

Reply to RC1

Manuscript information:

- Title: Testing the altitude attribution and vertical resolution of AirCore measurements with a new spiking method
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We would like to thank Pieter Tans for the constructive comments. In the following document, the reviewers' comments are marked in *italic font* and indented, our answers are in regular font. Changes in the manuscript are marked-up in red and listed as framed screenshots below the respective comment. For clearness: The line numbers in the RC refer to amt-2020-461-manuscript-version2.pdf instead of the preprint version. The two version only differ by minor formatting aspects. The line numbers in our listed changes refer to the marked-up version of the revised manuscript, that is provided separately.

Point-by-Point reply

1. *Line 81: The internal diameter is what matters, so please provide that. I assume that the predicted results use the ID.*

Agreed. The internal diameters are used to model the vertical resolution.

85	Three thin-walled stainless steel tubes with different outer -diameters (<u>internal diameters: 1.76 mm, 3.6 mm, 7.6 mm; Ø-D</u> <u>outer diameters: 2 mm, 4 mm, 8 mm</u>), coated with silconert2000® and soldered together result in the 100 m long and coiled GUF AirCore. This design allows to rapidly collect air in the large-diameter tubing, which is then gradually pushed into the
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2. *line 137: The fill gas at the closed end of the tube will not be distributed as a Gaussian. It has to be asymmetric because the end is closed off. What it looks like depends on how much fill gas is left, but close to the end the spatial derivative of each gas fraction has to go to zero. If there is a lot of fill gas left, occupying several diffusion length scales, the fill gas fraction must approach 1 at the closed end. When the fill gas enters the analyzer the transition ought to be rapid, unless the tubing toward the analyzer and the analyzer cell itself cause a lot of mixing. Some years ago I analyzed experiments with "plug" transitions, sudden mole fraction shifts inserted very close to the Picarro. In the hypothetical case that the cell would be perfectly mixed all the time, the insertion of a plug should produce a negative exponential approach toward the new steady state. If there is plug flow within the cell, so that the rapid transition is mostly preserved, the approach to the new state should be linear. It turns out that the actual transition was in between these two cases. The "response function" of each analyzer will depend on pressure, cell volume, and shape of the cell. This subject comes back on line 156.*

Thank you for your thoughts on this. We agree, that the fill gas will not be distributed as a Gaussian, which however had been assumed in the past by Engel et al. 2017. That is why we decided to introduce the new approach to identify an accurate starting point by reconstructing the gas fraction time series. In order to make this clearer, we inserted an additional citation in the manuscript:

in the AirCore (high CO), which is fourthly followed by the stratospheric sample (STRAT, low CO). The resulting idealized CO mixing ratio time series including gradual transitions between gas fractions is shown in Figure 2a. In the past, a Gaussian distribution was fitted to the FG peak in the CO measurements (Engel et al., 2017). The half maxima of the fit were then considered the start of the AirCore, respectively the start of the STRAT, as described in Engel et al. (2017) Sect. 2.4.1 and 2.4.3. This FG peak however is partly mixed with PG, Cal gas and STRAT. In the new approach, we reconstruct the gas

In the cases that we tested with our measurement setup, the fitting algorithm with CDFs of a Gumbel distributions was able to reproduce the asymmetric shape of the fill gas peak and yielded satisfying results for each gas fraction transition – starting from very close to 0 (but not exact 0) and approaching 1 with increasing time (see Fig. 2c). We addressed this issue in the revised version of the manuscript:

where μ is the mode of the respective Gumbel distribution (i.e. the inflection point of the CDF) and β is a measure for the standard deviation. Strictly spoken, the CDF of a Gumbel distribution does not start from exactly 0 – which in reality should be the case for e.g. the FG fraction due to being closed off. Albeit, we found that it reliably generates satisfying results. For simplicity of the fitting process we decided to use the CDF of the Gumbel distribution instead of the CDF of the lognormal distribution. By altering the parameters μ_i and β_i simultaneously for each gas taking into account Eq. (85), a CO mixing ratio

The maximum of the fill gas fraction is indeed reduced below a value of 1 occasionally with our measurement setup. The idealized time series shown in Figure 2 was actually generated using data from an GUF AirCore measurement in Sodankylä in June 2018. Figure 2 reveals, that the measured mole fraction at the maximum of the fill gas peak is the result of mixing with both, Cal gas (due to Taylor dispersion in the transfer line and mixing in the analyzer cell) and stratospheric sample (due to Taylor dispersion, diffusion and mixing in the cell).

3. line 193: *I suppose the signal gas mixture is CO-in-natural air. If a larger spike is inserted, you don't want to alter the main gases of interest.*

Thank you for your input. The amount of CO₂ and CH₄ in the signal gas is not relevant for the evaluation of the altitude retrieval, which is the aim of this paper. We do not recommend using contaminated sections of the AirCore profile for atmospheric interpretation at all. That is why we decided not to mention the CO₂ and CH₄ content of the signal gas. If we were to permanently include the CO-spiking system in an AirCore setup, we could however reduce the number of CO-spikes e.g. to just one (I would then suggest at the top of the profile) in order to minimize the effect of contamination due to mixing with signal gas. We included a short statement and mentioned the new idea at the end of sect. 3.3 in the revised version of the manuscript:

other AirCore geometries. In particular, this relationship could also change for the same AirCore when a different drier is used with significantly different flow restriction. We do not recommend using contaminated sections of the AirCore profile for atmospheric interpretation. Nevertheless, the CO-spiking system could also permanently be included in a once fully characterized AirCore setup and used to insert only one spike at the top of the profile, in order to obtain highly accurate trace gas profiles (including other trace gases than CO measured with the Picarro analyzer) with minimal signal gas contamination.

4. line 234: *typo - should be June 17*

Yes, changed that. Thank you.

5. *line 245: "resulting calculated vertical profile" (this is for clarity)*

Done.

6. *line 350: I would be very surprised if junctions between sections could cause much additional mixing. The flow conditions in the tube are extremely far away from any kind of turbulence. My suggestion is to look further into the analyzer contribution to mixing.*

Thanks for pointing that out. We took this into account in the revised version of the manuscript:

profiles, however agrees well within less than 220 m throughout the profile. This small discrepancy is probably caused by the simplified assumptions guiding the model calculations, that neglect diffusion and Taylor dispersion during the AirCore sampling process. In addition, the effect of mixing in the analyzer cell may be underestimated in the model and the junctions between different diameter parts of the AirCore might induce a small amount of additional mixing. Regarding the uncorrected
385 vertical profiles for both measurement flights, this discrepancy becomes larger at high altitudes, where the peak shape is

7. *line 387: I suggest replacing "proof," with "prove"*

Done.