Polar Heterogeneous Processes in WACCM: A New Approach

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Breckenridge, CO

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Why modify the heterogeneous approach?

1. Representation of gas-phase HNO_3.
    - Early attempts (using SD-WACCM) at representing the gas-phase HNO_3 for the 2004/2005 winter showed too much de-nitrification in the model relative to observations.
2. Improve the representation of supercooled ternary solutions (STS) surface area density (SAD).
    - The model was underestimating the obs SAD.
    - This STS SAD is used in the rate constant derivation for six heterogeneous reactions.
    - The STS iteration logic was improved!

Outline

- Review stratospheric heterogeneous chemistry.
- Discuss the heterogeneous approaches available in WACCM4.
- Results – show chemical ozone loss for 2010/2011.
- Summary and Future Work.

Heterogeneous Chemistry

Most Important Heterogeneous Reactions:

- $ClONO_2 + HCl \Rightarrow Cl_2 + HNO_3$
- $ClONO_2 + H_2O \Rightarrow HOCl + HNO_3$
- $HOCl + HCl \Rightarrow Cl_2 + H_2O$

Rate constant for these reaction is derived from:

- $K = ¼ * V * SAD * γ$

V = mean velocity
SAD = Surface Area Density of PSCs
γ = reaction probability.

Polar Stratospheric Clouds: Observations

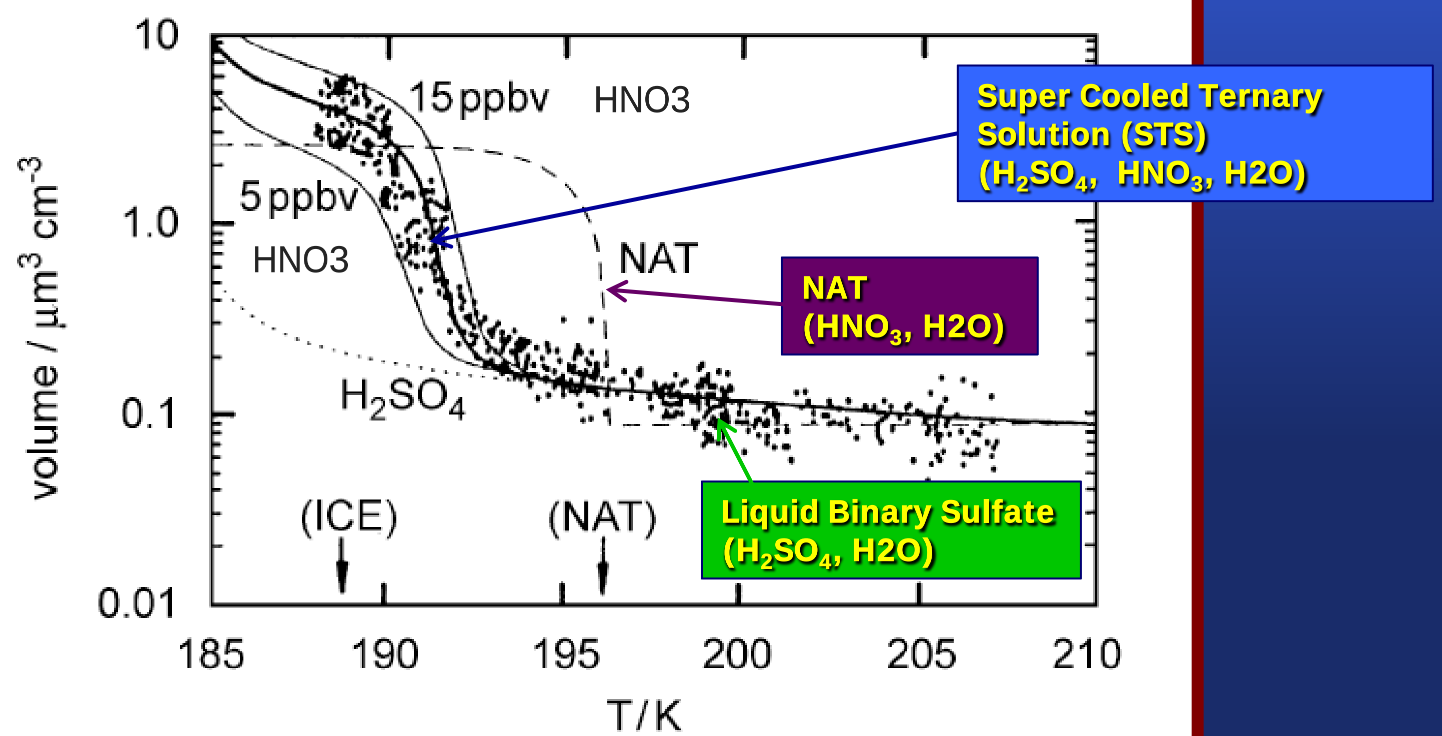


Fig. 2. Particle volumes of Dye et al. (1992) compared with model calculations. From Carslaw et al. (1994). Copyright 1994 American Geophysical Union. Reproduced with permission from American Geophysical Union.

Volume Density vs Area Density

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 103, NO. D5, PAGES 5773–5783, MARCH 20, 1998

Estimation of polar stratospheric cloud volume and area densities from UARS, stratospheric aerosol measurement II, and polar ozone and aerosol measurement II extinction data

Steven T. Massie, Darrel Baumgardner, and James E. Dye

Max STS SAD ~ 15 x 10^{-8} cm^{-2} cm^3

Max LBS SAD ~ 1 x 10^{-8} cm^{-2} cm^3

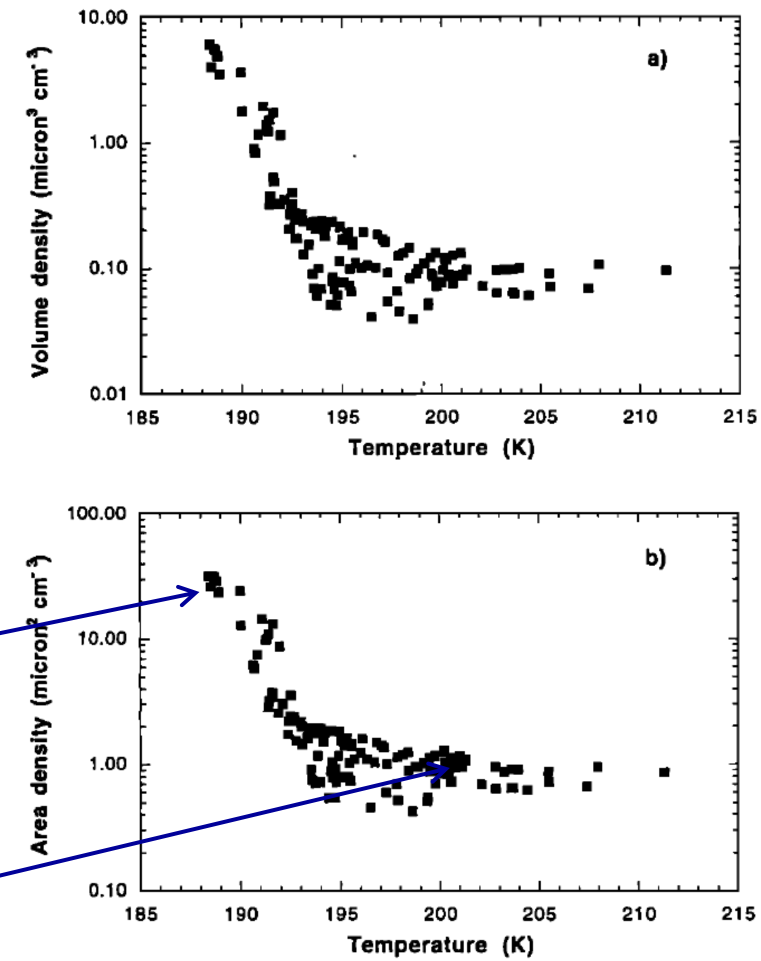
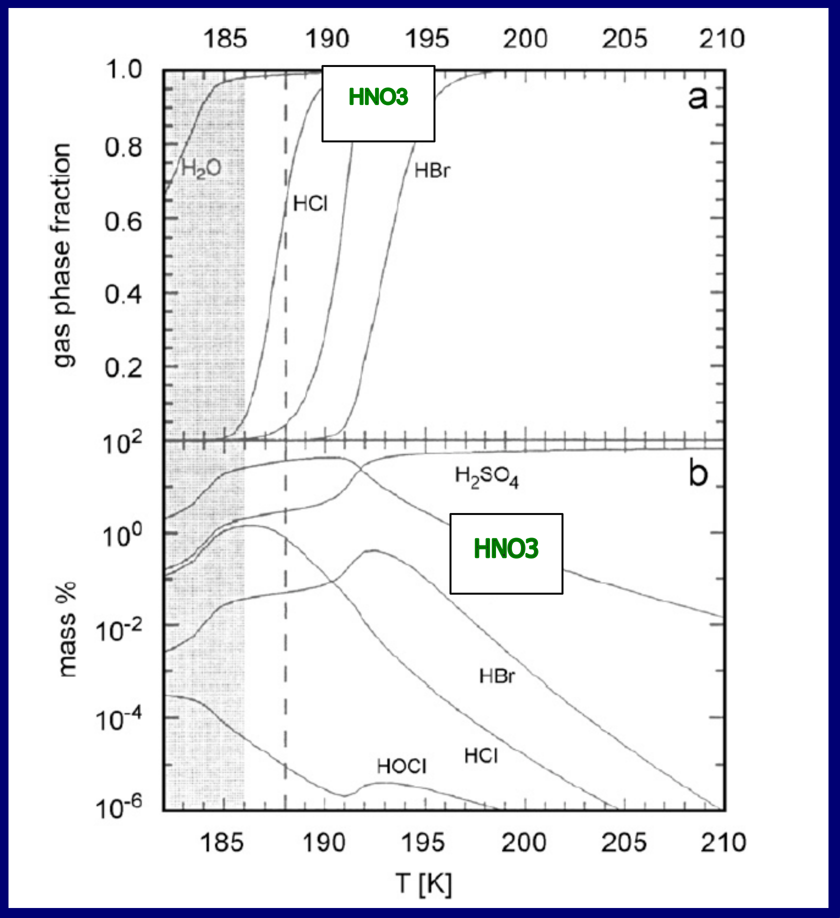


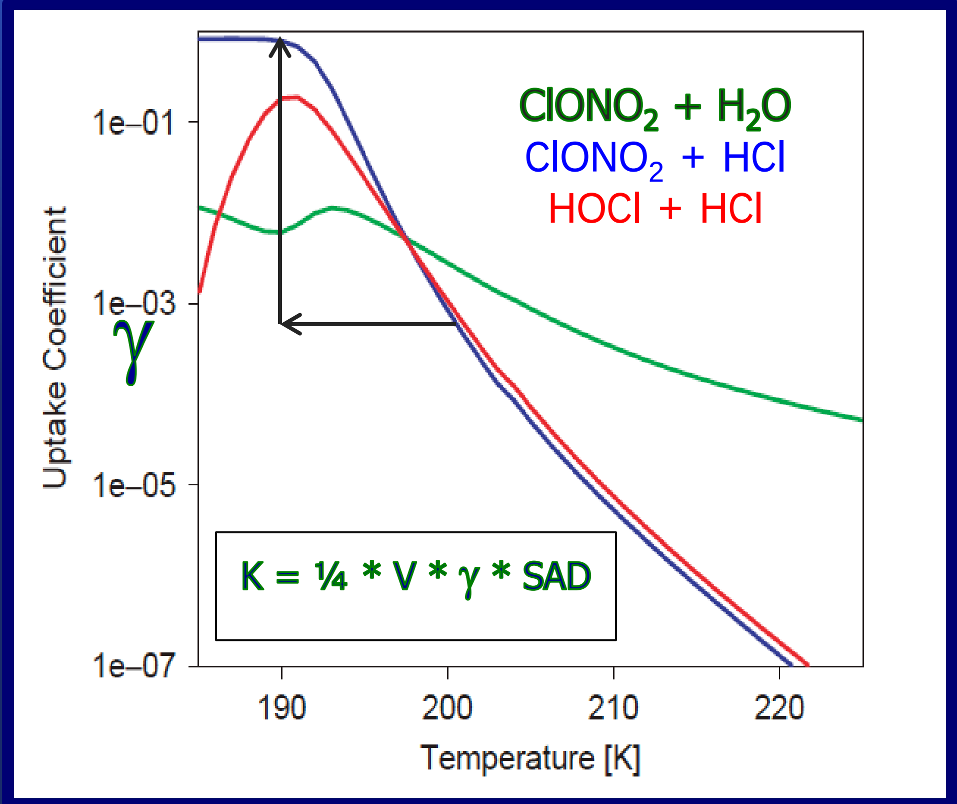
Figure 1. (a) Time-averaged forward scattering spectrometer probe (FSSP 300) volume densities from ER 2 flights 890119, 890120, 890124, and 890125. (b) Same as Figure 1a, except for the time-averaged area densities.

STS Model: Uptake HNO_3 and Reactivity

Update of HNO_3 in STS Aerosol



Increased reactivity



Temperature accuracy is important!

Lowe and MacKenzie, *J. of Atm. And Solar-Terrestrial Physics*, 70, 13-40, 2008.

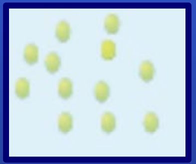
Outline

- Review stratospheric heterogeneous chemistry.
- **Discuss the heterogeneous approaches available in WACCM4.**
- Results – show chemical ozone loss for 2010/2011.
- Summary and Future Work.

WACCM4 PSC Equilibrium Approach - IPCC

200 K

LBS (H_2SO_4, H_2O)

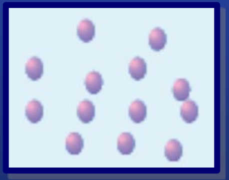


SAD = Observed

r = 0.01-0.1 µm; 10 particles cm^{-3}

~194 K

STS (HNO_3, H_2SO_4, H_2O)

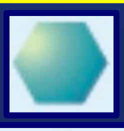


SAD = $(4\pi*\text{Pden})^{1/3} * (3*\text{Vden})^{2/3} * \exp(-\ln(\sigma)^2)$

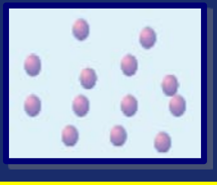
Assume Lognormal Size Distrib., 10 particles cm^{-3}, and width = 1.6; mass of H_2SO_4 derived from LBS

188 K

1) NAT (r>3µm); 0.01 particle cm^{-3}
Super saturation Ratio of 10



2) STS (HNO_3, H_2SO_4, H_2O)

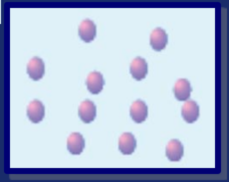


De-NOY occurs (too much)

Sedimentation Velocity:
$V = Vo*\exp(5*\ln(\sigma)^2)$

ICE
(r >10µm);
0.1 particles cm^{-3}

De-hydration



De-NOY occurs

“Standard” Approach

WACCM4 PSC Equilibrium Approach - Modified

LBS (H_2SO_4, H_2O)

SAD = Observed

r = 0.01-0.1 μm; 10 particles cm^{-3}

200 K

STS (HNO_3, H_2SO_4, H_2O)

SAD = $(4\pi * Pden)^{1/3} * (3 * Vden)^{2/3} * \exp(-\ln(\sigma)^2)$

Assume Lognormal Size Distrib., 10 particles cm^{-3}, and width = 1.6; mass of H_2SO_4 derived from LBS

~194 K

1) STS (HNO_3, H_2SO_4, H_2O)

2) NAT (r = 0.5 - 2μm); 0.1 particle cm^{-3}
Super saturation Ratio of 10

De-NOY occurs (less)

Sedimentation Velocity:
$V = Vo * \exp(5 * \ln(\sigma)^2)$

188 K

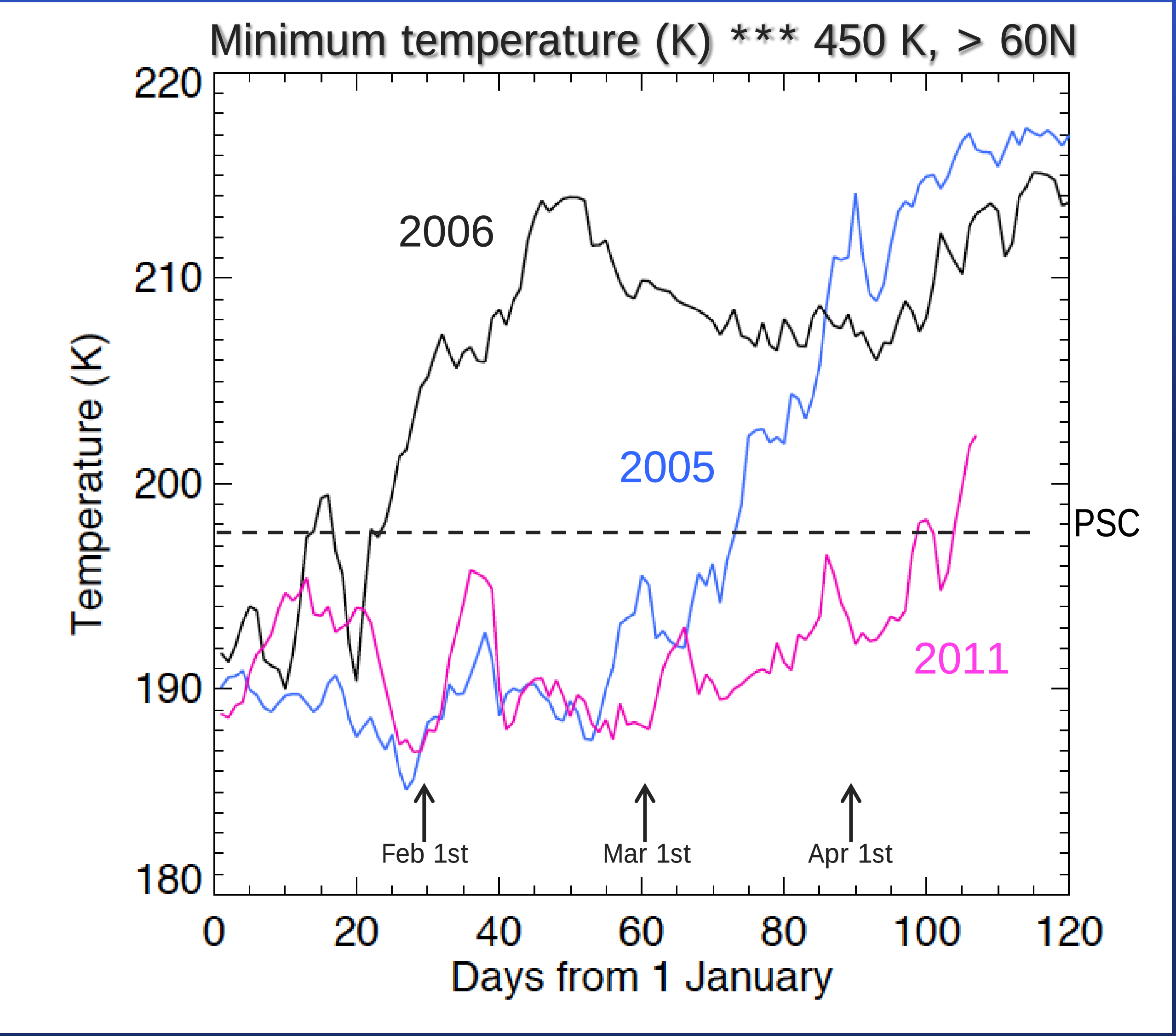
ICE
(r >10μm);
0.1 particles cm^{-3}

De-hydration

De-NOY occurs

"New" Approach

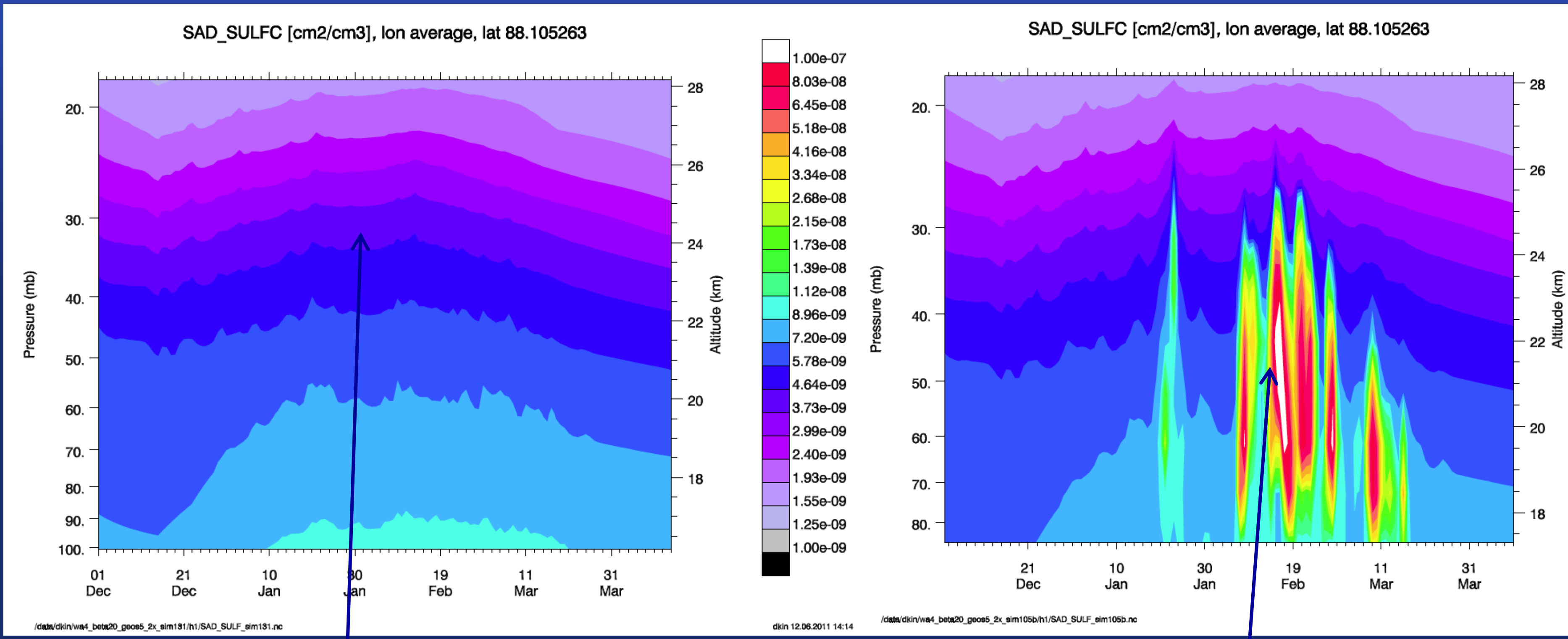
SD-WACCM / GEOS5 – 2010 / 2011



Sulfate SAD *** 88N, zonal mean

Standard (used in IPCC sims)

New (used in O3L sims)



Sulfate SAD ~ 1 x 10^{-8} cm^{-2} cm^{3}

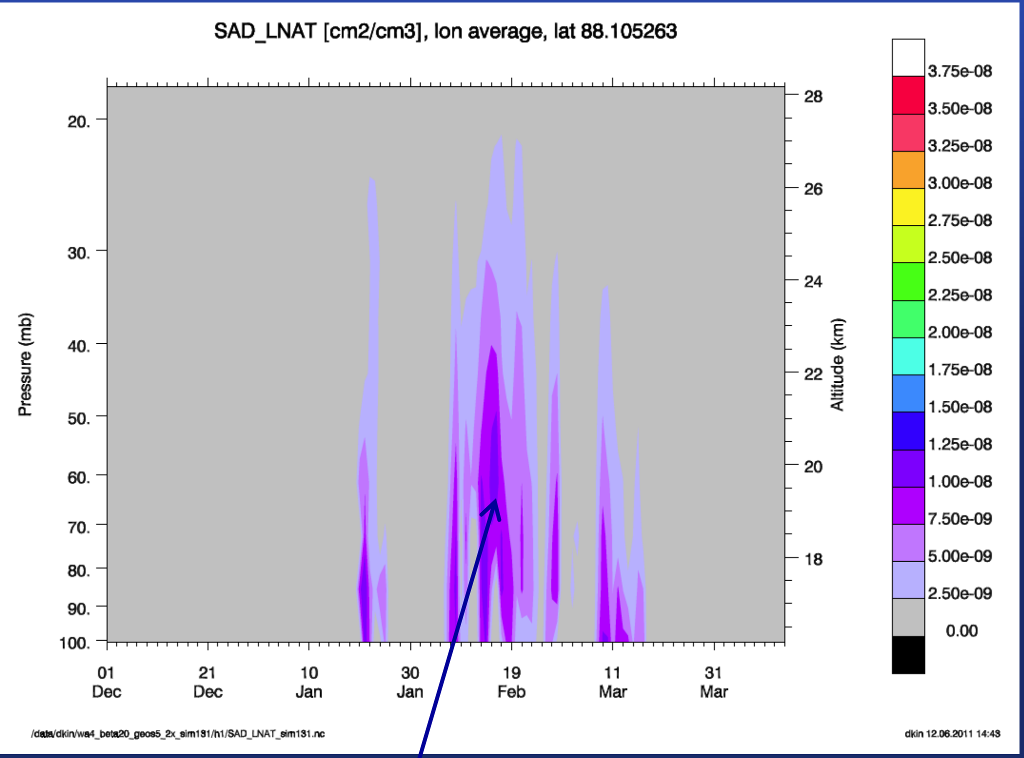
Sulfate SAD > 10 x 10^{-8} cm^{-2} cm^{3}

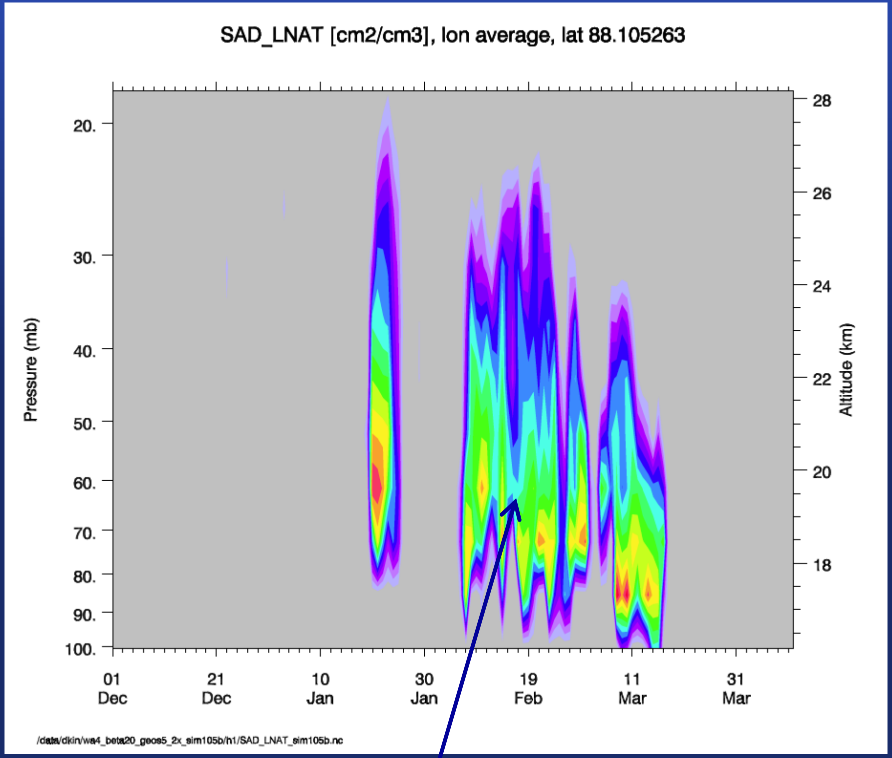
Allowing STS to see the available HNO_3 (before NAT) allows swelling to occur.

NAT SAD *** 88N, zonal mean

Standard (used in IPCC sims)

New (used in O3L sims)





NAT SAD ~ 1 x 10^{-8} cm^{-2} cm^3

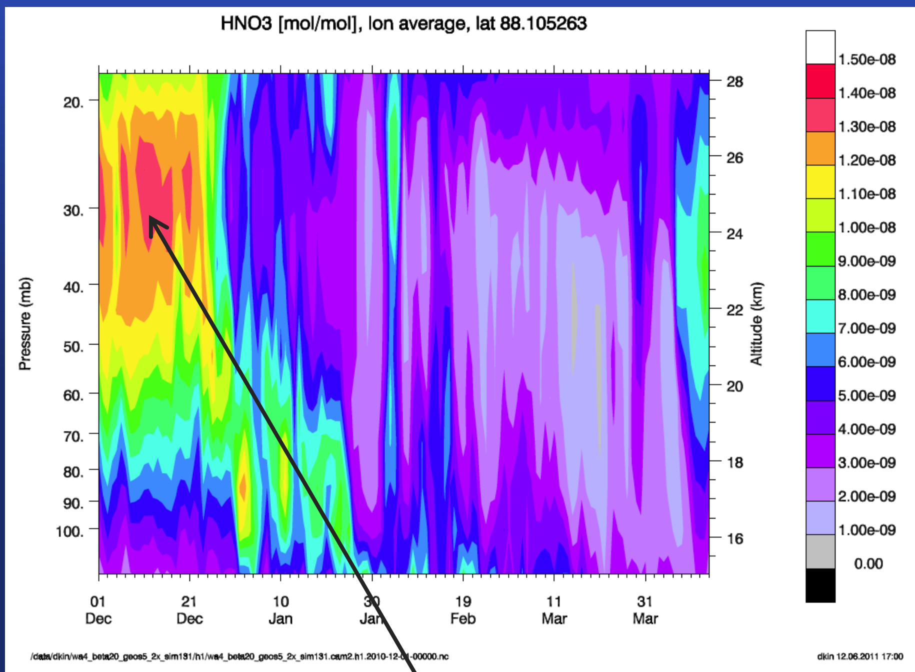
NAT SAD ~ 3 x 10^{-8} cm^{-2} cm^3

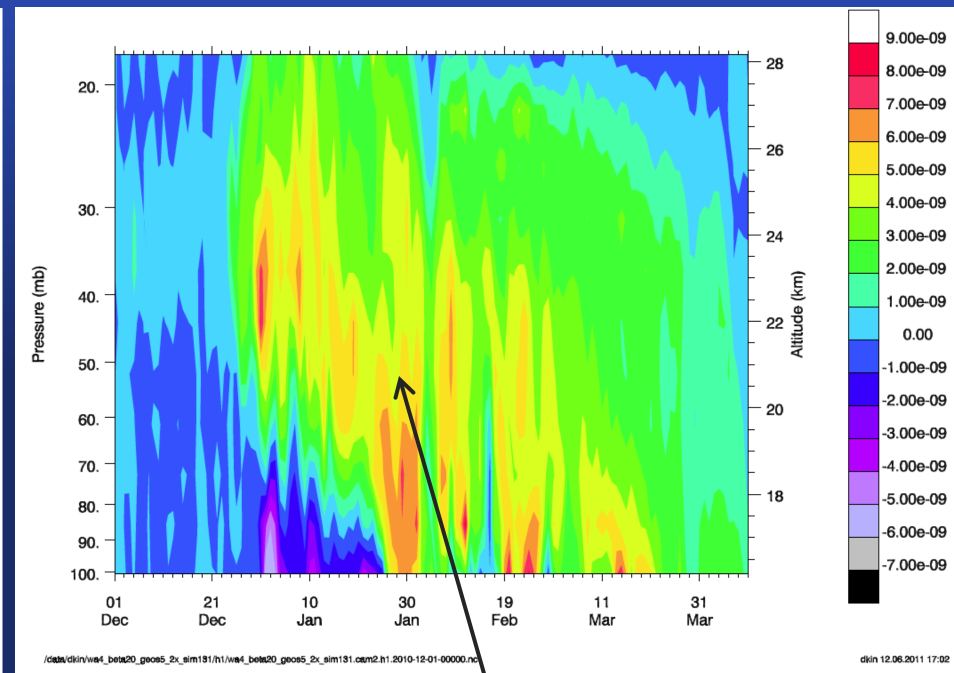
More NAT # density in "New", increased SAD. This will also decrease the effective radius and cause less denitifrication.

Total HNO_3 (ppbv) = Gas-phase + Condensed

Standard (used in IPCC sims)

New - Standard



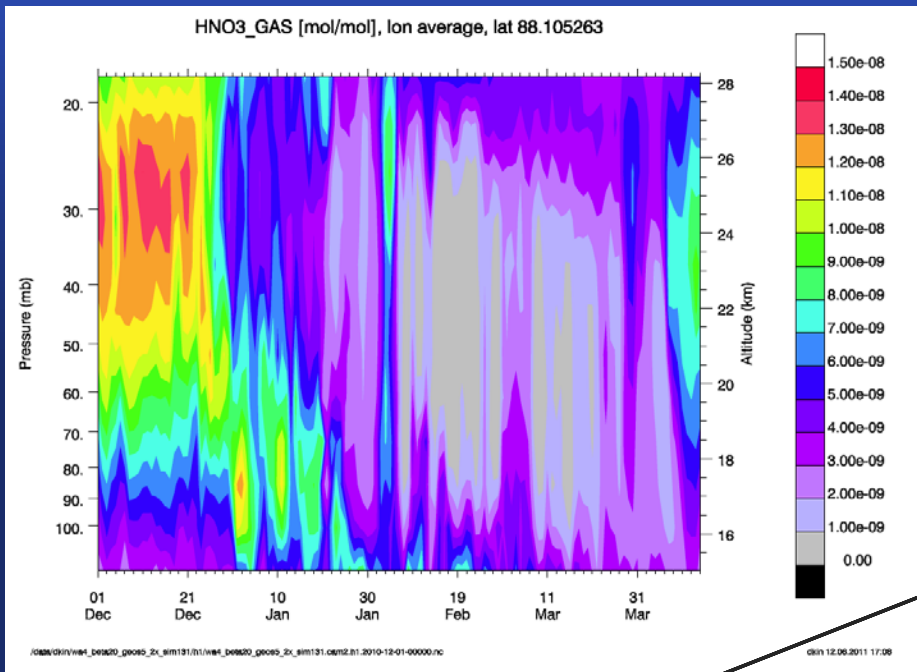


14 ppbv HNO_3 in December

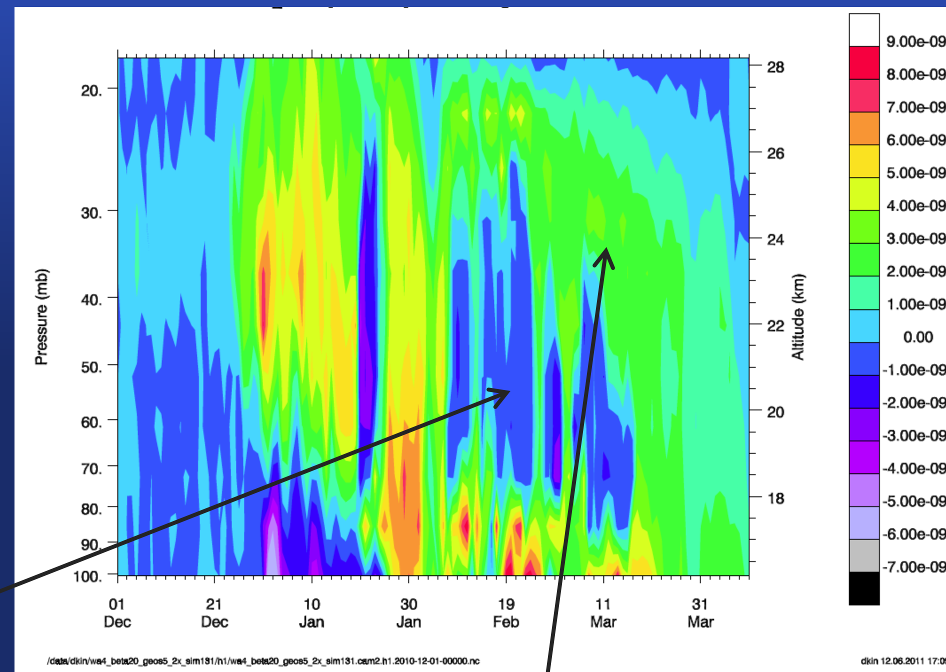
Less irreversible denitrification in “New”.

HNO_3 Gas-phase (ppbv) *** 88N, zonal mean

Standard (used in IPCC sims)



New - Standard

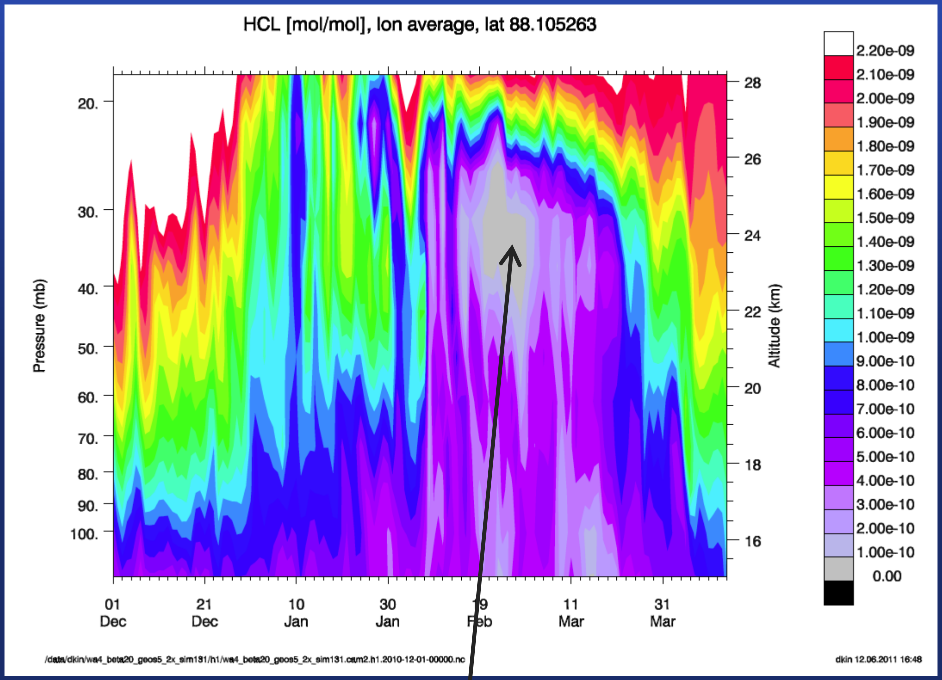


More HNO_3 in "New" STS and NAT condensed phases. The "New" approach does a good job of representing the gas-phase HNO_3 relative to MLS observations.

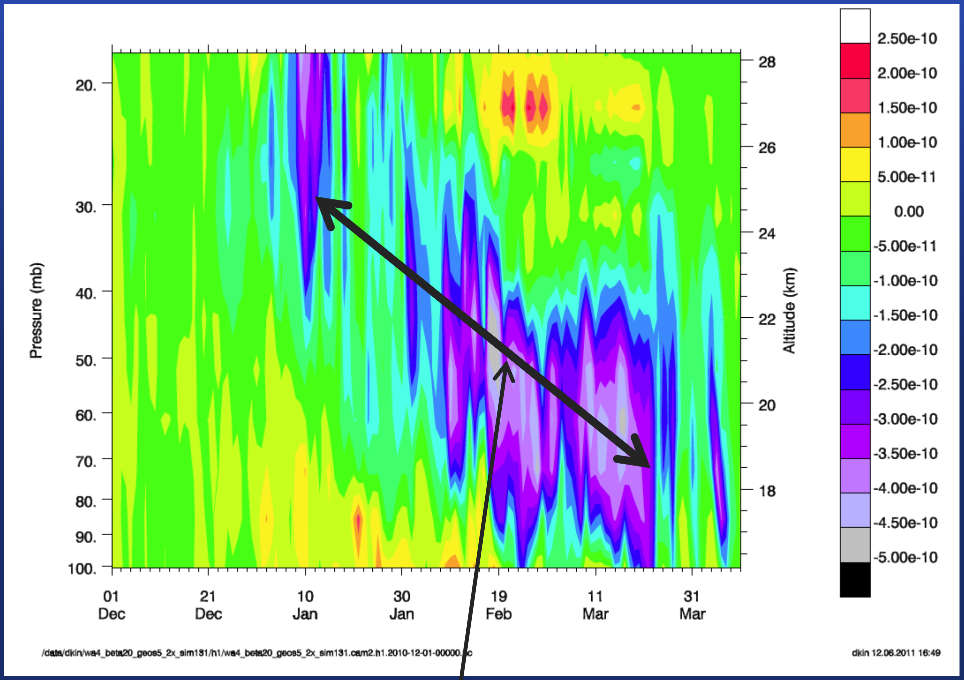
More HNO_3 in "New" in gas-phases (STS and NAT SAD is not present). Will affect ClOx abundance (see later slide).

HCl (ppbv) *** 88N, zonal mean

Standard (used in IPCC sims)



New - Standard



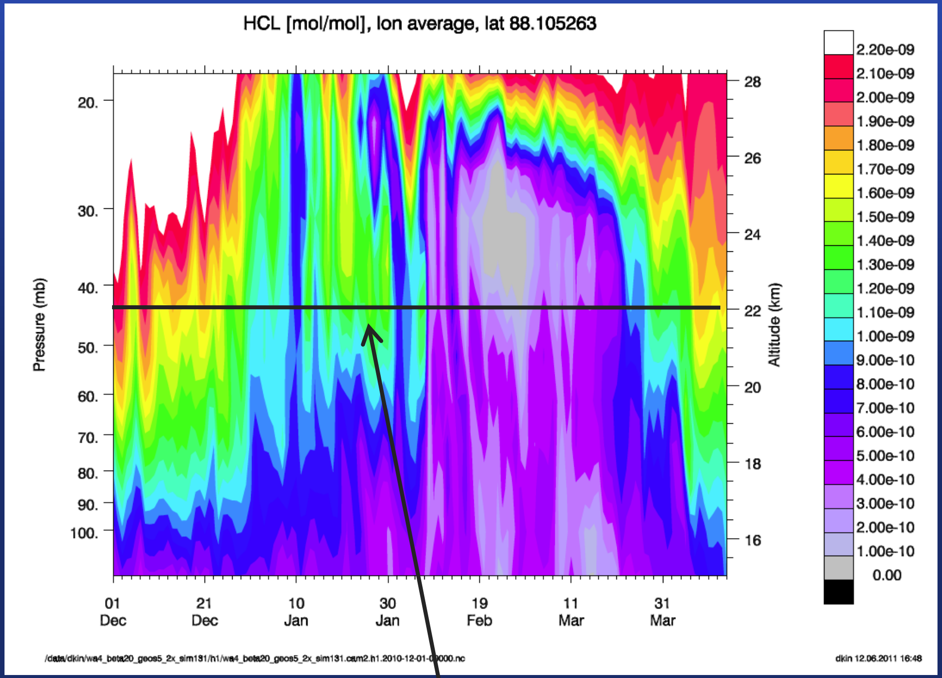
HCl is significantly depleted in this regions (~0.1 ppbv)

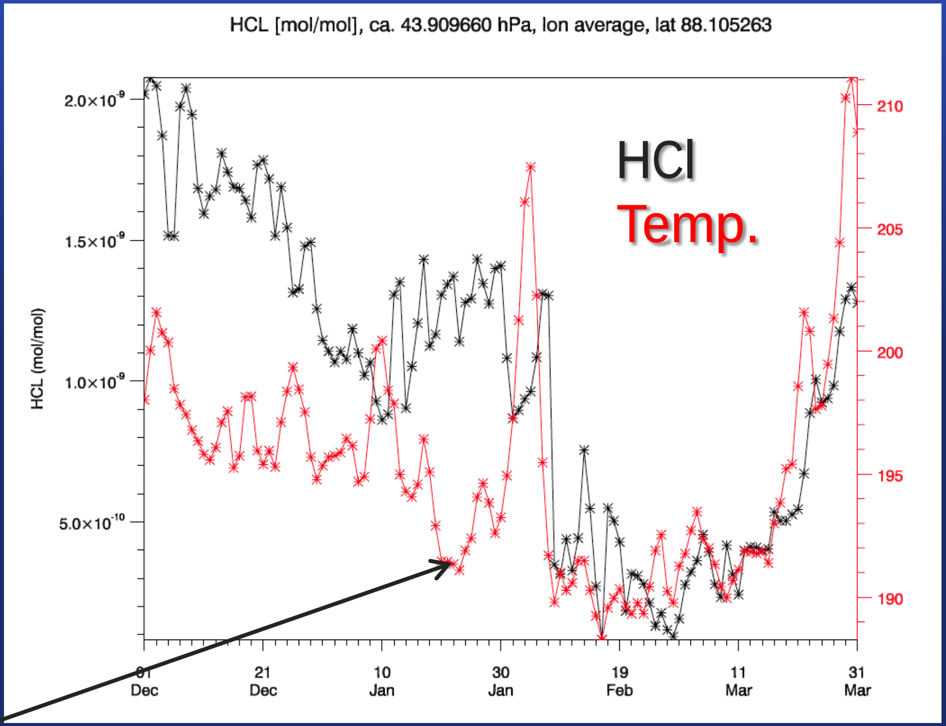
Region of enhanced STS and NAT SAD in "New". Additional HCl depletion of up to ~0.4 ppbv.

HCl (ppbv) *** 88N, zonal mean

Standard (used in IPCC sims)

43 hPa

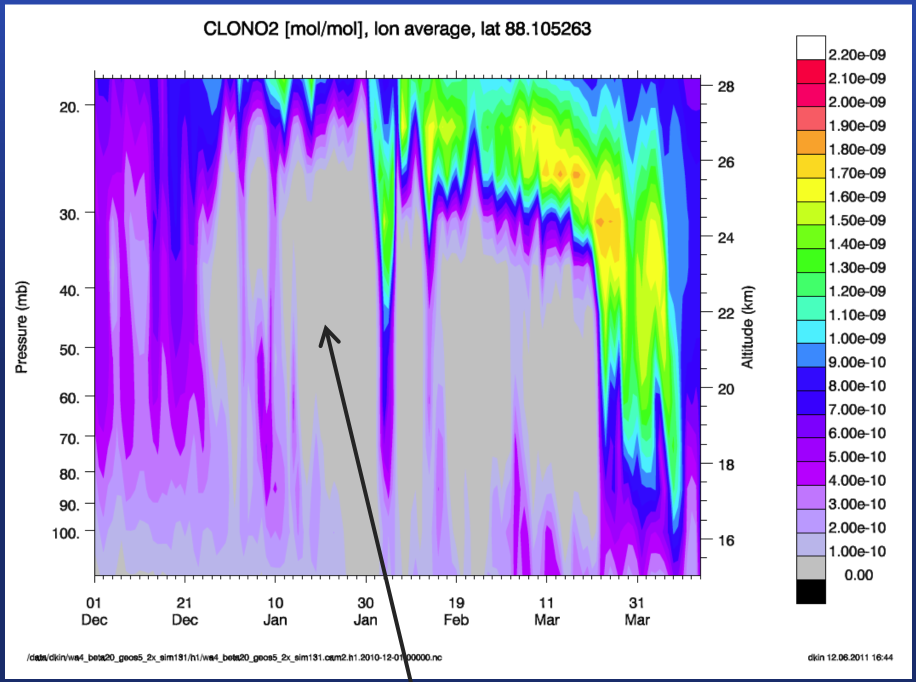




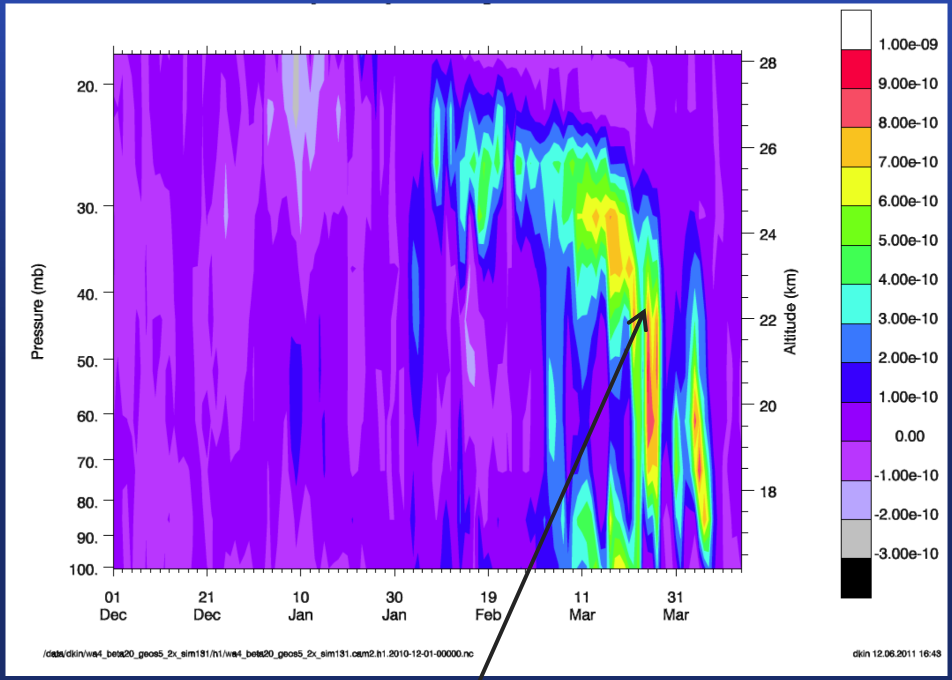
HCl "undepleted" even though T's are cold enough.

$ClONO_2$ (ppbv) *** 88N, zonal mean

Standard (used in IPCC sims)



New - Std

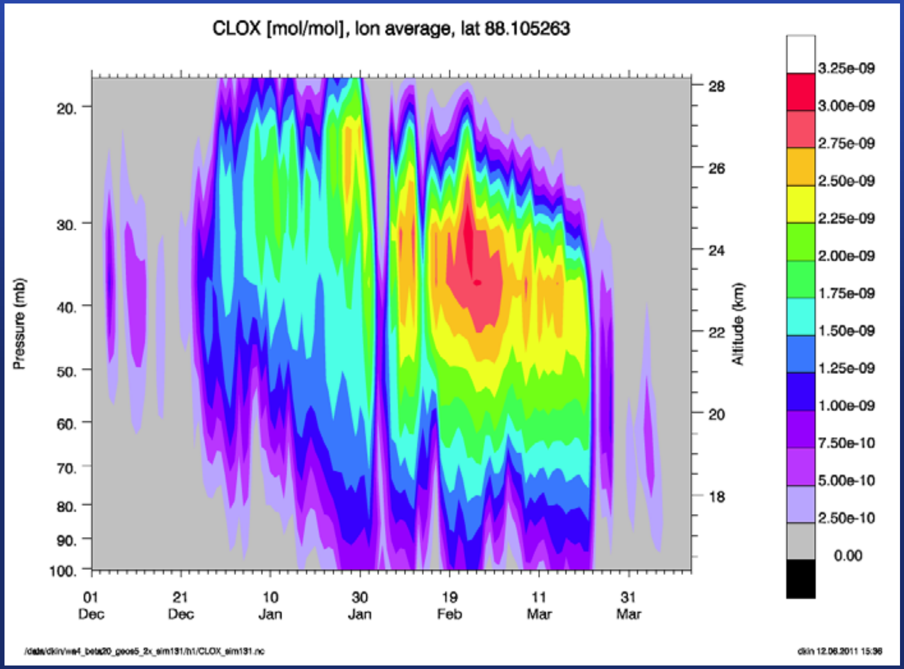


HCl depletion (ClO_X activation) is limited by the abundance of $ClONO_2$.

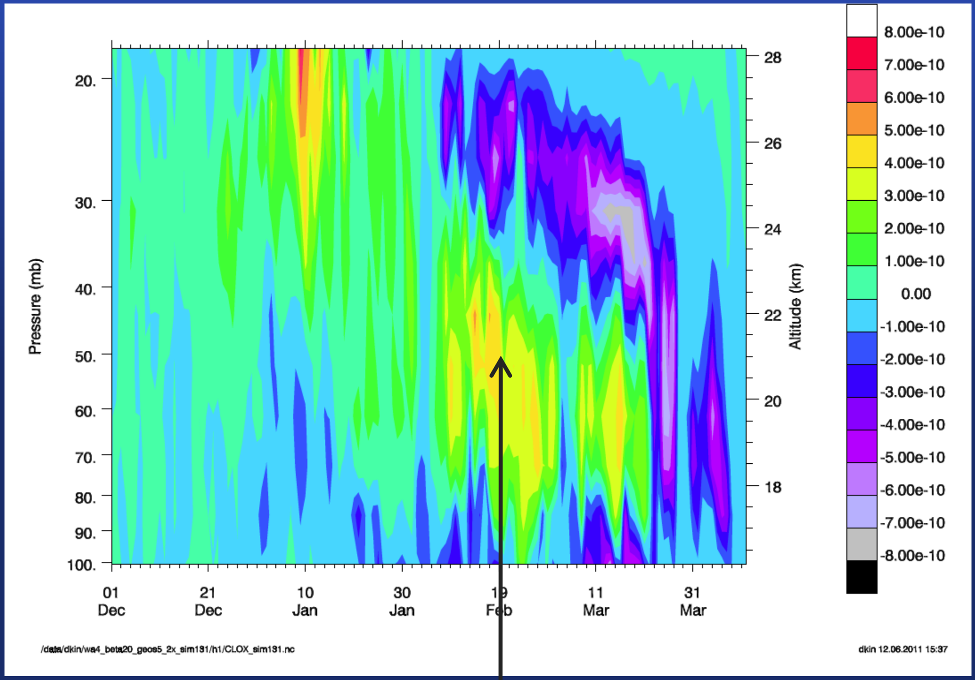
In the "New" approach, ClO_X activation is less due to reformation of $ClONO_2$.

ClOx (ppbv) *** 88N, zonal mean

Standard (used in IPCC sims)



New - Standard

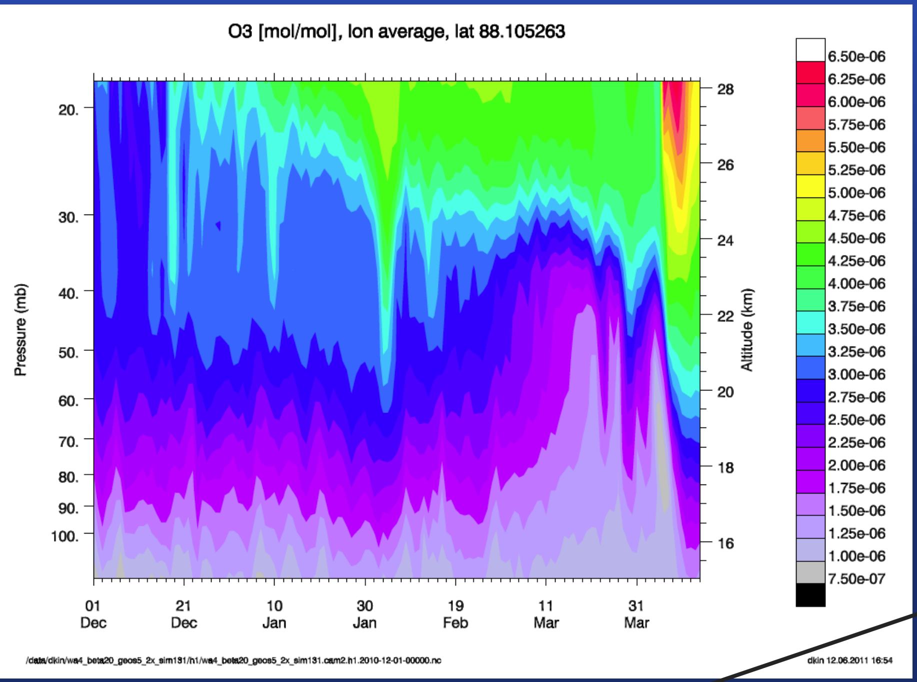


Higher STS and NAT SAD in "New" in this region.

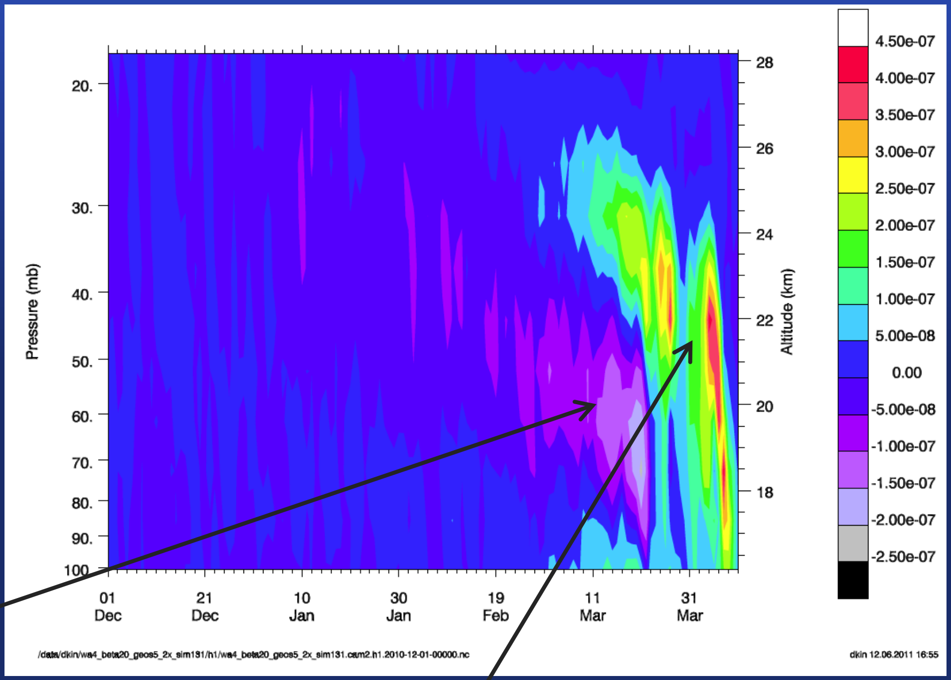
$ClOx$ = total inorganic chlorine – HCl – $ClONO_2$

Ozone (ppmv) *** 88N, zonal mean

Standard (used in IPCC sims)



New - Standard



In the "New" approach, ozone depletion is greater - due to enhanced STS and NAT SAD.

In the "New" approach, ozone depletion is less - due to reformation of $ClONO_2$.

Outline

- Review stratospheric heterogeneous chemistry.
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- **Results – show chemical ozone loss for 2010/2011.**
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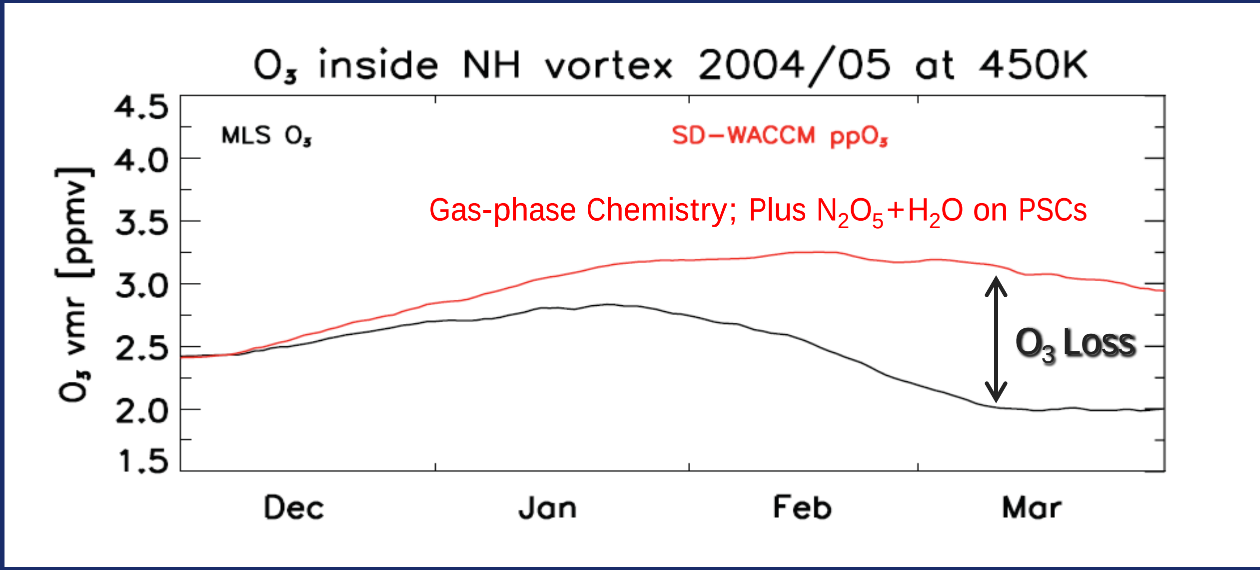
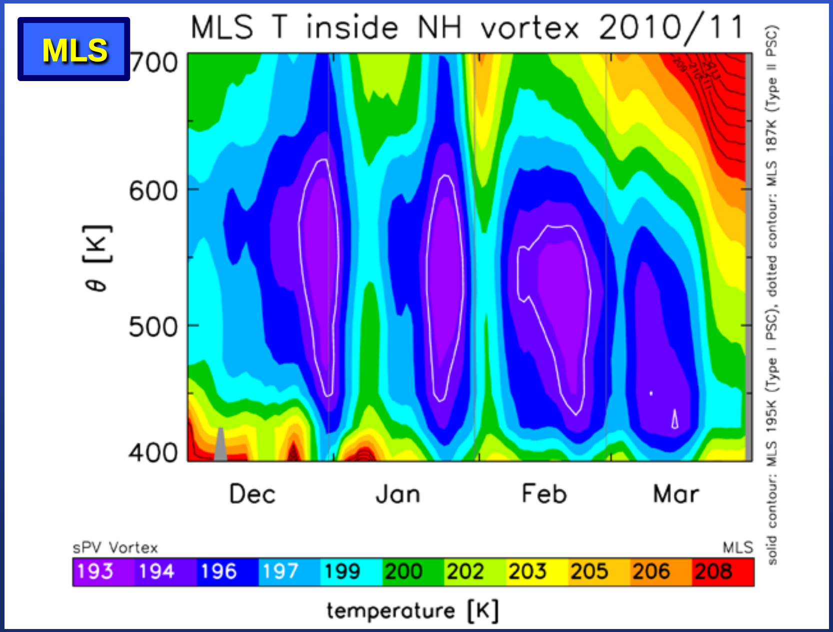
Model Simulations for Halogen Driven Chemical Ozone Loss

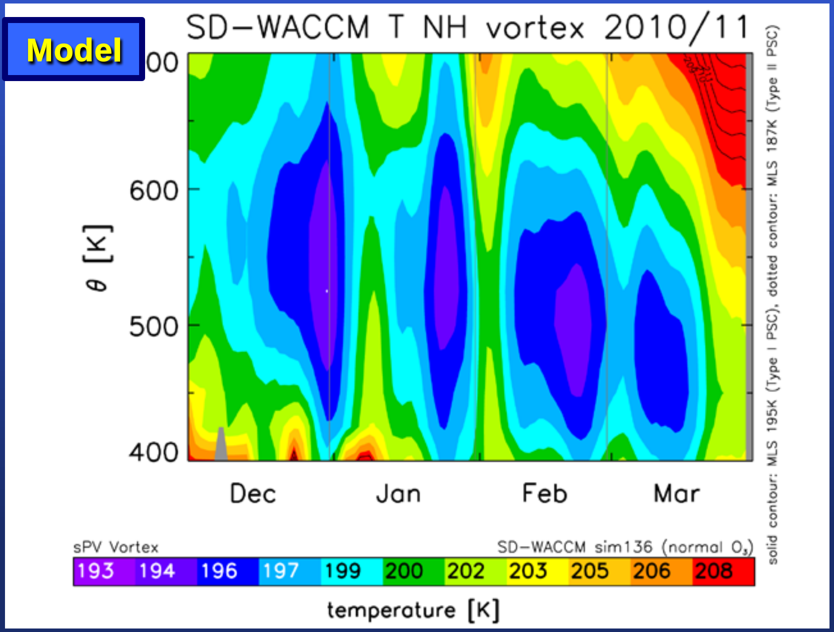
- **Initialize (with obs) on 1 December 2010.**
  - O_3, HCl, HNO_3, H_2O, N_2O
- **Run three simulations through April 2010…**
  - **Full Chemistry**
  - **Pseudo passive:**
    - Gas-phase chemistry only, plus N_2O_5 + H_2O on PSCs

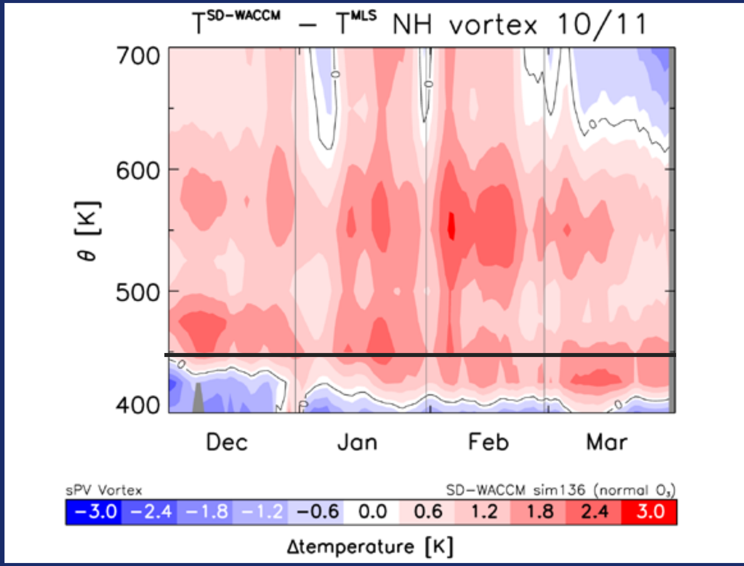


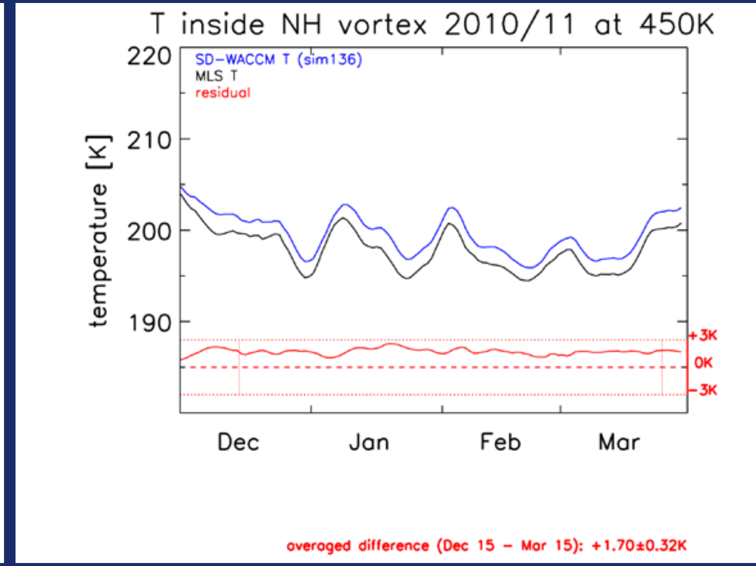
Reference simulation would have similar descent and mixing.

Temperature Evolution



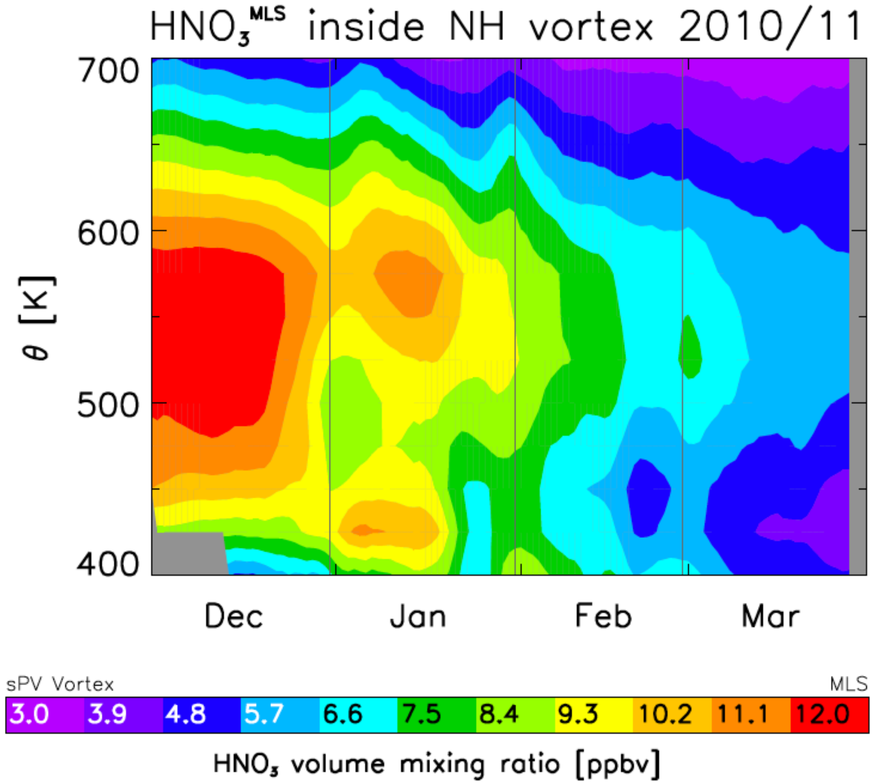




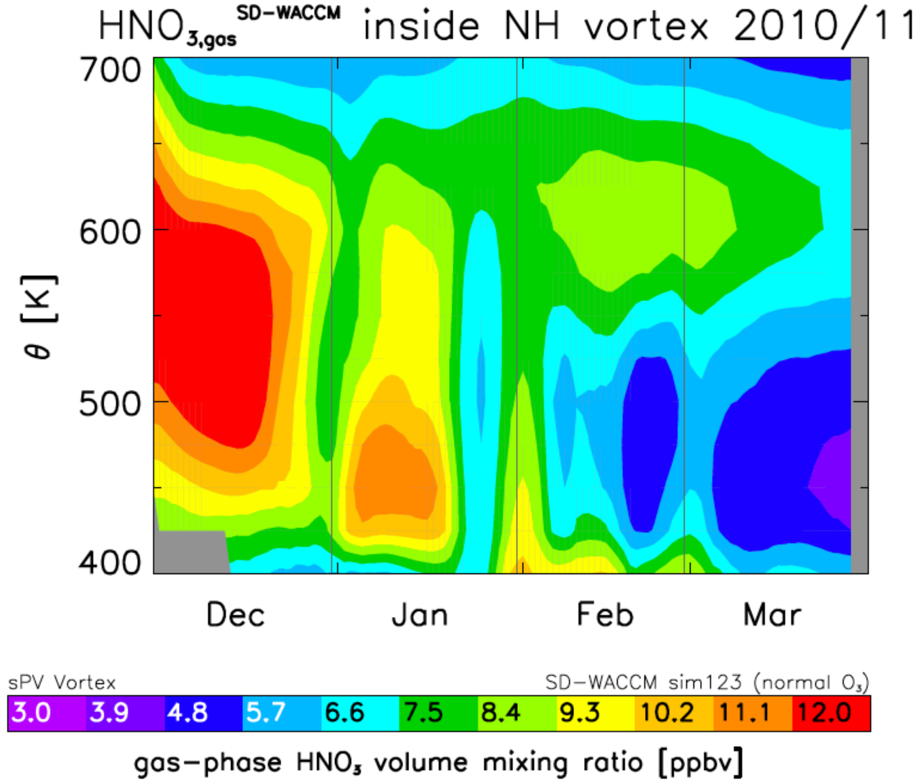


HNO_3 Evolution – no Bias

MLS

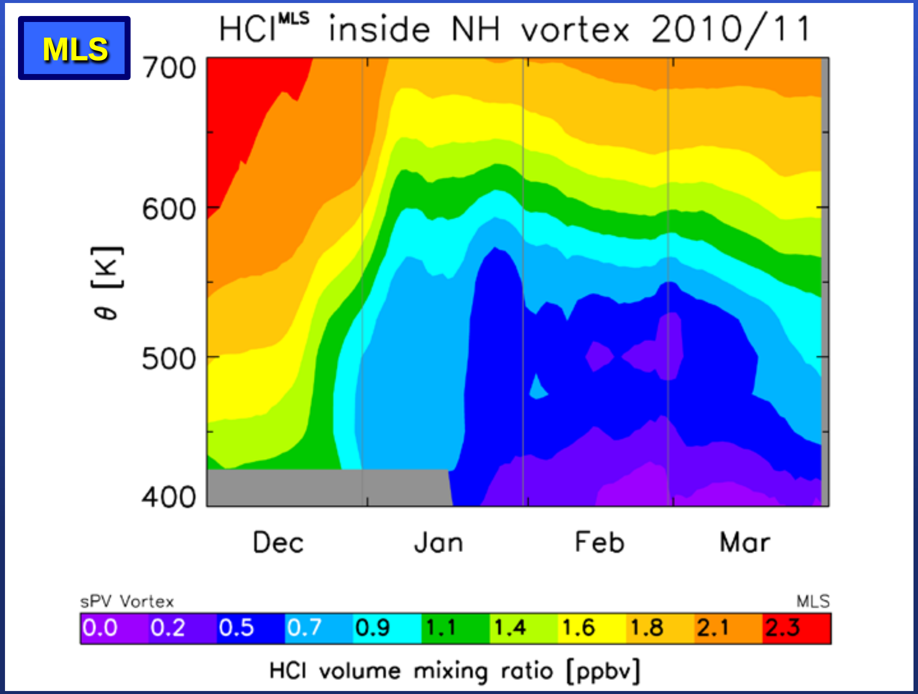


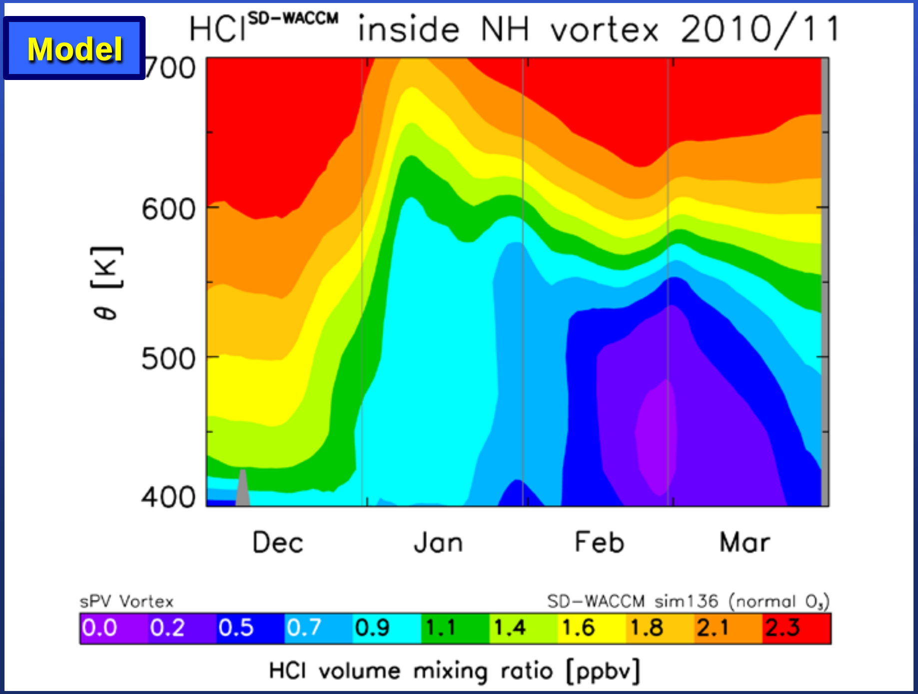
Model

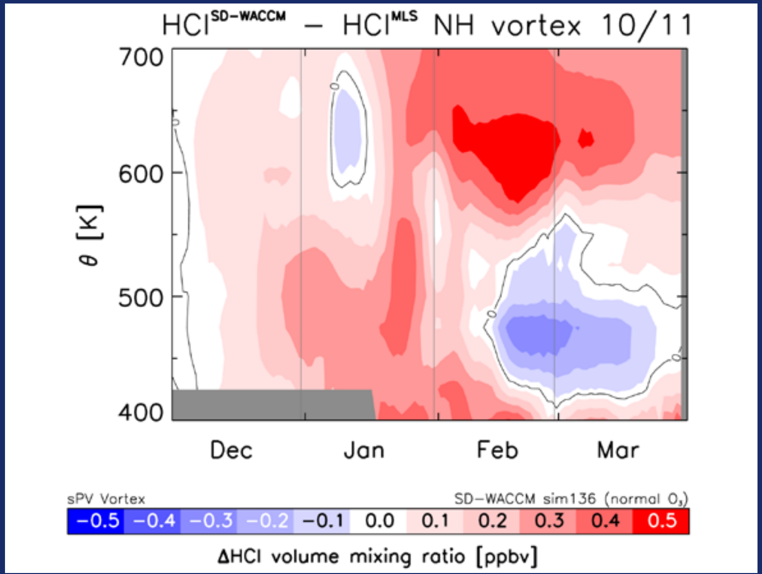


NOTE: These results are from the first attempt at representing 2011.

HCl Evolution – no Bias

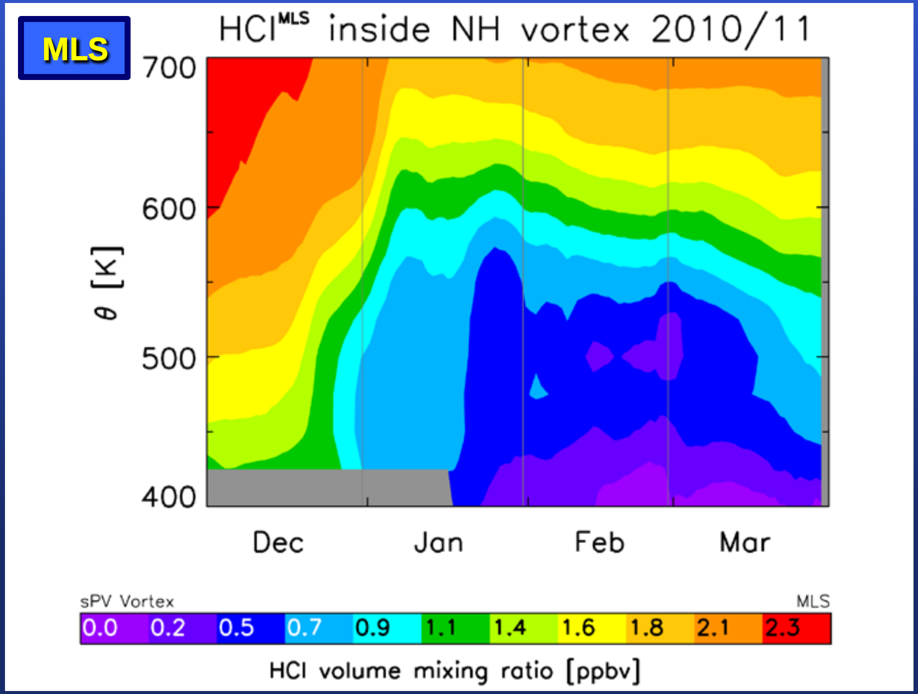


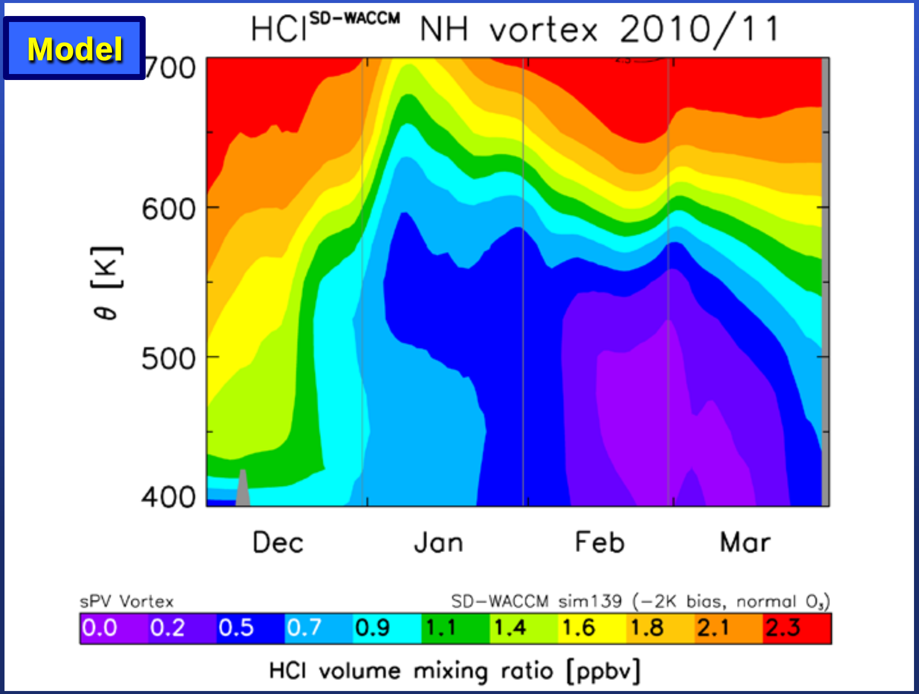


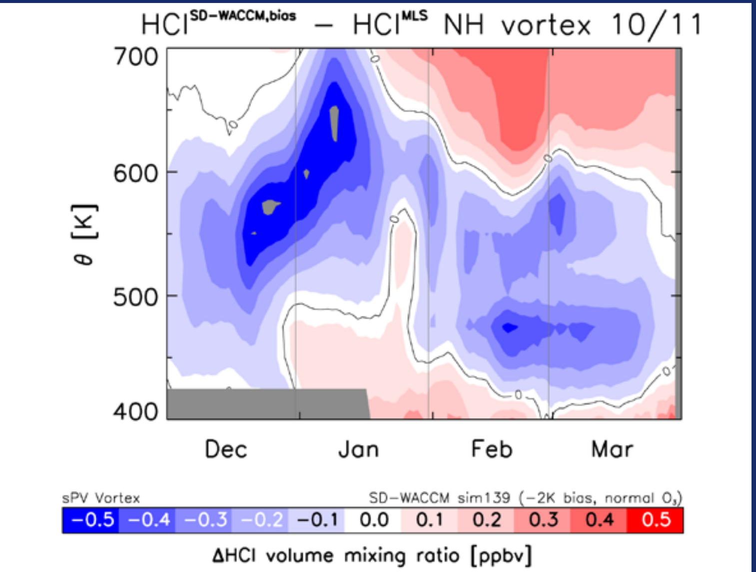


<= Model - MLS

HCl Evolution *** -2K Bias



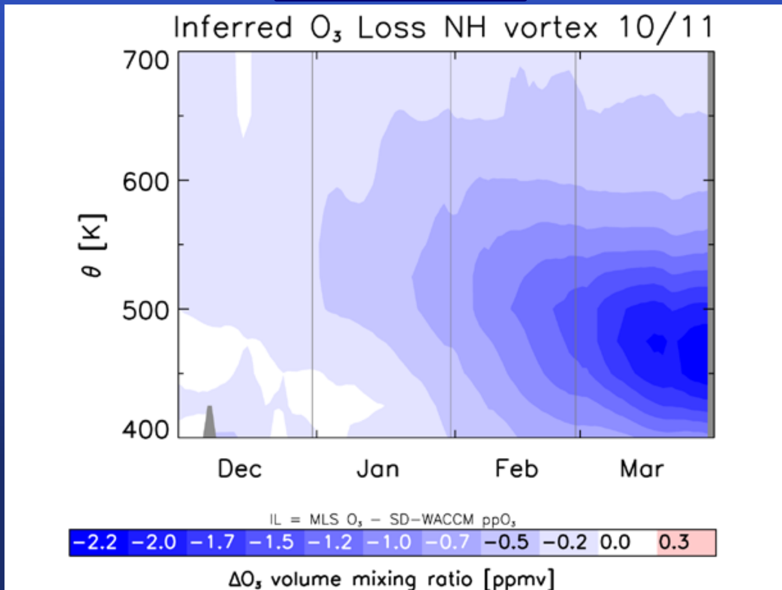




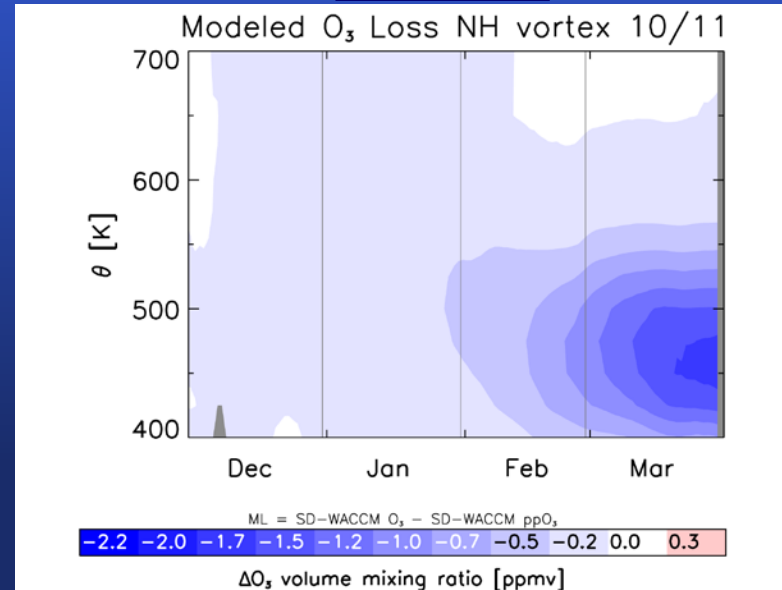
<= Model - MLS

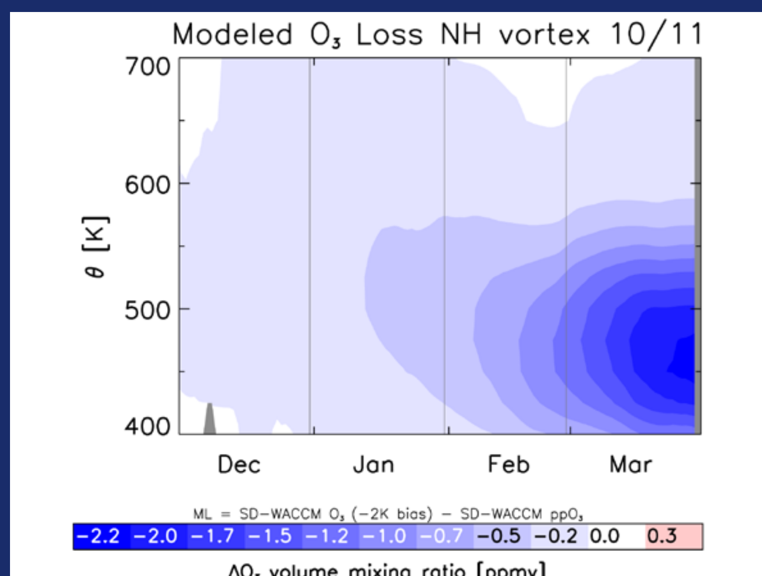
Chemical Ozone Loss

Inferred O3L



Model O3L

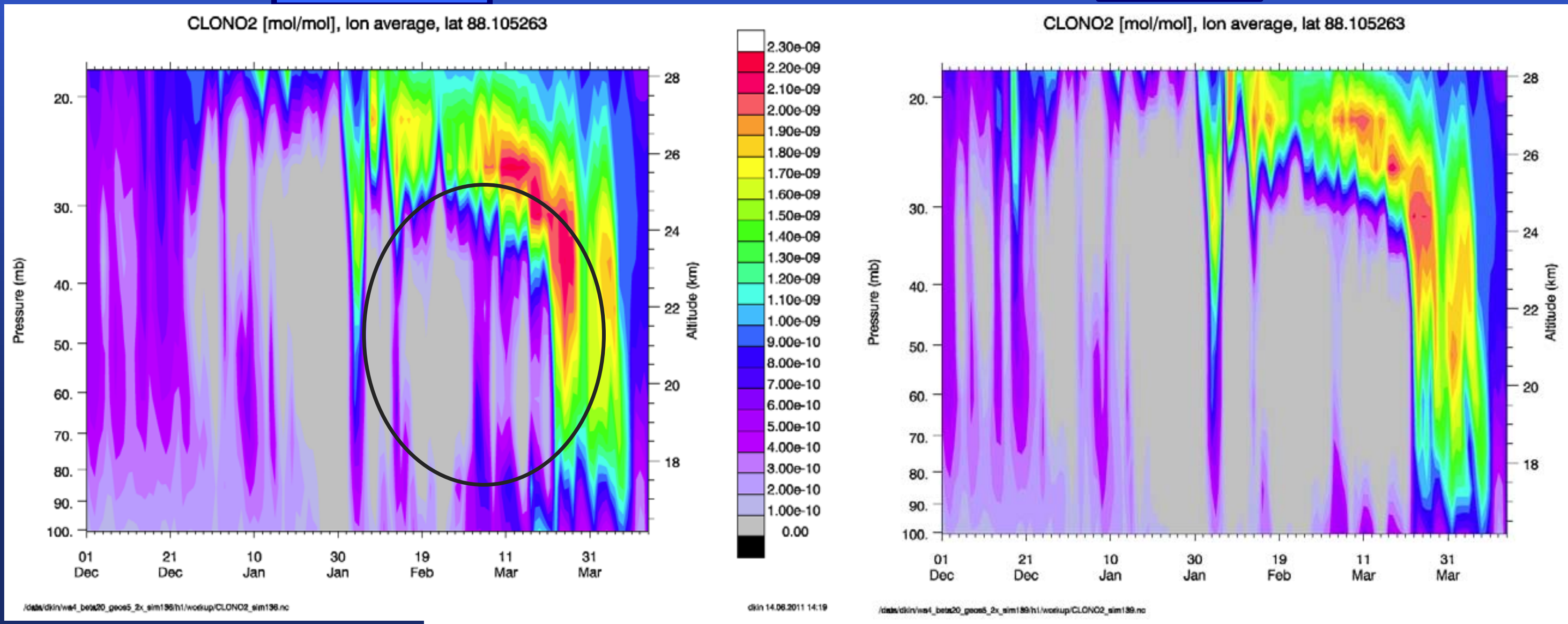


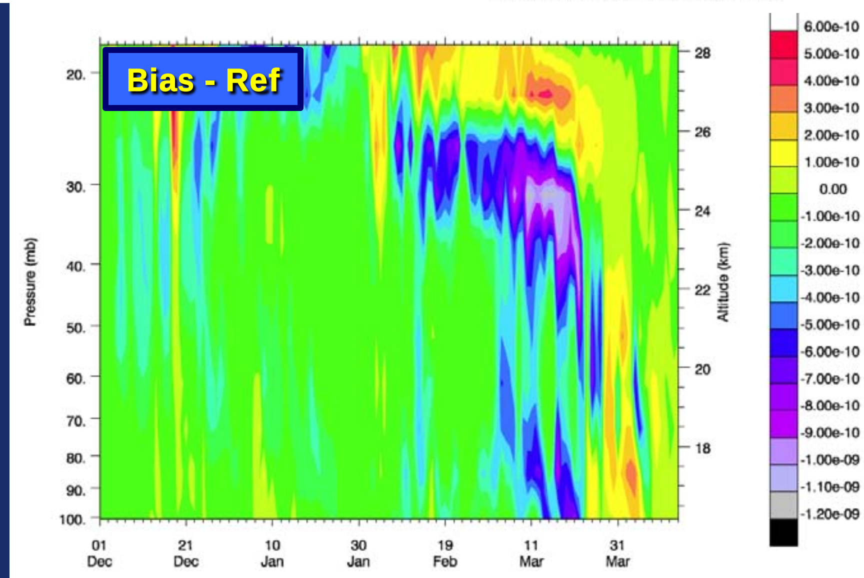


Model O3L with -2K Bias

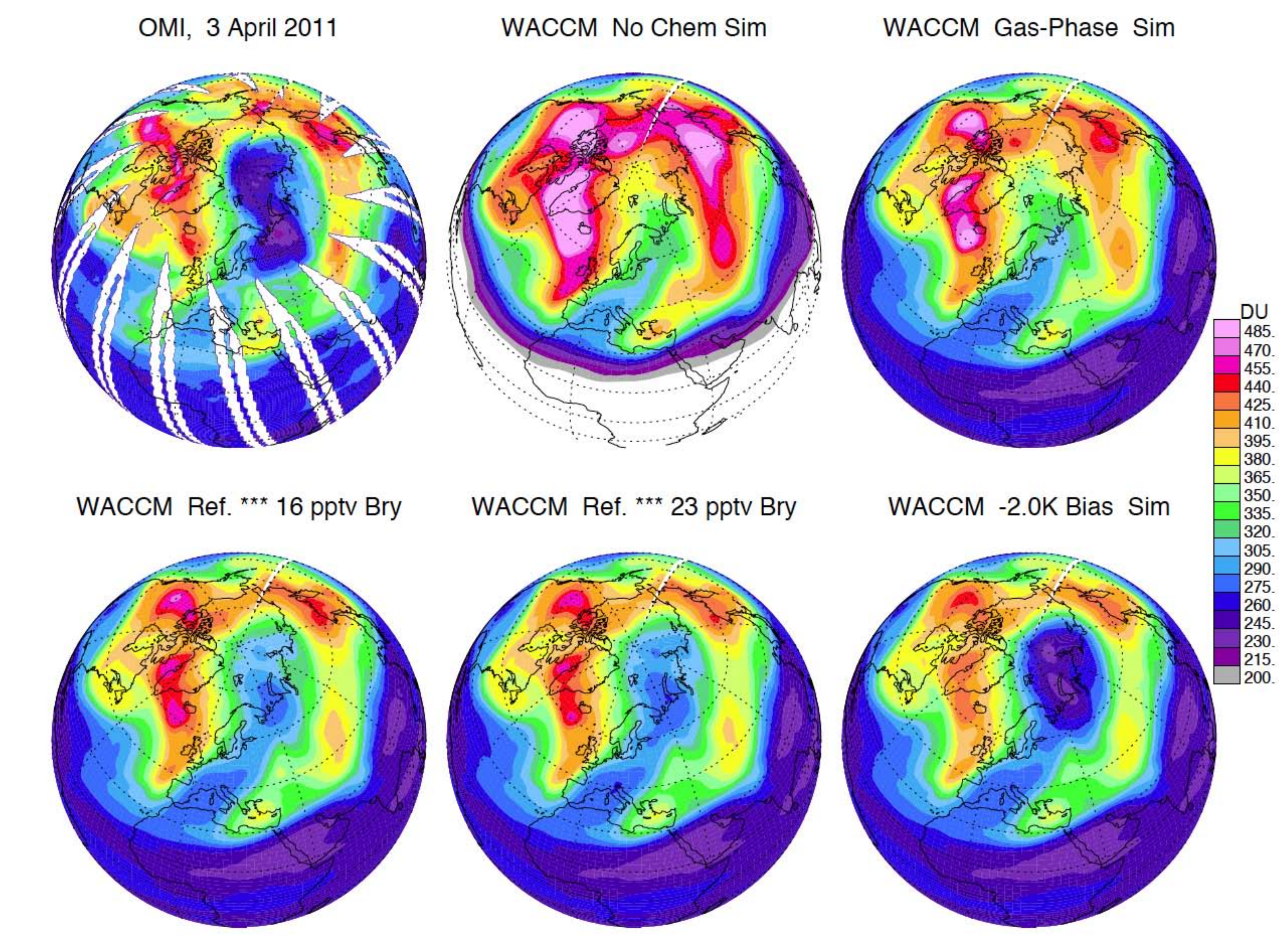
The Role $ClONO_2$ *** 88N, zonal mean

Reference **-2K Bias**





Total Column Ozone (DU)



Summary / Future work

New heterogeneous module…

- The “New” approach gives more STS (and NAT) SAD (relative to the IPCC simulation module).
- Ozone depletion in the “New” approach is > and < than the standard approach depending on period and altitude.
- Both SAD modules are available in the released version of WACCM.

2010/2011 Ozone Loss….

- In 2011, the MLS / model ozone and ozone loss comparisons are best represented when the model includes a -2K bias in temperature.

Next Step

New heterogeneous module…

- Write up the new heterogeneous approach in a WACCM chemistry update paper (Summer 2011).

2010/2011 Ozone Loss…

- Are we getting the correct heterogeneous processing on PSCs?
    - Examine the $ClONO_2$ (ACE-FTS) and HNO3 (MLS) recovery between model and observations.
- Is the temperature bias justified?
    - Evaluate model temperature profiles (with GPS and Sondes).
- Do other models give similar results?
    - Compare results to CLAMS simulations.

Thank you for your attention!