Authors' Response to Referee Comments

We appreciate the overall positive response of the Referee and we would like to thank for his constructive comments and helpful suggestions on the manuscript, which helped us to further improve the clarity of the paper. Below, we give detailed responses (in blue) where appropriate.

Alan Fried (Referee)

This paper is acceptable for publication with only 4 very minor points that this reviewer would like to see addressed.

1. It would be very informative to the reader if the authors could indicate how reproducible the Allan-Werle results of Fig. 4a were obtained. Does this figure represent typical performance or does this represent the best series of measurements? For that purpose, it would be very interesting to plot a histogram of the 1 second Allan-Werle deviations if the authors indeed recorded multiple plots.

We agree that is tempting to pick the most attractive time-series sequence to show the instrument performance. However, in our case, it is very difficult (as already mentioned in the manuscript) to maintain a constant concentration level, at the high precision level of the instrument, over prolonged period of time, because of the open-path configuration. Thus, the only well-controlled experiment took about 3 hours, from which 1.5 hours were spent for purging, and the other 1.5 hours are shown in Fig.4. For the 1 second Allan-Werle deviations, the long-term stability of the environment is not critical. Therefore, we analyzed our repeated overnight measurements, where the instruments simply measured laboratory air. We collected more than 30 hours of data and calculated the 1 s Allan-deviation of every 200 s period. The histogram plot of the corresponding 520 values is now is added to Fig.4a as an inset. It shows that the 1 s noise level is highly reproducible and adds confidence that the 1.5 hour Allan plot shown in Fig.4 is representative for the performance of the instrument.

2. Regarding the temperature sensitivity of their instrument, is there any possibility to further stabilize the temperature of the electronics and/or the entire optical system either actively or passively employing better insulation? Although the 4 ppb/K sensitivity (not 4 ppb/K-1) is quite good, 10 degree C temperature changes, as would be experienced by changing altitudes, seem to affect performance for time periods~20 - 30minutes (Fig. 5). It would be nice to mitigate this long equilibration time period.

Indeed, it is possible to develop additional solutions to achieve further thermal stabilization. In principle, there are two ways: active and passive. While the former can be very compact, it requires significant electrical power (e.g. TEC-based). A passive isolation can efficiently dampen sudden temperature fluctuations, but has limited flexibility for the range of supported temperatures. Thus, during cold season a good isolation can be beneficial, while on hot summer days can lead to thermal rollover. In our approach, we considered the most plausible applications (e.g. source identification and emission estimates), which require flight patterns that mainly involve close-to-surface surveillance or curtain-like profile flights. For these scenarios, the temperature fluctuations are less prominent. Nevertheless, for high-altitude flights, such as PBL determination, it is definitely advisable to adopt a more elaborated thermal stabilization scheme. Lately, we were successfully applied a combination of TEC and phase-change material (as thermal buffer medium) solution, but with considerable cost of weight and electrical power (see Graf *et al.*,

Compact and Lightweight Mid-IR Laser Spectrometer for Balloon-borne Water Vapor Measurements in the UTLS, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-243). This instrument demonstrates temperature stabilization in a range of nearly 80°C.

3. It would be useful to indicate the H2O sensitivity of their retrieved CH4 results since situations where the H2O mixing ratios can approach up to 3-4 times the 1% levels simulated.

We admit that having the H_2O sensitivity on the retrieved CH_4 concentrations would complete the overall characterization of our device, however such investigation is not trivial (see also our reply to comment #1). Furthermore, we do not expect that during the flight (~20 min) the water vapor would drastically change in the atmosphere. Since we consider calibrating our QCLAS instrument prior flights with a ground-based CRDS instrument, either in-situ or taking bag samples that are analyzed in the laboratory afterwards, potential biases due to water vapor are minimal.

4. The authors may wish to explain the slight UAV overestimate of CH4 relative to the CRDS in Fig. 8 at just after 08:00 at 12 m sampling height.

There are in fact significant discrepancies between the two data set at 8 AM and in the following hours. We are, however, confident that these variations are real inhomogeneities of atmospheric methane concentration. As we mentioned in our manuscript, in the close vicinity (~200 m) of the radio tower is a farmsteads (ruminants), which has significant methane emission (see Fig.8 top left, the farm is located at the right). The drone flights were conducted on the small-unpaved road that connects the farm with the tower. Because of communication difficulties with the drone in the proximity of the metal frame of the radio-tower, the flights took place about 20 m from the tower, down on the roadside in the direction of the farm. Immediately after sunrise, we observed slight winds that unfortunately had east-west direction, i.e. we were measuring downwind to the farm. Therefore, the early morning data (before sunrise) is much more representative for the nocturnal boundary layer.