Reply to RC2

We would like to thank William Sturges 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. 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. *My only comment would be that, for the sake of anyone **not** working directly on AirCores, this would benefit from having a little more explanatory text. E.g. under Section 2.1, a clearer summary of these steps that does not require reference to Engel et al. would make reading/understanding much easier.*

Thank you for your constructive comment. We added more explanatory text to Section 2.1 in the revised version of the manuscript:

ordered and refined compared to the four-stage process described in Engel et al. (2017). The overall concept of the retrieval remains the same. (i) The sampling of air during the balloon descent is calculated based on the ideal gas law. ~~(see Engel et al.~~

> ~~(2017) Sect. 2.4.2 for details).~~ (ii) The start and end times of the AirCore measurement in the analyzer time series are
> 120 determined. (iii) The sampling and the analysis can be matched based on the molar amount ~~following Engel et al. (2017)~~. Steps (i) and (iii) are still performed according to Engel et al. (2017) and are shortly described in the following.
>
> (i): Under the assumption of an instantaneous pressure equilibrium the molar amount n of an ideal gas within a constant AirCore volume V at the sampling time t can be described by the ideal gas law:
>
> $$n(t) = \frac{p(t) \cdot V}{R \cdot T(t)}, \tag{1}$$
>
> 125 where p and T are the atmospheric pressure, resp. the mean AirCore temperature at t and R is the general gas constant. The relative amount of gas n_{rel} is then described by:
>
> $$n_{rel}(t) = \frac{n(t)}{n(t_{close})} = \frac{p(t) \cdot T(t_{close})}{p(t_{close}) \cdot T(t)}, \tag{2}$$
>
> with the total molar amount of gas $n(t_{close})$ at the closing time t_{close} of the AirCore. The data evaluation software takes into account special cases, where air can be lost during sampling.
>
> 130 (iii): Since the analyzer is operated at a constant mass flow, the relative amount of measured gas $m_{rel}(t')$ at the elapsed measurement time t' can be described as
>
> $$m_{rel}(t') = \frac{t'}{t'_{end}}, \tag{3}$$
>
> with the total AirCore measurement time t'_{end}. By interpolating the meteorological and positional data collected during sampling from $n_{rel}(t)$ to $m_{rel}(t')$, it is attributed to the trace gas measurements $\chi(t')$.
>
> 135 In order to accurately derive $m_{rel}(t')$, the start and end points of the AirCore analysis in the continuous Picarro measurement time series need to be known (step (ii)). In Sect. 2.2 we present a new approach to determine the start point of the AirCore in the measured trace gas time series. This new approach has the advantage of providing an objective start point without the need for subjective judging.

We also updated the subsequent numeration of equations (not shown here).

2. L.15 add "positive" to "bias"

Done.

3. L.16 "shown" not "uncovered"

Done.

4. L.17 "to be represented by possible empirical"

Thanks for your suggestion. We decided to put the statement in different words to make it more accurate in the revised version of the manuscript:

> 15 within 250 m below 20 km. Above 20 km the positive bias becomes larger and reaches up to 1.2 km at 27 km altitude. Differences in descent velocities are ~~uncovered~~ shown to have a major impact on the altitude attribution bias. ~~We identified the time lag between the theoretically attributed altitude and the actual CO-spike release altitude to be a possible empirical correction parameter for our AirCore altitude retrieval across different flights.~~ We parameterize the time lag between the theoretically attributed altitude and the actual CO-spike release altitude for both flights together and use it to empirically correct
> 20 our AirCore altitude retrieval. Regarding the corrected profiles, the altitude attribution is accurate within ± 120 m throughout

5. L.19 is it +/- 120 m or +120 offset?

It's ± 120 m. Added this.

6. L.50 "needs to be attributed to positional data" – doesn't it just need altitudinal data? Lat/Long you'd get from GPS, wouldn't you?

Thanks for your suggestion. Indeed, the statement in the original manuscript was inaccurate. We decided to stick to the term "positional", since this statement is not restricted to passive AirCore sampling but also holds true for active AirCores. In all cases it's the molar amount of gas during the sampling process that is matched to the analysis time series. GPS altitude is not needed for this process, albeit it is one of the desired variables. It is not treated differently from Lat/Long data in this matching process. We clarified this in the updated version of the manuscript:

> 50 in 2015 and to small weather balloons flown in 2016.
> The wide range of platforms and fields of application concerning AirCore sampling (regardless of being active or passive) all have one in common: a continuous sample of atmospheric air is collected together with meteorological and positional data, that need to be attributed to the trace gas measurements of the sample after analysis. ~~a continuous sample of atmospheric air is collected that needs to be attributed to positional data.~~ Regarding vertical profiles from passive AirCores, an altitude attribution

7. L.90 what is the push gas made of?

We provided relevant information in the updated version of the manuscript:

> 90 comprises up to 8 temperature sensors, a pressure sensor, a GPS-antenna, an SD-card holder for data logging and controls the closing valve.
> Before launch, the AirCore is flushed with fill gas (FG, air standard with known trace gas mole fractions) and sealed at one end. During ascent it empties due to the decreasing ambient pressure with height. A small amount of FG remains in the AirCore.

And three lines below:

> push gas (PG) and analyzed with a Picarro G2401 CRDS continuous gas analyzer for H_2O, CO, CH_4 and CO_2 mole fractions at a constant rate. FG and PG are identical in the GUF setup. Figure 1 shows the analytical set-up for the measurement process.

8. Fig. 1 is quite tough to follow unless you have a little more background

Thanks for pointing that out. We added explanatory text in the updated version of the manuscript:

> push gas (PG) and analyzed with a Picarro G2401 CRDS continuous gas analyzer for H_2O, CO, CH_4 and CO_2 mole fractions
> at a constant rate. FG and PG are identical in the GUF setup. Figure 1 shows the analytical set-up for the measurement process.
> In the bypass/flushing position of the two position valve (Fig. 1a), PG is measured and the open transfer lines to the AirCore
> are flushed. After connecting the AirCore to the transfer lines, the two position valve is switched to measurement position
> 100 (Fig. 1b), so that the sample is pushed through the analyzer. Since June 17, 2019, our Picarro analyzer operated in the inlet

9. L.102 what does "PG resp. a calibration standard" mean? I didn't understand this.

We added labels to the transfer lines in Fig. 1a and referred to them in the caption in the updated version of the manuscript in order to make it more comprehensible:
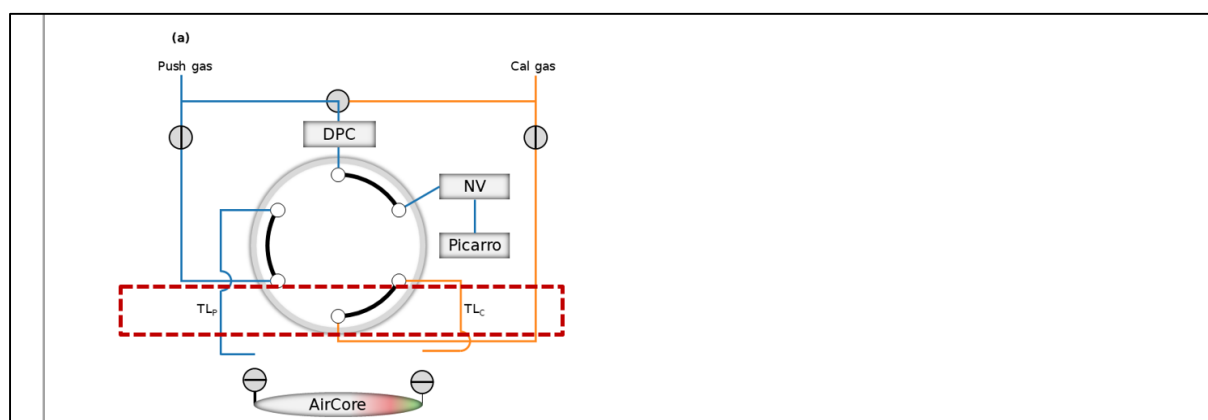
> **Figure 1:** Analytical set-up for AirCore measurements. Pressure is controlled by the digital pressure controller (DPC). Compared to the
> previously published set-up by Engel et al. (2017) the mass flow controller has been replaced by a needle valve (NV). The Picarro operates
> in inlet control mode. In the bypass/flushing position **(a)** push gas (PG) is measured bypassing the AirCore while the transfer lines (TL) are
> being flushed: TL_P with PG and TL_C resp. with a calibration standard (Cal gas). Tubing that contains PG is indicated in blue, Cal gas in
> 110 orange. In the AirCore measurement position **(b)** the PG is passed through the AirCore and pushes the air to the Picarro. Directly after

10. L104 I am not clear about "only tubing involved at the start of the AirCore measurement is coloured"; what is meant by "involved" – it's all involved isn't it?

Thanks for pointing that out. We rephrased it in the updated version:

> orange. In the AirCore measurement position **(b)** the PG is passed through the AirCore and pushes the air to the Picarro. Directly after
> switching to **(b)** a small amount of Cal gas is measured that has been enclosed by the transfer line TL_C. For clearness, in **(b)** only tubing
> involved containing gas that is measured at the start beginning of the AirCore measurement analysis is coloured (adapted from Engel et al.,
> 2017).

11. L.117 not clear what "starting point in the analysis" refers to.

Thanks for pointing that out. We rephrased it in the updated version, in order to make it clearer:

> 140 Membrive et al. (2017) stated that for their slowly descending high resolution AirCore the dominating uncertainty source in
> the stratosphere is related to the selection of selecting the start point of the AirCore analysis starting point in the continuous
> Picarro measurement analysis datatime series. They link this to the low amount of stratospheric sample compared to the
> tropospheric sample. For AirCores with less stratospheric sample the effect can be considered to be larger. Until now, the
> choice of the starting point relied on subjective judging (Engel et al., 2017; Membrive et al., 2017). In order to systematically
> 145 evaluate the altitude attribution procedure with the CO-spiking experiment presented in this paper, as many as possible
> subjective parameters need to be eliminated. We therefore decided to refine the process of selecting the start time of the
> AirCore and introduce a new approach to identify an accurate starting point without the need for subjective judging.

12. L.129 how high is high CO?

Good point. We added information about standard gas CO mixing ratios from recent campaigns for clarity and improved our description of the measurements in the updated version of the manuscript:

> For a regular GUF set-up AirCore flight analysis we first measure PG (high CO, 1.4 ppm in the recent GUF campaigns). We then switch the two position valve (see Figure 1b) like described in Engel et al. (2017) so that secondly the measurements
> 155 gradually approach the low CO mixing ratio of the calibration gas (Cal gas, 0.2 ppm in the recent GUF campaigns) that was left in the transfer line TL_C between AirCore and analyzer. the calibration gas (Cal gas) within the transfer line TL_C between

13. L.131 maybe explain how "Cal gas is used to distinguish between PG and FG"?

We extended the statement in the updated version of the manuscript:

> AirCore and analyzer is measured (low CO). Since in our setup one standard gas is used as both PG and FG, Thethe Cal gas
> is usedserves to distinguish between PG and FG in the measurement time series. Thirdly, it is followed by the remaining FG
> in the AirCore (high CO), which is fourthly followed by the stratospheric sample (STRAT, low CO). The resulting idealized
> 160 CO mixing ratio time series including gradual transitions between gas fractions is shown in Figure 2a. In the past, a Gaussian

14. L.189 what does "fastening valve" mean? I've not heard of this before (shutoff valve?).

Instead of "fastening type" we now call it "mounting hardware":

> **Figure 3:** Fastening typeMounting hardware for the SMLD 300G micro valve. (1) micro valve, (2) valve coil, (3) O-ring (material: viton), (4) inlet adapter, (5) valve holder. Fritz Gyger AG 2020.
> 215 The CO-spiking set-up consists of a signal gas reservoir, a micro valve and a connector which directly connects the micro valve to the open end of the AirCore in front of the sample drier. The adaptor is designed to have a negligible flow resistance for sampled air, while inducing only a minimal dead volume to the sampling system. We used the micro valve SMLD 300G by Gyger, which is light-weight and suited to dose signal gas volumes of around ¼ cm^3_n on the time scale of 20–100 milliseconds, thereby influencing the sampling process during descent as little as possible. Figure 3 illustrates the fastening
> 220 typemounting hardware for the micro valve.

15. Fig. 8 It took a while for me to realise that the steps in the curve related to the three diameters of tubing - maybe point this out from the start?

Thank you for your feedback. This has also been pointed out by Anna Karion. We added one sentence for clarification:

> **Figure 8:** Modelled (blue line), uncorrected (flight 1: dark grey circles, flight 2: light grey circles) and corrected (only flight 1: red triangles)
> 370 vertical resolution of GUF003.
>
> Taking into account the three different inner diameters and lengths of GUF AirCore tubing, the modelled vertical resolution exhibits two steps corresponding to the junctions between two adjacent parts of tubing. Figure 8 shows the modelled vertical resolution profile as a function of altitude, the Gaussian standard deviation of the respective peak and the Gaussian standard deviation derived from the Δt-corrected profile from June 17. Regarding the second flight on June 18, only data from the