Response to 'Comment on amt-2021-28' by Frans Harren

We thank Frans Harren for his constructive review of our AMICA manuscript. He identified some obvious mistakes and shortcomings and made important suggestions how to improve the paper. Below, we copy Frans Harrens comments in black and give our response to each of them in blue font. Significant changes to the text in the revised manuscript are shown here in purple font.

This is an interesting manuscript describing the technical set-up and calibration experiments of the AMICA instrument with demonstrating results for various aircraft campaigns. The technical part is described well, while the performance of the instrument and the molecular spectral interpretation could be better explained. As such I recommend substantial changes to the manuscript before accepting it for publication.

- In the abstract precisions are given for the various molecules, but within the text these precision data are not convincingly confirmed.

The discussion on accuracy and precision has been expanded both in the abstract and in the text, also in response to referee #2. The precisions given for OCS and CO in the abstract are now also given in the text together with a description on how they are derived from the Allen Plots in Figure 9:

> "For the OCS and CO observations made so far, respective precision estimates of 30 ppt and 3 ppb are made based on the higher two second value from the two experiments shown in Figure 9. For CO_2, both curves exceed 1 ppm for all averaging times, which is another reason (besides the issues described above) for currently not releasing AMICA CO_2 data for scientific use."

- Line 77 the description of the ICOS set-up refers to Fig.2. This an informative figure, but fails to give clearly optical beams paths from laser to detector.

The paths of the optical beams are included in a revised Figure 2.

- Line 258, the trace gas inlet is rear facing, considering the pressure problems at high altitudes (line 560), it could be better discussed why the inlet is rear facing. Taking into account the position is of the inlet on the aircraft cabin, distance of inlet to the cabin, boundary layer around the plane at high altitudes. Why not using a Pitot-like tube, taking advantage of the dynamic pressure?

Rear facing inlets are common for gas phase measurements, because they provide an easy way to discriminate against the inflow of particles. While particle discrimination has also been realized with forward facing inlets that allow for passive sampling by making use of the ram pressure (e.g. Gao et al., AMT, 2012), the implementation of such inlets is more complex, especially when they are shared by multiple instruments.

We add the following information to Section 2.4 in the revised manuscript:

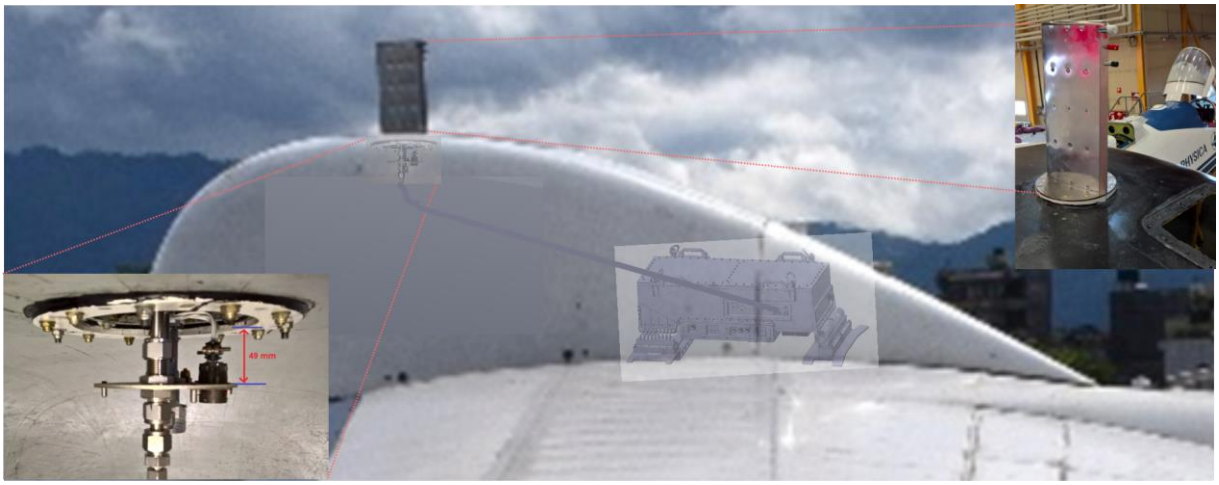
> "Both inlet systems are rear facing to avoid the intake of liquid water, ice and large aerosol particles (McQuaid et al., 2013). They are briefly described and characterized here."

- A proper Figure (picture) of the inlet and transfer line will be helpful.

A link is added to the HALO webpage, where the TGI is described in much detail:

> https://www.halo.dlr.de/instrumentation/inlets/inlets.html#TGI

The following figure of the AMICA inlet on M55 Geophysica is added to the supplementary material:



Line 130, next to the temperature control of the instruments, it is helpful to describe the heat transport resistance value of the foam, used on the ins of the enclosure walls, to keep the temperature of the AMICA within limits.

Relevant specs of the foam are added in Section 2.3 of the revised manuscript:

> "density: 2.2 kg m^{-3}, thermal conductivity: 0.06 and 0.05 W m^{-1} K^{-1} at 24 and -5 °C respectively"

- Line 460 and below: I to have objections with the word "absorption band" when an absorption line is meant. Maybe except for ozone all measured absorption features are single molecular lines.

This is, of course, correct, and the terminology is corrected throughout the revised manuscript.

- Line 460: The ICOS description for measuring OCS, CO_2, CO and H_20, is not convincingly describing the performance of the instrument. The mirror reflectivity is given in Table 3, what is the effective optical path length related to this? Taken the line strength and broadening effects into account and the noise of the detector, what are the expected detection sensitivities for these gases. How are these related to the real detection sensitivities (e.g. Fig. 7, 8) under lab conditions and flying conditions. What is the Noise Equivalent Absorptions Sensitivity of the instrument (see: Paul et al.)?

The effective optical path length $L_0 = L_{cav} / (1 - R)$ is added to Table 3 for each channel.

Laser, detector and mirrors for the 2050 cm^{-1} channel are the same as in the enhanced performance model of the commercial OCS analyzer by Los Gatos that has nominal 1 Hz precisions of 16 ppt OCS, 0.7 ppb CO and 0.4 ppm CO_2 and a NEAS of about 1 x 10^{-8} cm^{-1} $Hz^{-1/2}$.

Currently, AMICA sensitivity is significantly reduced (by roughly a factor of 10) due to the electrical noise caused by the ground loop at the preamplifier as explained in the paper. Once that issue is resolved, we expect sensitivity in the same range as the commercial analyzer (this was verified by measuring the detector signal with an external oscilloscope).

Note that the electrical noise is the same under lab and flying conditions. Because of the effective EMI isolation of the instrument electronics from the surroundings, we also expect the detector noise to be the same. The one thing that does get noisier under flying conditions particularly at higher altitudes is the cavity pressure (cf. Figures 4 and 13), but the effect on overall sensitivity should be small.

- The same clarifications should be made for the other two wavelength regions described in 4.2 and 4.3.

For the 1034 and 3331 cm^{-1} channels, a proper determination of sensitivity and precision has not yet been carried out because other issues will have to be resolved first as described in Sections 4.2 and 4.3.

For the 1034 cm^{-1} channel, however, a first estimate of precision for O_3 can be made based on the experiments shown in Figure 11. This is now stated in the text:

> "Based on the measurements shown in Figure 11, 0.5 Hz precision for O_3 is currently in the range of 20 to 40 ppb."

For NH_3, we reference once more the paper by Leen et al. (2013):

> "Ultimately, it should be possible to measure NH_3 with similar sensitivity as described by Leen et al. (2013)."

For the 3331 cm^{-1} channel, the low mirror reflectivity and effective path length do not allow measurements at atmospheric concentrations, so there is little point in giving precision data. Also following the suggestion of reviewer # 2, we stress in the revised manuscript that this is currently more a conceptual channel.

- Line 475: A discussion is made about the compromise of Leff, for strong CO_2 absorption and weak OCS absorption, and that the broadened CO_2 peak reduces sensitivity. It is unclear what the outcome of this discussion was and why a broadened CO_2 peak should reduce sensitivity?

This is already illustrated and described in some detail in the supplementary material (Figure S5).The sensitivity reduction a direct result of L_{eff} becoming a quasi-inverse function of A at high A and, so that the scaling of A with c in Eq. (2) becomes significantly less than linear. The additional broadening results from the sensitivity reduction being stronger near the centre of the absorption peak and less at the flanks. Unlike with Doppler and pressure broadening, this "cavity broadening" behaves in a way that peak area does not scale linearly with concentration anymore.

To make this clearer in the text, we change the end of the paragraph in Section 4.1 to:

"The consequence is a smaller change in absorption for a given change in concentration and thus a reduced sensitivity. Because L_{eff} varies with absorption over the broadened peak (L_{eff} is smaller at the peak centre where A is larger), the effect also introduces additional broadening that complicates the analysis. More details and an illustration on the sensitivity reduction for CO_2 are given in Figure S5 in the supplementary material."

- Line 480, a fit is made; is this a Voigt fit?

Indeed, a Voigt fit is made. This information was "hidden" in line 454, where it was stated that Voigt profiles are calculated from the HITRAN parameters. In the revised manuscript, we also add the word "Voigt" before fit in line 480.

- Line 536, a low mirror reflectivity is chosen for the setup, although HCN, C_2H_2 and N_20 concentrations are too low to be observed. What argument is made to install this low mirror reflectivity?

This highly experimental setup was implemented in a very short time frame for the StratoClim field campaigns. Because time was lacking to acquire and test an optimized HR mirror for this channel, a 3 µm mirror that had been sitting on the shelf was used, which unfortunately turned out to have a far too low reflectivity (clearly lower than we thought it would be).

When we chose to install this setup for the campaign, it was clear that we would not make measurements with this channel as we would make them with the operational 2050 cm^{-1} channel. However, we had hoped for a somewhat higher reflectivity and thus path length that would allow us to see a signal for elevated monsoon HCN mixing ratios when averaging spectra over longer time periods. As described in the paper, we ended up only observing laser ramps with absorption due to high water vapour at the ground, and can thus only demonstrate some sort of spectroscopy being done but anything close to a measurement of our target gases HCN, C_2H_2 and N_2O.

Note that it is likely that with sufficiently reflective mirrors, the laser power of 8 mW will be too low to couple enough light into the cavity and onto the detector. Lasers with up to 25 mW at 3 µm have now become available (see for example https://nanoplus.com/en/icl/) and we still hope to eventually acquire a set of mirrors with sufficient reflectivity (up to at least 0.9995 is available) and a new ICL with sufficient output power to turn the setup into an operational channel.

- Figure 10 lower panel, the fit deviates quite from the experimental observation, giving a systematic offset in the mixing ratio fits (Fig 11). Observing the spectrum, it seems that there is broadband background change in absorption over the wavelength range. Such a change is not taken into account in the fit? This would even make the fitted ozone concentrations lower than the standard.

The following discussion on the obviously poor fit to the ozone spectrum is added to the first paragraph of Section 4.2:

> "The spectral fit does not closely reproduce the observed spectrum. First, significant absorption up to 0.01 between peaks points to either a bias in the used baseline or trace gas absorption by lines or bands that are not included in the HITRAN data base. Second, the observed O_3 absorption peaks are broader than the fitted peaks. Possible explanations include inaccurate HITRAN parameters for the O_3 lines or cavity response broadening. Both, baseline offset and the broader peaks, will be further investigated in future laboratory experiments."

- Reading the manuscript from paper, it is very difficult to read the figures properly. The letter height of figures 5, 6, 7, 8, 10, 11 are too small (<1 mm, I had to use a magnifying glass). Besides the color of the figures could be also taking care of (yellow is very difficult to read).

Figure 5 is simplified (also following the suggestion of referee # 2) and the font of the remaining text is increased. The font size of axis titles/annotations and other labels in Figs. 6, 8, 10, 11 and 12 is increased, and the yellow colour in Figs. 6, 10 and 12 is replaced by a darker orange.

- For Fig 6, 10 and 12 the upper and lower panels have reversed wavenumber scales as compared to each other, which makes them difficult to compare.

This choice was made intentionally. In the top panel, the laser ramp is plotted in forward direction along the time axis; the wavenumber scale shown on the top axis is calculated, and the numbers are given as an indication and to illustrate that the wavenumber actually decreases along the laser scan. For the bottom panel, we chose to show the infrared absorption spectrum in the standard way against increasing wavenumber, i.e. with an increasing x axis scale. Also note that only a certain region (blue shading) from the top panel is shown here (absorption cannot be calculated for the ramp parts where the laser is off). We deemed this more logical and less confusing, and we do not see the added value of being able to visually draw direct lines from the ramp to the absorption spectrum. In addition, when the laser current ramp is linear as in Fig. 12, the relationship between ramp time and wavenumber is also not linear and one panel would have to be drawn with a distorted x-axis in order to provide for the direct comparison.

- For Fig. 6 lower panel, the difference between fit and data is very difficult to observe. I suggest also to show its difference in a separate panel (its residue)

In the revised manuscript, panels showing the residues are added to Figures 6 and 10.

Additional references cited in this response:

Gao, R. S., Ballard, J., Watts, L. A., Thornberry, T. D., Ciciora, S. J., McLaughlin, R. J., and Fahey, D. W.: A compact, fast UV photometer for measurement of ozone from research aircraft, Atmos. Meas. Tech., 5, 2201–2210, https://doi.org/10.5194/amt-5-2201-2012, 2012.