Response to Anonymous Referee #2 Comment on amt-2022-106 from 05 May 2022

The authors would like to thank the reviewer for their helpful comments that have helped improve the quality of this manuscript. The reviewer's comments are listed below in black with our responses given in red.

This paper describes the first results from a lidar system deployed onboard a research aircraft measuring atmospheric methane. The paper is well-written, and fits well within the scope of AMT. However, a few issues listed below should be addressed before the paper can be recommended for publication.

General comments:
Note that the term "mole fraction" is recommended rather than "mixing ratio", see e.g. https://www.empa.ch/web/s503/gaw_glossary#recommendations. I suggest simply replacing throughout the text. Also I would recommend to consistently use ppb for dry air mole fractions of CH4. Using both ppb and ppm (e.g. Fig. 10 (b)) is confusing to the reader.

We recognize this difference and appreciate the reviewer's input. We have changed references of "mixing ratio" to "mole fraction" throughout the paper. We adjusted the units in Fig. 10b to be completely in PPM.

Comparison to in-situ measurements: The deployment of the different aircraft sampling different altitude regimes really has potential, as indicated e.g. by Figs. 12 and 13 and the associated discussion. I suggest a simple combination of the in-situ measurements within the free troposphere from the C-130 aircraft, the boundary layer in-situ measurements from the B200 aircraft, and the estimate of the boundary layer height derived from the HSRL measurements onboard the C-130, to calculate a partial column XCH4 based on insitu observations that can directly be compared to HALO XCH4. The assumption is that CH4 is well mixed within the PBL and also within the free troposphere. Any advection of air masses with enhanced CH4 above the PBL would clearly stick out as differences between HALO XCH4 and the aircraft derived XCH4.

We recognize how the suggested analysis would be beneficial to understand the partial free troposphere vs boundary layer columns, however for the along track segments shown in Figs. 12 & 13 there are no in-situ spirals that can be leveraged to apportion the different parts of the column as suggested. In the future, we hope to have the necessary SNR from the lidar data to provide a robust assessment of the various partial columns for specific regions such as these. We will indicate, however, that we do show an in-situ spiral column in Figure 10, where the in-situ derived XCH_4 is calculated from various altitudes, so the total column and partial columns can be seen. Additionally, the analysis shown in Figure 18 demonstrates apportionment of the PBL and provides an indication of HALO vs. in-situ.

Dry air mole fraction - impact from H2O: In the in-situ measurement community there is much discussion on drying/conditioning samples before measurement vs. correcting based on simultaneous H2O measurement within the exact same sample. As the authors describe, MERRA humidity is used in the retrieval of XCH4 (the column average dry air mole fraction). The uncertainty in XCH4 introduced by this choice should be assessed, e.g. by comparing MERRA water vapor to that of the in-situ observations.

The topic of XCH_4 impact from H2O presence at the online/offline transmitted wavelengths was also brought up in Reviewer Comment #1. Due to the selection of our offline wavelength, the differential optical depth due to water vapor is minimized near the surface where most of the water vapor and water vapor variability resides within the troposphere. This method of minimizing the differential absorption as opposed to minimizing the differential cross section makes the impact of water vapor on the XCH_4 retrievals negligible. This implies that regardless of the use of MERRA or in-situ profiles of water vapor, the latter of which are only available during spiral ascent/descents, the total impact of using MERRA versus an in-situ profile is small on the total propagated impact on the XCH_4 retrieval.

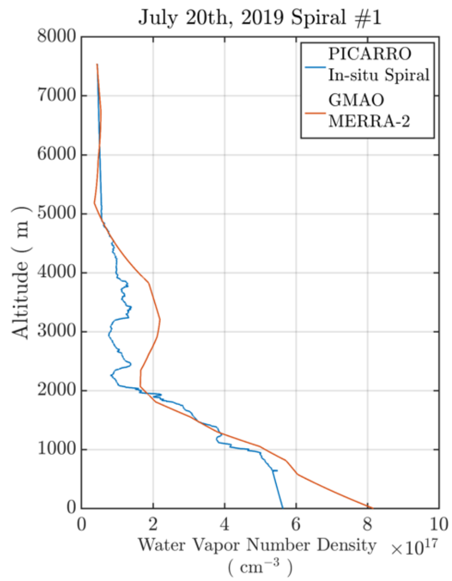

Figure 1 - Comparison of the in-situ spiral generated water vapor profile to the mean profile of water vapor number density generated with MERRA-2 reanalysis for the duration of the spiral overpass.

To support this conclusion, we have included some additional analysis here derived from the July 20th spiral profile shown in Fig. 10b of the manuscript. In Fig. 1 above we have taken the in-situ profile of water vapor number density generated during the spiral and compared to the mean MERRA reanalysis for the spiral overpass. Using these profiles to calculate the water vapor DAOD (pressure and temperature were utilized from each respective source for the computations) we calculate that the DAOD for in-situ is approximately 2.61e-4 and MERRA is 3.00e-4, both falling within 13% of each other, but ~0.09 % of the CH_4 DAOD. Utilizing Eq. 3 from the manuscript, the difference in water vapor DAOD to the XCH_4 retrieval between MERRA and in-situ yields an approximate 0.2 ppb difference. We feel that this is well within the random and systematic uncertainty bounds of our measurement capability and lends credence to the validity in utilizing the MERRA reanalysis for relative humidity inputs. It is necessary to note that under standard flight operations, void of spiral generated in-situ profiles, we report XCH4 using reanalysis atmospheric state to encompass all sources of error, this includes utilizing the relative humidity product.

The following text was added to Section 2.2 within the discussion of MERRA-2 atmospheric state:

"Comparisons of retrievals using MERRA-2 atmospheric state to those using in-situ profiles from spiral maneuvers indicate that differences are <1 ppb. Use of MERRA-2 under normal flight operations serves to then include atmospheric state error within the XCH4 retrieval, as expected for retrievals made in all regions without access to in-situ profiles."

Specific comments:
L271 "over samples" -> "oversamples"

Fixed

L280 "Altitude is used in lieu of MSL for all figures" this is not clear. May be "Altitude above MSL is used in lieu of Altitude above ground level"?

We have adjusted this for clarity.

L280: "post- flight reanalysis" may be drop "post-flight"? I guess reanalysis products are available only for past periods, i.e. after the flights, anyway.

The authors agree and have removed post-flight.

L331: "The superscript will be dropped for simplicity." Which superscript?

We have adjusted this for clarity: "The 'cal' superscript will be dropped for simplicity."

L362: the matrix T should contain the elements that the beta-vector elements are multiplier with, i.e. 0th, 1st 2nd and 3rdorder terms as formulated in the Eq. on line 361. May be simply write down the first and last row of the matrix, and the few elements

We recognize that this comment is the result of not enough information describing the matrix $\boldsymbol{T}$. We have taken the liberty to provide more description as follows:

" $\boldsymbol{T} = \begin{bmatrix} 1 & \delta\bar{\tau}_{CH_4} & \delta\bar{\tau}^2_{CH_4} & \delta\bar{\tau}^3_{CH_4} \\ \vdots & \vdots & \vdots & \vdots \\ 1 & \delta\bar{\tau}_{CH_4} & \delta\bar{\tau}^2_{CH_4} & \delta\bar{\tau}^3_{CH_4} \end{bmatrix}$ is the matrix composed of $\delta\bar{\tau}_{CH_4}$"

Fig. 6: please adjust color selection for the different gains so that color blind people can read the figure. To me HOLE and LOLE are identical, LOLE is very slightly different.

We appreciate the input here. We have changed the color diagrams for Figure 6 utilizing: https://yoshke.org/blog/essays/2020/07/colorblind-friendly-diagrams/ to accommodate color blind people (RGB [204,121,167];[0,114,178];[86,180,233]).

Fig. 6 caption: please explain DEM (I know what it is, but it should be mentioned once)

We have adjusted this for clarity, though it is also mentioned within the text.

Fig. 11: the Y-intercept is not clear. It should be negative, given the slope is larger than one, and the regression line crosses the 1:1 line at around 1900 ppb.

The reviewer is correct, since this the slope is greater than 1 the y-intercept from the fitting routine is negative. The fit provides y=1.0922x – 0.1755. In the scope of the XCH_4 measurements presented this y-intercept does not have a tangible meaning, so we had previously attributed the intercept to where the regression line crossed the plot axes. We have updated Fig. 11 to show the fit parameters directly.

L522: "PA region" – to make this clear to non-US readers (AMT it is a European journal) may be add a label to Figs 11 (a) and (c)

We have adjusted this for clarity in the text and provided an indication in Figure 12a,c.

L571: I don't see any cross-hatched area in Fig. 14, may be I am misunderstanding something

We have altered the wording here to be more descriptive of the region we wish to emphasize.

"Here all instruments register enhancements, emphasized within the lower flight track section of Fig. 14b."

L612 "spatial" -> "spatially" or drop

"spatial" was dropped due to redundancy.

L659: the dial DAOD estimated at SSE shown in the inset of Fig. 17 (b) (magenta symbol) is around 0.2875, not at 0.9243 as given in the text. Please clarify.

This was an error on our part, we have gone ahead and replaced the figure with the correctly indicated DAOD retrieval. We appreciate the reviewer catching this.

L661: “un-bias-corrected” may be use non-bias-corrected

Modified to read “non-bias-corrected”, which we agree is grammatically correct.

L736 “PBL fluxes” use PBL mole fractions or concentrations

We changed this to mole fractions for correct description.