

Dear Dr Li

Many thanks for submitting your further revised manuscript, which I am happy to accept for publication subject to a few minor revisions as outlined below.

- 1) Please integrate the supplement into the main text as appendix or appendices. The ACP manuscript guidelines require that "Supplementary material is reserved for items that cannot reasonably be included in the main text or as appendices. These may include short videos, very large images, maps, CIF files, as well as short computer codes such as matlab or python script. [...] Normal size figures, tables, as well as technical or theoretical developments that do not need to be included in the main text should be included as appendices."
- 2) Please remove the unnecessary approximations made in deriving equations 7 and 8, and make them consistent with Eq. 3. I have included the final steps of the calculation without approximation here (using the same δ and ε symbols as in the calculation I sent you during the discussion stage). Obviously, your steps up to the ratio R/R_2 are fine (except that I have used the inverse ratio, to simplify the final step of the calculation):

Mass balance:

$$f\delta(\text{NO}) + (1-f)\delta(\text{NO}_2) = \delta(\text{NO}_x)$$

$$f\delta + (1-f)(1+\delta_2) = \delta_x$$

$$\delta_2 - \delta_x = f(\delta_2 - \delta)$$

with $f = f(\text{NO})$

Steady-state assumption to derive Eq. 7:

$$\frac{R}{R_2} - 1 = \frac{1+\delta}{1+\delta_2} - 1 = \frac{1+A\alpha_1}{1+A\alpha_2+\varepsilon} - 1$$

$$\frac{\delta - \delta_2}{1+\delta_2} = \frac{A(\alpha_1 - \alpha_2) - \varepsilon}{1+A\alpha_2+\varepsilon}$$

$$\delta_2 - \delta = (1+\delta_2) \frac{(\alpha_2 - \alpha_1)A + \varepsilon}{1+A\alpha_2+\varepsilon}$$

Steady-state assumption combined with mass-balance to derive Eq. 8:

$$\begin{aligned} \delta_2 - \delta_x &= (1+\delta_2) f \frac{(\alpha_2 - \alpha_1)A + \varepsilon}{1+A\alpha_2+\varepsilon} \\ &= (1+\delta_2) [1 - f(\text{NO}_2)] \frac{(\alpha_2 - \alpha_1)A + \varepsilon}{1+A\alpha_2+\varepsilon} \end{aligned}$$

Setting $A = 0$ (very fast isotope exchange) immediately reproduces Eq. 3 for verification.

- 3) There is no need to repeat the reaction equations (R1 to R6, p. 5 of current supplement) or the definition of delta values (p. 6 of current supplement, lines from "Next, to calculate ..." to " $R_{\text{NO}_2}/R_{\text{NO}} - 1$ ") in the Appendix.
- 4) Please remove unnecessary factors ("1000 ‰") from your mathematical equations (e.g. l. 180 and 182).
- 5) Please also remove the unnecessary multiplication symbols (\times). These make the equations unnecessarily difficult to read. Such multiplication symbols are rarely required outside scientific notation, e.g. $8 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$.
- 6) L. 182: The quantity with the symbol ϵ should not be called "isotope enrichment factor" (which is a different kind of quantity). Please use the term "equilibrium isotopic fractionation" instead (as you do on l. 89 and elsewhere).
- 7) L. 188 & 193: The quantities should be enclosed in parentheses so that the unit applies to value and uncertainty, e.g. " $(-58.7 \pm 0.8) \text{ ‰}$ "

Kind regards
Jan Kaiser
Editor ACP