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Atmospheric Measurement Techniques
Discussions

Interactive comment on "The development and evaluation of airborne in situ N₂O and CH₄ sampling using a Quantum Cascade Laser Absorption Spectrometer (QCLAS)" *by* J. R. Pitt et al.

Anonymous Referee #2

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General Comments:
In this paper, Pitt et al. describe the use and characterisation of a Quantum Cascade Laser Absorption spectrometer (QCLAS) on the UK FAAM large research aircraft for in situ measurements of methane (CH_4) and nitrous oxide (N_2O). The atmospheric importance of these two species is well known and is covered appropriately in the Introduction. Continuous analysers already exist for measuring atmospheric methane mole fractions and have been deployed on this aircraft (such as the cited Fast Greenhouse Gas Analyser (FGGA), which also measures carbon dioxide). The successful deployment of an instrument for measuring atmospheric concentrations and fluxes of N_2O is especially welcome. Its installation on an aircraft allows the estimation of N_2O fluxes from important disperse or area sources, such as wetlands (for CH_4) or agriculture (both CH_4 and N_2O).

Two main instrumental issues were investigated: (1) the need to correct for the effect of water vapour on the spectral absorption lines, and (2) the origin and correction of pressure effects. The availability of the Fast Greenhouse Gas Analyser (which has been previously calibrated to WMO standards) and the use of reference gas mixtures of known concentrations has allowed the precision and accuracy of the QCLAS to be established.

Overall, the paper is well written and should be published after addressing the specific and technical comments below.

Specific Comments:

In the Introduction (p. 8862), many of the cited papers on airborne measurements refer to methane. It would be useful to know which greenhouse gases were measured in the different studies. This would also help to emphasis the lack of measurements on N_2O (and hence the selection of N_2O for the case study).

Water–Vapour correction
As is standard, a water–vapour correction (Section 2.4) is applied to report the measurements as dry–air mole fractions and thereby remove any effects arising from large changes in ambient water vapour concentrations. The first approach used built on work by the same group using the FGGA, as reported in the cited paper by O'Shea et al. (2013b). In that paper, the use of a nafion drier is also described and the decision taken to remove it. In the current paper, there are clearly issues with long–term stability, with the July 2014 calibration results differing significantly from

the initial set made in November 2013. I did not altogether find the the discussion on this completely clear. Are the authors sugesting that the spectral intensity of the laser is changing? There are no spectra presented to show that the absorption lines are distinct and therefore give a good measure of the baseline. It is simply stated that a region between 1275.3 and 1275.8 cm^{-1} contains ro-vibrational transitions of N_2O, CH_4 and H_2O (p. 8865).

A second method is investigated based on the vendor TDLWintel software. This makes a significant improvement to the retrieved mole fractions. Why was this not used initially? What are the advantages and disadvantages of the two approaches? More discussion is needed here. If I understand correctly, the water broadening coefficients of 1.6 and 1.8 for CH_4 and N_2O, respectively, are not absolute values (in which case, the units are missing) but relative to the air-broadening coefficients. Is this correct?

Pressure effect

A pressure effect is observed leading to large changes in retrieved atmospheric mole fractions over short periods of time. This is linked to changes in cabin–air pressure and further tests suggest that the changes in cabin–air pressure affect the optical alignment of the spectrometer. This was found to be a major term in the error budget. An approach is developed which effectly removed this effect, albeit at the expense of data capture. Calibration measurements made at the relevant pressure levels are used and as the authors acknowledge can be several hours apart from the actual measurements. Clearly, more frequent calibration would help to address this issue. Are there reasons why this was not done? Further work (outwith this paper) is needed on this topic as the authors acknowledge that potentially useful data are removed when rapid pressure changes occur, e.g., during vertical profiling or aircraft manoeuvres.

The authors should clarify 'these campaigns' (p. 8874, line 22). Presumably, the pressure was not recorded during the 2014 campaigns.

Case Study

An illustrative case study is included (Section 4). As discussed in the paper by the same group (cited paper by O'Shea et al., Atmos. Phys. Chem., 2014), the FGGA methane measurements were used to estimate a regional–scale methane flux from Scandinavian wetlands. I would very much have liked the case study presented here to have been expanded to give some idea of the fluxes. This would have required discussion of the methods (possibly involving atmospheric chemical transport models). At a minimum, it would be useful to know (with assumptions made on wind speed and boundary-layer height), the minimum detectable fluxes of nitrous oxide (and methane) implied by the QCLAS precision and accuracy and how these compare with emission estimates from the UK national inventory for this region.

Technical comments:

There is a tendency to use the name of the instrument manufacturer as an adjective describing the instrument (e.g., "Aerodyne Research Inc. Quantum Cascade Laser Absorption spectrometer" or "Los Gatos Fast Greenhouse Gas Analyser"). This is very clunky. It is more normal practice to describe the instrument and put the manufacturer and model in brackets. This is done elsewhere in the paper, for example, thermoelectrically cooled photovoltaic detector (Vigo Systems, Poland).

There are minor typographical errors and other comments:

- p. 8862, line 17: "top-down" measurement could insert atmospheric to re-inforce the point that atmospheric measurements can be used to constrain surface emission fluxes.
- p. 8863, line 17: "small" in "small instrument response time" should be replaced with "fast" or similar.
- p. 8863, line 18: "optics and detectors available at near-IR wavenumbers of 6000

cm^{-1}" should be rephrased to, e.g., "optics and detectors available in the near-IR region around 6000 cm^{-1}". One could also give the wavelength.

- p. 8874, line 26: "was flown to further understanding of the underlying issues" needs to be rephrased.
- Figures 7 and 8: Add flight number to figure captions

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 8859, 2015.