## Response to Referee 1:

- 1. Page 1, Line 13. The abbreviation of MLS is incorrectly marked here.
  - 1.1 Response to Referee: The parenthetical MLS was intended to specify which satellite measurements were compared with our observations. After reading the reviewer's comment, we agree that could be clearer. We've updated the text accordingly.
  - 1.2 Changes to Manuscript: On page 1/lines 13-14, the text is now, "The observations agreed with nearby satellite measurements made by the Earth Observing System Microwave Limb Sounder within 10 % on average."
- 2. *Page 3, Line 67. The response time is related to the volume of the cell and sample flow rate. This argument needs the support of the detailed information of these parameters.* 
  - 2.1 Response to Referee: For our instrument, we do state the dimensions of our cell, the sample flow rate of the pumps, and the net flush rate for the cell under normal operation (Section 2.2). The commercial HCl ICOS spectrometers sold by Tiger Optics and Los Gatos Research do not have detailed descriptions published in open literature. Hagen et al., 2014 does report that their CRDS instrument operates at 8 sLpm and that the cavity is 90 cm. However, they do not provide the diameter of their mirrors (and their instrument schematic suggests the cell is not a simple cylinder anyway; Hagen et al., 2014, Figure 3). As such, the volume of their cell is unclear. Therefore, we are unable to provide most of these parameters for the other instruments not developed by us.

What they all do provide is the response time, though, which allows for the most direct comparison with our instrument. The response time for an analyte like HCl is more complicated than the flush rate, which could be determined by the cell volume and sample flow rate. Unlike instruments measuring gases like CO<sub>2</sub>, the response time of an HCl instrument also depends on factors such as heating of the cell and the material that the cell is composed of. This is because HCl is especially prone to being scavenged by surfaces. As such, the response time is the culmination of a large portfolio of factors beyond cell volume and sample flow rate—many of which are not explicitly provided for the other cited instruments. As such, the simplest and most direct comparison is the one we provide.

- 2.2 Changes to Manuscript: None
- 3. Section 2.2, page 5. What is the bandwidth of the detector? The mirror reflectivity or effective cavity length was determined by the ring-down measurement. What is the ring-down time of the empty cavity? By using re-injection performance, more light will enter the cavity. It is no longer appropriate to use the base length divided by 1-R to express the effective optical path, which is usually used for laser beam one-time injection into the cavity.
  - 3.1 Response to Referee: The bandwidth of the detector and pre-amp used during flight is 1 MHz. The Stirling-cooled detector and pre-amp has a bandwidth of 1.4 MHz. The manuscript has been revised to include this information.

The ring-down time, which we called the cavity time constant, is 7.9 microseconds for an empty cavity. We have added that number to the manuscript and clarified that we are referring to the ring-down time.

The average, effective optical path is still dictated by the base length divided by 1-R as long as the analyte of interest is not present outside the cavity (if it is present in the extra-cavity volume, then some of the light will be absorbed as it reflects between the first cavity mirror and the re-injection mirror, RIM). More light entering the cavity after first being reflected by the RIM does not affect that additional light's average lifetime in the cell once it enters the cell. That is strictly dictated by the length of the cell and the reflectivity of the mirrors.

Additionally, the effective cavity time constant would not be affected the presence of the RIM, as the time taken for the light to reflect off the RIM is orders of magnitude shorter than the amount of time the light spends in the optical cavity.

3.2 Changes to Manuscript: On page 5/line 155-157, the text is now, "During the HUSCE balloon flight, the detector used was a four-stage thermoelectrically cooled MCT detector that, coupled with a pre-amplifier, had a bandwidth of 1 MHz (Vigo, PVI-4TEMXPXX-F)."

On page 8/line 248-249, the text is now, "A two-stage pre-amplifier and anti-aliasing filter collectively adjusts the gain to 5 x  $10^5$  V A<sup>-1</sup>, with the detector and pre-amp resulting in a bandwidth of 1.4 MHz."

On page 5/lines 144-146, the text is now, "The mirrors have a light loss of 200 ppm at  $3.34 \mu m$  (R = 0.9998, or 99.98 % reflective), which is determined by pulsing light into the cell and measuring the e-folding time for decrease in light intensity (the ring-down time, which is 7.9 microseconds for an empty cavity)."

- 4. Section 4. What kind of interference does "balloon interference" mean? What is the ascent and descent speed of the balloon? Will the release of the helium affect the measurement of HCl? Is the pressure of the sample cell kept constant or the same as the ambient pressure? Will the residence time of the sample in the cavity change?
  - 4.1 Response to Referees: Balloon interference refers to observed air having physically interacted with the surface of the balloon. This is common in balloon-borne campaigns. When air interacts with the balloon surface, the air usually gets 1) some water vapor that outgasses off the balloon, and 2) some thermal energy from the balloon due to its surface being heated by solar radiation. Evidence of balloon interference, therefore, manifests as elevated and highly variably water vapor and temperature levels (stated on page 12/lines 360-362 in the manuscript). This is discussed in more detail in the citation provided in the manuscript: Kräuchi et al., 2016. We've modified the manuscript to better clarify the purpose of this citation. The ascent rate was approximately  $3 \text{ m s}^{-1}$  on average. The descent rate was approximately 2 m s<sup>-1</sup> on average and never rose above 5 m s<sup>-1</sup>. We agree the descent rate should be in the manuscript and have revised accordingly. The release of helium would not affect the measurements of HCl since they were made on descent, and the gondola was approximately 50 meters below the balloon. Also, the release of helium was gradual and sporadic. We have updated the manuscript to better convey the location of the gondola relative to the balloon. The pressure of the cell is kept constant at 53 hPa when feasible—at atmospheric pressure = 60 hPa and above (stated on page 10/lines 309-310).

The pumps are constant volume displacement, so residence time is not significantly affected by pressure changes. The reporting time of 30 seconds for our data would certainly extend beyond any slight change that may occur in residence time.

4.2 Changes to Manuscript: On page 12/line 360-362, the text is now, "There is evidence that balloon interference may have impacted portions of the mid-stratospheric descent, based on anomalous readings from the diagnostic water vapor measurement and the ambient temperature measurement (for more detailed discussion of balloon interference, see Kräuchi et al., 2016)."

On page 10/line 299, the text is now, "The descent rate of the balloon was adjusted in real time, averaging 2 m s<sup>-1</sup> and never rising above 5 m s<sup>-1</sup>."

On page 9/line 279-281, the text is now, "The HCl instrument was secured within a sealed cylindrical pressure vessel and mounted to a gondola suspended approximately 50 meters below the balloon, to separate the instrument platform from the wake of the balloon."

- 5. During the flight, will the changes in the atmospheric temperature affect the performance of the cavity?
  - 5.1 Response to Referee: The cell is temperature-controlled and sealed in a temperaturecontrolled pressure vessel that maintained a constant pressure, so the instrument is isolated from atmospheric temperature variability (page 9/line 281-283 of the manuscript). For that reason, atmospheric temperature changes do not perceptibly affect the performance of the cavity.
  - 5.2 Changes to Manuscript: None