

We are grateful to the reviewer for the valuable suggestions and/or comments which improve the manuscript significantly. Below we list the detailed responses to the reviewer's suggestions and comments. The comments are listed in italics, followed by the response in normal font with changes highlighted in blue.

Response to Referee #1

Ice core nitrate isotope compositions may be useful to reconstruct atmospheric nitrate isotope compositions in the past with paleoclimate implications. Previous studies have investigated impacts of post-depositional processing on isotope compositions of nitrate preserved in snow and ice. In this work, Jiang et al. revised the TRANSITS model (a one-dimension snow photochemistry model) to calculate atmospheric nitrate deposition flux and isotope compositions based on snow records. Exemplary applications were applied to Summit and Dome C data and the calculated results were compared with measurement data. Although future efforts are needed to further evaluate and improve this model, this work is an important step towards a more precise interpretation of ice core nitrate isotope data. The equation derivation appears correct, and the model logic looks scientifically reasonable to me. However, the writing of this manuscript is too technical to readers outside the small community of post-depositional possessing of nitrate isotopes, and one may need to read all papers written by the authors previously to understand this work. This writing style is not easy for casual readers (especially for atmospheric scientists who do not work in cryospheric sciences and isotopes) to follow. I spent considerable time to digest the manuscript, even though I am kind of familiar with topics discussed in this manuscript. I therefore have some suggestions that aim to improve the clarity of this manuscript.

Response: We really appreciate the reviewer's time and effort on evaluating the manuscript, and thanks for the positive comments. Indeed, the topic of the manuscript is for a relatively small community and would be difficult to understand without sufficient background. In the revised manuscript, we have followed the reviewer's suggestion and try to improve the readability of the manuscript.

Line 29: Please define F_{pri} at the very beginning. For readers who did not read the authors' previous papers, they would not understand what it is.

Response: We have added the following text at the beginning of the abstract: "...reconstruct primary nitrate flux (i.e., the deposition flux of nitrate to surface snow that originates from long-range transport or stratospheric input) and its isotopes..."

Lines 263-265: Please rewrite this sentence. It is not clear.

Response: The original sentence "...The subsequent conversion of NO₂ to nitrate would also determine 1/3 of the oxygen atom of the newly formed nitrate." has been rephrased as follow:

"...During the subsequent oxidation of atmospheric NO₂, one more oxygen atom inherited from the oxidants (e.g., OH or BrO) is incorporated into one newly formed

HNO₃ molecule. Thus, $\Delta^{17}\text{O}(\text{FP})$ can be represented by 2/3 of $\Delta^{17}\text{O}(\text{NO}_2)$ plus 1/3 of $\Delta^{17}\text{O}(\text{oxidant})$ ”

The definition listed in Table 1 is unclear. For example, I do not understand what “d15N of archived nitrate flux” means. Does a flux have a d15N value? Does it mean d15N of archived nitrate? In my opinion, these definitions make the manuscript difficult to follow.

Response: Thank you for pointing this out. We follow the reviewer’s suggestion to avoid the term “ $\delta^{15}\text{N}$ of xx nitrate flux” and substitute it to “ $\delta^{15}\text{N}$ of xx nitrate” in the revised manuscript. In this case, “ $\delta^{15}\text{N}$ of archived nitrate flux” was revised as “ $\delta^{15}\text{N}$ of archived nitrate”.

There are 13 input parameters listed in Table 1, but only 6 are described in Table 2. It is difficult for readers to check everything throughout different parts of the manuscript and from different papers.

Response: Thanks for this point. We have added the omitted parameters in Table 2 to make it accordant with Table 1. The new Table 2 are shown as follows:

Table 2. Values of major parameter used in the model simulations at two different sites.

Parameter	Dome C, Antarctica		Summit, Greenland	
	Value	Reference	Value	Reference
FA	$1.3 \times 10^{-7} \text{ kgN m}^{-2} \text{ a}^{-1}$	Erbalnd et al. (2013)	$6.7 \times 10^{-6} \text{ kgN m}^{-2} \text{ a}^{-1}$	Jiang et al., (2022)
$\delta^{15}\text{N}(\text{FA})$	273.6 ‰	Erbalnd et al. (2013)	0.6 ‰	Jiang et al., (2022)
$\Delta^{17}\text{O}(\text{FA})$	26.0 ‰	Erbalnd et al. (2013)	27.9 ‰	Jiang et al., (2022)
A	$28 \text{ kg m}^{-2} \text{ a}^{-1}$	Erbland et al. (2013)	$250 \text{ kg m}^{-2} \text{ a}^{-1}$	Dibb et al., (2014)
ρ	300	Erbland et al. (2013)	380	Geng et al. (2014)
TCO	175-300 DU	Erbland et al. (2015)	228-494 DU	Jiang et al., (2021)
Φ	0.015	Adjusted ^a	0.002	Jiang et al., (2021)
σ	Wavelength dependent	Berhanu et al. (2014)	Wavelength dependent	Berhanu et al. (2014)
ε_d	+10 ‰	Erbland et al. (2013)	+10 ‰	Erbland et al. (2013)
$\Delta^{17}\text{O}(\text{NO}_3^-)$ of FP	Observed atmospheric $\Delta^{17}\text{O}(\text{NO}_3^-)$	Erbland et al. (2013)	Calculated	Jiang et al., (2021)

f_c	0.15	Erbland et al. (2015)	0.15	Erbland et al. (2015)
f_{exp}	0.2	Erbland et al. (2015)	0.35	Jiang et al., (2021)

^aAdjusted according to the best fit of snowpack nitrate $\delta^{15}\text{N}$ profile at Dome C (Appendix C).

Section 3: It is recommended to explicitly describe what parameters were used to do the calculation and what parameters were used to compare with the model results at each site. For example, it is stated that weekly data from Jiang et al. (2022) were used (Section 3.1). I need to re-read Jiang et al. (2022) to understand what these data are. In addition, I do not fully understand how the authors tested their model results. Did they use the atmospheric nitrate data reported in Jiang et al. (2022) or the surface snow as described in page 12? Where are the data from?

Response: Thanks for this suggestion. We have added more detailed explanations regarding the model input data (i.e., parameters used to do model calculations) and the observational data used to compare with the model outputs. In particular, for Summit, the snowpack nitrate concentration and isotope data with weekly resolution compiled in Jiang et al. (2022) were used as model input values (i.e., archived snow nitrate properties with weekly resolution). To compare with the model outputs, i.e., the model calculated atmospheric nitrate isotopes based on observed the snowpack data, we used the observed atmospheric nitrate isotopes at monthly resolution reported by Jiang et al. (2022). For Dome C, we mainly used the annual skin layer and atmospheric nitrate isotopic observations from Erbland et al. (2013) as observational constraints to compare with the model calculated skin layer and atmospheric nitrate isotopes, where the snowpack nitrate isotopes below the photic zone were used as model input values.

In the revised manuscript, in section 3.1, we added: “...The snowpack nitrate concentration and isotope data with weekly resolution at Summit compiled in Jiang et al. (2022) were used as initial model input values to represent the archived snow nitrate signals.”

At the beginning of section 4.1.1, we rephrased the original sentence to: “Currently there are no skin layer nitrate isotope observations at Summit, so we used the monthly atmospheric nitrate isotopic data from aerosol observations at Summit reported by Jiang et al. (2022) to compare with the modeled atmospheric nitrate isotopic variations...”

Lines 387-389 and 397-398: I would not say that the seasonality of modeled $d15\text{N}$ agrees well with observation based on Figure 3. As noted by the authors, the model cannot capture the seasonal variation from September to April.

Response: We have weakened our statement in the revised manuscript: “...As shown in Fig. 3, the modeled seasonality in atmospheric $\Delta^{17}\text{O}(\text{NO}_3^-)$ generally agrees well with the observed seasonal variations, while for $\delta^{15}\text{N}(\text{NO}_3^-)$, the model

predicted a similar seasonality as the observations, though in the winter half year the model underestimated the absolute values in comparison with the observations...”

Lines 428-430: Could the authors tune the epsilon value in the model, give a quick estimation what epsilon value may reproduce the observational data, and briefly discuss if this epsilon value is reasonable? This test should be straightforward.

Response: Thanks for this suggestion. In principle, the modeled atmospheric $\delta^{15}\text{N}(\text{NO}_3^-) = \text{modeled } \delta^{15}\text{N}(\text{FD}) - \epsilon_d$. In the model, the ϵ_d was set as +10 ‰ and this leads underestimations of atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$ in winter months compared to observations. In the above equation, if we replaced the **modeled** atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$ with the **observed** atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$, then the ϵ_d should be the epsilon values needed to reproduce the observations. In the following figure (Fig. 1), we plotted the epsilon values needed to reproduce the observations by applying $\epsilon_d = \text{modeled } \delta^{15}\text{N}(\text{FD}) - \text{observed atmospheric } \delta^{15}\text{N}(\text{NO}_3^-)$. As shown in the figure, the ϵ_d is generally close to the model default value of +10 ‰ during the summer half year, while in winter half year the ϵ_d is lower than +10 ‰ (except in March and December). This is consistent with what we speculated in the manuscript, i.e., the ϵ_d in winter is lower than that in summer and this is probably the reason leading to the model underestimation in winter months.

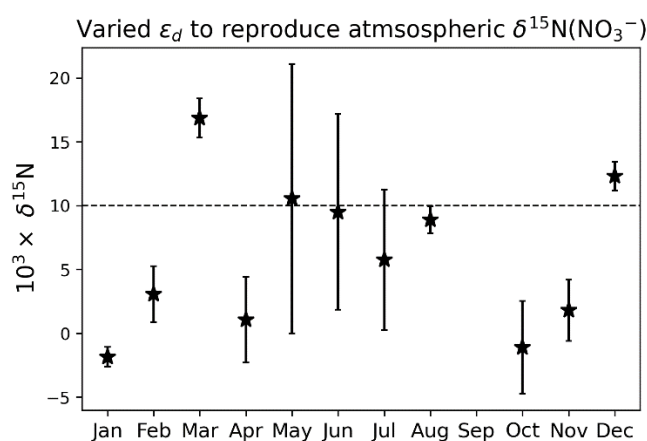


Figure 1. The calculated ϵ_d reproducing the observed atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$ at Summit using $\epsilon_d = \text{modeled } \delta^{15}\text{N}(\text{FD}) - \text{observed atmospheric } \delta^{15}\text{N}(\text{NO}_3^-)$.

Figure 5: I am confused what the results of “inverse model” mean. The inverse model used measured isotope values as input parameters. So I guess the model “results” plotted in this graph are the isotope values of “deposition nitrate” calculated from the model or the model input (calculated averages of measured values?). Please clarify.

Response: In this figure, we plotted the observed and modeled nitrate concentration and isotopes in the photic zone. The modeled results are from the TRANSIT model and the inverse model. The differences between the two modeled results are that the forward model (TRANSITS model) uses the prescribed **isotopes of the primary nitrate as model inputs** to calculate snow nitrate concentration and isotopes **in the photic zone and the archived layers**, while the inverse model uses **the archived**

snowpack nitrate concentrations and isotopes (i.e., observed snowpack data well below the photic zone) as model inputs to calculate snow nitrate concentration and isotopes **in the photic zone including surface snow, and those in the atmosphere**. This is saying, both models are capable of simulating the snowpack nitrate profiles in the photic zone, and these are what plotted in Figure 5.

To make it more clear, in the revised manuscript, we have added the following statements at the end of section 2.3:

“...The archived nitrate profile could be dated by using various types of seasonal markers, such as the $\delta^{18}\text{O}$ of H_2O , the ion concentrations or their ratios, and the snow accumulation rates (Hastings et al., 2004; Furukawa et al., 2017; Dibb et al., 2007). As long as the archived snow nitrate profiles (i.e., snow nitrate concentration and isotopes below the photic zone) are given, the model can calculate nitrate concentrations and isotopes throughout the photic zone, and those in the atmosphere. The latter is considered as the atmospheric signals before being affected by post-depositional processing.”

Figure 6: Is it possible to show a similar figure for Summit so that readers can better understand how the model behaves if we just look at the annual average?

Response: This is a good suggestion. But unfortunately, at Summit there is no skin layer observations so we can only compare the modeled and observed atmospheric values. The results are presented below as Fig.2. Overall, the modeled annual average values are in good agreement with the observations. For $\delta^{15}\text{N}(\text{NO}_3^-)$, the modeled and observed average values are $-17.5 \pm 3.0 \text{ ‰}$ and $-14.8 \pm 7.3 \text{ ‰}$ respectively, while for $\Delta^{17}\text{O}(\text{NO}_3^-)$ the values are $28.8 \pm 2.6 \text{ ‰}$ and $28.6 \pm 3.2 \text{ ‰}$. The small departure in $\delta^{15}\text{N}(\text{NO}_3^-)$ are likely caused by the assumed constant depositional fractionation factor used in the model as had been intensively discussed in the main text. Nevertheless, we think it’s a good idea to provide comparison on the annual average values at Summit and we have added the following text in the revised version (but we didn’t add the figure):

“...which is close to the value of 0.19 ‰ predicted by the TRANSITS model (Jiang et al., 2021). At an annual scale, the modeled and observed average atmospheric $\delta^{15}\text{N}(\text{NO}_3^-)$ are $-17.5 \pm 3.0 \text{ ‰}$ and $-14.8 \pm 7.3 \text{ ‰}$, while for $\Delta^{17}\text{O}(\text{NO}_3^-)$ the values are $28.8 \pm 2.6 \text{ ‰}$ and $28.6 \pm 3.2 \text{ ‰}$ respectively, suggesting that the inverse model reproduced the atmospheric observations quite well...”

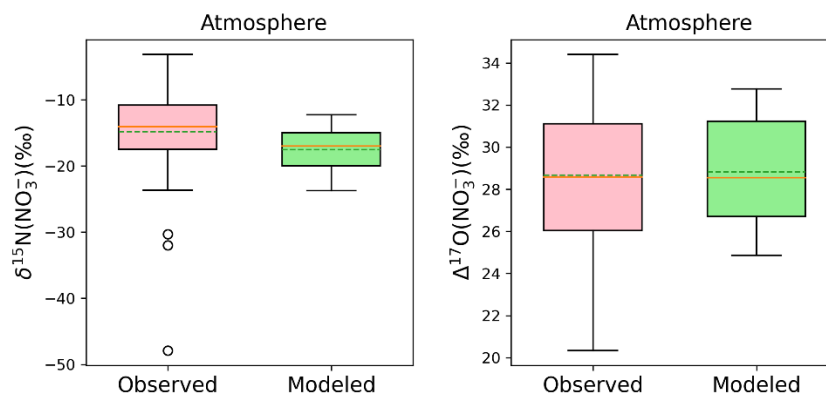


Figure 2. Comparison between the observed and modeled annual averages of $\delta^{15}\text{N}(\text{NO}_3^-)$ and $\Delta^{17}\text{O}(\text{NO}_3^-)$ in the atmosphere at Summit, Greenland. The atmospheric observations are adapted from Jiang et al. (2022). The solid line in the box plot indicates the median value, while the dash line represents the average value.

The authors may notice the new work by Shi et al. (2023), which is highly relevant to this manuscript and was just published after the submission of this manuscript. Please cite this work during the revision: Shi, G., Buffen, A. M., Hu, Y., Chai, J., Li, Y., Wang, D., & Hastings, M. G. (2023). Modeling the complete nitrogen and oxygen isotopic imprint of nitrate photolysis in snow. Geophysical Research Letters, 50, e2023GL103778

Response: Thanks for this reminder. We have added this citation in the revised version.