Development of a mechanistic representation of snow-atmosphere exchange of reactive compounds for implementation in large-scale models

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NSF’s Arctic System Science Program:
Collaborative research: A synthesis of existing and new observations of air-snowpack exchanges to assess the Arctic tropospheric ozone budget

develop, implement, and evaluate a representation of the key processes governing impacts of surface exchange over snow on tropospheric ozone simulated by chemistry-climate models.

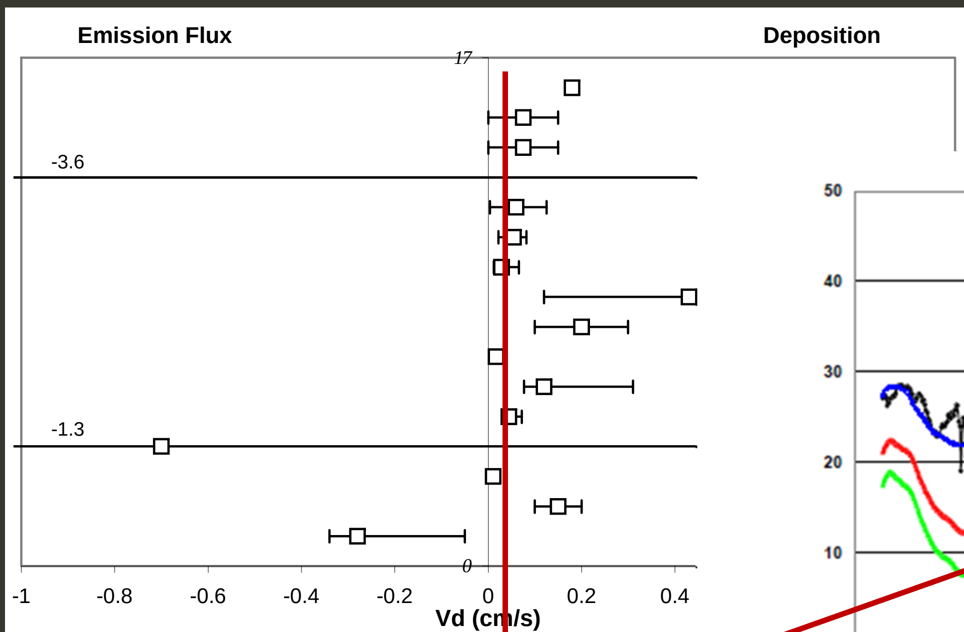
- Review and synthesis results from prior field studies relevant to O_3 and NO_x exchange fluxes
- New field studies to fill key knowledge gaps, especially those related to the dependence of vertical O_3 fluxes on height above snow and sub-snow surface type
- Incorporate parameterizations of snowpack and sub-snow processes into a single column model (SCM) version of a chemistry-climate
- Evaluation of model
- Provide a first estimate of the total impact of current snow- and ice-cover upon tropospheric O_3 in subarctic and arctic and subarctic regions.

Modeling objectives

- To develop & evaluate a process-based representation of snowpack O_3 and NO_x exchange for implementation in global chemistry-climate models
- To determine key O_3 and NO_x chemical reactions in the snowpack
- To better describe the connections between air-snow O_3 and NO_x exchange on tropospheric O_3 budget
- To assess the potential future consequences of climate change on cryosphere-atmosphere exchange of NO_x and O_3 and high-latitude photochemistry
- Feedbacks?
  Coupling the representation of cryosphere-atmosphere exchange to climate model simulations of cryosphere physical properties

Motivation

Observed V_{dO3} (F/C) over Snow



~0.05 cm s^{-1}

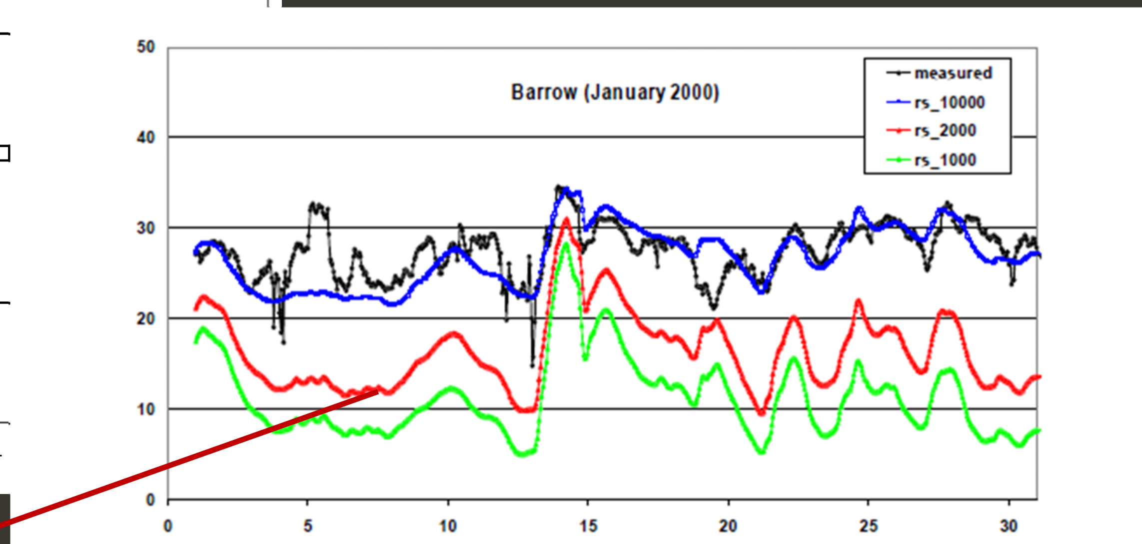


Fig. 7. Comparison of hourly ozone data (in ppbv) at Barrow for January 2000 in comparison with MATCH simulations at three different ozone-to-snow uptake resistances (Rs=10 000 [blue], 2000 [red] and 1000 m s^{-1} [green]).

Helmig, D., **L. Ganzeveld**, T. Butler, and S. Oltmans, The role of ozone atmosphere-snow gas exchange on polar, boundary-layer tropospheric ozone – a review and sensitivity analysis, *Atmospheric Chemistry and Physics*, 7, 15-30, 2007.

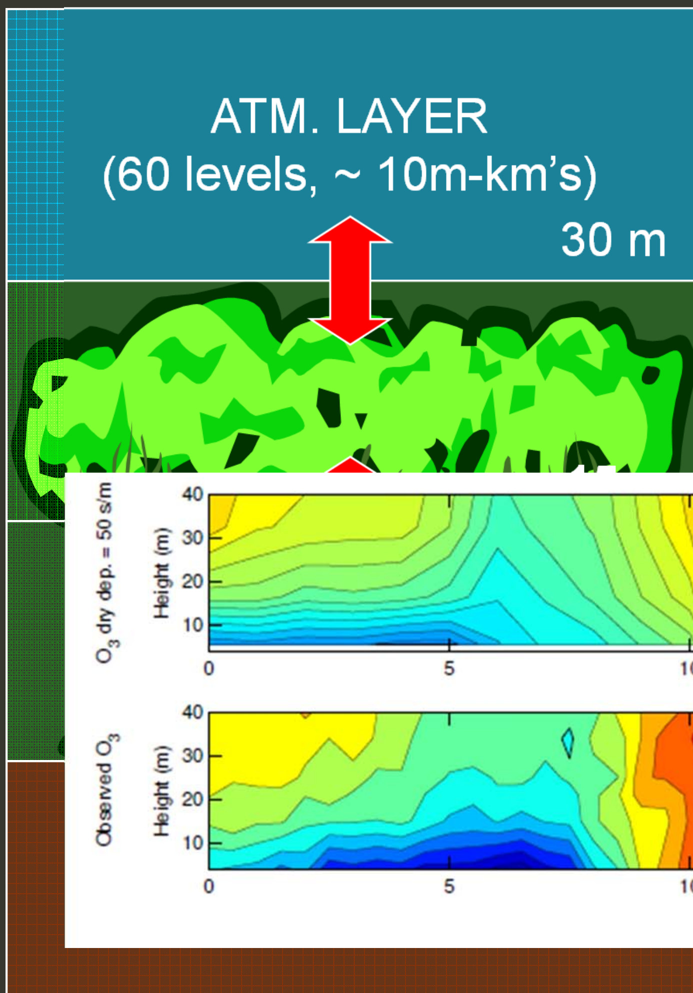
Model description

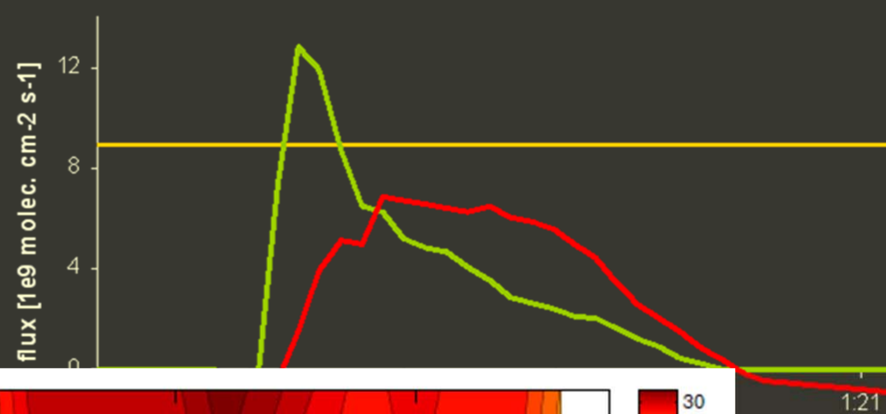
- Single-Column Model (SCM)
  - 1D model + time dimension
  - Based on
    - ECHAM4 (General Circulation Model) & RACMO (Regional Atmospheric Climate Model) physics
    - ECHAM4 atmospheric chemistry scheme considering natural and anthropogenic emissions, gas-phase and cloud water chemistry, turbulent & convective tracer transport, wet & dry deposition
  - Uses
    - ECMWF (European Center for Medium-Range Weather Forecast) Re-analysis Data
      - For considering role of advection of u, v, T, q and LWC
    - Free troposphere initial concentrations/observed concentrations
      - For considering role of advection of long-lived tracers

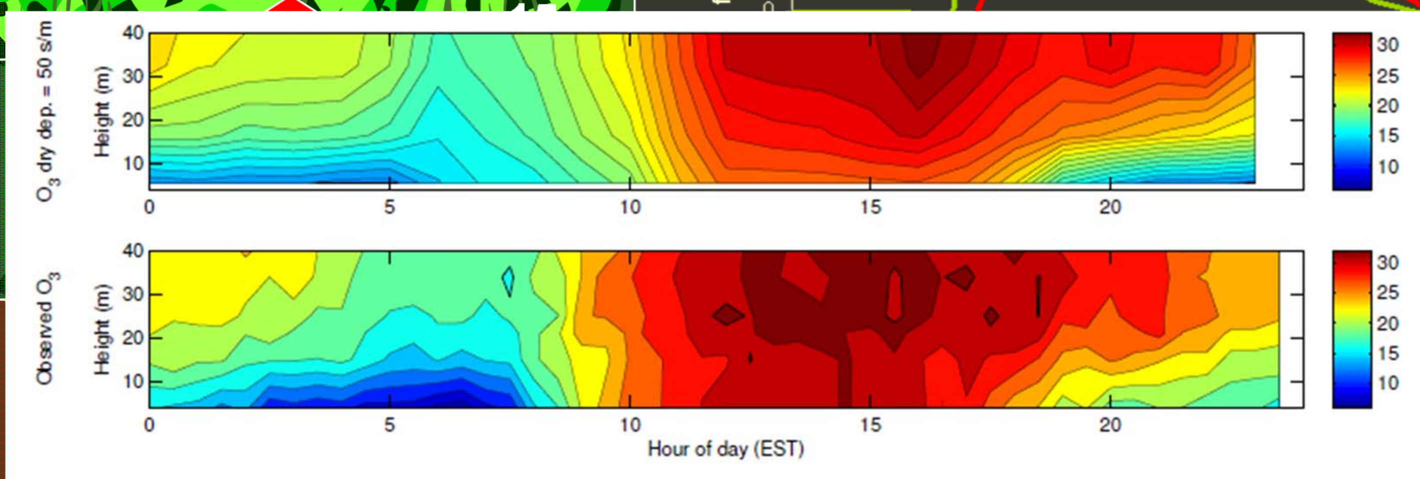
Model schema

Canopy layout well tested…

Should work as well for snowpack



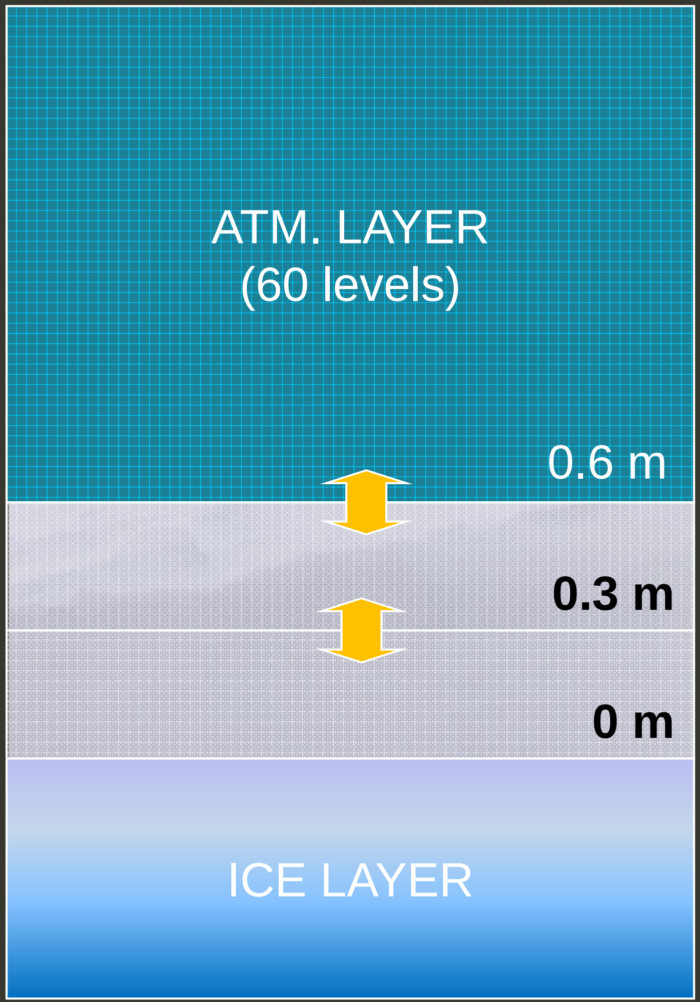




mixing

pts
45 m
ayer

Model schema: Advantages / Disadvantages



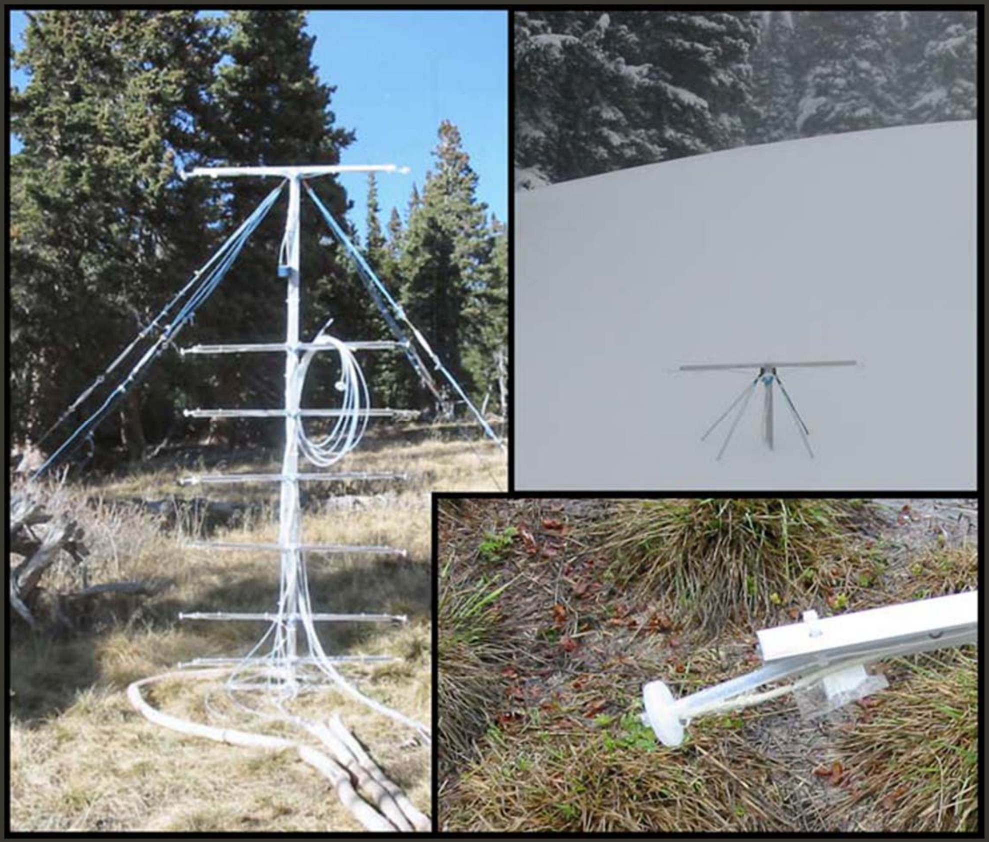
Advantages of 2-layer representation

- Transport between each layer can be solved analytically
- Slightly easier to debug, less code
- SPEED, much faster
  - Huge benefit when module is integrated into a 3D model

Disadvantages

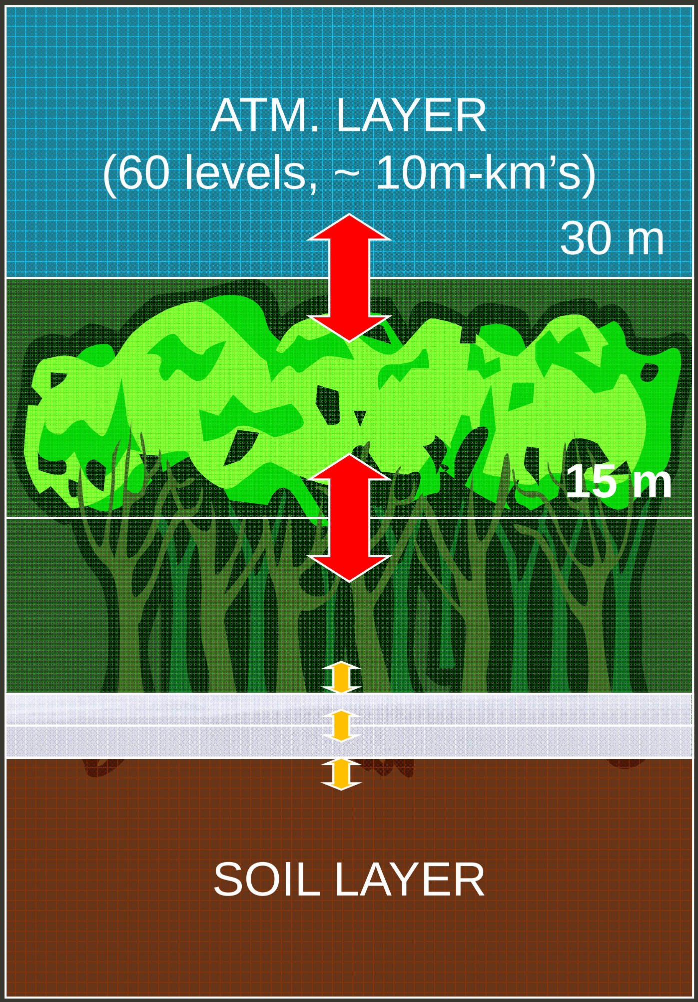
- Too simple?
- Lacks resolution
  - May miss out capturing some processes that can only be observed at higher resolution
  - Is it necessary?

Model schema

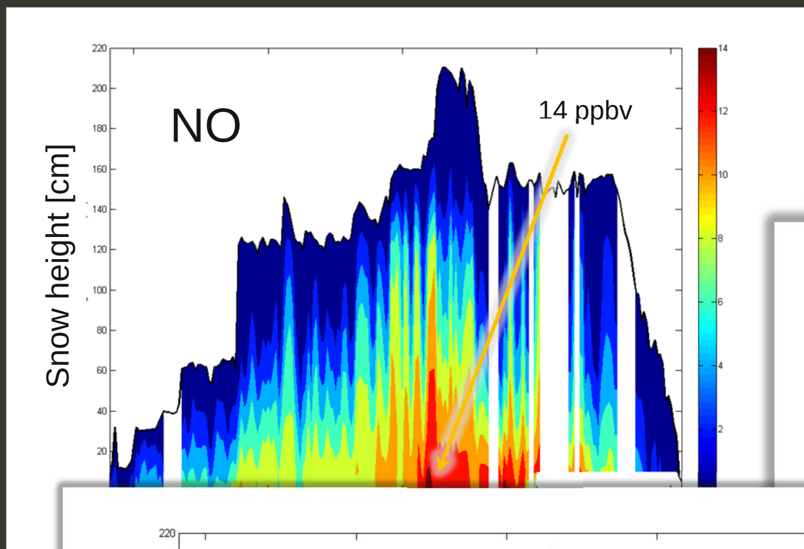


An automated system for continuous measurements of trace gas fluxes through snow: an evaluation of the gas diffusion method at a subalpine forest site, Niwot Ridge, Colorado, Brian Seok, et al., Biogeochemistry, 2009

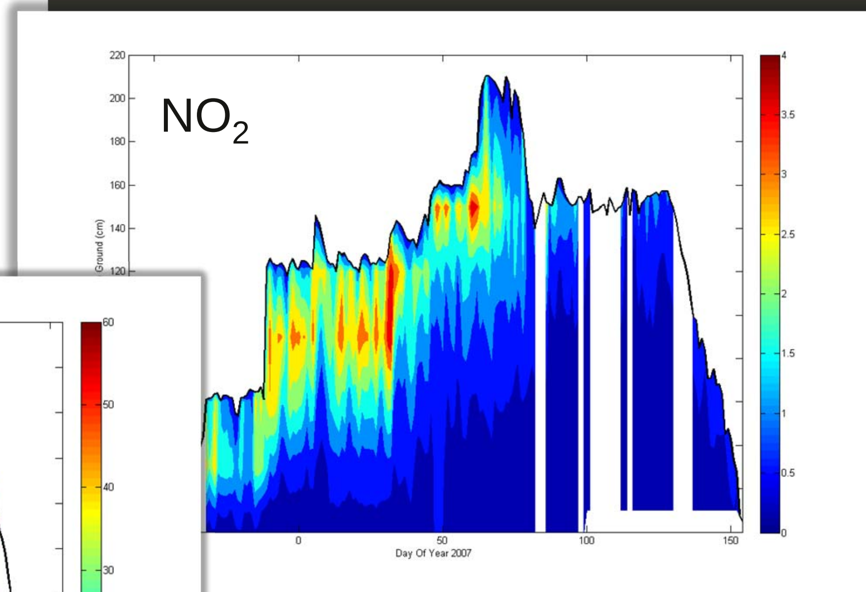
Snow cover under the canopy

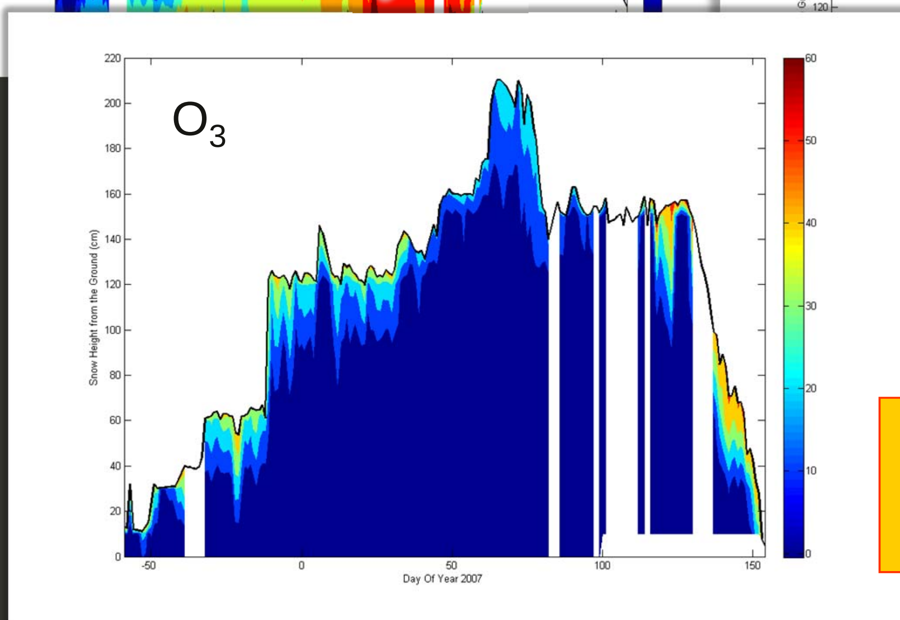


Model: snow under the canopy



Fluxes and chemistry of nitrogen oxides in the Niwot Ridge, Colorado, snowpack, Detlev Helmig, Brian Seok, Mark W Williams, Jacques Hueber, Robert Sanford, Biogeochemistry (2009)

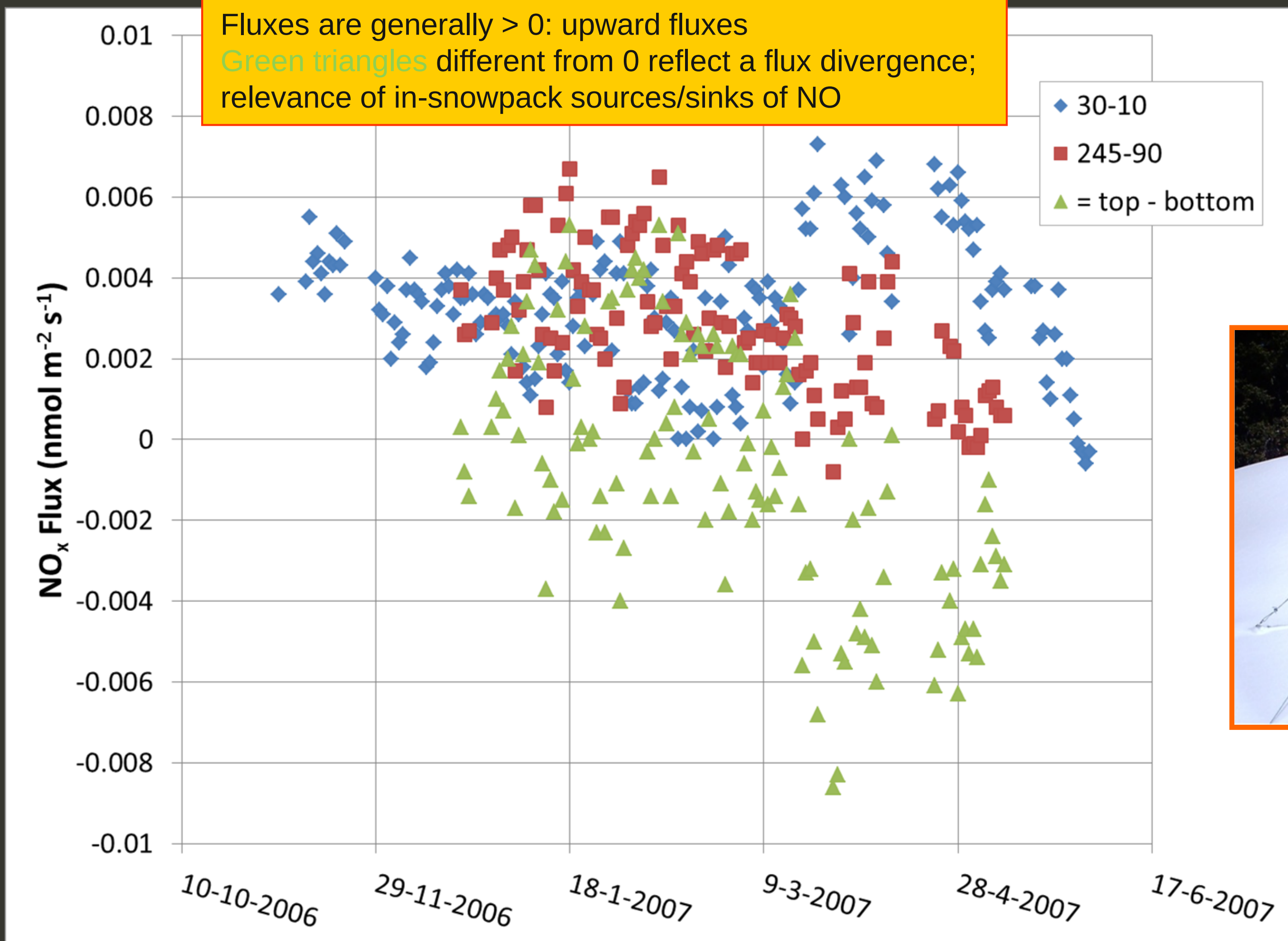




- How much of the soil NO_x and CO_2 is effectively emitted into the canopy trunkspace/atmosphere?
- How does the below canopy snow-cover affect (O_3) dry deposition?

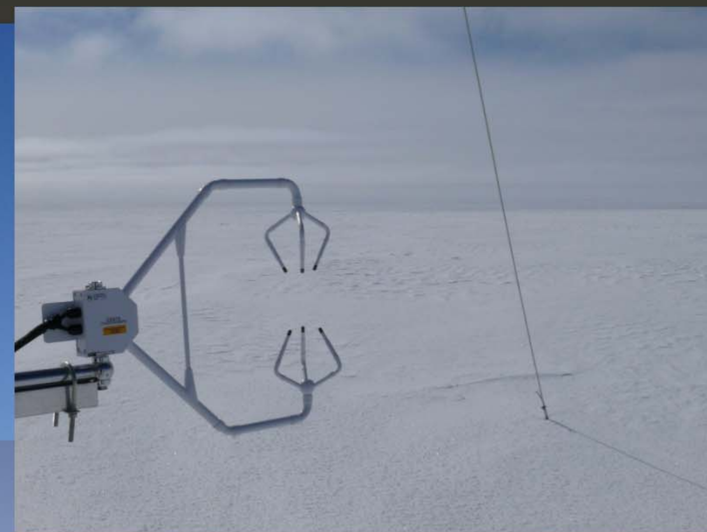
Model: snow under the canopy

Fluxes are generally > 0: upward fluxes
Green triangles different from 0 reflect a flux divergence; relevance of in-snowpack sources/sinks of NO



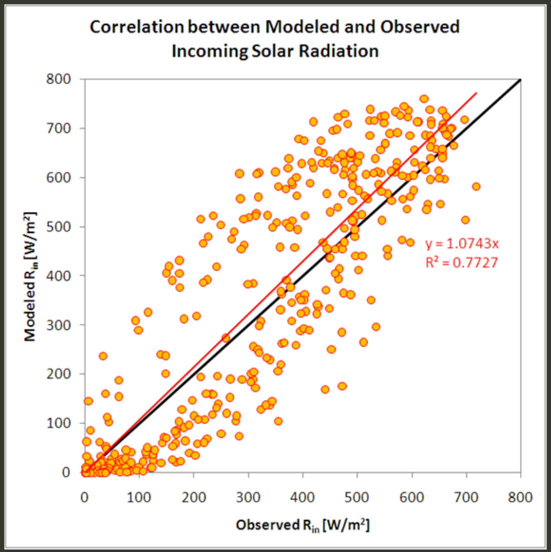
An automated system for continuous measurements of trace gas fluxes through snow: an evaluation of the gas diffusion method at a subalpine forest site, Niwot Ridge, Colorado, Brian Seok, et al., Biogeochemistry, 2009

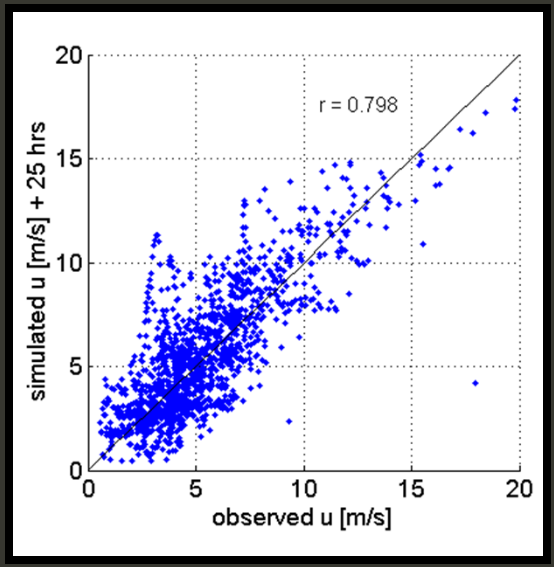
2-years of NO_x and O_3 concentration and flux measurements at Summit

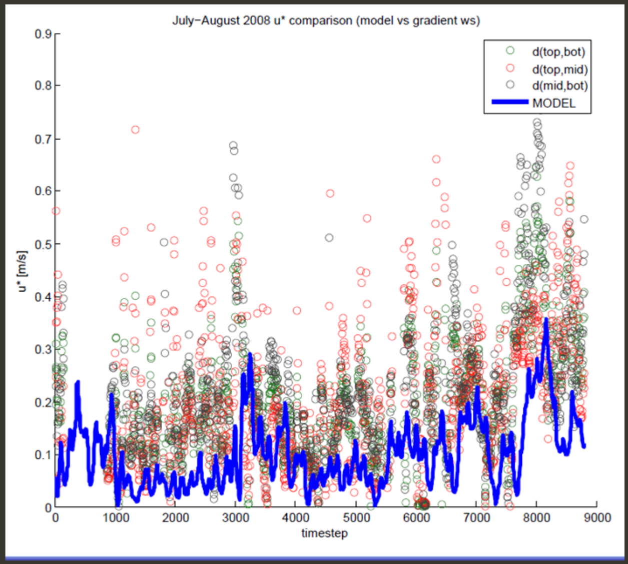


Micro/BL meteorology validation

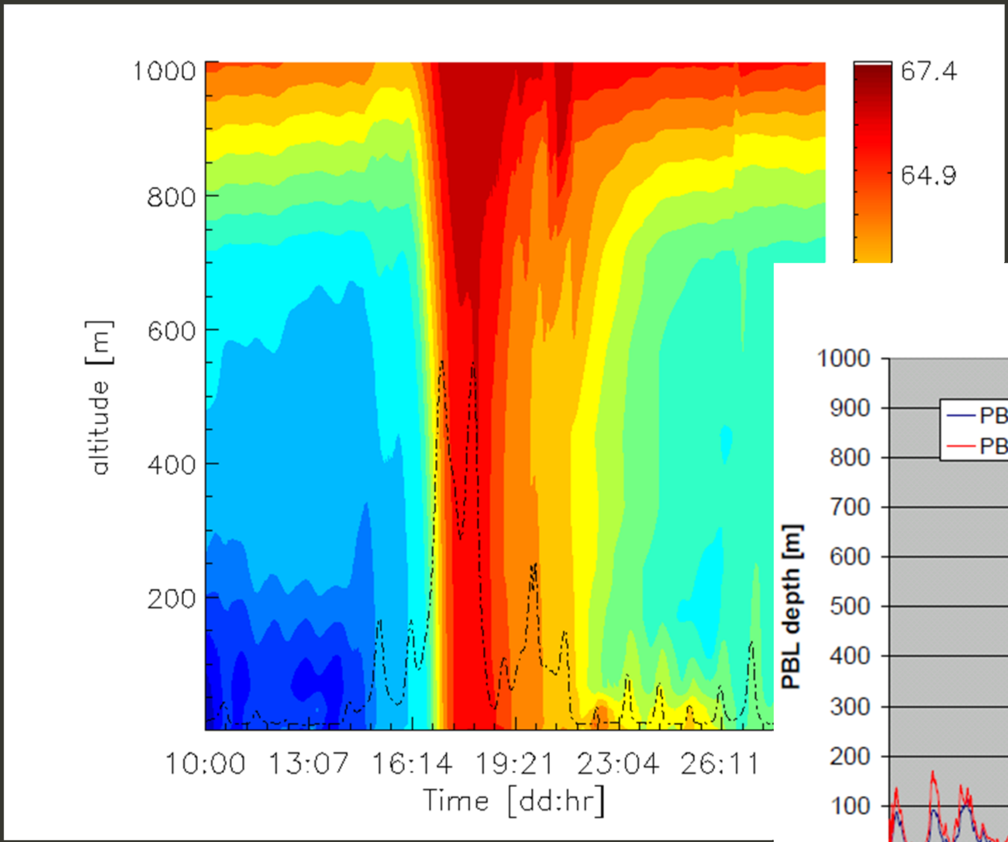
- To properly simulate concentrations and fluxes, the micrometeorology needs to be correct

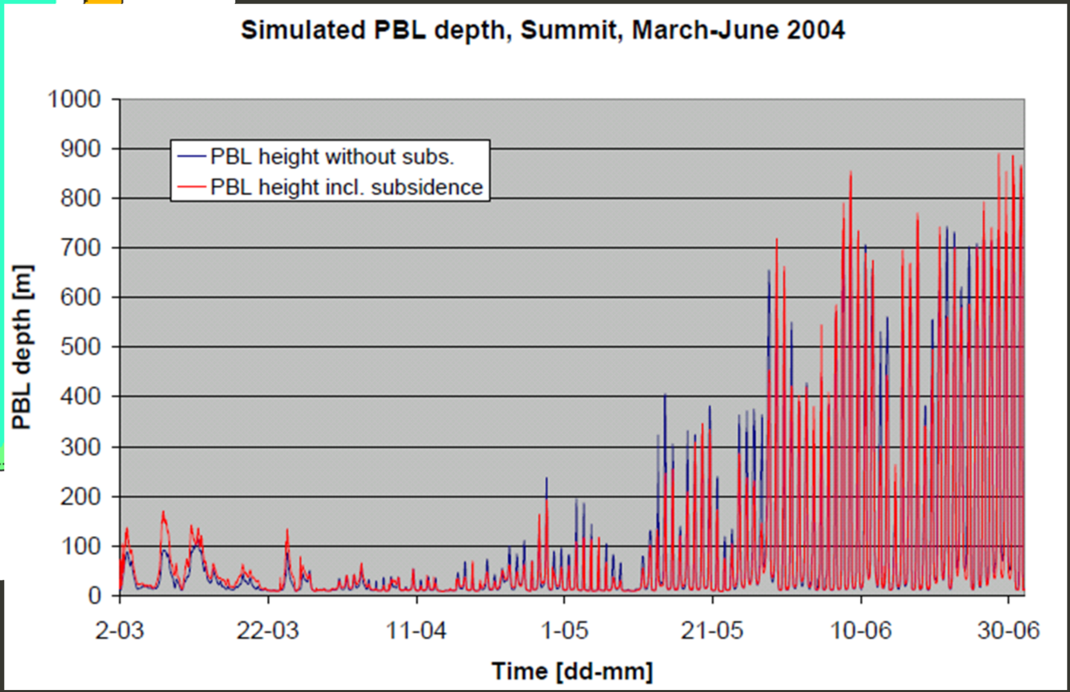






Micro/BL meteorology validation





- To be evaluated by comparison with SODAR observations at Summit

Chemistry: Model initialization

- Initial [NO_3^-] and J_{NO3_NOx} taken from Honrath et al 2002
  - Next slide
- Snow surface microtopography (for windpumping)
  - Relief height, length, width (“guessed”): 0.23, 2.2, 1.3 m (Liao/Tan 2008, Antarctica)
  - Jennie’s Thomas estimates: 0.015, 0.03, 0.03 m
- Ice pack temperature, 263 K (from meas.)
- Snow density (bulk), 0.3 g/cm^3 (from meas.)
  - Grain diameter, permeability, etc. calculated based on relationship with density (Domine et al 2008)
- Albedo, 0.89 (from meas.)

Chemistry:
Nitrate concentrations in snow

$[NO_3^-]$ for our model	Unit	$J_{NO3\text{-}NOx}$	Unit	LOC	REF	NOTE
2.6E+20	molec/m²	N/A	1/s	Svalbard	Beine et al 2003	total conc
6.9E+20	``	``	``	Dome C, Antarctica	Frey et al 2009	range min
9.4E+20	``	``	``	``	``	range max
1.8E+21	``	8.3E-07	``	Summit	Jacobi et al 2007	conc value from Dibbs et al 1998
3.6E+20	``	N/A	``	``	Dibbs et al 1998	average inventory value
7.2E+20	``	``	``	``	``	average snow surf conc (snow surf = top 50 cm)
7.2E+20	``	N/A	``	``	Honrath et al 2002	min obs conc
2.6E+21	``	1.3E-06	``	``	``	average conc and photolysis from HNO3 to NOx
4.8E+21	``	3.5E-06	``	``	``	max observed conc; max photolysis during the day
3.6E+21	``	N/A	``	South Pole	Liao et al 2008	value from personal comm with Dibbs
2.1E+21	``	``	``	Summit	Jennie's AGU slide	lower end from Dibbs et al measured in summer 2008
3.0E+21	``	``	``	``	``	upper end ``

Values used for model initialization highlighted in dark-tan.

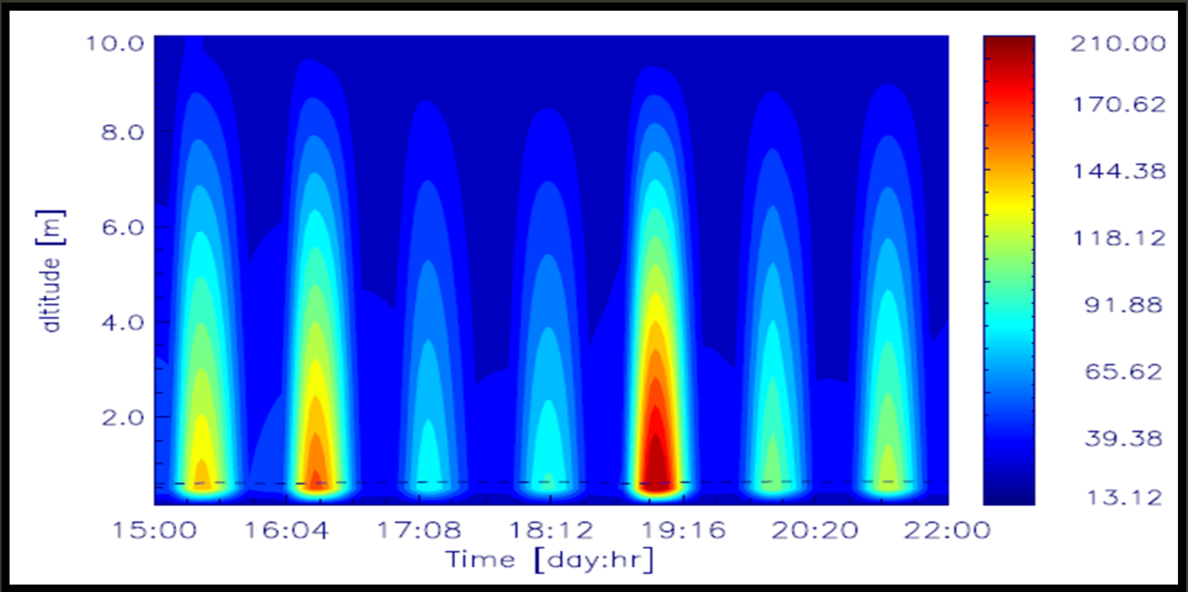
Chemistry: Experiments

1. Test if $[NO_3^-]$ and J_{NO3_NOx} result in proper order of magnitude [NO], $[NO_2]$ in snow
   – Compare simulated results against measured in 14-20 April 2009
2. Test if NO_x chemistry (gas-phase only) is sufficient enough to explain most of the O_3 removal in snow
   – "Trial & error", found V_{dO3} = ~$5e^{-4}$ cm/s for proper O_3 gradient b/w surface and in-snow

Chemistry: Experiment 1

- Test if $[NO_3^-]$ and J_{NO3_NOx} result in proper order of magnitude [NO], $[NO_2]$ in snow
  - Compare simulated results against measured in 14-20 April 2009

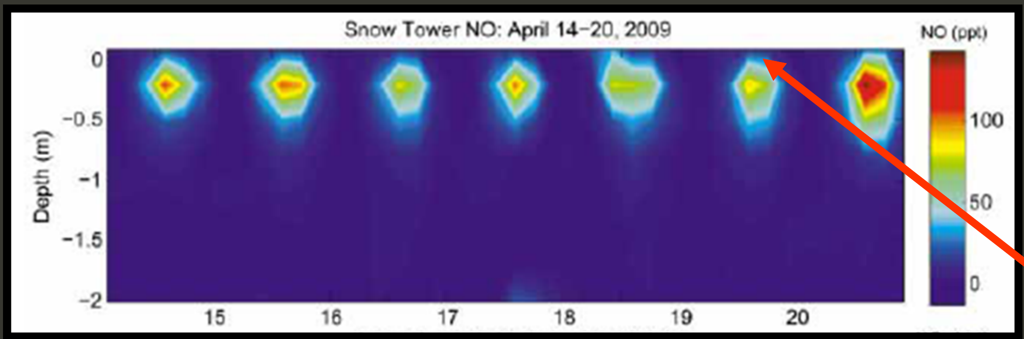
Chemistry:
Exp 1 results (NO)



Simulated NO
[ppt]

14-20 April 2009

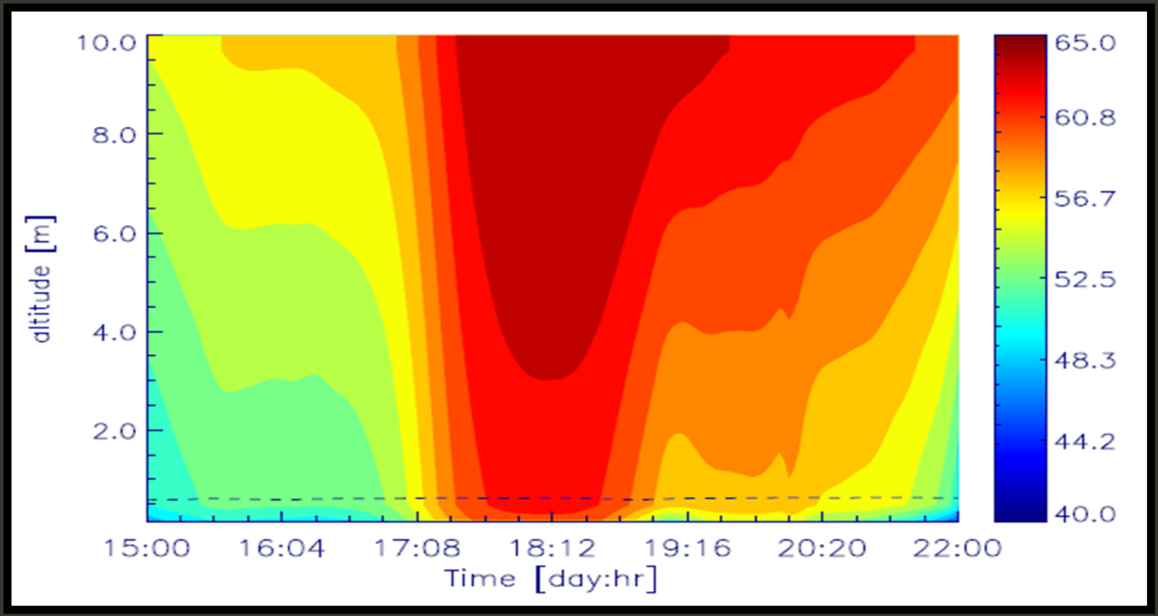
NOTE: discrepancy between time axis model vs observations; leap year...



Measured NO
[ppt]

Is observed NO above the snow indeed <<50 ppt or is this a plotting interpolation issue?

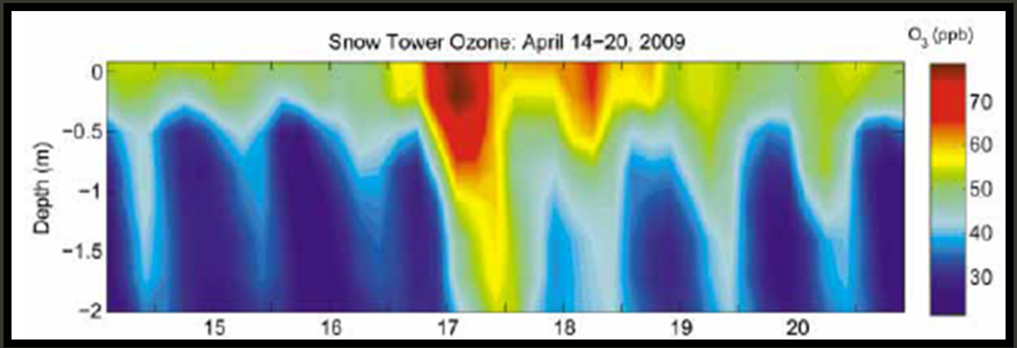
Chemistry: Exp 1 results (Ozone)



Simulated O_3
[ppb]

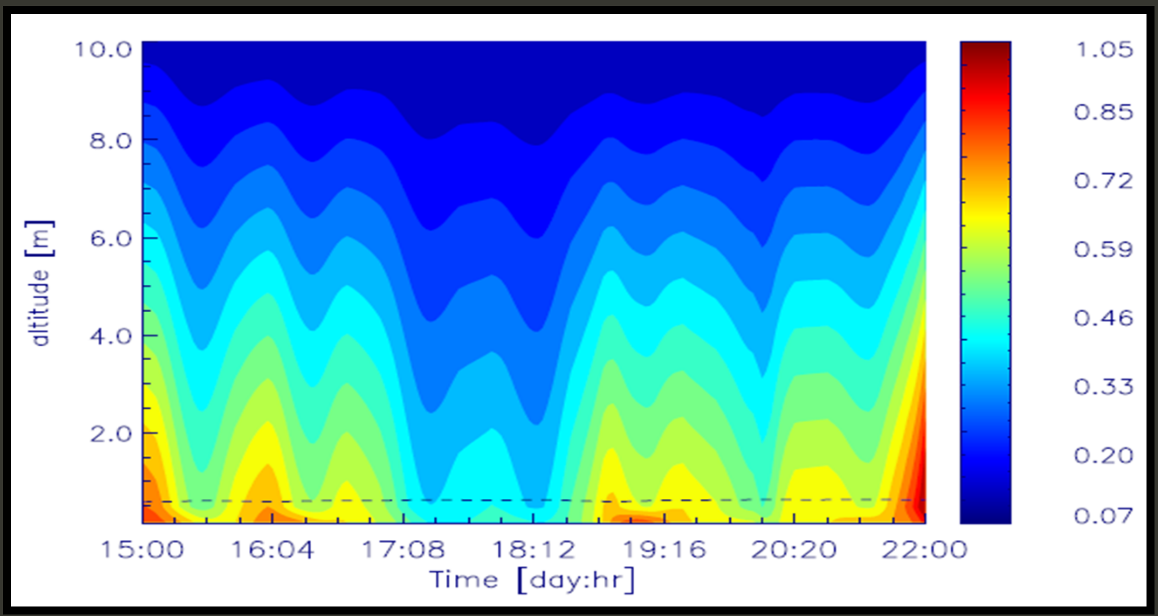
14-20 April 2009

NOTE: discrepancy between time axis model vs observations; leap year...



Measured O_3
[ppb]

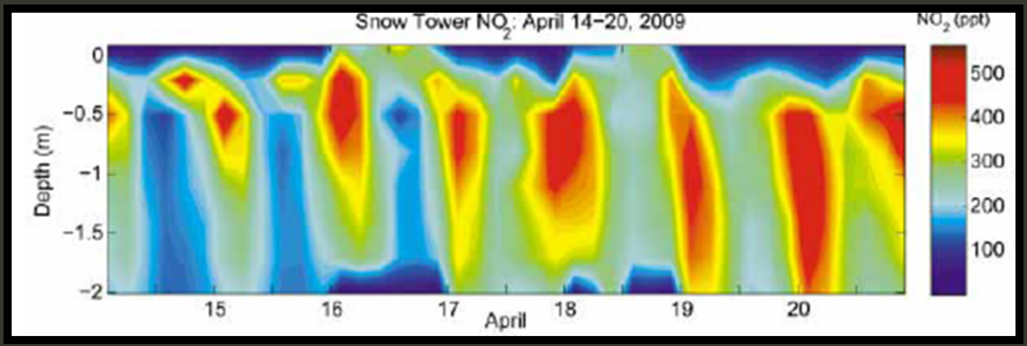
Chemistry: Exp 1 results (NO_2)



Simulated NO_2 [ppb]

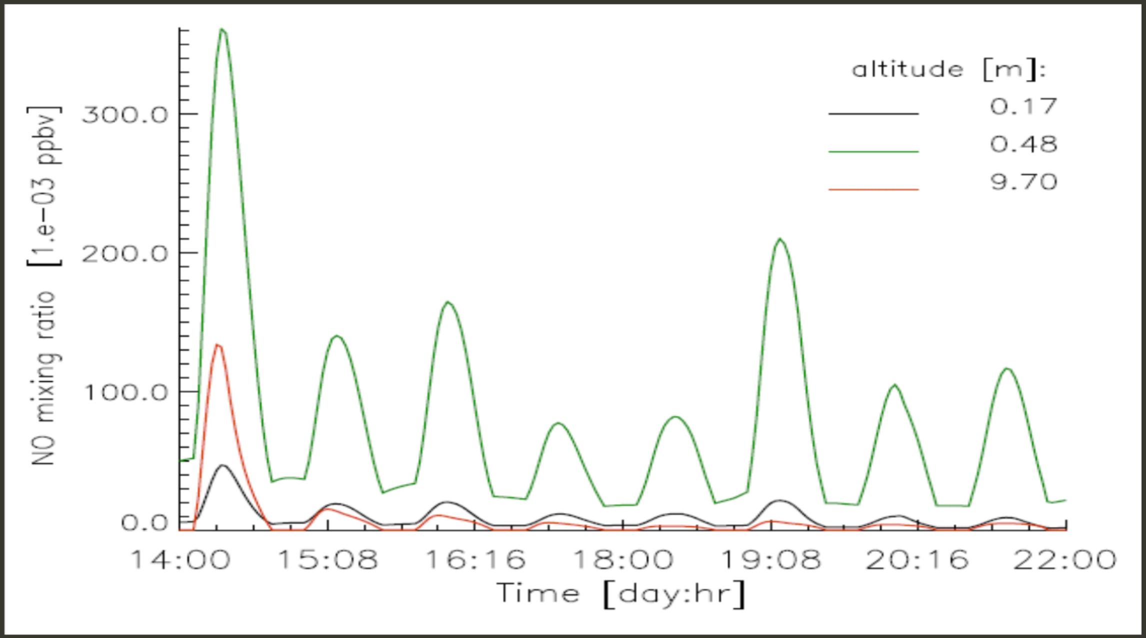
14-20 April 2009

NOTE: discrepancy between time axis model vs observations; leap year...



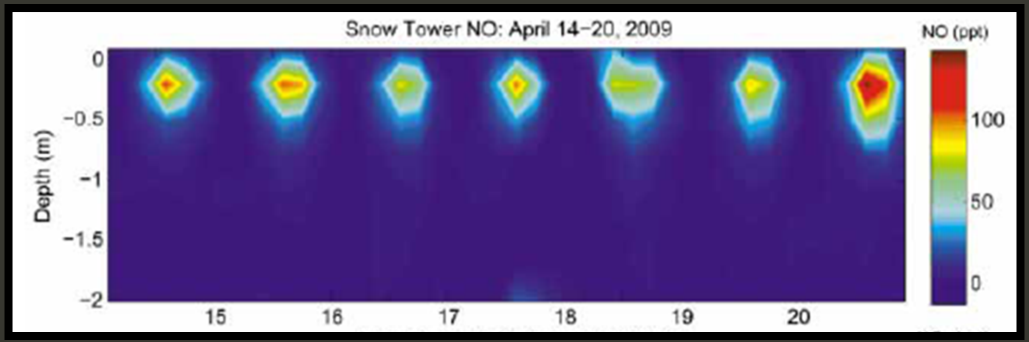
Measured NO_2 [ppt]

Chemistry:
Exp 1 results (NO)



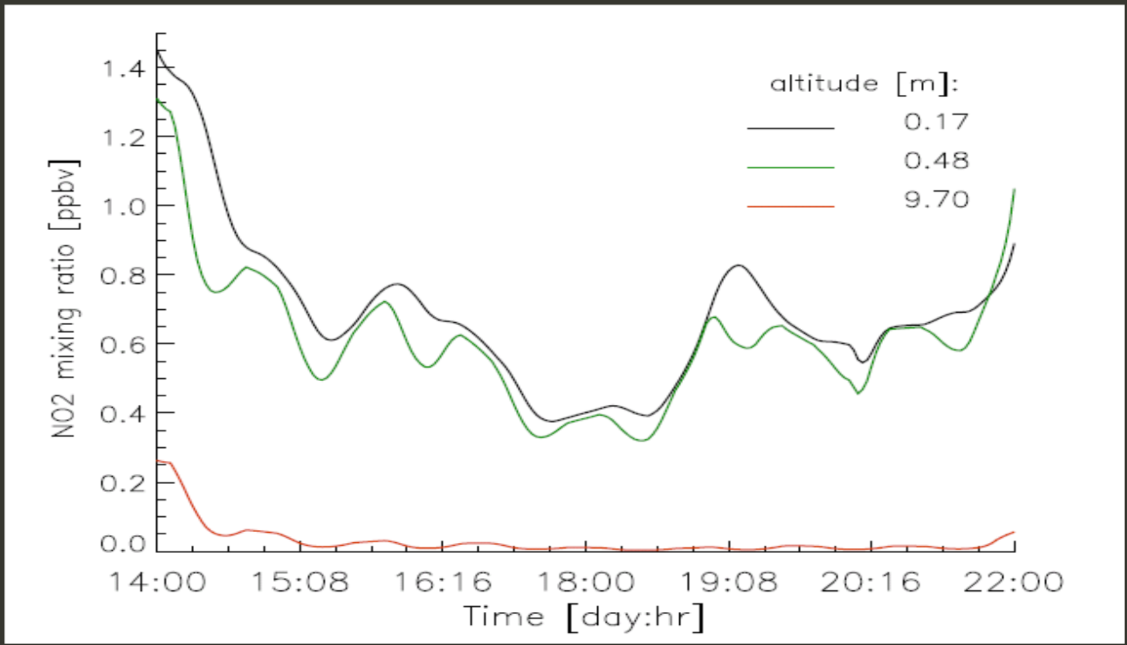
Simulated NO
[ppt]

14-20 April 2009



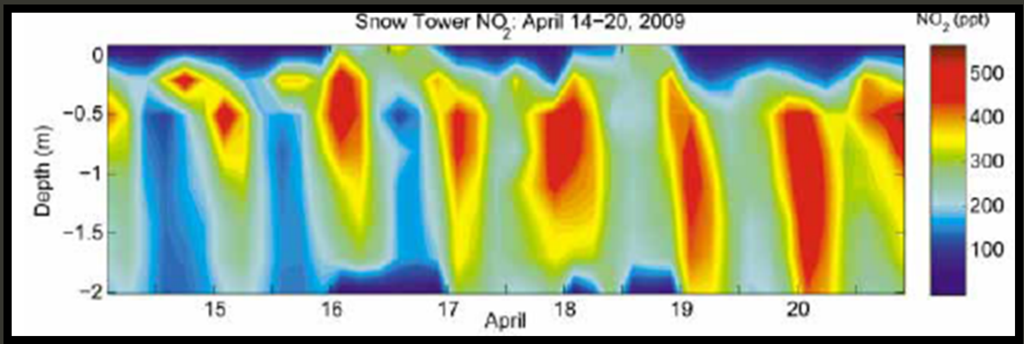
Measured NO
[ppt]

Chemistry: Exp 1 results (NO_2)



Simulated NO_2 [ppb]

14-20 April 2009



Measured NO_2 [ppt]

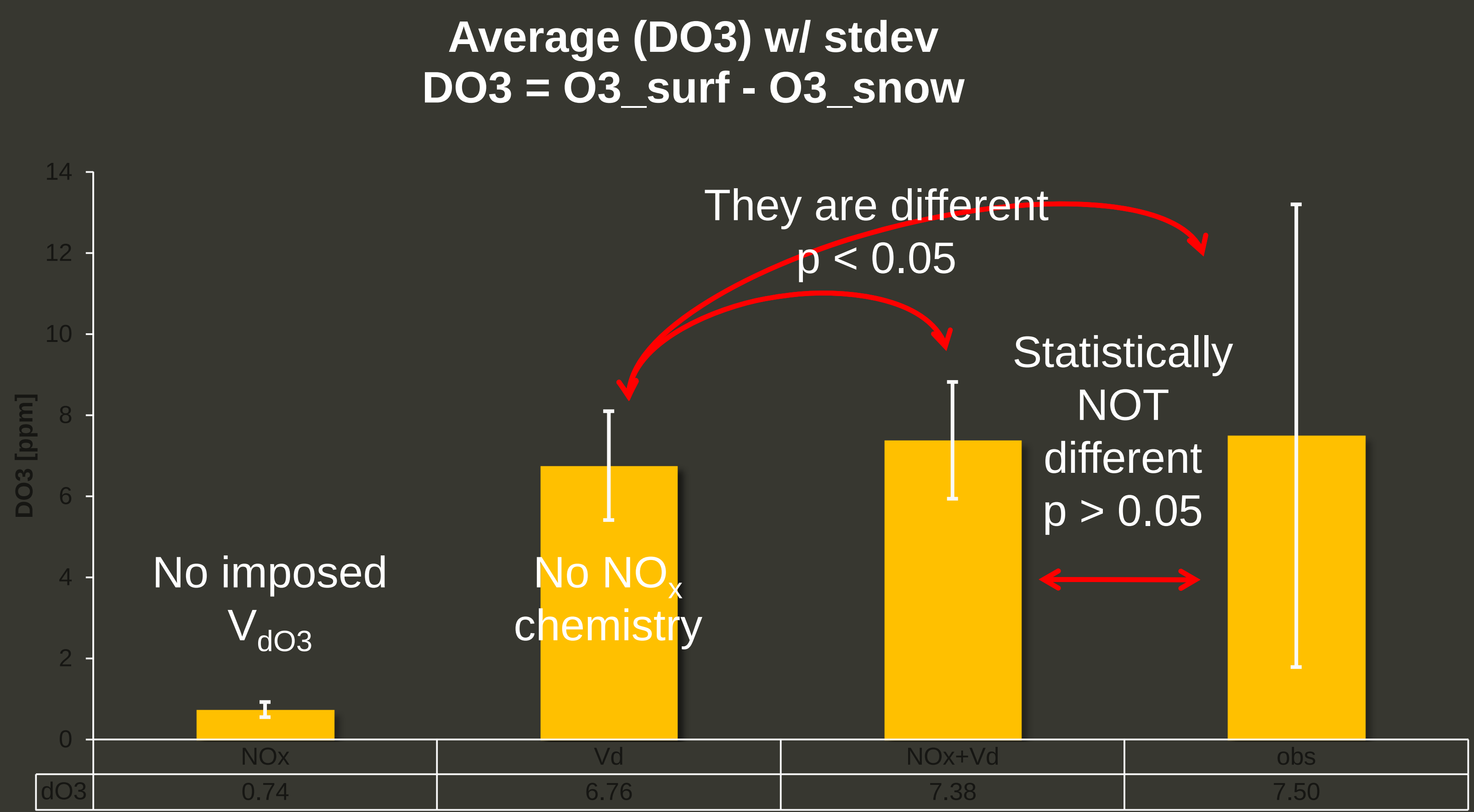
Chemistry:
Exp 1 discussion

- Using Honrath et al 2002 average $[NO_3^-]$ and J_{NO3_NOx} values seem to be OK
- [NO] is slightly overestimated
- $[O_3]$ is slightly underestimated
- Diurnal signals captured overall pretty good
  - NOTE: This run included NO_x chemistry and imposed $V_{dO3} = 5e^{-4}$ cm/s
    - Explained in experiment 2

Chemistry: Experiment 2

- Test if NO_x chemistry (gas-phase only) is sufficient enough to explain most of the O_3 removal in snow (hypothesis; NO-O_3 titrat.)
  - "Trial & error", found V_{dO3} = ~5e^{-4} cm/s for proper O_3 gradient b/w surface and in-snow

Chemistry:
Exp 2 results (Ozone gradients)

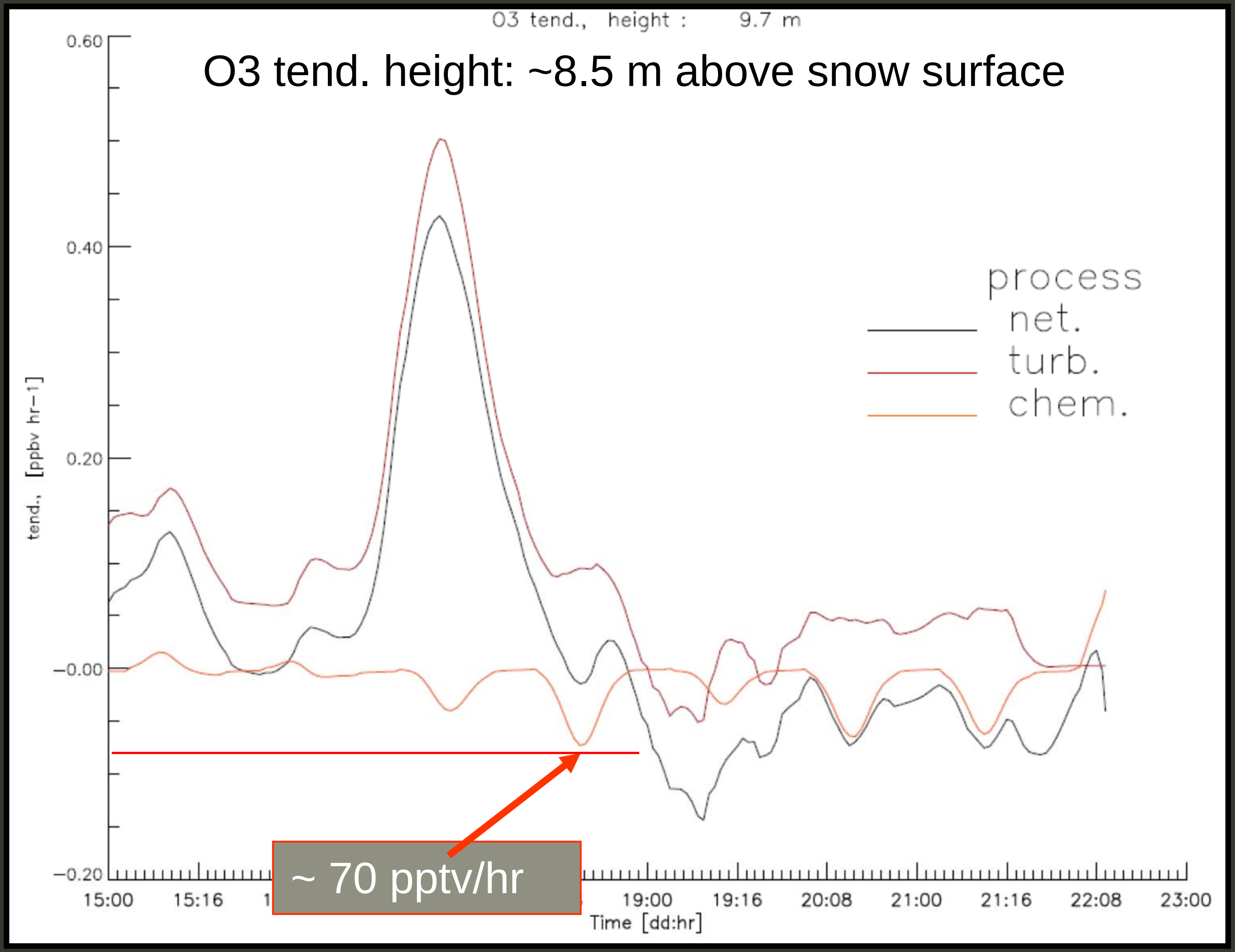


Set V_{dO3}	0.0 cm/s	$5e^{-4}$ cm/s	$5e^{-4}$ cm/s
Result V_{dO3}	$1.1e^{-3}$ cm/s	$3.6e^{-4}$ cm/s	$>3.6e^{-4}$ cm/s

Chemistry:
Exp 2 discussion

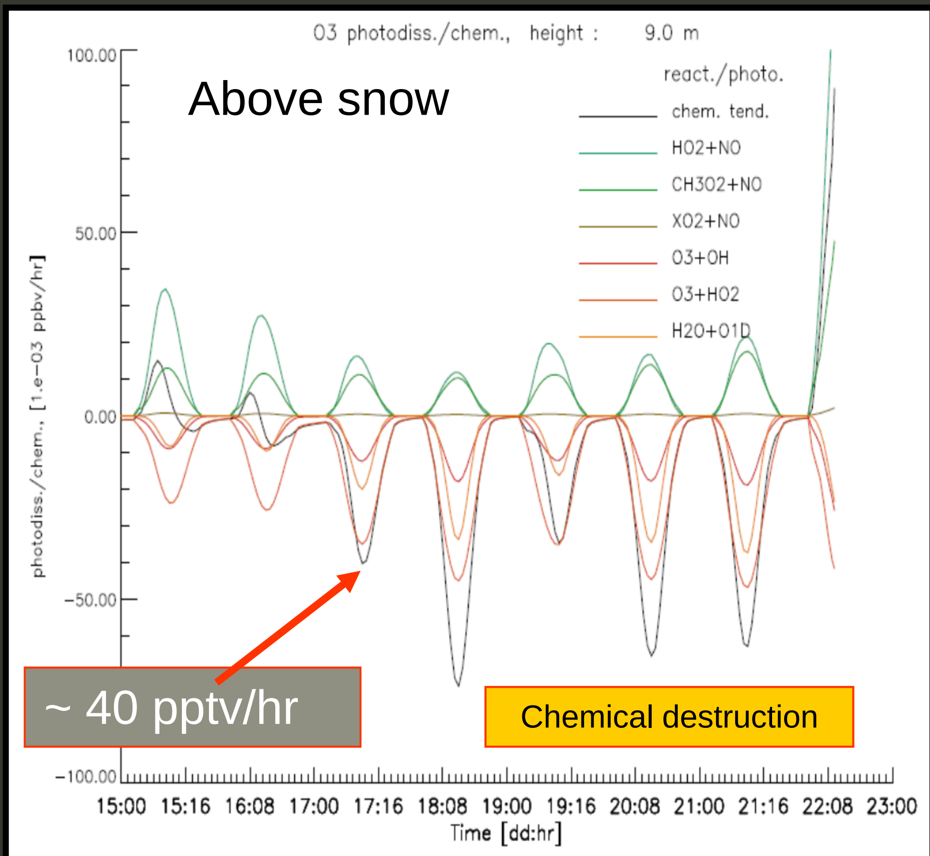
- NO_x chemistry (gas-phase only) alone does NOT explain most of the O_3 removal in snow
- We will have to look into heterogeneous (QLL) chemistry.
- Looking at tendencies
  - What rxns destroy/produce O_3 in the snow?
  - How much is it chemical vs physical?

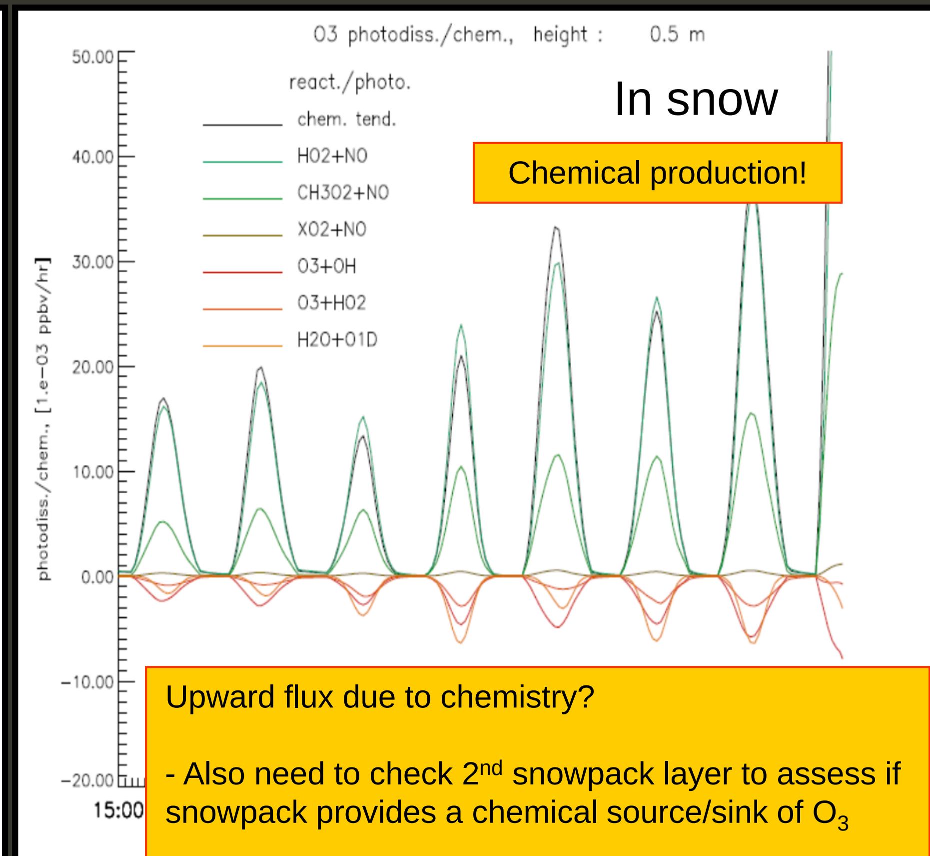
Chemistry:
Exp 2 tendencies (O_3)



April 2009

Chemistry: Exp 2 tendencies (Rxns → O_3)





Upward flux due to chemistry?

- Also need to check 2nd snowpack layer to assess if snowpack provides a chemical source/sink of O_3

- Missing reactions? NOx-O3

April 2009

Conclusions and outlook

- NO_x gas-phase chemistry alone does NOT explain O_3 removal in snow
- We will have to look into heterogeneous (QLL) chemistry
  - Based on the aqueous-phase chemistry scheme of 1D model or....
  - Jenny Thomas's model to assess role of BrO in snowpack O_3 destruction?
  - Physical sorption process?
- Further validation
  - micromet. and BL structure
  - photolysis rates (data?)
  - Mid-latitude snowpack simulations, Michigan forest, Niwot Ridge