Investigating the Impact of Snowpack Photodenitrification on Polar Atmospheric Chemistry Using Results from a Snowpack Radiative Transfer Model

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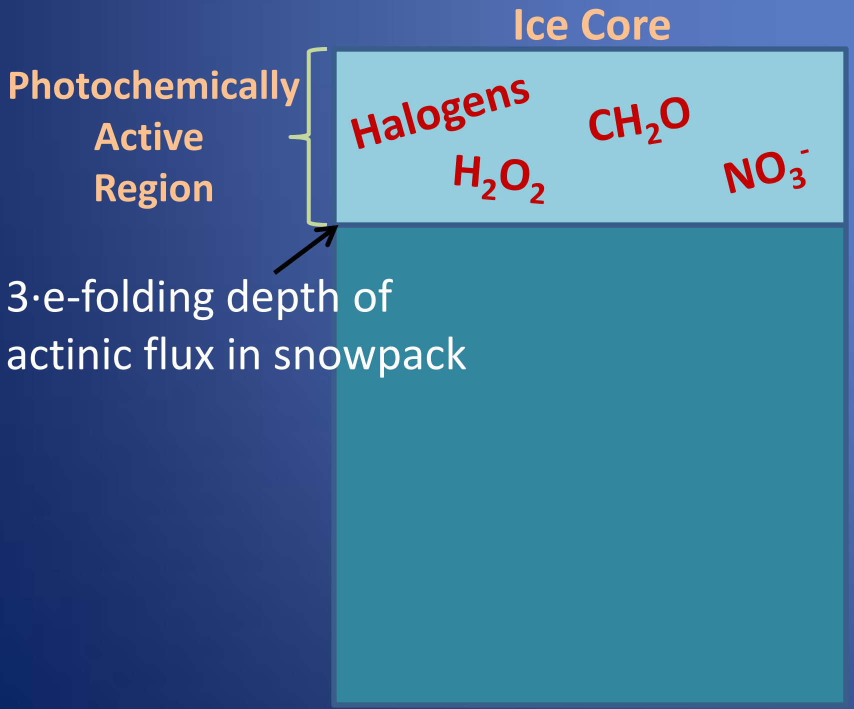
Snowpack Photochemistry

GOAL: Incorporate process-based study of snowpack photodenitrification into a global chemical transport model (GEOS-Chem) to determine spatial redistribution of nitrate

-collaborators using GEOS-Chem adjoint model to determine sources of nitrate to Antarctica



This Study

-Snowpack radiative transfer model with updated optical properties of ice in UV used

-Sensitivity of snowpack actinic flux profile to snowpack physical and optical properties explored

-τ_{NOx} in snowpack interstitial air against physical (τ_{escape}) and chemical ($\tau_{chemical}$) loss determined

-parameterization for actinic flux in snowpack developed for use in large scale models

Snowpack Radiative Transfer Model

Wolff et al. [2002]

Snowpack Radiative Transfer Model [*Grenfell,* 1991]

e-folding depth of actinic flux in snowpack of 3.7 cm at Neumayer

This Study

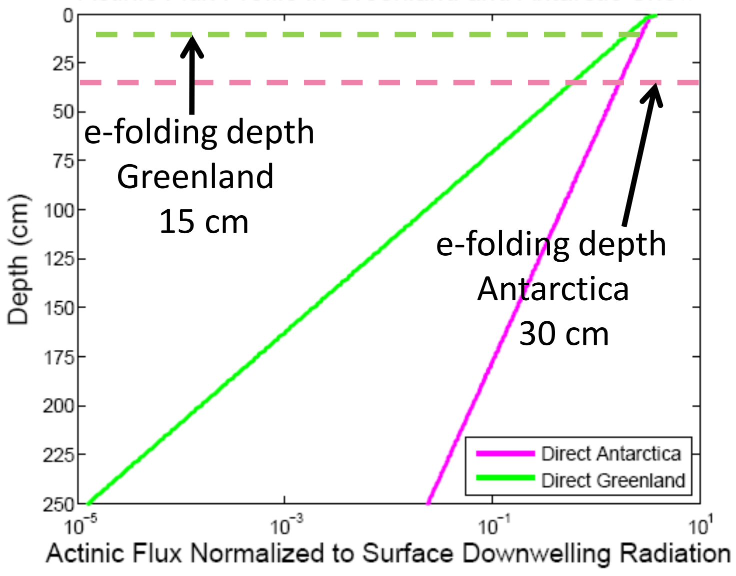
Snowpack Radiative Transfer Model [*Grenfell,* 1991]

-Updated optical properties of ice [*Warren and Brandt*, 2008]

-Inclusion of soot and dust as dominant absorbers

e-folding depth of actinic flux in snowpack of 30 cm at Neumayer

Profile of Actinic Flux in Snowpack



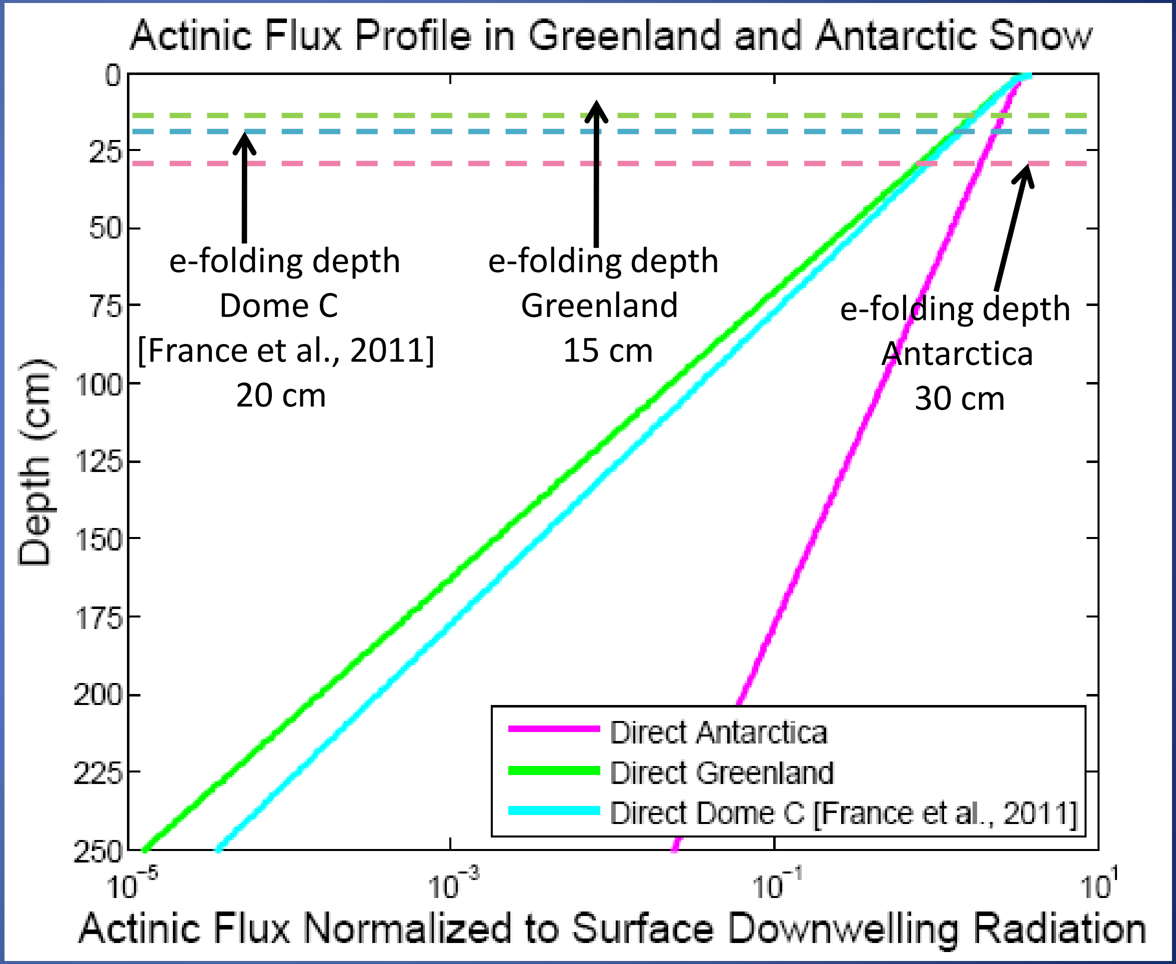
e-folding depth Greenland 15 cm

e-folding depth Antarctica 30 cm

Snowpack Radiative Transfer Model

Our parameterizations agree with recent e-folding depth observations from France et al. [2011] when same snow conditions used at Dome C

Soot concentrations observed by France et al. [2011] (~3 ppb) greater than concentrations in clear air sectors in Antarctica (0.3 ppb) [*Warren and Clarke*, 1990]



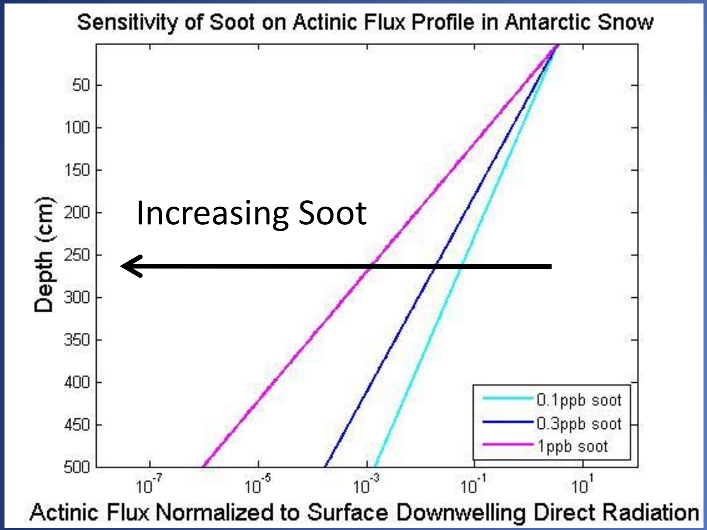
e-folding depth
Dome C
[France et al., 2011]
20 cm

e-folding depth
Greenland
15 cm

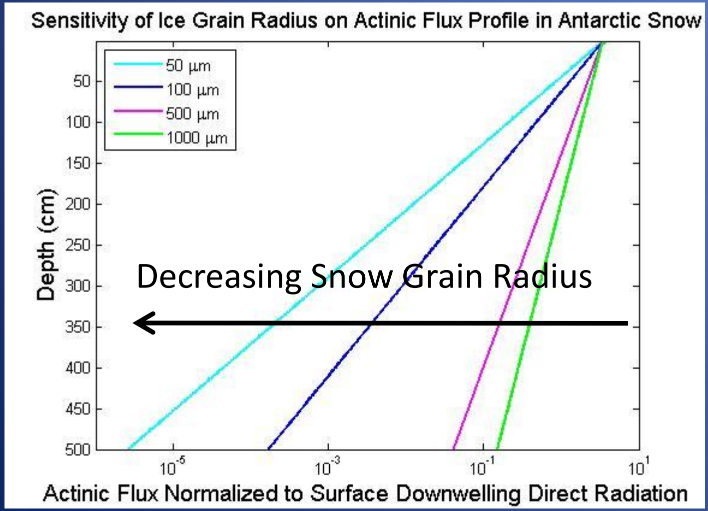
e-folding depth
Antarctica
30 cm

Actinic Flux Profiles in Snowpack

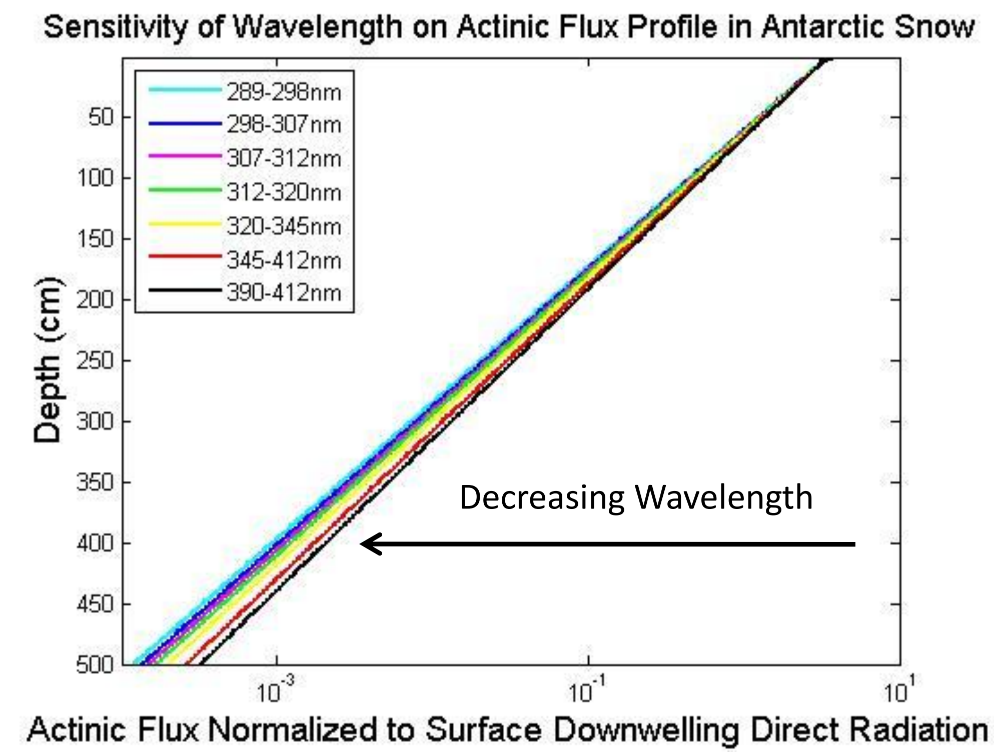
Diffuse vs. direct radiation, solar zenith angle, wavelength, effective snow grain radius [*Hansen and Travis*, 1974] , density, soot concentration and dust concentration


Sensitivity of Soot on Actinic Flux Profile in Antarctic Snow

Increasing Soot


Sensitivity of Ice Grain Radius on Actinic Flux Profile in Antarctic Snow

Decreasing Snow Grain Radius


Sensitivity of Wavelength on Actinic Flux Profile in Antarctic Snow

Decreasing Wavelength

Actinic flux in deep snowpack most influenced by soot concentration and effective snow grain radius

Will NO_x produced at a given depth in snowpack be released into atmosphere?

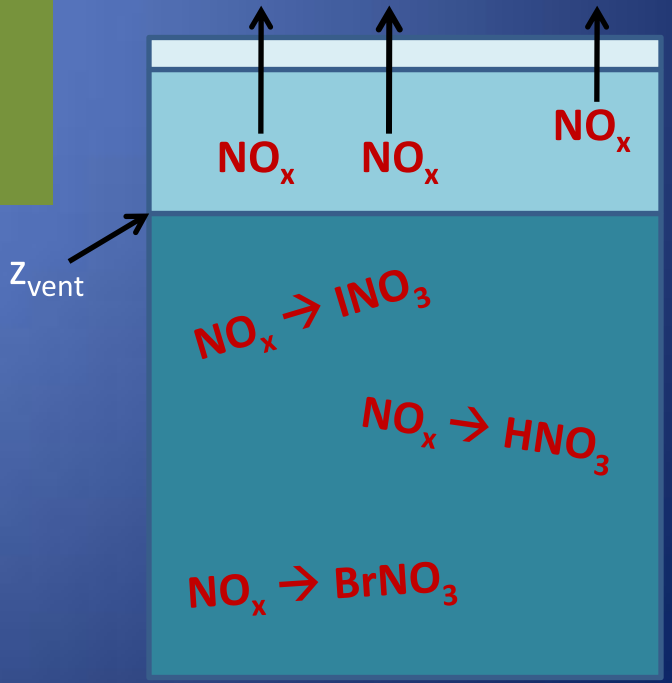
Determine ventilation depth by comparing the lifetime (τ) of NO_x against:

$$\left.\begin{array}{l}\text{Diffusion}\\ \text{Wind pumping}\end{array}\right\}\tau_{escape}$$

$$\text{Conversion to } HNO_3 \}\tau_{chemical}$$

Ventilation depth (z_{vent}): depth below which NO_x produced is unlikely to escape to atmosphere before reacting to form HNO_3.

NO_x Escape vs. Conversion

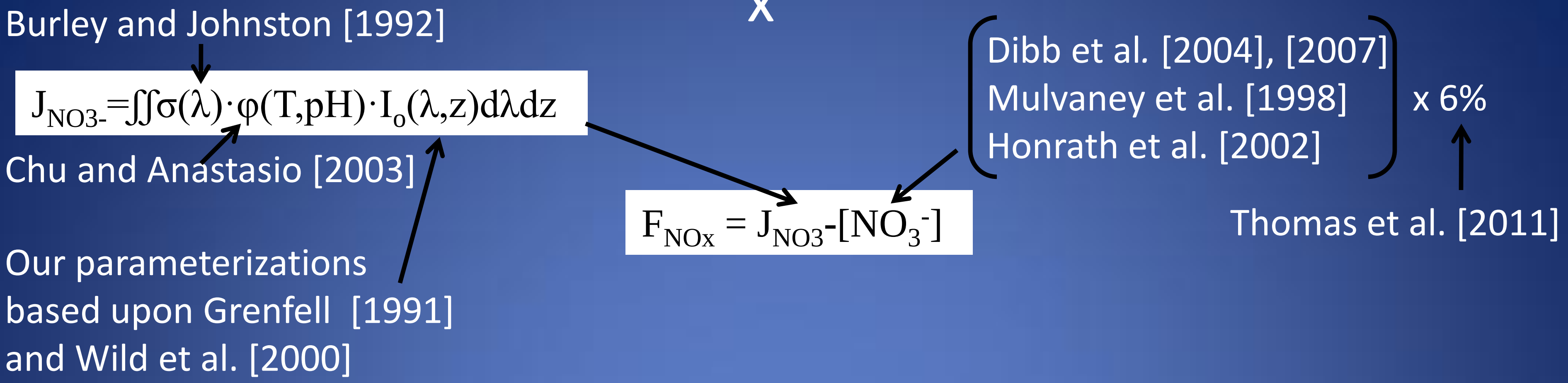


Conversion of NO_x into HNO_3, $BrNO_3$, and INO_3 in snowpack prevents NO_x ventilation to atmosphere.

$NO_2 + OH \rightarrow HNO_3$

$$\left.\begin{array}{l}NO_2 + BrO \rightarrow BrNO_3\\ NO_2 + IO \rightarrow INO_3\end{array}\right\}\xrightarrow{\text{hydrolysis}} HNO_3$$

NO_x Fluxes



Burley and Johnston [1992]

$J_{NO3^-}=\iint \sigma(\lambda)\cdot\varphi(T,pH)\cdot I_o(\lambda,z)\,d\lambda dz$

Chu and Anastasio [2003]

Our parameterizations
based upon Grenfell [1991]
and Wild et al. [2000]

$F_{NOx} = J_{NO3}\text{-}[NO_3^-]$

Dibb et al. [2004], [2007]
Mulvaney et al. [1998]
Honrath et al. [2002]

x 6%

Thomas et al. [2011]

NO_x Flux (molecules cm^{-2} s^{-1})			
Locations	**This Study**	**Observations**	**Reference**
Neumayer (coastal)	7.1×10^7-2.9×10^8	1.3-2.5×10^8	Jones et al., 2000 Jones et al., 2001
South Pole (continental)	1.0-1.8×10^8	2.2-3.8×10^8	Davis et al., 2004 Oncley et al., 2004
Halley (coastal)	9.2×10^7-3.7×10^8	1.7-3.8×10^8	Jones et al., 2007 Bauguitte et al., 2009
Summit (continental)	9.5×10^7-1.8×10^8	2.5×10^8	Honrath et al., 2002

NO_x fluxes calculated in this study are compared to NO_x flux observations at South Pole, Neumayer, Halley, and Summit

Snowpack Actinic Flux Parameterizations

For direct radiation at the surface:

$$\left[\frac{I_o(\lambda, z=0)}{\mathrm{F_{inc}}(\lambda_\downarrow)}\right]_{direct} = \left[\frac{0.577 + \mu_o}{0.577 \cdot \mu_o}\right] \cdot Corr(\mu_o)$$

correction factor because no exponential decay in top 2 cm

solar zenith angle

For direct radiation below z_{ref}:

2 cm

$$\left[\frac{I_o\left(\lambda, z \geq z_{ref}\right)}{\mathrm{F_{inc}}(\lambda_\downarrow)}\right]_{direct} = G\left(z_{ref}, \mu_o\right) \cdot e^{-0.60 \cdot c\varpi_{eff}^{\frac{1}{2}} \cdot Kext_{tot} \cdot (z - z_{ref})}$$

$$G\left(z_{ref}, \mu_o\right) = 3(0.577 + \mu_o) \cdot e^{-0.60 \cdot c\varpi_{eff}^{\frac{1}{2}} \cdot Kext_{tot} \cdot z_{ref}} \cdot Corr(\mu_o)$$

For diffuse radiation:

$$\left[\frac{I_o(\lambda,z)}{\mathrm{F_{inc}}(\lambda_\downarrow)}\right]_{diffuse} = 3.831 \cdot e^{-0.60 \cdot c\varpi_{eff}^{\frac{1}{2}} \cdot Kext_{tot} \cdot z}$$

Actinic Flux

$$I_{o_{combined}}(\lambda, z) = \left\{\left[\frac{I_o(\lambda, z)}{F_{inc}(\lambda_\downarrow)}\right]_{diffuse} \cdot (f_{dif}) + \left[\frac{I_o(\lambda, z)}{F_{inc}(\lambda_\downarrow)}\right]_{direct} \cdot (1 - f_{dif})\right\} \cdot [F_{inc}(\lambda_\downarrow)]_{tot}$$

Extinction coefficients and co- albedo for single scattering

$$Kext_{snow} = \frac{3\, Qext \cdot \rho_{snow}}{4\, r_e \cdot \rho_{ice}}$$

[soot] in snow

$$Kext_{soot} = \frac{\beta_{soot} \cdot L_{soot} \cdot \rho_{snow}}{c\varpi_{soot}}$$

[dust] in snow

$$Kext_{dust} = \frac{\beta_{dust} \cdot L_{dust} \cdot \rho_{dust}}{c\varpi_{dust}}$$

$$c\varpi_{eff} = \frac{[c\varpi_{snow} \cdot Kext_{snow} + c\varpi_{soot} \cdot Kext_{soot} + c\varpi_{dust} \cdot Kext_{dust}]}{Kext_{tot}}$$

$$Kext_{tot} = [Kext_{snow} + Kext_{soot} + Kext_{dust}]$$

The equation for actinic flux in snowpack can be integrated over wavelength and depth

Conclusions and Future Research

- The calculated e-folding depth of actinic flux is ~30 cm in Antarctic snowpack and ~15 cm in Greenland snowpack.

- Ventilation depths are most sensitive to sastrugi dimensions, [BrO], and [IO]. In absence of a ventilation depth, use 3·e-folding depth. (effect z_{vent} = 90 cm in Antarctica and 45 cm in Greenland)

- We calculate a wide range of NO_x fluxes based upon variations in ventilation depths and [NO_3^-] in snow which are in agreement with observations in Antarctica and Greenland

- We have developed simple and broadly applicable equations to calculate depth dependent actinic flux in snowpack. These equations can be incorporated into global models and adjusted to represent all snowpack types by varying relevant parameters (e.g. soot, dust, snow density).

- Our next step is to incorporate our methods and results into the GEOS-Chem global chemical transport model to investigate the impacts on polar nitrogen and oxidant budgets.

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