

Response to L.E.C. Christensen:

1. *Abstract should be clearer: 26 pptv was demonstrated in lab not flight.*
 - 1.1 Response to Referee: We'll clarify that 26 pptv refers to a laboratory assessment, not the precision in-flight.
 - 1.2 Changes to Manuscript: On page 1/Lines 11-12, the text is now, "Laboratory assessments demonstrated that the spectrometer has a 90 % response time of 10 s to changes in HCl and a 30 s precision of 26 pptv."
2. *Line 92: Add O₃ to list of species*
 - 2.1 Response to Referee: We'll update this as well.
 - 2.2 Changes to Manuscript: On page 3/lines 91-92, the text is now, "When selecting the line, the spectral interference from several common stratospheric molecules was considered, including: CH₄, CO, CO₂, H₂O, O₃, NO, and N₂O."
3. *Line 101: Add instantaneous linewidth and SMSR of ICL laser*
 - 3.1 Response to Referee: The SMSR of the ICL has been added to the manuscript. The FTIR spectrometers used to characterize the laser don't have a sufficient resolution to measure the instantaneous linewidth (both of them had a resolution of 0.125 cm⁻¹). However, we see no error associated with linewidth during measurements at such low pressure that Doppler broadening dominates. We find no measurable difference between the calculated and observed Doppler width, so we believe the instantaneous linewidth to be 0.001 cm⁻¹ at most. Because we did not rigorously measure this, though, we do not present this metric in the manuscript.
 - 3.2 Changes to Manuscript: On page 4/lines 101-102, the text is now, "The laser emission is centered at 2963 cm⁻¹, or 3.37 μm, and has a side-mode suppression ratio greater than 25 dB (Borgentun et al., 2015)."
4. *Line 114: You sure this is heating? The main lasing mode can get pulled red from feedback which looks like heating.*
 - 4.1 Response to Referee: We're providing a general description of the problem of laser feedback. We did not experience these issues for the HCl instrument, as we started with the optical isolator to avoid such feedback. We'll update the manuscript to be clearer about this.
 - 4.2 Changes to Manuscript: On page 4/lines 113-114, the text is now, "The polarizer and quarter-wave plate (shown collectively as (2) in Fig. 3) serve as an optical isolator that prevents laser light from ultimately reflecting back into the laser housing, which is known to cause unwanted feedback."
5. *Line 161: What pressure did you regulate the cell to during lab experiments – those expected during balloon flight? Later in the manuscript, you mention no regulation during flight experiment below 60 mbar.*
 - 5.1 Response to Referee: This is a good point, thank you. Unless otherwise stated, the cell pressure in lab was regulated to 53 hPa—the same as during the balloon flight, when feasible.

5.2 Changes to Manuscript: On page 6/line 164-165, the following sentence has been added. “Unless otherwise specified, cell pressure in the laboratory was regulated to 53 hPa.”

6. *Do you have any laser spontaneous emission issues?*

6.1 Response to Referee: After receiving this review, we evaluated whether we experience this issue. There are very weak methane absorption lines in our laser scanning region. We created theoretical spectra that showed we could saturate several of these lines if we flowed pure methane through the cell. We did this and found that 4 % of the light was not absorbed at these wavelengths. While the effect is fortunately minor, we very much thank the reviewer for bringing this up. We have re-fit our flight data for HCl, adjusting the calculated laser power impinging on the detector to remove 4 % of the light that is from spontaneous laser emissions. Figures 10 and 17 have been updated with this new analysis. Some of the discussion in the main text that quantifies these comparisons was also modestly affected by this correction and has been updated as well. We have also increased the uncertainty in our accuracy by 1 % to account for the uncertainty in the percentage of measured light that is from spontaneous emissions. The general conclusions regarding the instrument validation via MLS and O₃/HCl correlation remain the same. We plan to add a bandpass filter to the optical path to block these spontaneous emissions in future campaigns that involve this instrument. Thank you again for bringing this to our attention.

6.2 Changes to Manuscript: On page 9/lines 273-281, the text is now, “Separately, we evaluated to what extent the laser was emitting light through amplified, spontaneous emission (ASE), incoherent light emitted by the laser at a broad array of wavelengths. While this light from ASE should be orders of magnitude lower than light at the lasing frequency, light that is at wavelengths outside the cavity mirror reflective coating will pass through the cavity and create an apparent offset in the total laser power. To determine whether ASE was an issue in the instrument, very weak methane lines in the scanning region were saturated with pure methane and showed that 4 % of the light is from ASE, which is corrected by decreasing the calculated laser signal by 4 %. This approximately translates to a 4 % increase in HCl mixing ratios that were determined without this correction. The uncertainty in accuracy is 8 % (5 % from uncertainty in spectral parameters; 2 % from uncertainty in ICOS mirror reflectivity and HCl-surface interactions; 1 % from uncertainty in ASE signal). In-flight accuracy is given a conservative upper bound (11 %) due to less stable in-flight conditions.”

On page 11/lines 340-342, the text is now, “The spectrum in Fig. 9 (bottom panel) is a 30 s averaged spectrum that corresponds to an ambient mixing ratio of 1.19 ppbv HCl, measured at atmospheric pressure of 26.3 hPa.” (Fig. 9 caption was similarly updated)

On page 11/lines 352-353, the text is now, “All observations were greater than $3 \times dl$, where dl is the detection limit, defined as the noise-equivalent absorption for a 30 second average (70 pptv).”

On page 14/lines 451-452, the text is now, “HUSCE HCl and O₃ measurements in the lower stratosphere (65-80 hPa) yield a slope of 0.00057 ± 0.00007 (Figure 17).”

In Table 1, reported uncertainty in accuracy for our instrument has been increased by 1 % for both columns.

Figure 10 has been remade with corrected flight data and corrected percent differences compared to average MLS observations (though the mean absolute percent difference is still 8 %).

Figure 17 has been remade with the corrected data, updated regression coefficient (rising from 0.00055 to 0.00057), and updated 95 % CI for that coefficient.

7. *Line 285: Did pre- and post- ringdown times agree? Why isn't ringdown time measured periodically during flight – is it needed?*

7.1 Response to Referee: While we collected ringdown spectra at the beginning of the flight for the wavenumber affiliated with the HCl absorption feature, every spectrum in flight includes an off period at the end that can serve as a ringdown spectrum. A ringdown measurement is therefore performed with every scan, and they indicate the cavity time constant was consistent throughout the flight. We agree this can be clarified in the manuscript and have updated it accordingly.

7.2 Changes to Manuscript: On page 10/lines 303-305, the text is now, “The instrument was powered on for 26 minutes prior to recording spectra to 1) allow time for system controls to stabilize, and 2) record ringdown spectra to determine the reflectivity of the ICOS mirrors for the wavelength at which HCl absorbs, $R = 0.9998$. The measured reflectivity in flight agrees with the measurements made in the laboratory. Ringdowns were also recorded at the end of every spectrum, when the laser is turned off, that was collected during the balloon descent and showed no significant change in the mirror reflectivity throughout the campaign.”

8. *Line 320: I'm not following the lineshape in Figure 9. Figure shows water is to blue of HCl while the tail in Figure 9 is red. Is this due to instrument function?*

8.1 Response to Referee: The theoretical spectrum provided in Figure 2 shows H_2O^{18} absorption at 200 ppmv. This is to illustrate that even extremely elevated H_2O levels only subtly interfere with HCl. However, the H_2O^{18} absorption line blue of the HCl line is not perceptible during the flight. H_2O occasionally--and transiently--reaches up to 150 ppmv, but it is usually closer to 20 ppmv or lower. We did not detect significant water vapor levels when the spectrum in Figure 9 was collected. The red tail on the absorption feature is not H_2O^{18} but rather an instrument function idiosyncratic to ICOS. The red tail is due to the cavity time constant being comparable to the tuning rate of the laser (i.e. the intensity of light entering the cavity at any given time is still decreasing exponentially as the laser scans through the subsequent wavelengths, resulting in a red skew in absorption features). This is taken into account when fitting. Thank you for pointing this out; we will update the description to clarify.

8.2 Changes to Manuscript: On page 11/lines 343-344, the text is now, “The absorption feature has an asymmetric skew due to the cavity time constant being comparable to the tuning rate of the laser. This skew, characteristic of all ICOS spectra, is taken into account when fitting the HCl absorption feature.”

9. *Line 339: Did you have an inlet tube during flight? Did you also Silco coat it? Where was the opening placed – e.g. below the gondola?*

9.1 Response to Referee: Yes, we did. We'll add a brief description of the in-flight inlet in the manuscript that answers these questions.

9.2 Changes to Manuscript: On pages 9-10/lines 293-296, the following text was added.
“The instrument inlet during flight protruded from the center of the pressure vessel, parallel to the length of the vessel. The inlet was a SilcoNert2000 coated steel tube with an inner diameter of 1.1 cm and a length of 62 cm, with the tip being 54 cm away from the side of the gondola. This length was chosen to ensure sampled air was sufficiently separated from the gondola. The inlet was also heated to 310 K and insulated.”

Note that referenced line numbers may differ slightly from the original manuscript due to added text in response to comments.