

We appreciate the reviewer for the time and efforts to review this manuscript, and for the suggestions/comments which improve the manuscript significantly. Below we list detailed responses to the reviewer's suggestions and comments. The comments are listed in italics, followed by the response in normal font.

Comments

In this study, the authors carried out a model study, trying to reveal the post-depositional processes of snow nitrate and isotopes at Summit, Greenland. This study addressed the question for the snow nitrate regarding the ice core study and in the scope of The Cryosphere.

The model was proposed by Erbland et al. (2015) and has been applied for the investigation of the post-depositional process of snow nitrate in Antarctica. The field data used in the model was taken from the previous studies. The present study demonstrated the significant redistribution of nitrate in the upper snowpack due to the photolysis in the high accumulation site. In addition, the effects of the post-depositional process on the isotopes ($\delta^{15}\text{N}$ and $\Delta^{17}\text{O}$) were investigated in a quantitative way. Thus, the present study has novelty and impact to be published in this journal after revising.

The methods were clearly written and suitable for this study. However, some assumptions in the model were not discussed, such as the effect of the wavelength, the wind blowing, the temperature, and the evaporation as mentioned in the specific comments. In addition, there is a lack of evaluation of the present study comparing to the previous model studies as mentioned in the specific comments.

Response: Thank you for the comments, we responded in details in the specific comments below.

Specific comments

Line 60-63: This sentence was supported by field observations (Erbland et al. 2013; Noro et al. 2018)

Response: Thanks for this suggestion. We have added these references in our main text accordingly.

Line 95-96: The e-folding depth depends on the wavelength (Noro and Takenaka 2020). How did you obtain the e-folding depth of each wavelength from 280-350 nm?

Response: Thank you this point. The e-folding depth does vary with wavelength, but here in the text we only report the value at 305nm to compare with other results at the same wavelength. To get the e-folding depth from 280nm to 350 nm, we used the method as follows: the attenuation of actinic flux in snowpack was modelled by using a two-stream snow radiative transfer model TARTES (Libois et al., 2014). TARTES calculated the radiative transfer by using the specified snow properties such as LAI contents, SSA, snow grain size, snow density etc. The wavelength-dependent inherent optical properties (extinction coefficients σ_{ext} and absorption coefficients σ_{abs}) was calculated according to the complex refractive index of impurities and ice. The e-folding depth **for each wavelength** was calculated according to the normalized

actinic profile and the result in 305 nm was chosen to compare with previously published results at Summit. The calculated e-folding depth showed a gradually increasing trend towards longer wavelength, similar to the field observation at Summit (Galbavy et al., 2006) but in contrast with Noro and Takenaka. (2020). We have made this process more clearly by adding additional details in the revised manuscript.

Line 191-192: Mean values of the accumulation data were used to avoid the negative values induced by the wind blowing in the present study. However, Pham et al. (2019) demonstrated that the wind blowing dominates the removal of the photodegradable organic contaminants from the surface snow in Antarctica (Pham et al. 2019). Therefore, the effect of the wind blowing should be discussed (I do not mean that authors have to conduct the model study which includes the wind blowing.).

Response: We thank the reviewer for this comment. But we think these are two different scenarios. “Wind blowing” in our manuscript was referred to wind blowing snow that remove both snow and nitrate in it, when such thing occurs, both snow nitrate and its products leaves, and it should have null effects on the leftover. Since here are only two weeks among 52 weeks that displayed slightly negative accumulation rate, we think the impact of using average value should be minor.

Line 238: Jarvis et al. (2009) reported the surface snow $\delta^{15}\text{N}(\text{NO}_3^-)$ only for 5 months (March to July). How did you obtain the annual data? In addition, if you have each data point of Jarvis et al. (2009), please indicate in the same manner as observation (plots and lines) in Fig. 1.

Response: Jarvis et al. (2009) also reported seasonal mean $\delta^{15}\text{N}(\text{NO}_3^-)$ of surface snow samples covering two years as listed in Table 2 of the that paper. These were what we used to get the annual average as well as seasonal variations that used to constrain the model for sensitivity test.

Line 274-275: please add citations for the wavelength dependent of ε_p .

Response: Thanks for pointing this, we have added Berhanu et al. (2014).

Line 274-275: Does this sentence means that the wavelength change in season affects the ε_p , resulting in the peak of the FP ($\delta^{15}\text{N}$) in mid-summer? In this case, please show the data for the wavelength change.

Response: Yes, as modeled (Frey et al., 2009) and experimental determined by Berhanu et al. (2014), ε_p is sensitive to wavelength. This is why in Figure 3a the calculated ε_p varies seasonally from -60 to -90 per mil as the spectra reaching surface differs at different time.

Fig. 2: Please explain why the FD ($\delta^{15}\text{N}$) is changing.

Response: FD is a mixture of F_{pri} and the snow-sourced nitrate (FP). In the model, $\delta^{15}\text{N}$ of F_{pri} varied seasonally, and the $\delta^{15}\text{N}$ of FP also varied seasonally due to the variations in ε_p . In addition, the relative contributions of F_{pri} and FP to FD were also

different at different time. So $\delta^{15}\text{N}$ of FD varied with time. This has been discussed in the manuscript (the second paragraph of section 3.2).

In regard to evaporation/volatilization: The effect of evaporation was neglected in the present study. Shi et al. (2019) demonstrated that 38% of nitrate was lost from the snow sample at -4°C for 14–16 days (Shi et al. 2019). Moreover, the temperature of the surface snow is closed to 0°C in the daytime in summer in the Antarctic coastal site (Noro et al. 2020). Thus, the potential impacts of evaporation should be discussed in the present study.

Response: We thank the reviewer for this comment. But we don't think evaporation is significant. The Shi et al. 2019 experiment was not a fair design. They collected Dome A snow and put it in an open-door room at Zhongshan Station (a coastal site). Dome A snow is of much higher concentrations than in the coast (500 ppb at Dome A while 30 ppb at the coast, Shi et al., 2015), a re-equilibrium between snow and the overlying air will be established which artificially enhances the physical release. In addition, Erbland et al. 2013 suggested that the apparent ε_p became closer to zero at higher snow accumulation rate site is due to enhanced role of evaporation. However, this conclusion is questionable as the derived apparent ε_p is also influenced by snow nitrate that has not experienced photolysis, the higher of this fraction (at high accumulation rate site), the closer ε_p to zero.

What is more, in both evaporation experiments (including Erbland et al. (2013)), when mass loss is significant at higher temperature, the fractionation factor is very small. At Summit, even using a 25 % mass loss (the maximum loss fraction, given by Dibb et al. (2007)), and a ε_p of 3.6 ‰ (Erbland et al., 2013), only a 1 per mil difference in $\delta^{15}\text{N}$ can be induced.

In regard to the positioning of the model compared to the previous studies: The model studies have been reported, related to the post-depositional process of nitrate in Greenland (e.g. Zatzko et al. 2016). The advantages and the disadvantages of the models proposed in the previous studies and the present study should be described to demonstrate the positioning of the present study as a paragraph in the Introduction or as a section in the Results and discussion.

Response: Thank you. The Zatzko et al. (2016) study using a global 3-D chemical transport model (GEOS-Chem model). But the model treated snowpack as a whole and didn't specify the behaviors of nitrate at different depths in the photic zone, and can't distinguish seasonal differences. In addition, it didn't incorporate isotope fractionation associated with photolysis, but instead using a fixed fractionation constant and a Rayleigh fractionation model to calculate the changes in isotope with mass loss. While the TRANSITS model is a layer specific model and treats the reaction and recycle of nitrate step-by step, and it can predict the changes of isotopes in each defined layer (e.g., the seasonal changes). We have added this briefly in the introduction.

Technical corrections

Line 20 and any other pars: Space is not needed before “%” and “‰”.

Response: Thanks for this comment, but according to IUPAC recommendation, the dimension quality of physical quantities such as “%” and “‰” should be treated as units, thus a space is necessary when writing “%” and “‰” after a number.

Line 32 and many other parts: “Minus” should not be indicated as “-” but “–”.

Response: Thank you and we have revised in the manuscript accordingly.

Line 110: $J(\text{NO}_2) \rightarrow J_{(\text{NO}_2)}$

Response: Revised accordingly.

*Line 215: *won't* → *will not**

Response: Revised accordingly.

*Line 279: *ware null* → *were negligible**

Response: Revised accordingly.

Fig. 3: Please spell out F_{pri} in the caption.

Response: Did you mean in Figure 2? We have spelled it out.

Fig. 3: Remove the frame border of the legend.

Response: Revised accordingly.

Reference

- Berhanu, T. A., Meusinger, C., Erbland, J., Jost, R., Bhattacharya, S., Johnson, M. S., & Savarino, J.: Laboratory study of nitrate photolysis in Antarctic snow. II. Isotopic effects and wavelength dependence, *J. Chem. Phys.*, 140, 244306, <https://doi.org/10.1063/1.4882899>, 2014.
- Dibb, J. E., Whitlow, S. I., & Arsenault, M.: Seasonal variations in the soluble ion content of snow at Summit, Greenland: Constraints from three years of daily surface snow samples, *Atmos. Environ.*, 41, 5007-5019, <https://doi.org/10.1016/j.atmosenv.2006.12.010>, 2007.
- Erbland, J., Vicars, W., Savarino, J., Morin, S., Frey, M., et al.: Air–snow transfer of nitrate on the East Antarctic Plateau-Part 1: Isotopic evidence for a photolytically driven dynamic equilibrium in summer, *Atmos. Chem. Phys.*, 13, 6403-6419, <https://doi.org/10.5194/acp-13-6403-2013>, 2013.
- Frey, M. M., Savarino, J., Morin, S., Erbland, J., & Martins, J.: Photolysis imprint in the nitrate stable isotope signal in snow and atmosphere of East Antarctica and implications for reactive nitrogen cycling, *Atmos. Chem. Phys.*, 9, 8681-8696, <https://doi.org/10.5194/acp-9-8681-2009>, 2009.
- Galbavy, E. S., Anastasio, C., Lefer, B. L., & Hall, S. R.: Light penetration in the snowpack at Summit, Greenland: Part 2: Nitrate photolysis, *Atmos. Environ.*, 41, 5091-5100, <https://doi.org/10.1016/j.atmosenv.2006.01.066>
- Jarvis, J. C., Hastings, M. G., Steig, E. J., & Kunasek, S. A.: Isotopic ratios in gas-phase HNO_3 and snow nitrate at Summit, Greenland, *J. Geophys. Res. Atmos.*, 114, D17301,

<https://doi.org/10.1029/2009JD012134>, 2009.

Libois, Q., Picard, G., France, J., Arnaud, L., Dumont, M., Carmagnola, C., & King, M.: Influence of grain shape on light penetration in snow, *The Cryosphere*, 7, 1803-1818, <https://doi.org/10.5194/tc-7-1803-2013>, 2013.

Shi, G., Chai, J., Zhu, Z., Hu, Z., Chen, Z., et al.: Isotope fractionation of nitrate during volatilization in snow: a field investigation in Antarctica, *Geophys. Res. Lett.*, 46, 3287-3297, <https://doi.org/10.1029/2019GL081968>, 2019.

Zatko, M., Geng, L., Alexander, B., Sofen, E., & Klein, K.: The impact of snow nitrate photolysis on boundary layer chemistry and the recycling and redistribution of reactive nitrogen across Antarctica and Greenland in a global chemical transport model, *Atmos. Chem. Phys.*, 16, 2819-2842, <https://doi.org/10.5194/acp-16-2819-2016>, 2016.