

Interactive comment on “Probing stratospheric transport and chemistry with new balloon and aircraft observations of the meridional and vertical N₂O isotope distribution” by J. Kaiser et al.

J. Kaiser et al.

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We thank the referee for the constructive comments on our manuscript. The technical corrections pointed out by the referee have been taken into account in the revised manuscript. In the following, we respond to the specific comments, which were made.

1. It is true that the ASA 2002 data for mixing ratios below 150 nmol mol^{−1} are close to the generic climatology. However, they deviate for mixing ratios greater than 200 nmol mol^{−1}. This applies to other balloon profiles as well. The magnitude of the deviation is less likely to depend on latitude, but rather on the historic development of N₂O and CH₄ mixing ratios relative to each other, as we have pointed out in the paper. We think that a detailed discussion is beyond the scope of the present paper and have chosen

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to change the last phrase of the paragraph to “This may explain why some samples fall above or below the generic N_2O - CH_4 climatology.”

2. We have now added a black rim around the symbols for Kiruna 02/92, Kiruna 03/95, ASA 09/93 and India 04/99 to help distinguish the symbols in all figures.

3. This is a misunderstanding. The age difference of 4 months refers to the maximum age difference that could be caused by using either the Boering et al. (1996) or the Engel et al. (2002) N_2O -age relationships. For clarity, we have changed the sentence to “The age difference between the parameterization of Engel et al. (2002) and that based on the data of Boering et al. (1996) is at most 4 months, which can be neglected for the present application.”

4. In our opinion, the advantages of a second-order polynomial fit in itself (better description of individual profiles, less bias towards the most enriched samples) are sufficient to merit its application. However, for illustrative purposes, we also give an example of the improvement achieved by a second-order polynomial now and have added the sentence “For illustration, the average regression coefficient R^2 for ^{15}N Rayleigh plots improves from 0.988 (linear fit for samples with $\ln(\mu/\mu_{\text{T}}) > -0.6$) and 0.997 (linear fit for samples with $\ln(\mu/\mu_{\text{T}}) < -0.6$) to 0.999 for a quadratic fit. Higher-fitting orders do not lead to a significant improvement of the goodness-of-fit.”

5. The numbers we quote from Toyoda et al. (2004) were taken from their Table 3 and according to their paper have been calculated using their Eq. 12. It has not been pointed out by Toyoda et al. (2004) that using the intrinsic photochemical fractionation constants in combination with the apparent stratospheric fractionation constants does not consider the effects of diffusion and mixing. We acknowledge that our discussion on the relative contribution of photo-oxidation in Section 3.5 is rather condensed. We will extend it in the revised version of the paper, backed up with equations, an additional figure and calculations of the relative contribution of photo-oxidation for different N_2O mixing ratios.

6. We have added vertical bars to indicate absolute amounts.

7. We agree that other authors have considered mixing and its influence on isotope ratios in other contexts and environment. However, we couldn't find a concise derivation of the $\delta-\mu^{-1}$ relationship we are using anywhere in the existing literature. The paper by Bender (1990), which we are citing, came closest to our needs, but had a sign error. We therefore think that it is useful to leave this derivation for use by future readers. Reference is now made to the colloquial name "Keeling plot" for $\delta-\mu^{-1}$ plots, even though the 1958 paper by the late Dave Keeling, which is often cited in this context, only discusses the mathematical relationship between δ and μ^{-1} , but doesn't show a corresponding plot.

8. We have added an inverse mixing ratio x-axis in the revised paper.

9. We have deleted "the photochemistry of".

10. We have now added an explanation why we think that a plot of the residuals is more meaningful than simple R^2 values, but also note what the change in R^2 is: " R^2 values are less indicative of the suitability of the fit than a possible pattern in the residuals. For completeness, we note that R^2 improves from 0.997 to 1.000 (SOLVE) and from 0.993 to 0.994 (EUPLEX) upon change of the lower boundary from 170 nmol mol⁻¹ to 200 nmol mol⁻¹."

11. Fig. 6 shows that the EUPLEX measurements form a "curve" below the simulated red dotted line. Fig. 13b shows how the EUPLEX results "bend away" from the simulations for $r = 0$. We think that this illustrates the curvature sufficiently. Also, Table 2 shows the increase of the apparent fractionation constants with altitudes. Finally, please note that the figures will approximately double in size in the ACP version compared to the present ACPD version and can be viewed at great magnification using a PDF reader. We do not want to inflate the paper with an additional figure and therefore prefer to refrain from adding a magnified figure to the paper. The statement that the continuous weak mixing model successfully simulates the increase of the apparent

fractionation constant with altitude was probably too strong. What we meant is that its qualitative behavior, i.e., an increase of the apparent fractionation constant with altitude is correct. However, it cannot be expected to simulate the behavior quantitatively, as it is only a very simple conceptual model. We will clarify this in the version of the paper.

12. It is correct that the uncertainties stem from the fitting procedure. We have added an explanation to the Table caption to indicate where the uncertainties stem from.

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