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Atmospheric Measurement Techniques Discussions

Interactive comment on "An airborne infrared laser spectrometer for in-situ trace gas measurements: application to tropical convection case studies" *by* V. Catoire et al.

Anonymous Referee #2

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Catoire et al., "An airborne infrared laser spectrometer for in-situ trace gas measurements: application to tropical convection case studies"

This manuscript describes the design and initial flight performance of a quantum cascade laser-based sensor for detecting CO2, CO, CH4, and N2O. The lasers are multiplexed together into the same optical cell / detector with about 25% duty cycles each. Measurements are obtained for each species every 1.6 seconds. The instrument conducted flights onboard the DLR Falcon and the data were examined to show how boundary layer air was transported into the outflow of deep convection. Accurate measurements, and improved instrumentation, for upper tropospheric/lower stratospheric

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sciences are critical for understanding dynamics and photochemistry in this region. While a manuscript on sensor development/testing for these topics is well-aligned with the scope of AMT, I do not recommend publication of this manuscript at this time. As noted below, the manuscript in its current form is incomplete both in the description of the system performance (lab and field) and in its presently demonstrated flight capabilities for UT/LS science. For AMT, there should be a robust sensor/testing/deployment component and at least some demonstration that the technique/sensor/system has the capability to do interesting science (even if the science is not developed in the AMT manuscript; but the current "science" examples seem insufficient for most UT/LS science - and too long anyway relative to the sensor part).

1. Sensor/measurement/field demonstration

For AMT, I expected far more (and more rigorous) experimental / laboratory / field demonstrations/analyses of sensor performance. For example, the Allan deviation plots in Fig. 7 only extend to 360 s, barely past the local minimum. For airborne CH4, CO2, and N2O measurements, where very small changes are observed on relatively large backgrounds (particularly in the UT/LS), the drift at timescales beyond a few minutes is absolutely critical toward the ability for its use in scientific analyses. It also provides a measure of how often one needs to calibrate to remove long-term drift sources. With turnarounds in the Allan plots near 100 s – a timescale which is a bit sooner than most diode laser-based sensors - it is even more important to characterize the extent of drift at longer timescales. As discussed in the science demonstration part of my review (point #2), drift greater than 1 ppbv N2O is already troublesome for UT/LS science. It is expected to be worse at longer timescales based upon typical laser-based sensor performance, but one doesn't know with the data presented here.

For a flight-based instrument, the frequency of calibration is not reported. Was the sensor calibrated in flight at all? And, if not, couldn't a calibration gas be flown and its output directed into the sample cell under flight conditions? Can results be shown before and after a given flight? Can the second cell (laser tuning) be used as a proxy of

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flight performance? Just calibrating with a WMO standard doesn't mean the instrument maintains that calibration for longer times. What is meant by "850 measurements" – 850 different calibrations at different dates for each gas? If so, this is a rich dataset and should be plotted vs. time (reported/actual vs. time). More information (results and discussion) on the frequency of calibrations is needed for an airborne-based system.

Spectroscopy: why wasn't the N2O absorption line near 2179.3 cm-1 probed by the CO laser? This (the CO one) QCL should be able to scan over this range (via continuous scanning by current or jump scanning). This N2O line is $\sim$ 1.5X stronger than the one the authors chose near 1249.67 cm-1. It may help improve the sensitivity to N2O.

Also for spectroscopic considerations, what is the influence of the nearby CO peak on the retrieved CO2 peak near 2064.40 cm-1? This seems like it may impact the fit at the 0.3 ppmv level (CO2 isn't discussed much after the Allan plot, which is presumably due to the laser malfunctioning – also an indication that the manuscript is incomplete). How does a change of, e.g. 20 ppbv CO, impact the retrieved CO2 mixing ratio? There is no discussion of this potential "cross-talk" between the measurements in the manuscript.

For N2O, CH4, and CO2 in particular (where especially high precisions/accuracies are required), does water vapor broadening need to be considered (my guess is not since the measurements are at reduced pressure) – please quantify/state if this is correct.

A full statistical error accounting needs to be conducted. Pressures have uncertainty, the calibrations have uncertainties, the temperature is uncertain, the spectral fits are uncertain. How accurate is the laser tuning rate known? What about the leak rates? What about the incomplete flush rate for a 6 second reporting period? What is the total error budget with all of these included? It seems like many of the smaller though perhaps significant uncertainties – though stated - are ignored in the overall error discussion. Perhaps a table can be added with a full, statistical error accounting.

Why does the alignment need to be adjusted every 10 minutes in-flight? How much does the raw detector signal change before it re-aligns? Does the misalignment impact

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the fringing significantly (i.e. not constant over time)? To what extent is the alignment changing (angle, distance, etc. – whatever units are best to describe this)? 9174, line 20

How bad are the fringes without the third actuator (i.e. turned off)? By saying they are “attenuated”, do the authors mean that they are below a certain value? If so, what value in optical absorption units? 9173, line 25

How often is the second reference channel added for absolute wavelength calibration? Can a high concentration cell (to achieve comparable absorption as in the sample cell) be added in flight to at least estimate the in-flight performance in terms of optical absorption noise equivalence? P. 9173, line 5

A measurement time of 6.4 s and flush time of 8.8 seconds suggests that each individual measurement is slightly dependent upon the last measurement. 9176, line 10

2. Scientific capabilities

AMT is for sensor development/methodologies, and the current manuscript is too focused on the “science” applications, i.e. field data and interpretation. With that said, I’m not sure the instrument performance can really do UT/LS science well. Yes, the authors demonstrated some case studies where it can see convective outflow – but most UT/LS changes in N2O, CH4, and CO2 are comparable to or below the precisions/accuracies are needed. A clear example is the statement on p. 9182, line 15 where the authors state that several flights during SHIVA showed no CO enhancements. Is this really because these were not fresh convective clouds (what evidence suggests this besides speculation)? Or was it because the CO enhancements were just not evident from the sensor precision? I also noticed that no flight N2O data was presented in the figure. The methane mixing ratios are largely within the error bars of one another. I believe the CO sensor can be useful for science in the UT/LS; I’m not as convinced the N2O/CH4/CO2 measurements are as useful there for high-quality

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science.

The in-flight CO validation with the mountain measurements looked good, but the other species' measurements were not intercompared at all. In the absence of this, more data is needed to convince readers that the calibrations on the ground are still valid when in flight.

3. Other

The introduction is too long and too general – TDLAS is a well-known and applied technique; QCLs have been often applied for atmospheric science, even airborne campaigns (in contrast to the statement on p. 9169, line 4-6). A review of the overall field is not necessary. What is more needed is specific context to state-of-the-art CO2/CO/N2O/CH4 airborne-based measurements (and particularly laser-based methods). What are the specifications and methods used in the Aerodyne/Harvard QCLS system? What about the Diskin/Sachse DACOM sensor often onboard the NASA DC-8 aircraft? What about researchers who fly Picarro/Los Gatos/Aerodyne systems on research aircraft for UT/LS work? What does this system bring to the community that the others do not? Is it smaller? More compact? More sensitive? More stable? More adaptable for including other species? Easier to deploy? I note that SPIRIT only measures all gases in 6.4 seconds whereas other sensors do this at 0.1-1 Hz. In other words, what makes this sensor unique compared to these other systems? More elaboration on the motivation of this sensor and its relationship to published sensors is needed.

Discussion of the limit of detection is not terribly relevant for CH4, CO2, and N2O – given that they rarely go below 1800 ppbv, 390 ppmv, and 320 ppbv in the troposphere and perhaps only slightly less in the lower stratosphere. The precision is the more relevant factor. See, for example, Fig. 2 in Wofsy et al. Phil. Trans. Royal Society A, 2011, doi:10.1098/rsta.2010.0313, p. 2073-2086. Same with the discussion on the upper limit of detection – optical saturation will not occur in the UT/LS.

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p. 9169, lines 1-5: ICLs are not limited in their use because of their 3-6 micron spectral region availability. Indeed, the fundamental bands of methane, N2O, and CO all lie in this region! They are limited because they are less mature of a technology than QCLs – their packaging isn't as good (lens coatings), only one real vendor (Nanoplus), and their field performance is still being improved (thermal regulation, optical fringing).

Like in the introduction, the results from SPIRIT should be compared/discussed in detail after the sensor performance is described. What are the benefits of SPIRIT vs. those other systems?

Overall: The novelty of the three QCLs coupled into the same optical cell/detector and for use in atmospheric science is a topic certainly worthy of publication, but it has to be tested/explained/demonstrated rigorously. And one needs to show that it can address a key atmospheric question of interest with whatever capabilities are demonstrated. This manuscript does neither in its current form (a commercial CO sensor could do the science observed in the later sections). I will acknowledge that comparing new technologies, especially flight-based ones, to those sensors with years/decades of development/maturation is not entirely fair, but it is the authors who are trying to make such strong claims to make very challenging measurements, on an airplane no less. Perhaps the application/platform should be re-considered or additional data/experiments presented for relevance to UT/LS work. The revision of this manuscript would be major and may want to be considered as a new submission.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 9165, 2015.