From: Getachew Agmuas Adnew and co-authors

Anonymous Referee #1

We thank referee 1 for the constructive feedback and suggestions on how to revise the manuscript. The answers to the questions/ comments and suggestions are stated below each comment in blue. *Paragraphs that are modified in the revised manuscript are shown in blue (italics).*

This work presented high-precision measurement results of CO2 triple oxygen isotopes from upper troposphere lower stratosphere air samples up to 21 km collected during past aircraft campaigns. The results are interesting as it showed distinct relationship between triple oxygen isotopic compositions and N2O for air in the upper troposphere vs. lower stratosphere. Such observation is critical to enable CO2 triple oxygen isotopes as a tool to understand stratosphere-troposphere exchange, as well as global carbon cycle. This work highlighted the importance of high-precision triple oxygen isotopes measurements during quantification of the downward net isoflux of O-MIF signal. While the presentation of the results is clear, interpretation of the data is mostly adequate, I have a few minor general comments:

We appreciate your kind words about our work presented in the manuscript and your valuable comments.

1. It would be great if there is more discussion about the de-coupling of chemical mechanisms of CO2-O17 generation and N2O loss in the stratosphere due to stratospheric dynamics (Lines **325 - 344**). The CO2-O17 signal is originated from ozone chemistry, therefore the path history (O1D abundance vary greatly in the stratosphere) and age of the air parcel are both important; while N2O is more sensitive to altitude as the photochemical lifetime of N2O decrease exponentially in the stratosphere. Therefore, air parcels that are relatively “young” but have been to mid-stratosphere (~30 km) could have significant N2O loss but low O17, and vise versa. Clarifying some of these mechanisms could be useful.

In section 2.2 of the revised manuscript, we included the following paragraphs for clarification:

The isotopic composition of CO_2 in the upper stratosphere and mesosphere provides a unique tool for studying atmospheric transport and chemistry [Wiegle et al. 2013; Liang et al., 2007, 2008, boering et al., 2004]. The $\Delta^{17}O$ of CO_2 is primarily modified by $O(^1D)$, which is produced photochemically by O_3 photolysis. However, the relevant isotope effects occurring in the stratosphere are still not yet well enough understood [Wiegle et al.2013; Liang et al., 2007, 2008]. Nevertheless, an empirical estimate of the isotope flux *from the stratosphere can be derived from measurements near the tropopause, like the ones presented here*

In the stratosphere CO_2 and N_2O isotopes are influenced by different processes. N_2O is mainly destroyed by N_2O photolysis but is also affected by $O(^1D)$ in the lower stratosphere and upper troposphere. Since N_2O photolysis and O_3 photolysis occur at different wavelengths, the relationship between $\Delta^{17}O(CO_2)$ and N_2O contains valuable information about atmospheric chemistry and transport. The lifetime of N_2O varies with altitude. In the upper stratosphere and mesosphere, the N_2O lifetime decreases, leading to greater scatter between the two tracers$\Delta^{17}O(CO_2)$ values increase with altitude as N_2O mixing ratios decrease below ~70 km. However, above 70 km, $\Delta^{17}O(CO_2)$ begins to decrease with further decreases in N_2O mixing ratios. However, in the lower most stratosphere and upper troposphere, where the lifetime of N_2O against photolysis is longer than the transport time, the scatter in N_2O values remains low. The $\Delta^{17}O$–N_2O correlation remains consistent both spatially and temporally in the lowermost stratosphere and upper troposphere (Liang et al., 2007, 2008). Since the net isotope flux of $\Delta^{17}O$ is derived from samples from the lower stratosphere and upper troposphere, the observed variability (scatter) in the stratosphere does not affect the global average $\Delta^{17}O$–N_2O slope used to estimate the flux of $\Delta^{17}O$ from the stratosphere to the troposphere.

2. More discussion may be needed to support the argument that the slope from CARIBIC samples can represent a “global average” N2O-O17 slope. Because of the observed potential “de-coupling” of N2O-O17 slope, it could be useful to discuss what are the potential factors that could result in different slopes. If the well-mixed upper trop air from CARIBIC represents global average slope, StratoClim gives you “below average” slope, where can you anticipate “higher than average” slopes? How will such variations impact the uncertainty of the global average slope?

   See our reply to the previous comment.

3. If the uncertainty in “global average” slope changed because of 1) and 2), how does it impact the uncertainties in global estimation of O17 isoflux?

See previous comment

More detailed comments:

Sections 2.2 & 2.3: since these were not mentioned until section 5, maybe considering moving these down (after 2.5) a little bit?

Thank you very much for your suggestion. In the revised manuscript we moved section 2.2 and 2.3 to appear after 2.5.

Line 174: maybe briefly mention how age of air is calculated?

In the revised manuscript we included a reference to the publication that was used to calculate the age of air.

The age of air was calculated using SF6 measurements as described in detail by Krol et al., 2018.

Line 249: CO2 not CH2.

Thank you very much for spotting this. In the revised manuscript the typo is corrected.

Figure 5: subpanel titles (a, b, c, d) are not lined up.

In the revised manuscript the subpanel title is aligned.

Figures 3-8: $\Delta'^{17}O$ is used in your text but in figures you used "$\Delta^{17}O$", please consider using consistent notations.

In the revised manuscript we have changed the $\Delta^{17}O$ in the figure label to $\Delta'^{17}O$.

Line 368: uncertainty inconsistent with figure.

Thank you very much for pointing out the inconsistency, in the revised manuscript the uncertainty indicator in the figures has been corrected to be consistent with the text.

Lines 388 & 394: repetitive sentences.

In the revised manuscript the repeated sentence has been deleted.

References

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Liang, M.-C., Geoffrey, A. B., Brenton, R. L., and Yung, Y. L.: Oxygen isotopic composition of carbon dioxide in the middle atmosphere, PNAS, 104, 5, 2007.

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