Responses to the Referee #1 Comments

Thank you very much for your significant and useful comments on the paper "Stratospheric $\delta^{13}CO_2$ observed over Japan and its governing processes and potential as an air age tracer" by Sugawara et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows. The line numbers denote those of the revised manuscript.

This manuscript presents a novel data set of stratospheric δ13CO2 dating back to 1985 and examines the processes controlling its distribution and its potential use as an age tracer. I think this is a very useful dataset. The results presented will be of interest to ACP readers, and I think they will be acceptable for publication after relatively minor changes.

My main concern with the manuscript is the discussion of the δ13CP age results. I think the authors are overstating the agreement with the CO2 age estimate and the potential use of the δ13CP age estimate. Specifically, in the abstract it is stated that "the mean age derived from δ13CP was consistent with that derived from the CO2 mole fraction, suggesting its usefulness for further investigation of stratospheric transport processes.", while in the conclusions it is started that "it [δ13CP] should prove a useful tool for investigating stratospheric transport processes" and "d13CP age is slightly larger than CO2 age ... by about 1.1 years on average".

The stated consistency occurs only because there is a very large (30%) uncertainty in δ13CP, and consistency is in terms of the mean over all data. Figure 10 shows that there is no statistical agreement for many individual measurements. Also, I don't think an average bias of 20% (which is what 1.1 yrs is) can be considered small. Given this large uncertainty and bias, I am doubtful that δ13CP age is a useful estimate/tool, as stated in the abstract and conclusions. It has the potential to do this only if the uncertainty can be greatly reduced.

I think the text needs to be modified to better indicate that there is large uncertainty and bias, and this needs to be reduced if this is to be a useful age tracer.

We have deleted the descriptions about usefulness of $\delta^{13}C_P$ as an age tracer throughout the paper and clearly stated that the $\delta^{13}C_P$ age has large uncertainties and it cannot be used to improve mean age observations at present in Conclusions. Along with this, we have changed the manuscript title to "*Stratospheric $\delta^{13}CO_2$ observed over Japan and its governing processes*" by removing "*potential as an*

air age tracer". The related changes are as follows.

Lines 27-30: The sentences in Abstract "*used it to estimate the mean age of stratospheric air. Despite large uncertainties, the mean age derived from $\delta^{13}C_P$ was consistent with that derived from the CO_2 mole fraction, suggesting its usefulness for further investigation of stratospheric transport processes*" have been replaced with "*we found that $\delta^{13}C_P$ in the mid-latitude mid-stratosphere decreases over time with an about 5-year lag relative to the tropical upper troposphere. This fact strongly supports that stratospheric $\delta^{13}CO_2$ variations are governed by the airborne production of ^{13}C-depleted CO_2 by CH_4 oxidation, the gravitational separation, and the propagation of the decreasing tropospheric $\delta^{13}CO_2$ trend into the stratosphere*"

Lines 505-524: We have removed many sentences about $\delta^{13}C_P$ age in Section 3.5 and moved the minimum necessary descriptions to Appendix C. We have deleted sentence "*However, the result that $\delta^{13}C_P$ age and CO_2 age are roughly consistent implies that $\delta^{13}C_P$ age could be used in addition to SF_6, halocarbons, etc., for better multi-component age estimations (Umezawa et al., 2024).*" and add "*At present, the $\delta^{13}C_P$ age is subject to larger uncertainties, making unsuitable for use as an additional constraint for age estimation. However, considering that the average $\delta^{13}C_P$ age was estimated to be 5.5 ± 1.6 years, the concept of stratospheric $\delta^{13}C_P$ itself would be valid.*".

Lines 539-541: We have deleted sentence in Conclusions "*our results showed that stratospheric $\delta^{13}C_P$ can serve as an additional age tracer alongside CO_2 and SF_6 mole fractions, and it should prove a useful tool for investigating stratospheric transport processes*" and add "*Because the $\delta^{13}C_P$ age has larger uncertainties than the CO_2 age at present, it is difficult to refine the mean age estimation. However, $\delta^{13}C_P$ in the mid-latitude mid-stratosphere decreased over time with a time delay and it was found to be quasi-conservative in the stratosphere.*".

Following on from this, I think there could be more discussion of potential errors. On line 549 it is stated that "it is likely that d13CP age is overestimated because the effect of methane oxidation was underestimated in the calculation of d13CT." Why was methane oxidation underestimated, and can an estimate of the impact of this be made?

Lines 681-687:We have added some descriptions about underestimation of the effect of methane oxidation in Appendix C. Note that the related sentences have been moved from Conclusions to Appendix C, in relation to the deletion of the descriptions about usefulness of $\delta^{13}C_P$ as an age tracer. We have added sentences "*The $\delta^{13}C_P$ age is larger than the CO_2 age (4.4 ± 0.6 years) by about 1.1 years on average. In*

this regard, $\delta^{13}C_T$ was calculated from the observed CH_4 mole fraction and its $\delta^{13}C$, assuming a closed system (Eq. 9). However, actual chemical processes do not occur in a closed system, and atmospheric mixing processes always result in apparent fractionation being smaller than true fractionation (Rahn et al. 1998; Kaiser et al. 2002; Toyoda et al., 2018). In this study, the isotopic effect of CH_4 oxidation was calculated based on the apparent fractionation factor. This would result in an underestimation of the CH_4 oxidation effect in Eq. 9 and an overestimation of the $\delta^{13}C_P$ age. To solve this problem, it is necessary to explicitly incorporate the isotope effect of CH_4 into the model, which will be a future challenge."
Along with this, we have added some references.

Finally, the age calculation from δ13CP is buried at the end of Appendix C. Rather than having this appendix focused on CO2 and then meaning the new aspect (δ13CP) at the end, both tracers should be discussed at the start. Highlighting when differences in approach, or steps were could be large uncertainty.

Lines 644-717: The title of Appendix C has been changed to "*Age calculation*" and totally rearranged descriptions following your suggestions. We first explained the age estimation methods common to both components and then described for $\delta^{13}C_P$ age. Figure of convolutions for $\delta^{13}C_P$ has been moved to C1. Accordingly, the order of the figures C1 – C4 has been changed.

Other revisions

Fig. 3

Because NOAA GML and INSTAAR have kindly provided us with updated $\delta^{13}C$ data at MLO in addition to well-organized data repository (Michel et al., 2025), we have replaced Fig.3 and replaced references. This change does not affect our results. Accordingly, the old organization name "ESRL" in NOAA was changed to the new name "GML".