changed to read:

“Pressure RCS (RCS_p) corrections need to be applied as cell pressure during sample and calibration measurements differ up to 100 hPa prior to cell pressure and flow decoupling (Fig. 4a). Experimental determination of RCS_p **is performed by taking repeated measurements of dry cylinder air (usually the TC or WS) at different cell pressure, at stepped pressure increments, spanning the cell pressure operational range (see table 1). Other factors such as cell flow rate and cell temperature are held as constant as possible. Multiple measurements per pressure step are taken and averaged. The RCS_p is the gradient from a simple linear regression of the retrieved dry mole fraction (response) to the cell pressure (predictor). The linear regression includes errors in the measured pressure and dry mole fraction measurement spread. For example, Fig. A1b displays the retrieved N₂O dry mole fraction as a function of cell pressure from tests conducted in December 2013, the resulting RCS_p is 0.005 ± 0.0008 ppb hPa⁻¹ (from table 1).**”

P18L3: retrieved dry mole fractions *to* that of the assigned

Thanks for spotting this. The grammatical error in the manuscript has been fixed on L2 (not L3).

The manuscript has been changed to read:

“From the measurements of the calibration gas an instrument response function (IRF) is constructed to map the retrieved dry mole fractions **to** that of the assigned value.”

P18L5: Do you mean to refer to Eqn (2)?

Yes, most definitely. Thanks for spotting this error.

The manuscript has been changed to read:

“The FTIR has been shown to have a linear response (H13) thus the IRF can be approximated by a first-degree (linear) polynomial, as in Eq. (2).”

P19L28: A step change is an indication **of** an acute incident

Thanks for spotting this. The grammatical error in the manuscript has been fixed.

The manuscript has been changed to read:

“A step change is an indication **of** an acute incident in the FTIR, FTIR acquisition procedure or a WS change”

P22L27: Suggestion: "Our approach is to take regular measurements..."

Thanks for the grammatical suggestion, it reads a lot better (eliminates the double use of the word 'take').

The manuscript has been changed to read:

“**Our approach is to take regular measurements** of a target cylinder.”

P26L16: This approach would **be* need*ed** when comparing...

Thanks for the grammatical suggestion.

The manuscript has been changed to read:

“This approach would **be needed** when comparing the measurements taken in conditions of high variability (i.e. during nocturnal boundary layer inversion events).”

Figure captions: Please make the figure captions self-explanatory. For example, Fig 10 shows scaling factors, but does not discuss what is being scaled.

We also took the referee’s advice to review all figure captions (taking into account information provided in figure legends) to provide better clarity and consistency.

For the example given in figure 8, the term calibration is added in front of ‘scale factor’. Also, the abbreviation ‘SF’ is not mentioned/referenced in the manuscript. ‘SF’ has been replaced with ‘A_{sf}’, which is referenced in the manuscript (section 5.8). In the figure 8 a, b legends, ‘SF’ is also replaced with ‘A_{sf}’. A new figure 8 has been inserted into the manuscript.

The caption for Figure 8 has been changed to read:

“Figure 8. (a) CH₄ 7-day running mean **calibration** scale factor (**A_{sf}**). Black data points are the drift corrected **calibration** scale factors. Uncorrected **calibration** scale factors are shown as grey data points. The vertical dashed red line indicates WS replacement and (b), CH₄ scale factor uncertainty. (c and d) same as (a and b) but for CO. (e and f) same as (a and b) but for N₂O.”

The caption for Figure 3 has been changed to read:

“Figure 3. Cell temperature measurements. From **January 2007 to** September 2010 cell temperature measurements were made with an **LM335** integrated circuit sensor attached to the outside of the cell. The invitro PT100 temperature measurements started in September 2010 and **then** replaced with a Type-J thermocouple in April 2013 (measurements outside the range 31-35 °C were filtered out). Box plots provide a statistical summary prior and post LM335 temperature sensor change. Vertical grey

dashed lines indicate an event in which changes to FTIR hardware, operating conditions or analysis were made (FTIR instrument events explained in Sect. 5.11).”

The caption for Figure 4 has been changed to read:

“Figure 4. (a) Cell pressure (black) and cell flow rate in flow mode (red) during air sample measurements. After the April 2013 upgrade the flow rate is set to 0.5 Lmin⁻¹ and cell pressure is set to 1100 hPa. The sudden drops in flow rate on three occasions (post upgrade) are due to MFC power supply faults. Data taken during such faults is filtered out. **Overlaid are box plot statistical summaries for cell pressure and flow rate prior to the April 2013 upgrade.** (b) Difference between air sample and WS cell pressure.”

The caption for Figure 7 has been changed to read:

“Figure 5. (a) **Bruker IRcube** interferogram ZPD signal and the mean signal level of the associated spectra calculated over the range 2450-2550 cm⁻¹. (b) Spectra SNR over the range 2450-2550 cm⁻¹. The 2450-2550 cm⁻¹ region was selected due to a lack of absorption features and is representative of the **spectrum** continuum **level**. (c) Fitted spectra phase and FOV.”

The caption for Figure 9 has been changed to read:

“Figure 9. Complete-IRF **linear** fit residuals (with 1 σ uncertainty bars) from measurements of multi-tank suites N14, N15 and W10.”

The caption for Figure 10 has been changed to read:

“Figure 10. (a) The difference between calibrated CH₄ measurements of the three multi tank suites (N14 black, N15 red and W10 blue) against assigned tank values with 1 σ uncertainty bars. The coloured diamonds are the assigned WS dry mole fraction used to calibrate each respective set of suite measurements using the scale factor method. The ~~coloured~~ dash-dot-dot lines are the estimated concentration dependent biases (CDB) arising from applying the scale factor method, for each measurement suite. The grey shaded area indicates the typical baseline concentration range at Lauder. (b and c) the same as (a) but for CO and N₂O respectively.”

The caption for Figure 12 has been changed to read:

“Figure 12. (a) Calibrated time series of CH₄, (b) CO and (c) N₂O **for all processed data (grey data points), quality-controlled data (black data points) and quality-controlled data during baseline conditions (red data points).**”

The caption for Figure 15 has been changed to read:

“Figure 15. (a) Baseline CH₄, (b) CO and (c) N₂O FTIR measurements and flask samples. FTIR trend analysis fit and the trend analysis linear fit component are over plotted **in red.**”

The caption for Figure A1 has been changed to read:

“Figure A1. Retrieved N₂O dry mole fractions as a function of cell pressure from tests conducted in December 2013. (a) **Region 1 (2150–2320 cm⁻¹)** N₂O spectral analysis (with 1 σ uncertainty bars). (b) Same as (a) but for **Region 4 (2097–2242 cm⁻¹)** N₂O spectral analysis.”

The caption for Figure B1 has been changed to read:

“Figure B1. A typical background spectrum (**black line**) taken on 8 August 2014 (cell pressure of 1.6 hPa) and corresponding background spectrum (**red line**) with water absorption spectral features removed. MALT spectral fit regions are shaded in grey.”

The caption for Figure E3 has been changed to read:

“Figure E3. Difference between the FTIR MKS 902 **cell pressure sensor** and external PTB110 pressure sensor prior to any calibration adjustments. Comparisons are conducted at a cell pressure of approx. 960 hPa (atmospheric pressure). “

Lastly, nine figures contain grey vertical dashed lines corresponding to instrument events. Only in the first instance (fig. 3) is an explanation given in the figure caption. The subsequent eight figure captions omit a description. We decided that repetition was not needed, as it explained in the first instance and explained in the manuscript (section 5.11). Upon the editors’ decision, a repetitive descriptor per figure can be added.

Fig 17: I had trouble seeing the blue triangles. Could you make them bigger?

Sure. The blue triangle (flask data) symbol size is increased by 33% in Figs. 15 a, b, c (any larger they could be disproportionate). The new figures are incorporated into the revised manuscript.

As an example, the new Fig 15a is displayed directly below, and the old fig15a beneath it (for comparison).

Fig15. (A) new:

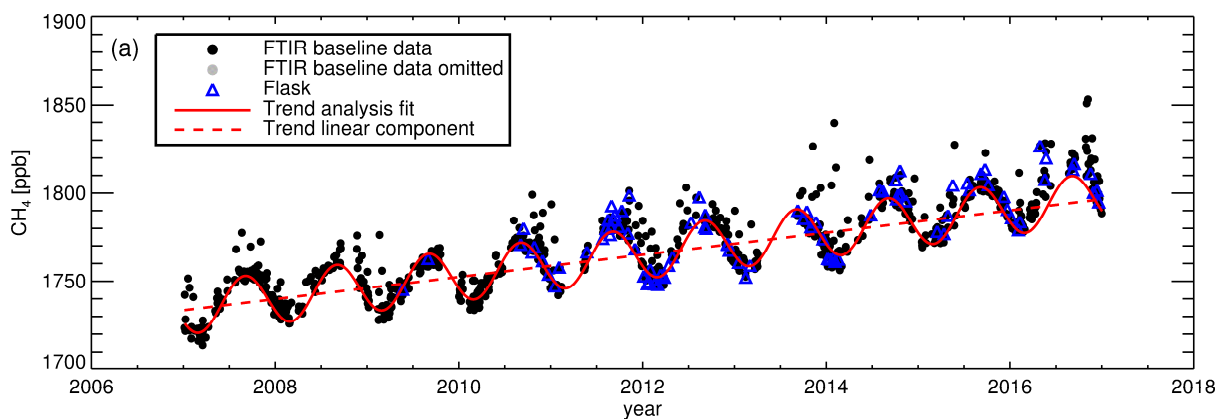
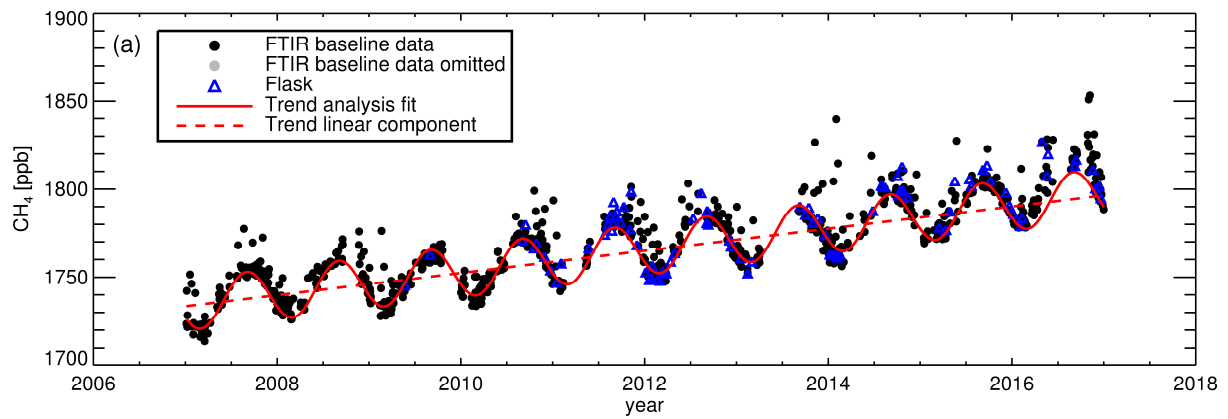


Fig15. (A) old:



References mentioned:

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Hammer, S., D. W. T. Griffith, G. Konrad, S. Vardag, C. Caldw, I. Levin, Assessment of a multi-species in situ FTIR for precise atmospheric greenhouse gas observations, *Atmos. Meas. Tech.*, 6, 1153–1170, 2013

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