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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Development of an atmospheric N_2O isotopocule model and optimization procedure, and application to source estimation" *by* K. Ishijima et al.

Anonymous Referee #2

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Review for the following manuscript Journal: ACP Title: Development of an atmospheric N2O isotopocule model and optimization procedure, and application to source estimation Author(s): K. I. Ishijima et al. MS No.: acpd-15-19947-2015 MS Type: Research Article

I have major criticisms on technical justifications. The model sections were written in a way that I cannot tell whether the approaches they adopted are reasonable or not. Interpretation was not well presented and what the new things the current modeling is delivering are not clear. Details are listed below.

Major concerns:

1. Section 4.1.2 Given that the photolysis in the stratospheric is critical for N2O fractionation. One has to examine carefully the constants used in the simulation. Without correct realization, the results derived and conclusions drawn are weak. It is known that the degree of fractionation is a strong function irradiation wavelength. Indeed wavelength dependent photodissociation is account for for normal species (though at limited resolution) but not for isotopic species, if my understanding is correct. Single epsilon is obtained for each isotopocule. Mixing/transport and photon attenuation affect the distributions of the N2O isotopocules. Simply fitting the isotopocules observed in the stratosphere to get epsilon's is not acceptable. Though optimization is done (though not clear to me how this is done, given that the data are limited and variable), the correction is applied uniformly to all latitudes/altitudes. I'd suggest examine wavelength dependence carefully in the model before optimization starts.

Lower stratospheric N2O concentrations are critical for cross-tropopause exchange, i.e., >200 nm photolysis where you have 3 bins only. An attempt to estimate the error due to poor simulation with 3 bins is required. To make the model more applicable, finer spectral resolution is preferred.

2. Next critical parameter is transport Two transports are crucial for N2O modeling. One is the transport in the stratosphere and the other is cross-tropopause exchange.

The former can be done by trying to model the so-called age of air in the stratosphere. ECMWF-Interim does a good job but other new reanalyses (NCEP R1/R2 and ERA40 are bad) are unknown to me. The authors have to show and demonstrate the robustness of the transport they used. I'm not convinced with the authors saying in section 5.1 that $\sim$2 year underestimation is not important.

The latter can be checked against, for example, data made in the mid-troposphere, if not upper-troposphere. To my knowledge, CARIBIC project (see, e.g., Assonov et al. 2013) provides a good dataset for the verification.

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The above three are critical before any solid conclusion can be given.

3. 50-yr spun-up. Given that the lifetime of N2O is >100 yr in the atmosphere. The mode spun-up should be longer than that. Please explain.

4. Despite being with such a more complicated 3D model framework, I don't see what new things learned from the exercise. The authors conclude that 3D model agrees with updated box-model estimation.

5. For trends in each hemisphere, given the long lifetime in the troposphere I expect the trends are similar. Moreover given the facts of long N2O lifetime and short cross-hemisphere mixing/transport, why there's noticeable difference in trend between stations?

Other comments:

6. Section 4.2.1 is not clearly written I was lost initially. After reading through it once more more carefully, I can follow. I don't have good suggestions how to reorganize it at the moment. The authors should do some work polish/rephrase.

7. The validity of linear assumption for Equations 10-11. Typical photolytic fractionation can be described by Rayleigh distillation, which is not linear. How the resulting parameters become linear, with respect to variations in input variables, like E. Linear assumption may be a good approximation for small varying variables, but this has to be shown, though in the first sentence of section 4.2.1 it's written that this is confirmed. Given the number of N2O dissociative photons is limited, the total destruction rate is not constant, due to additional UV absorption from other molecules. To what extent the linear approximation is valid has to be shown.

8. Seasonal cycles It would be good if the authors also present a comparison of the model seasonal cycles with the observed ones, in a table.

9. Please show spatial homogeneity of N2O in figures for each hemisphere.

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10. Age of air to verify the transport Stratospheric transport is crucial for N2O modeling. Please verify your transport against the determined age of air. I know ECMWF-interim reanalysis is good but am not sure JMA reanalysis.

11. Too busy fig 4 If possible, please separate 3 variables into 3 figures.

Same for fig 10, difficult to compare model with data.

12. Section 4.1.3 that a single scaling factor was used, inconsistent with Fig S1 that latitudinal profiles between the 2 cases in each hemisphere are not off by a single factor. Please check. I may misunderstand the plot.

13. Section 5.1, to scale the fractionation to reproduce stratospheric observations requires a modification beyond the uncertainly of von Hessberg et al. Please compare explicitly the values of the two.

14. Why not include Toyoda et al. (2013) data? Please also state clearly why not include the new data for model optimization. Anthropogenic is a component of the model.

15. Latitudinal gradient in section 5.3 should be in unit of nmol molˆ-1 per deg latitude. Or you meant hemispheric difference?

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