

### Overall Summary:

This paper is within the scope of AMT and should be published. There are some confusing sections, discussed further below, but overall the paper does a nice job of explaining a new technique that will improve our understanding of stratospheric circulation and add important measurements for long-term climate relevant gases. They prove that this technique works well and appears easy to add on to already existing infrastructure, and is cost-effective. There is nice agreement with aircraft measurements and any discrepancies are reasonably explained. I look forward to seeing more StratoCore-GC-ECD data in the future. Given that only one section of this paper needs restructuring, it should be accepted with minor revisions.

### General comments:

The main section of the paper that needs improvement is section 2.3 and relevant figures. Section 2.3 was the most confusing of the entire paper and also critical to the verification of this new system. It was hard to follow the steps of the different experiments. In part this was a challenge because of the different language used (test, experiment, etc) along with the two tests (but maybe more?) to study three scientific objectives. This section is important and a restructure would be helpful for the reader to follow the logic. For example, line 160 mentions 'both experiments' and then line 192 mentions 'another set of tests'. Was that related to the first two tests mentioned at the start of the section? If so, set doesn't make sense since was only two tests. Overall, this section is content heavy and the reader needs a clearer path to follow the important discussion.

Related to Section 2.3, it is really hard to understand what figures 4 and 5 are trying to show. How does figure 4 show us that there is no contamination? What does the CO<sub>2</sub> and N<sub>2</sub>O tell us in 5a? This figure and/or the relevant text to it needs to be reconsidered so the reader doesn't need to spend the majority of their time trying to understand it.

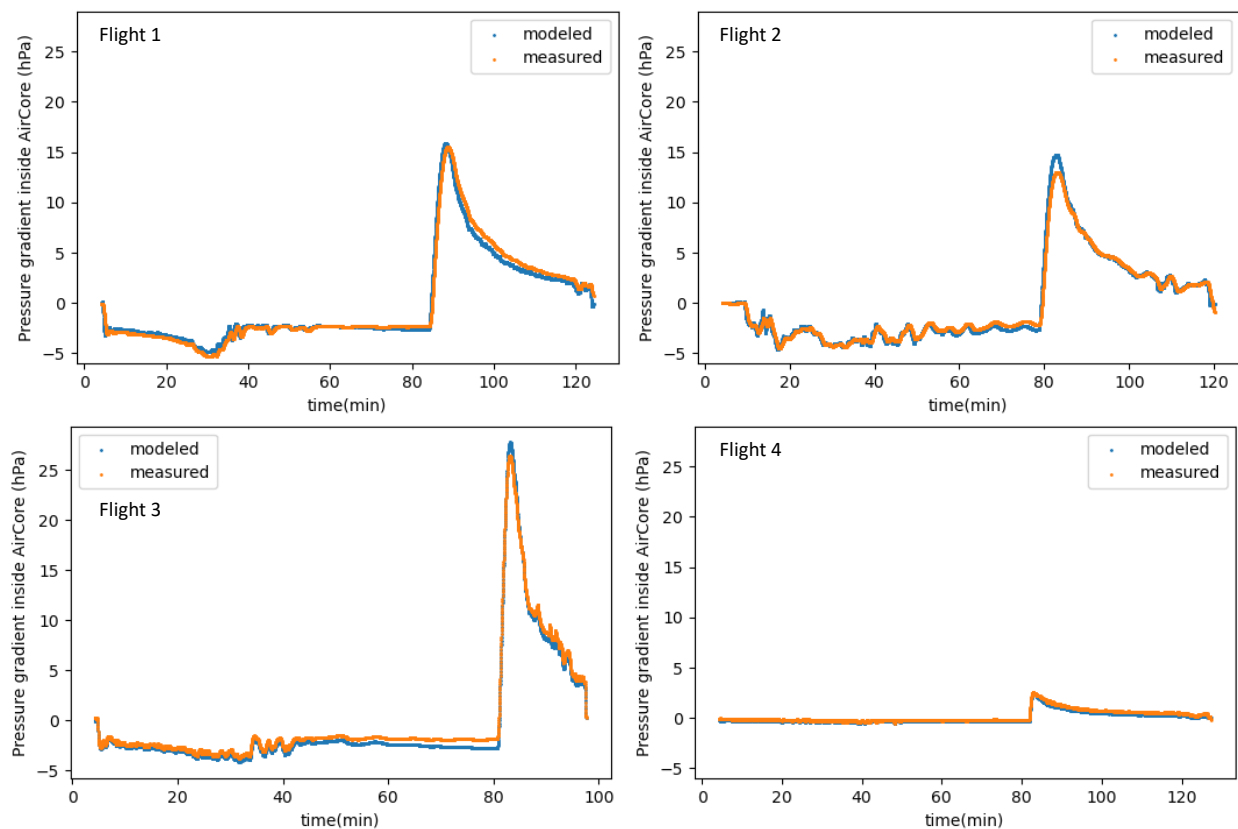
To enhance the clarity of Section 2.3 and to better convey the results of each test and their implications, we have re-organized this section. We have also highlighted the significance of each figure in the main text to facilitate easier comprehension of the content. First, we flushed the AirCore zero-grade air, then stored overnight (~14 hours), and subsequently analyzed using the StratoCore-GC-ECD system, during which a standard gas of typical tropospheric composition was used as the push gas following the stored sample. Figure 4A shows the results: the first 164 mL of injection, corresponding to the gas inside the AirCore, we can see that the dry mole fractions of all target molecules were below the StratoCore-GC-ECD detection limit for these species in the entire, suggesting during the 14 hours storage did not have any contamination. Figure 4B was similar, the only difference is that we flushed the AirCore with standard gas, to investigate if the AirCores can act as sink of some molecules. Our results again showing that the inside of AirCore will not absorb our target molecules.

Section 3 and Figure 6: Is there a reason why the parachute is red? What is the 'lightweight material'? Is the fact the balloon is off-white important?

Thank you for the comments. We revised Figure 6, deleted irrelevant information on the figure to make it more concise and easier to follow.

Line 268: “data not shown in figure” – Could the comparison between model and observed be done in a supplement if no space in the paper? It is referenced in the conclusion that there is good agreement between model and observations but there is nothing that directly shows that for the reader. The RMSE is given but more information would be nice.

Thank you for the comment. We agree that the comparison is useful for readers to better understand the performance of the model. Since this model was discussed in depth in Tans (2022), we originally did not include this part into our manuscript. Here we are attaching the model-measurement comparison in this document.



Lines 275-289: Vertical references are given as hPa but the plots are shown as km which is challenging to compare the figure to the text. Perhaps add a second y axis to Figure 9 with hPa?

Thank you for the comment. We agree that using hPa in the text but km in the figures can be confusing. Therefore, in the main text we replaced “hPa” to “km” to avoid such ambiguity.

Technical Corrections:

Line 20: We then launched

We have corrected this grammar error.

Line 44: chlorofluorocarbonsmolecules

We corrected this typo.

Line 114: add degree symbol to 38C

We corrected this typo.

Line 255: “after the descent” – during the descent? After the initial descent? The phrasing as written implies once it is on the ground

We changed this phrase to “after the balloon cutaway” to clarify.

Line 299: in Kansas

We corrected this grammar error.

Line 301: Are you saying the StratoCore data can get higher than the ER-2 or that it covers more of the stratosphere than the ER-2?

Here we are trying to express that the StratoCore sample can reach 25-28 km ASL, above ER-2 altitude, meaning it may cover more of the stratosphere in a cost-effective method. The two sentences here are repetitive so we deleted one of the sentences.

Figure 9: Is it possible to outline the symbols to make them easier to see? Or make them larger like in Figure 10.

We revised Figure 9, increased the size of the markers so they are easier to see. In addition, we added 3 panels in Figure 9 showing the vertical profiles of CO<sub>2</sub>, CO and CH<sub>4</sub> measured on the four flights to provide more information.