

**Manuscript structure:** To avoid fragmentation and for ease of readability, tables S1 and S2 as well as Figures S3 to S9 should be moved to the main part of the paper. They are key parts of the Results and Discussion sections and in many cases referred to repeatedly in the text. Since Table S3 only adds one additional column with new values compared with Table 2, it should be merged with Table 2.

**Data availability:** Please upload all data used for the study to a publicly accessible archive and provide a Digital Object Identifier (DOI). It is not sufficient to have them available on request from the lead author. For details, please refer to [https://www.atmospheric-chemistry-and-physics.net/about/data\\_policy.html](https://www.atmospheric-chemistry-and-physics.net/about/data_policy.html)

**Mixing calculation:** There is an error in your "Estimates of the combined effects from STE and soil emissions on  $\delta^{15}\text{N}^{\text{SP}}$ ". The effect on  $\delta$  due to mixing with stratospheric air is about three times larger than the 5 ‰ value you used. In contrast, the effect due to soil emission is too big and should be about three times smaller. However, overall, the combined effect is too small to be measured.

I think this is an important calculation and you should move it from the supplement to the main text (after correcting it as explained in the following).

*Mixing with stratospheric air:* The effect on tropospheric air can be approximated using the apparent isotopic fractionation  $\epsilon_{\text{app}}$  (Kaiser et al. 2006). You can readily derive this from the slope of the Rayleigh fractionation equation at  $y = y_{\text{T}}$  (where  $y_{\text{T}}$  is the  $\text{N}_2\text{O}$  mole fraction at the point the of mixing between troposphere and stratosphere):

$$\delta = (1 + \delta_0) f^{\epsilon_{\text{app}}} - 1 \quad \text{with } f = \frac{y}{y_{\text{T}}}$$

$$\frac{d\delta}{df} = (1 + \delta_0) \epsilon_{\text{app}} f^{\epsilon_{\text{app}} - 1}$$

$$y_{\text{T}} \frac{d\delta}{dy} = (1 + \delta_0) \epsilon_{\text{app}} \left( \frac{y}{y_{\text{T}}} \right)^{\epsilon_{\text{app}} - 1}$$

This gives

$$\lim_{y \rightarrow y_{\text{T}}} \frac{d\delta}{dy} = \frac{(1 + \delta_0) \epsilon_{\text{app}}}{y_{\text{T}}}$$

The derived slope is in agreement with the compact relationship between  $\delta$  and  $\text{N}_2\text{O}$  mole fraction plotted in Fig. 5 of Kaiser et al. (2006).

For the "site-preference"  $\delta^{15}\text{N}^{\text{SP}}$ ,  $\epsilon_{\text{app}}$  needs to be replaced with the difference between the  $^{15}\text{N}/^{14}\text{N}$  isotope fractionations at the central and terminal N atoms,  ${}^2\epsilon_{\text{app}} - {}^1\epsilon_{\text{app}}$  (or  ${}^\alpha\epsilon_{\text{app}} - {}^\beta\epsilon_{\text{app}}$ ).

For the lower stratosphere,  $\epsilon_{\text{app}}(^{15}\text{N}^{\text{SP}}) \approx -15$  ‰ (Kaiser et al. 2006), i.e. about three times larger than your value of 5 ‰.

*Effect of soil emissions:* I am not sure what you mean by the sentence "The isotopic effect of soil emission (−30‰), which is mainly attributed to the switch from nitrification to denitrification, is taken from Sutka et al. (2006)." It sounds as if you have taken the difference between the  $\delta^{15}\text{N}^{\text{SP}}$

values of denitrification (0 ‰?) and nitrification (+30 ‰?). However, the relevant quantity here is the difference between the  $\delta^{15}\text{N}^{\text{SP}}$  values of soil emissions (+7.3 ‰; Table 3) and tropospheric air (+18 ‰; Fig. 2), i.e. -10.7 ‰, i.e. three times smaller than your estimate.

*Overall effect:* Since your estimate  $\text{N}_2\text{O}$  mole fraction enhancement due to emissions (0.15-0.2  $\text{nmol mol}^{-1}$ ) is about twice the  $\text{N}_2\text{O}$  mole fraction depletion due to mixing with stratospheric air (-0.35-0.4  $\text{nmol mol}^{-1}$ ), the net effect is  $[(-0.4 \text{ nmol mol}^{-1})(-15 \text{ ‰}) + (0.2 \text{ nmol mol}^{-1})(-10.7 \text{ ‰})] / (330 \text{ nmol mol}^{-1}) = 0.01 \text{ ‰}$ . In other words, the effect due to STE dominates, but is too small to be measured.

#### **Other corrections:**

Röckmann, not Rockmann

Units and chemical symbols should not be mixed. Please express mass fluxes as "Tg  $\text{a}^{-1}$  N equivalents" or "Tg  $\text{a}^{-1}$  N" with a short explanation what this means upon first usage.

ACP requires adhering to the International System of Units (SI) and the Recommendations in the IUPAC Green Book ([https://www.atmospheric-chemistry-and-physics.net/for\\_authors/manuscript\\_preparation.html](https://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html))

Please use a suitable (single-letter) quantity symbol for the troposphere-stratosphere air exchange flux (e.g.  $F_{\text{TS}}$  or  $F_{\text{ex}}$ ).

I. 174 & 175: Please use SI units for mole fractions, i.e.  $\text{nmol mol}^{-1}$  (not ppb).

I. 301: Ditto, but " $\text{pmol mol}^{-1}$ ".

I. 240: What is the remaining 1 ‰? Even with rounding errors, the values of 78 ‰ and 21 ‰ cannot add up to 100 ‰.

I. 673: Please replace "species" with "deltas".

Table S2: The uncertainty range for the stratosphere-troposphere exchange rate is too wide. In particular, it cannot be negative.

Tables 2 & S3: You should add the estimates of Röckmann et al. 2003 to the table since you cite our study on various occasions. You may need to make an assumption on the modern tropospheric values on international scales (e.g. use your measurements). An alternative idea would be to express the source signatures of each study relative to the modern value, to avoid biases due to different isotope calibration scales. The source vs. troposphere  $\delta$  differences are more robust than the absolute values.

Tables 2 and S3: The internationally accepted abbreviation for "year" is "a" (from Latin annum). You actually use the symbol "a" elsewhere in the manuscript.

Your responses to queries 19 and 20 of referee #2 are missing.