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Impacts of the photo-driven post-depositional processing on snow nitrate and its isotopes at Summit, Greenland: a model-based study

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Wavelength-dependent ε_p and its impact on δ^{15}N of FP.

In TRANSITS, the time step was set to one week. However, the actinic flux also fluctuates within one week. To calculate the integral effect in this time step, we first computed the actinic flux at different solar zenith angle (50 to 90 ° at Summit) by setting a constant TCO value form observation:

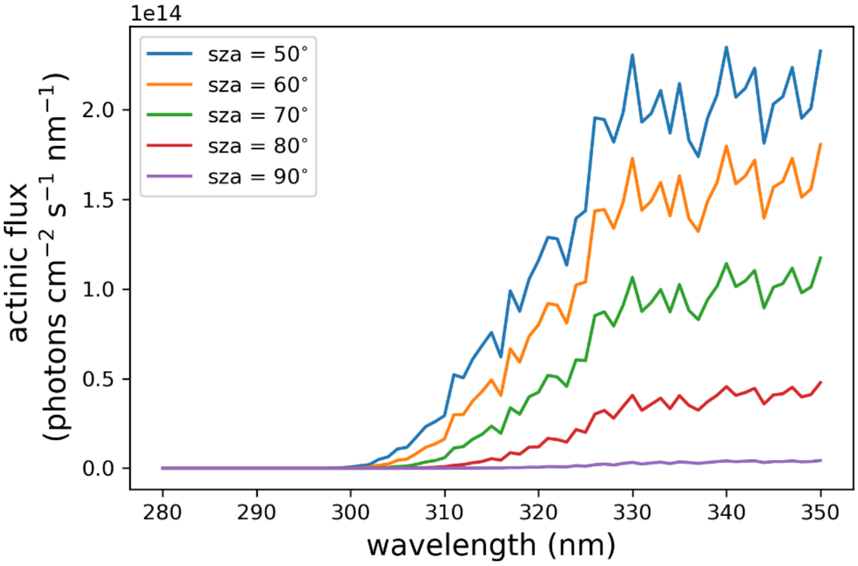

Figure S1. The computed actinic flux at snow surface under different solar zenith angle at Summit with a TCO of 300 DU as example.

Then we calculated the photolysis rate constant and fractionation factor at each solar zenith angle by:

$$^{14}J(sza) = \int_{280\,nm}^{350\,nm} \Phi(\lambda) \times {}^{14}\sigma_{NO_3^-}(\lambda) \times I(z,\lambda)\, d\lambda$$

$$^{15}J(sza) = \int_{280\,nm}^{350\,nm} \Phi(\lambda) \times {}^{15}\sigma_{NO_3^-}(\lambda) \times I(z,\lambda)\, d\lambda$$

$$\varepsilon_p = \frac{^{14}J(sza)}{^{15}J(sza)} - 1$$

The calculated results are shown below:

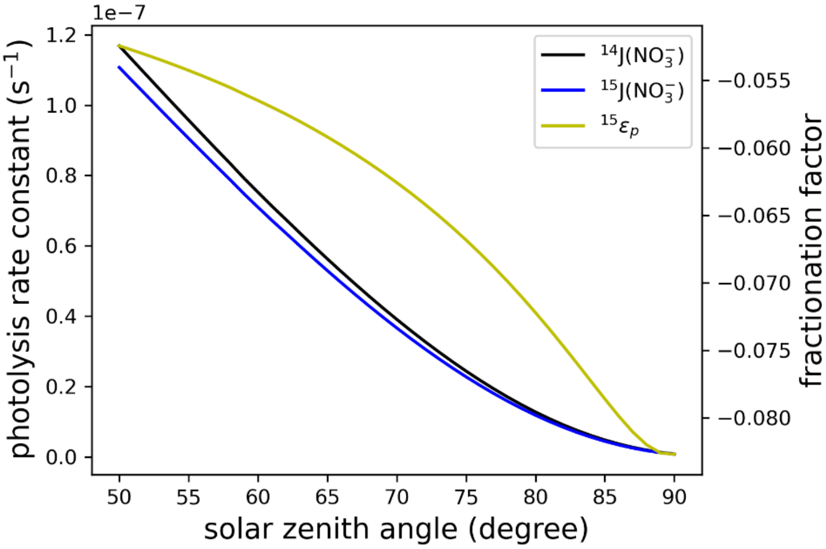

Figure S2. The computed nitrate photolysis rate constant and fractionation factor (ε_p) under different solar zenith angle at Summit with a TCO of 300 DU as example.

The overall effect in one week can be calculated as below:

$$\bar{J} = \frac{\int J(sza) \times t(sza) \times d(sza)}{\int t(sza) \times d(sza)}$$

and:

$$\bar{\varepsilon_p} = \frac{\int \varepsilon_p(sza) \times t(sza) \times d(sza)}{\int \varepsilon_p(sza) \times d(sza)}$$

As can be seen in the above equations, the photolysis fractionation factor (ε_p) is closely linked with the mean solar zenith angle in each week. Although the $\delta^{15}N(NO_3^-)$ of the bulk snowpack would also change owing to mass balance, its effect was muted by the large variation in ε_p, and $\delta^{15}N$ of FP was completed controlled by the variation in ε_p.

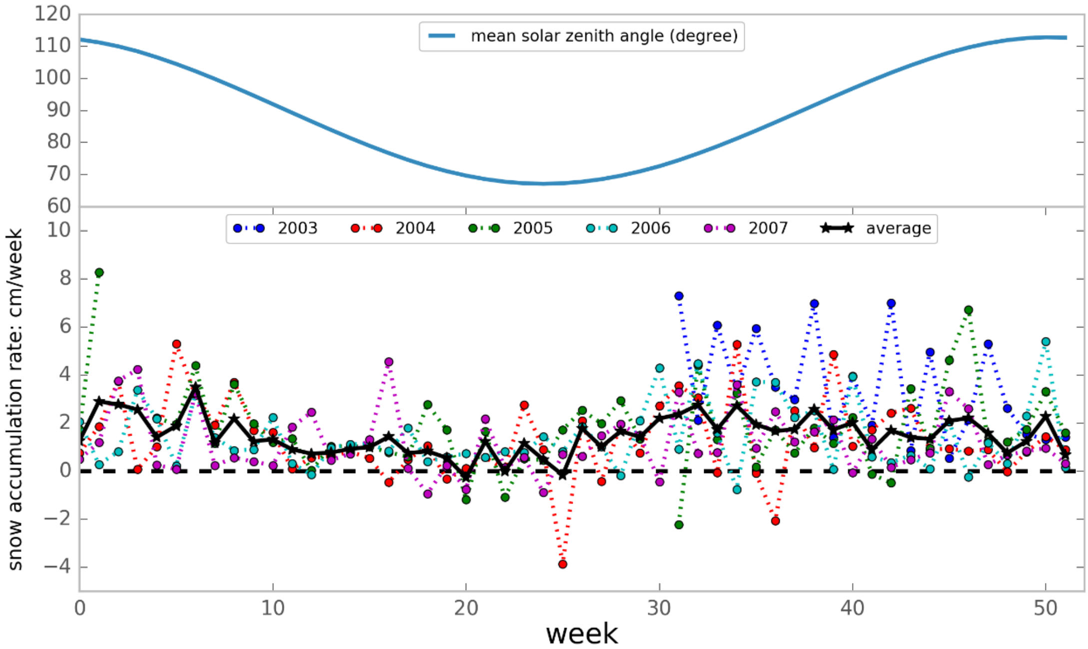

Figure S3. Solar zenith angle and the weekly snow accumulation observations at Summit, Greenland. The thick black curve represents the average over the 5 years, and others were the observations in each year from 2003-2007.

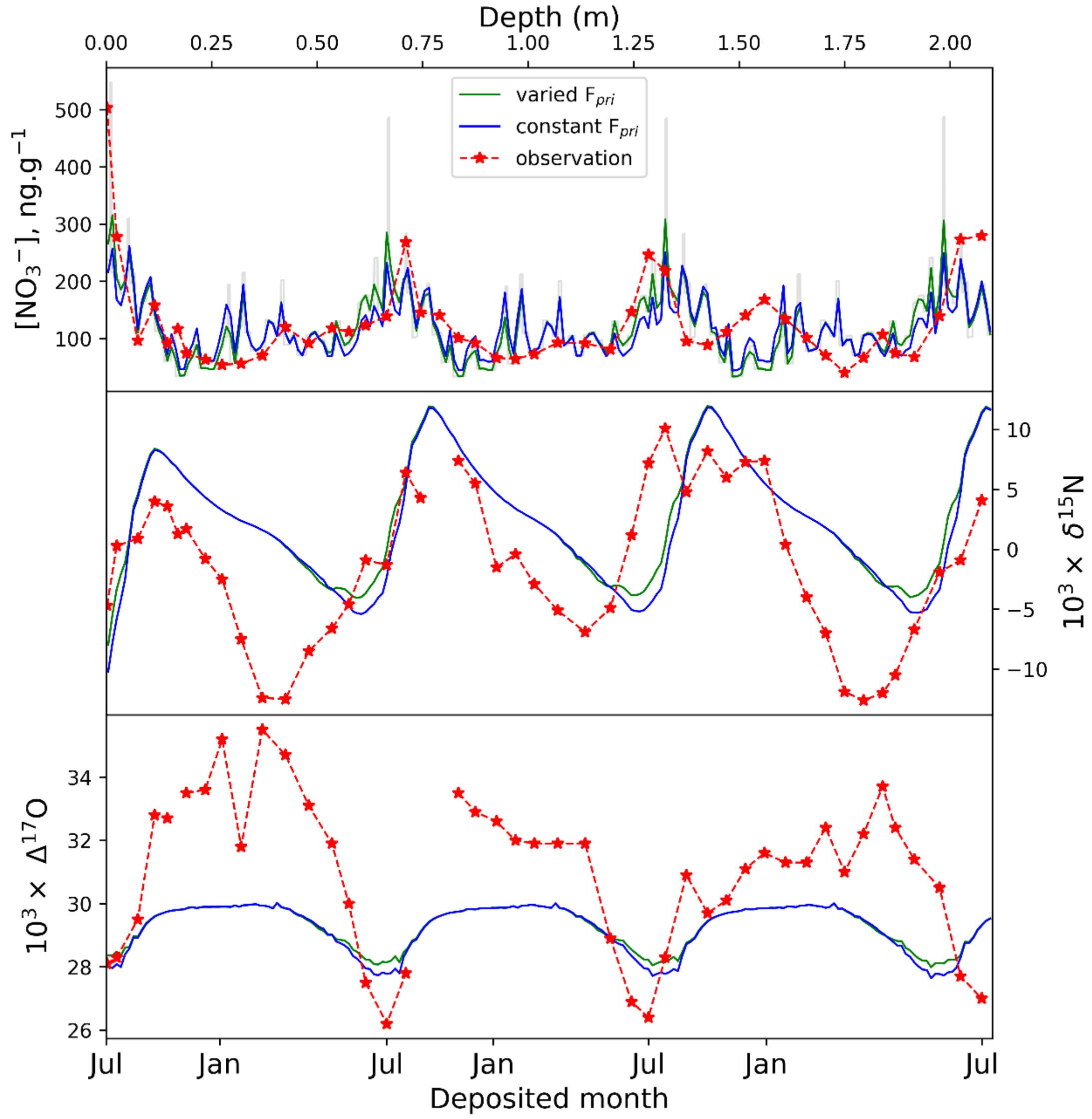

Figure S4. The impact of choosing different F_{pri} on modeled snow nitrate profiles. Varied F_{pri}: using seasonally-varied F_{pri} as seen in Figure 1a. Constant F_{pri}: using constant F_{pri} throughout the year. Note the annual amount of F_{pri} was kept same in these two scenarios.

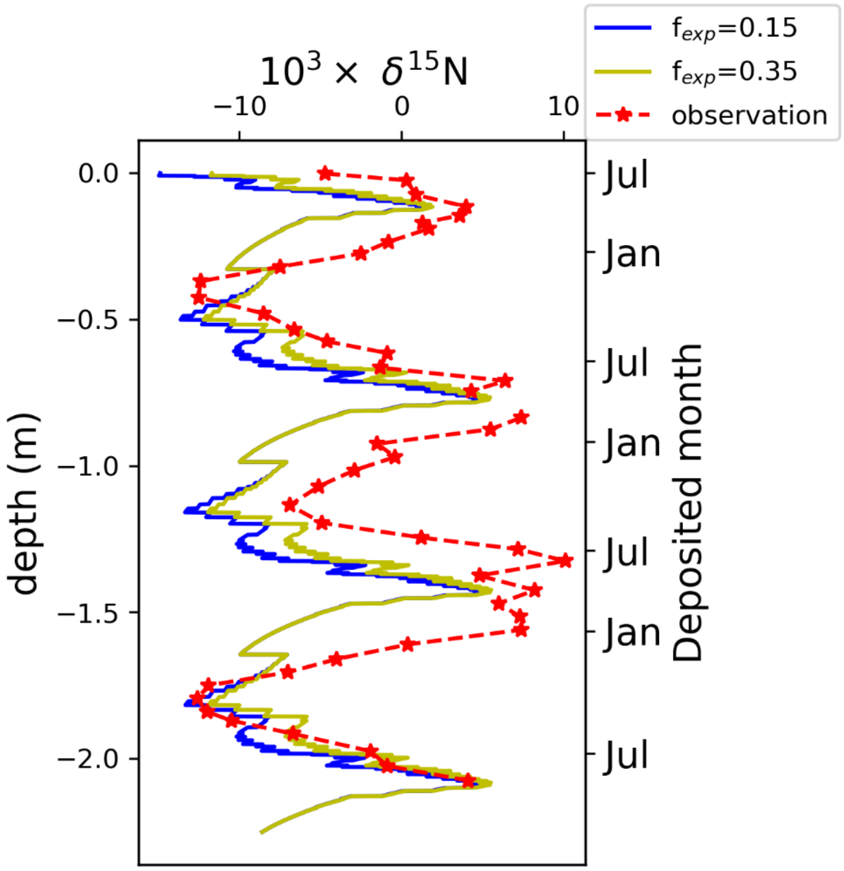

Figure S5. The modelled seasonal $\delta^{15}N(NO_3^-)$ with different f_{exp} (the export fraction).

Table S1

parameter	description	value	reference
h	Boundary layer height	156 m	Cohen et al., 2007
T	Temperature	-	NOAA observation
P	Pressure	-	NOAA observation
TCO	Total column ozone		NOAA observation
O_3	Ozone concentration		NOAA observation
BrO	BrO concentration	2 pptv	Fibiger et al., 2016
$OH/HO_2/RO_2$	$OH/HO_2/RO_2$ concentration	-	Sjostedt et al., 2007
f_{exp}	Nitrate export fraction	0.35	calculated
A	Snow accumulation rate	250 kg m^{-2} a^{-1}	Dibb et al., 2004
ρ	Snow density	0.35 g cm^{-3}	Geng et al., 2014
Φ	Quantum yield	0.002	[1]Scaled.
σ	Nitrate cross section	-	Berhanu et al., 2014
f_{cage}	Cage effect	0.15	Erbland et al., 2015
F_{pri}	Primary input nitrate	6.6×10^{-6} kgN m^2 a^{-1}	[2]Iizuka et al., 2018

ε_d	Nitrate deposition fractionation factor	+10 ‰	Erbland et al., 2013

*1. The quantum yield of nitrate photolysis was scaled by comparing the measured surface $j_0(NO_3^-)$ (Galbavy et al., 2007) with the modelled actinic flux at Summit.

*2. The magnitude of F_{pri} were estimated from the observed snowpack from Geng et al. (2014). The seasonal variation of F_{pri} were scaled according to Iizuka et al. (2018).