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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 #1

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Review of "An airborne infrared laser spectrometer for in-situ trace gas measurements: application to tropical convection case studies"

Overview: Catoire et al. present a new laser absorption spectrometer name SPIRIT for airborne observations of CO, CH4, N2O, and CO2. They present data from the lab and in-flight, and use flight data of CO and CH4 to estimate the fraction of boundary layer air lofted to altitude during convection. This manuscript is well placed in AMT. Much of the manuscript is clear and adequately describes the instrument. However, there remain some major gaps and flaws that need to be addressed. I outline these concerns below (broken into major and detailed issues). Given the importance of my major

concerns, I cannot recommend publication at this time. Once these concerns outlined in detail below have been addressed sufficiently, I would reconsider the manuscript for publication in AMT.

Major Issues: The author's never state whether they dry air prior to sampling. From what I read, I assume they author's neither dry nor measure water vapor. This presents a major problem in interpreting measured quantities-particularly in applications such as convective studies in the tropics. For all the trace gases the authors report, global standard is to report dry air molar fraction (which is not always identical to volume mixing ratio). It is essential that the authors report what they are doing regarding water vapor. If a correction is to be applied, it can be complicated and needs to be discussed (see Rella et al., Atmos. Meas. Tech., 6, 837–860, 2013). To present an instrument that measures CO, CH4, N2O, and CO2, it is essential that dry air molar fraction on a recognized reported scale are given. Just to put numbers on this dilution impact (which doesn't include the impact of interferences)-2% water vapor changes methane diluted versus dry molar fraction by $\sim$30 ppb.

Following this thread-there is also a major gap in calibration. Standard practice is to calibrate the instrument in-flight with a span of gases covering the dynamic range of interest, or demonstrate why this is not needed-neither is presented in the manuscript. This is particularly important for flight instruments were a number of variable factors can cause drift, and of crucial importance for long-lived trace gases where it is essential to report values onto known global scales. There appears to have been no calibration, or other standards flown in flight-this would cause all the data to have much greater uncertainty and be unable to be placed on a known calibration scale. The authors do report large ranges for accuracy from spectroscopy, but do not investigate drift that may occur in flight. This is linked to the limited discussion of pressure and temperature control in-flight. These must be held very stable to ensure no drift in the complex aircraft environment. Regular or continuous flow of a standard gas while profiling in the aircraft would provide definitive information on how the instrument performs

in this challenging environment. Without this information or similar demonstration of stability of performance we cannot know the instrument is indeed accurately reporting concentrations while in flight.

There is almost no discussion of CO2 given the laser broke early in the experiments. The authors should remove CO2 entirely from the manuscript since there really is not demonstration of a functioning airborne CO2 instrument that has necessary precision and accuracy to be of scientific value.

The overall instrument performance falls far below most contemporary standards for CO2, CH4, and N2O. Airborne signals of N2O are typically only on the order of 1ppb or so (Kort et al.,), and the instrument presented here has neither the precision nor accuracy to measure this (> 4ppb). Airborne methane is regularly measured with precision and accuracies better than 2ppb (Peischl et al. JGR 2013, Karion et al. GRL 2013) and placed onto global standards, much better than reported here (> 8 ppb).

To address these major issues the author's need to clearly state what they are doing with regard to water vapor. They need to generate dry air molar fractions and explain how these values are generated. They need to state what scale they are reporting on. They need to demonstrate that the airborne instrument shows no artifacts during profiling or when T or P are changing dynamically. Finally they need to accurately present the capabilities of the instrument relative to community standards and uses. As reported, this instrument performs almost an order of magnitude worse than demonstrated flight QCL (Santoni et al.) or cavity ring-down systems (Rella et al., Karion et al., Peirschl et al.). For scientific analyses of long-lived greenhouse gases, much better precision and accuracies are typically needed. As presented, this instrument would only have value in applications where accuracy and high precision are not needed-and this must be stated explicitly.

Detailed Issues: It is misleading to imply it is easy to change lasers and study other gases with excellent performance-this is made clear by the loss of the CO2 laser and

lack of replacement.

p. 9168 Line 29: Actually Yacovitch et al. uses a near-IR DFB diode laser and not a QCL-this should be corrected. This also is true in a more recent study not cited Smith et al., 2015 ES&T. Another recent relevant citation would be Richter et al., Appl. Phys. B (2015) 119-131, where a DFG source is used.

p.9174 Line 4. Teflon is a very risky material to use if studying CO2-and should be avoided.

p. 9174 Line 12: Manually actuating a valve should not be a part of any airborne system-this will almost guarantee problems with pressure control on vertical profiles.

p. 9178 line 1: See Santoni et al-N2O is not necessarily a non-sticky molecule as when measured with high precision some 'stickiness' has been observed. It may simply be hard to observe when instrument noise is so large (9 ppb).

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