

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.

Anonymous Referee #2

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Kaiser et al. report a comprehensive isotopic data set of stratospheric N₂O which includes valuable vertical profiles over tropics as well as intensive Arctic observations. Making careful and precise analyses of their data, they demonstrate that N₂O isotope ratios can be used as fingerprints of chemistry and transport in the stratosphere. They first find that the isotopic data could support “continuous weak mixing between intravortex and extravortex air”, which will stimulate the researches on modeling of atmospheric circulation. The paper is well structured and clear, although some parts are a little lengthy. I recommend publication after minor revisions.

Specific comments

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1. p. 4283, line 18. The authors seem to attribute the departure of tropical data of 1987 from the generic N₂O-CH₄ relationship to different evolution of the two species during the 6-7 years before the base year for the relationship (1993-1994). If it would be the case, why does the latest data, e.g., ASA 2002 is more than 6 years apart, lie very close to the tracer relationship? Is the magnitude of the deviation expected to depend on latitude? Need more explanation.

2. p. 4284, line 4. I cannot follow the feature of “Kiruna 03/95” because data plots for Kiruna in Fig.5 are difficult to distinguish each other except for March 2003, due to the use of similar colors. This is also the case for Figures 3, 4, 6, 9, and 10.

3. p. 4287, line 11. Why do the authors adopt the age difference between tropopause and tropospheric air of 0.8 years despite the fact that they base on the work of Boering et al. (1996) in which the age difference is ~ 4 month (0.33 years) as described in the preceding sentence?

4. p. 4288, line 8. Although the authors state that a second-order polynomial fit to the Rayleigh plots has advantage, neither the basis for the choice of order (why second?) nor excellence of the fitting (e.g., R^2) is shown.

5. p. 4289, line 15. I suppose that the authors misread the Toyoda et al. (2004) paper. “Contribution of photo-oxidation between 70-80%” does not seem to be the conclusion of the cited authors as they noted the significance of the factors other than photochemistry and discuss them separately. In addition, the authors don't show the degree of photo-oxidation in Sect. 3.5 while they state it is much less than 70%.

6. p. 4289, line 24. Since epsilon takes negative value, ambiguous expression should be avoided. “ $\epsilon(15\text{N})_{\text{app}}$ values are far below -48 permil” means that the absolute value of ϵ is below 48 permil, doesn't it?

7. p. 4291, Sect. 3.3.1. Evidence of “end-member mixing” was discovered about 50 years ago (Keeling, 1958) although the author did not explicitly derive the relationship

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between delta and inverse mixing ratio using mathematical equations. The relationship has been known as “Keeling plot” among many atmospheric scientists. Why do the authors ignore the previous work and describe lengthy equations? In addition, it is certain that mathematical derivation of two-component mixing was published not later than 1970’s (e.g., G. Faure (1977), Principles of Isotope Geology, John Wiley & Sons). Incidentally, index “A” to “x” is missing in equation (4).

8. p. 4292, line 14. Discussion relevant to Figs.9 and 10 is not easy to follow. Additional horizontal (not inversed) concentration axis would help.

9. p. 4292, line 24. Delta-inverse mixing ratio relationship is not based on the photochemistry but mixing, although it may be affected by the photochemistry.

10. p.4293, line 20. I don’t think Fig.11 is of significance because the stated trend of data plots for $mew > 170$ nmol/mol in the right panel seems not clear. If the authors would like to demonstrate the suitability of linear regression, why don’t they just compare the R^2 of the two cases: linear fitting for 200-320 nmol/mol range and 170-320 range?

11. p. 4297, line 24. “Curvature of the EUPLEX measurements” is not clear in Figs 6 and 13b. Also I cannot see whether the continuous week mixing model really simulates the increase of the magnitude of epsilon with decreasing N_2O mixing ratio. More magnified figure would help.

12. p.4309, Table 2. Uncertainty shown with plus-minus is associated with linear or polynomial fitting?

Technical corrections

13. p.4285, line 7. I suppose “below 70 nmol/mol” should be “greater than ...” and “greater than ...” (next sentence) should be “below”

14. p. 4285, line 12. “To be invoked the explain...”: awkward phrase.

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15. p.4301, line 9. Mixing ratios in the lower stratosphere should be GREATER THAN 200 nmol/mol.

16. p.4313, Fig. 4. Unit of the vertical axis must be MICRO mol/mol.

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