

## ***Interactive comment on “Evaluation of the Airborne Quantum Cascade Laser Spectrometer (QCLS) measurements of the carbon and greenhouse gas suite – CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and CO – during the CalNex and HIPPO campaigns” by G. W. Santoni et al.***

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Reviewer 1:

Comment: "Table 2b: The first column name seem rather cryptic, are they needed?"

\*Response: We have added the following to the caption to explain the syntax: "The naming corresponds to the usage of each cylinder as follows: H1, HIPPO-I; H2, C4567

HIPPO-II; H3, HIPPO-III; etc.; CN, CalNex; LS, Low Span; HS, High Span; REF, Reference."

Comment: Figures hard to read.

\*Response: All figures noted by Referees were updated with more readable versions.

Reviewer 2:

Comment: "With a small time shift, the reference/zero signals shown in Fig 8 seem to be anti-correlated with the ambient pressure. This is not a problem as long as the reference/zero frequency is high enough to capture the features. However, the question is whether every 15 minutes is frequent enough (when a profile takes about 15 minutes as well). Clearly the frequency is not high enough, as the zero/reference signals reached different maximums and minimums during each pressure cycle (assuming that pressure variations cause the zero/reference fluctuations), and thus a bias may have been introduced. What are the uncertainties related with this type of bias, and for each species? Furthermore, can the authors explain why the zero/reference signals are anti-correlated with the ambient pressure?"

\*Response: It should first be noted that the relationships shown in this flight are not the same relationship exhibited in other flights. We have examined the relationship between pressure (including atmospheric pressure as measured by GV sensors, inlet pressure measured by the QCLS, cabin pressure measured by GV sensors, etc.) and the reference-concentration and found no consistent relationship. While this particular flight shows one relationship, other flights have either no relationship or the opposite relationship (when pressure is regressed against reference tank concentrations, as sampled by the QCLS). We agree that 15 minutes on this flight is not frequent enough, but the physical constraints of the HIPPO campaign (e.g. refills of the gas decks are only possibly in New Zealand and Alaska) limited the frequency of calibration. There is always a trade-off that must be made in terms of duty cycle versus calibration cycle as it takes time to equilibrate a stream of zero/reference gas in the cell. Fifteen minutes

was determined to be the optimal frequency: sufficiently frequent so as to account for Allan drift but also sufficiently infrequent so as to maintain gas deck pressures to last one entire southbound or northbound leg of each HIPPO mission. Gas decks with pressures reaching < 500psig have exhibited drift. It should also be noted that the cabin pressure, which is really what drives the alignment of the lasers, and consequently the overall measurement accuracy, typically lags ambient atmospheric pressure.

Comment: P9693/L21-25, the CO<sub>2</sub> external absorption path is flushed with pure nitrogen. How about the external absorptions of CH<sub>4</sub>, N<sub>2</sub>O, and CO? How large is the influence of external absorptions on CH<sub>4</sub>, N<sub>2</sub>O, and CO measurements?

\*Response: QCLS-CO<sub>2</sub> is purged with Nitrogen because the cabin of the plane has elevated and changing concentrations of CO<sub>2</sub> and the external absorption of CO<sub>2</sub> is not negligible as the path length within the sampling cell is of the same order of magnitude as the path length of the light external to the cell (i.e. it is a single-pass spectrometer).

In contrast, the QCLS-DUAL is not flushed with Nitrogen for several reasons. The path length of the light outside of the sampling cell is ~1m, while the effective path length of the sampling cell is 76m (i.e. it is a multi-pass spectrometer). The absorption in the external path is much smaller and pressure-broadened and this effect is compensated for by pulse-normalization. The pulse-normalization path (at aircraft cabin pressure) is designed to be nearly identical in length to the sample beam-path (excluding the beam-path of the measurement cell), also at aircraft cabin pressure. There are no sources of CH<sub>4</sub>, N<sub>2</sub>O, or CO on board the plane and therefore the concentration changes are very small. Taken together, these measures make the effect of the absorption external to the sampling cell negligible for the QCLS-DUAL.

Comment: P9695/L2-9, were the measurements made during ascending/descending or during constant ambient pressure? This may be a place to demonstrate whether ambient pressure has an impact or not on the measurements.

\*Response: Measurements were made continuously with gaps every 15 minutes for

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calibrations. We did not want to further bias the results by having calibrations occur, for example, on ascending segments while measurements were made only on descending or level-flight segments.

Comment: P9699/L14-16, what are the mean and standard deviation of the differences between gasdeck calibrations and assigned values? This will give readers an idea about possible discrepancies, at least for the gasdeck that has a chance to be calibrated.

Response: Direct gasdeck versus source tank comparisons were available in a few instances. The range of difference between the gasdeck values, as measured directly against the primaries, and the fill tank values, as measured directly against the primaries were typically within 1 SD of the NOAA calibrated uncertainties, for the instances in which we were logistically able to make both measurements. This was only possible before the southbound leg of HIPPO, after the northbound legs of HIPPO (when gasdecks had been used to ~500 psig, down from fill values of ~2000psig) and on two instances during CalNex.

As an example, we can look at the GasDeck calibrations of CH<sub>4</sub>, N<sub>2</sub>O, and CO for H4a (southbound) which were made on 2011-05-09:

CH<sub>4</sub> N<sub>2</sub>O CO

2209.80 353.83 329.05 (HS)

1803.85 331.54 144.64 (PRI)

1672.59 301.15 58.86 (LS)

And compare the gas deck values to the secondary fill tanks used to fill the Gasdeck four months earlier:

—————CH<sub>4</sub> N<sub>2</sub>O CO

CAL\_NewOnly\_2011-01-12\_CC62384 2210.50 353.96 328.91 (HS)

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CAL\_NewOnly\_2011-01-14\_CC56519 1803.68 331.65 146.47 (PRI)

CAL\_NewOnly\_2011-01-14\_CC37815 1672.87 301.29 58.57 (LS)

We used the gasdeck calibration values only to verify that no contamination occurred during the flushing and filling of the gas deck. If the differences were larger than  $3\sigma$  the uncertainty assigned to the NOAA primary tanks (from Table 2a), the gas deck bottle in question (e.g. GasDeck\_LS) was purged, refilled, and recalibrated.

Comment: P9700/L22, I do not see an advantage of the normalized range here. Why not show the difference in real ppb directly?

\*Response: Without normalized ranges the transitions would appear exactly the same as these are square wave switches from a tank with a known quantity of CH<sub>4</sub>, N<sub>2</sub>O, and CO and are transitioning to an ultra-zero tank. N<sub>2</sub>O for instance is transitioning from a concentration of 319.3 ppb to a concentration of 0.00 ppb and without a normalized range would look perfectly flat. It is the surface effects that occur with N<sub>2</sub>O that cause a potential bias, and the normalization in the figure allows us to show that explicitly.

Comment: P9710/L2-8, what are the differences between "airborne measurement compatibilities" and "long-term compatibility" reported here? Are they derived from the same dataset?

\*Response: Those are the same.

Additional changes to be made in the revised paper:

\*Acknowledgements will be updated to read as follows:

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\*Affiliations for two people need to be corrected: #2 - RJ: Universidad Nacional de Colombia - Bogota, Department of Chemical and Environmental Engineering, Bogota, Colombia

#3 - EAK: University of Michigan, College of Engineering, Department of Atmospheric, Oceanic and Space Sciences, Ann Arbor, Michigan, USA

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