Author response to Anonymous Referee #1.

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We thank Anonymous Referee #1 for their postive review of the manuscript. Below are our responses to their comments. Referee comments are *bold and italicised* with our responses below. Where it makes sense we have broken up the referee's specific comments into smaller chunks to hopefully make the responses clearer.

5 1 Specific comments:

Why was HITRAN 2008 used?

Here and in the past we have used HITRAN 2008 because of the speed of the computational calculations and working under the assumption that any biases, which might change with different spectroscopy, can be accounted for by corrections such as those detailed here.

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How does the bias compare using 2016?

We've gone ahead and re-fitted some of the spectra using HITRAN 2016 instead of HITRAN 2008. While this process is still ongoing, initial results suggest that HITRAN 2016 makes a difference of approximately 1%, bringing the uncorrected (via comparison to in situ measurements) retrieved CO_2 and CH_4 closer to the in situ scale. We have re-fitted the period of overlap

15 between the open-path FTIR and in situ measurements, and are re-fitting the entire time series for all gases and will update the results with these.

In summary, HITRAN 2016 reduces the bias bewteen open-path FTIR and in situ measurements by about 50% for CO_2 and CH_4 , but has little effect for O_2 . The 2.67% bias for CO_2 is reduced to 1.41%, for CH_4 the bias reduces to 1.61%, while for O_2 it increases slightly to 3.24%. The uncertainties as assessed by the standard deviations barely change, but appear to be slightly bigher with HITRAN 2016 then HITRAN 2008 for CO_2 and O_2 and O_3 and slightly reduced for CH_4 .

20 higher with HITRAN 2016 than HITRAN 2008 for CO_2 and O_2 and slightly reduced for CH_4

What about using independently measured line strengths (e.g., Long et al, GRL, 2020, doi:10.1029/2019GL086344)?

Thanks for bringing this paper (Long et al., 2020) to our attention, it's nice to know about. In this case, however, their work is not relevant as they re-measure linestrengths for the bands used by the Total Carbon Column Observing Network at 6228
cm⁻¹ (30012 <- 00001) and 6348 cm⁻¹ (30013 <- 00001), but here we use a stronger band near 4980 cm⁻¹ (20012 <- 00001).

What is the magnitude of the short-path correction? From Figure 2 it is apparent that the short-path + temperature effects combined are large, how much of this is attributable to the short-path correction?

Typically the short-path (hereafter 'SP') spectrum is about 1-2% of the intensity of the long-path (LP) spectrum. The SP 30 spectrum is essentially independent of the LP spectrum; that is, it simply adds to it.

A reference SP spectrum was recorded after any operator intervention or realignment. The relative intensity can vary, but the shape changes are minimal with time. A scaled reference SP spectrum is subtracted from each recorded spectrum, as described in section 2.1.1 (line 117). Its magnitude (or scaling factor) is determined for every measured spectrum. This is done by a least-squares fit of the reference SP spectrum in the 5000-5500 cm⁻¹ region, where there is no contribution from the LP because it is

 $_{25}$ blacked out by H_2O . After subtraction, the corrected spectrum has zero intensity in the blacked out bands of the LP spectrum. This is illustrated in Figure 1.

The reduced continuum intensity after SP subtraction leads to an increased retrieved amount of trace gas, dependent on the relative contributions of the SP and LP spectra. This is because the trace gas absorptions retain the same depth in the corrected spectrum while the continuum decreases. For weaker bands, for example CH_4 , the effect is approximately proportional to the SP/LP fraction, but amplied for stronger bands such as the CO_2 bands at 4850 cm⁻¹ and 4977 cm⁻¹ in Figure 1.

The temperature correction, using the spectrum-fitted temperature rather than the measured temperature, usually decreases the retrieved mole fractions and therefore works in the opposite direction to the short-path correction. The fitted temperatures are typically 3-10 °C lower than locally measured, varying with time of day and solar radiation. The magnitude of this correction is influenced by two factors:

- 45 1. the lower measured air temperature leads to a higher calculated air density, and therefore a proportionally lower mole fraction of the trace gas for a given column of trace gas; and
 - 2. the retrieved column amount of the trace gas depends in a complex way on the temperature dependences of the absorption lines contributing to the spectrum.

In general, the spectroscopic effect is smaller than, but of the same sign as, the density effect.

- A series of figures show four retrievals for CO_2 (Figure 2), CH_4 (Figure 3) and O_2 (Figure 4) for one day, October 22, 2018. The different retrievals are:
 - 1. the uncorrected retrieval using HITRAN 2008 (red);

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- 2. the uncorrected retrieval using HITRAN 2016 (black);
- 3. the short-path corrected retrieval using HITRAN 2016 (green); and
- 4. the short-path and temperature-corrected retrieval using HITRAN 2016 (blue)

As mentioned in the response above, from HITRAN 2008 to HITRAN 2016, the 2.67% bias for CO_2 is reduced to 1.41%, for CH_4 the bias reduces to 1.61%, while for O_2 it increases slightly to 3.24%, while the uncertainties do not appear to change.

Why was O_2 not used to correct for pressure variations?

60 If used, O_2 would act as an alternative measure of the air density instead of using P/RT; however, the precision of the O_2 measurement is less than that of the pressure and temperature. It therefore considerably increases noise in the derived trace gas mole fractions. Note this case is different from TCCON, where O_2 is used to calculate the total air column.

The running mean of the well-mixed time series also removes potential sensor drift. Can you verify that the observed variability was indeed due to atmospheric variability and not drifting bias? Could you also add the Allan deviation plot?

On the time scales at which we have looked at data with the running mean, the bias relative to in situ measurements is constant within uncertainties.

Here we include some Allan deviation plots for the night of November 23-24, 2018 (17:15 - 04:30). For all gases, the determined Allan deviations are in agreement with the repeatability estimates given in Table 3 of the manuscript: 0.24 ppm (AD) vs 0.26 ppm for CO₂ (Figure 5); 2.0 ppb vs 2.0 ppb for CH₄ (Figure 6) 17.6 ppb vs 17 ppb for CO (Figure 7); and 22.2 ppb vs 21.7 ppb for N₂O (Figure 8).

I didn't understand the argument for the SNR scaling with path length. If anything, I would think that turbulence should cause additional beam spreading and power loss. Could you explain that more? Do you know the size of the beam at the reflector array? If it is smaller than the array, could this explain it?

We propose the following re-wording of the paragraph at L187 in the article:

- For a (constant) detector-noise limited spectrum measurement, increasing pathlength from 600m to 1110m to 1500m
 would decrease spectrum signal and SNR as the inverse square of the pathlength, while in practice we observe an approximately inverse linear falloff. This is consistent with a component of noise proportional to signal, presumably due to a combination of turbulence and photon noise. The depth of absorption lines increases in proportion to pathlength, so the net absorption:noise ratio (and thus measurement precision) remains roughly constant with pathlength.
- 85 While we don't have an accurate measurement of the size of the beam at 1500 m, we observed that it was significantly larger than the retro-reflector array.

There seem to be step changes in CO and N2O (near 2018-09 and 2018-10, respectively). Do you know what caused those?

90 Yes, these were caused by author stupidity. The CO and N_2O weren't originally fitted for the period at 600 m one-way path, resulting in 2749 missing points at the start of their time series. However, in the plot displayed previously they were plotted as if they started at the same time as the other gases, and therefore the time axes for them are incorrect. This was addressed prior to all analysis, but somehow that figure was not updated. With some embarrassment, we've fixed this figure in the revised manuscript.

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The step-change in N_2O therefore occurs over the data gap in October, when the system was modified to use a 12" telescope. The step-change in CO occurs when we shifted from 1110 to 1500 m pathlength. Both N_2O and CO are currently retrieved with limited signal and we are not sure what caused the step-changes, though in both cases the later periods appear to correspond to more realistic background southern hemisphere mole fractions of the gases.

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Did you correct the open-path measurements for water content?

Yes, we did apply a correction to dry-air mole fractions to account for the retrieved H_2O content. We've clarified this in the revised manuscript at around line 108 by adding the text "The retrievals are corrected to dry-air mole fractions using the retrieved H_2O ."

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Do you see any difference in bias for the different path lengths?

Unfortunately we do not have in situ data that overlap with the open-path measurement periods at different pathlengths, so we can't assess this. We have performed a preliminary assessment of the bias after setting the instrument up elsewhere over a 1.3 km one-way path and it is consistent with that determined here. Ideally we would perform co-located in situ measurements at

110 a variety of different pathlengths and locations (similar to the aircraft/AirCore comparisons done by TCCON) to determine the instrument's consistency between deployments and under a range of conditions.

What is the expected sensitivity for CO and N2O with different reflectors?

Excellent question! We haven't rigorously investigated this, but with some alternative reflectors that were lying around the lab
we see throughput at these frequencies of three times the current reflectors. This would presumably lead to at least improvement of √3 in the measurement repeatability for CO and N₂O, to approximately 10 ppb and 13 ppb, respectively. Clearly this is not sufficient to be useful for detecting atmospheric signals except under exceptional circumstances; however, there are a couple of strategies that could be employed. We could either seek reflectors designed for maximum signal at this range and create a retro-reflector array alternating reflectors with peak performance for CO/N₂O and CO₂/CH₄, and/or further increase the size
of the current array to allow for more returning signal at pathlengths where the beam overfills the current array size. For all situations, we need to refine our setup perform further tests to assess these possibilities.

2 Technical corrections:

Lidar (DIAL and IPDA) should be added to the list of open path techniques. Probably this should be with TDLAS under a
general heading of single-frequency laser techniques. In addition, the CLADS technique should be mentioned under this heading (e.g., Plant et al, Sensors, 2015 doi:10.3390/s150921315)

We've refined the section discussing open-path technologies following these suggestions and included reference to Plant et al. (2015) and Nikodem et al. (2015).

130 The Oueisser 2016 ref is DIAL, not DOAS.

Noted and corrected. Thanks.

For the LIDAR refs, here is an IPDA system for CO2, CH4, and H2O: Wagner and Plusquellic, Appl Opt, 2016, doi:10.1364/AO.55.006292

135 Thanks, we have included this (Wagner and Plusquellic (2016)).

The Waxman 2017 ref should also be include in the frequency comb part of the introduction

We have made reference to this paper (Waxman et al. (2017)) in the introduction as well.

140 What was the size of the retro array?

The size of the final array was 600 (h) x 625 (w) mm. We've added this information to the manuscript around line 88: with a total dimension of 600 mm by 625 (height x width).

While similar to Griffith 2018, it would be nice to have the full spectrum shown here to provide clarity. It would also be 145 nice to add the short-path spectrum to this figure.

The full spectrum is shown below in Figure 9, along with the short-path spectrum and a zoom (x20) of the short-path spectrum for clarity. We've added this figure to the revised manuscript with reference to it at the end of the section describing the shortpath correction (around line 122, section 2.1.1).

150 Could you label the direction towards potential sources on the polar CH4 plot?

We've avoided this to try to limit misinterpretation of the polar plot radial axis as being distance rather than wind speed. Many of the potential sources are also about the same distance from the open-path as the pathlength itself, and therefore marking them on this polar plot is potentially confusing. Instead, we have marked the location of CSG well and grazing areas on the map in Figure 7. The updated Figure 7 (now Figure 8) from the manuscript is shown below in Figure 10. We also added text to the manuscript referring to the indicated sources at the end of the paragraph discussing the polar bivariate plot (around line 226).

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What averaging time was used for the precision numbers?

The averaging time varied depending on the period used. In each case we picked a period where the atmosphere was wellmixed and there were no obvious changes in trace gas mole fractions due to local sources. These correspond to the periods in 160 Table 1.

Can you also add the O2 time series to Figure 5?

We don't have an in situ measurement of O_2 , so we've not added this to that particular time series plot. The O_2 mole fraction should not vary significantly from 0.2096.

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References

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Figure 1. Long-path spectrum in the 4700 - 5600 cm^{-1} region before (red, as measured) and after (blue, corrected) correction for the contribution from a short measurement path resulting from reflection directly off the secondary mirror.



Figure 2. Uncorrected CO₂ retrieved using HITRAN 2008 (red), and HITRAN 2016 (black), and after the short-path correction only (green) and both short-path and temperature corrections (blue) with HITRAN 2016, for October 22, 2018.



Figure 3. Uncorrected CH_4 retrieved using HITRAN 2008 (red), and HITRAN 2016 (black), and after the short-path correction only (green) and both short-path and temperature corrections (blue) with HITRAN 2016, for October 22, 2018.



Figure 4. Uncorrected O_2 retrieved using HITRAN 2008 (red), and HITRAN 2016 (black), and after the short-path correction only (green) and both short-path and temperature corrections (blue) with HITRAN 2016, for October 22, 2018.



Figure 5. CO₂ Allan deviation plot from 17:15 November 23 - 04:30 November 24.



Figure 6. CH₄ Allan deviation plot from 17:15 November 23 - 04:30 November 24.



Figure 7. CO Allan deviation plot from 17:15 November 23 - 04:30 November 24.



Figure 8. N₂O Allan deviation plot from 17:15 November 23 - 04:30 November 24.



Figure 9. Example long-path spectrum (23:48 Dec 5, 2018, black) and original (red) and 20x zoom (pink) short-path spectrum (21:48 Dec 2, 2018).



Figure 10. Map of the 1.55 km path at EMAI and corresponding elevation profile. The OP-NIR and stationary Picarro were located at the left end of the path and the retroreflector array at the right end. The mean elevation across the 1.55 km observation path is 118 metres above sea level an average of 23 metres above surface elevation. Map © Google Earth.

Period	Start Date/Time	End Date/Time	Num Points
1	20180804 00:00	20180804 05:00	100
2	20180813 08:00	20180813 20:00	239
3	20180821 17:00	20180822 02:00	179
4	20180915 20:00	20180916 06:00	199
5	20181123 18:00	20181124 04:00	199

Table 1. Periods used for calculating the measurement repeatability for each instrument setup.