FLEXPART along flighttracks

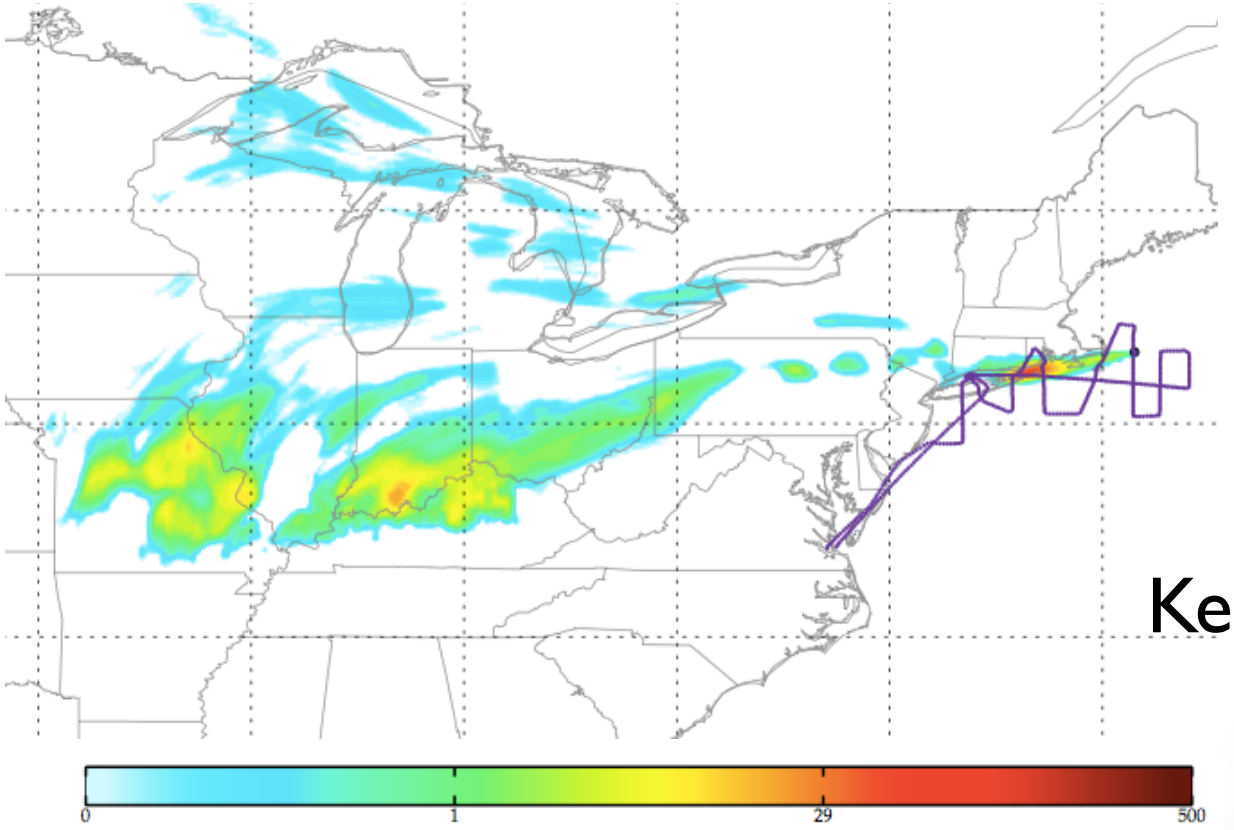
★Lagrangian particle dispersion model

★Use 0.5° GFS met fields

★Calculate Footprint Potential Emission Sensitivity (PES) in lowest 100 m

★Multiply by emissions (NOx) to get potential source contributions

★Example of PES based on 2-day backtrajectories for RF03:



Kelsey Larson

Modeling of emissions, chemistry, and transport of nitrogen oxides during WINTER

Lyatt Jaeglé, Viral Shah, Jessica Haskins, Kelsey Larson
Joel Thornton, Felipe Lopez-Hilfiker, Ben Lee
University of Washington, Department of Atmospheric Sciences
and WINTER Science Team

- *How well do we know anthropogenic emissions of NOx?*
- *What is the partitioning of reactive nitrogen species? How is it affected by day/night/multiphase chemistry?*

The GEOS-Chem model

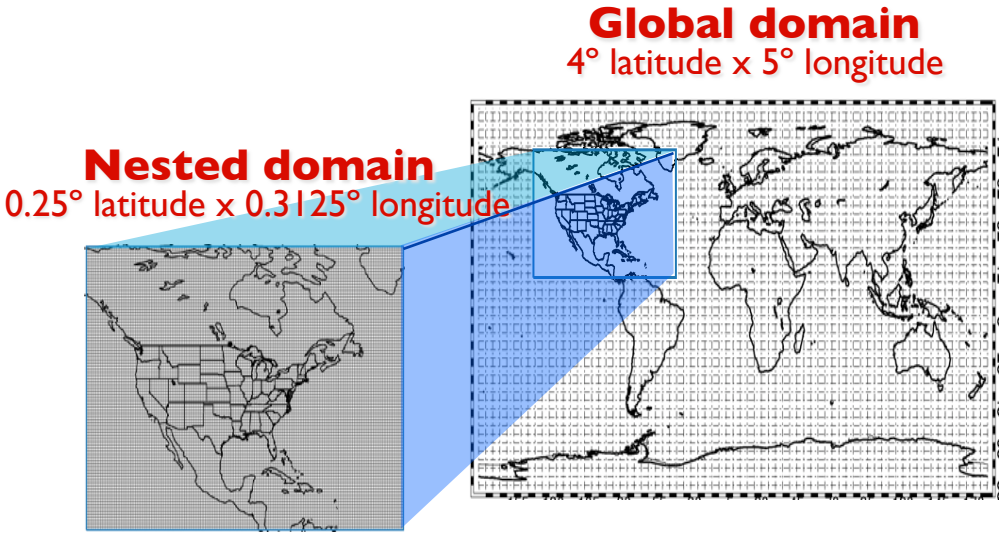
★Global 3D model 4°x5° resolution, 47 vertical levels

★Nested grid simulation 0.25° x 0.3125° (~25 km) over N. America

★GEOS-FP assimilated meteorology NASA GMAO

★Aerosol-oxidant simulation (GEOS-Chem v10.01)

- Oxidant chemistry: HO_x-NO_x-O_3-VOC-BrO_x
- Aerosols: SO_4^{2-}-NH_4^+-NO_3^-, organic carbon (POA, SOA), BC, dust, sea salt
- Emissions: Anthropogenic (NEI2011), vegetation, soils, fires, lightning

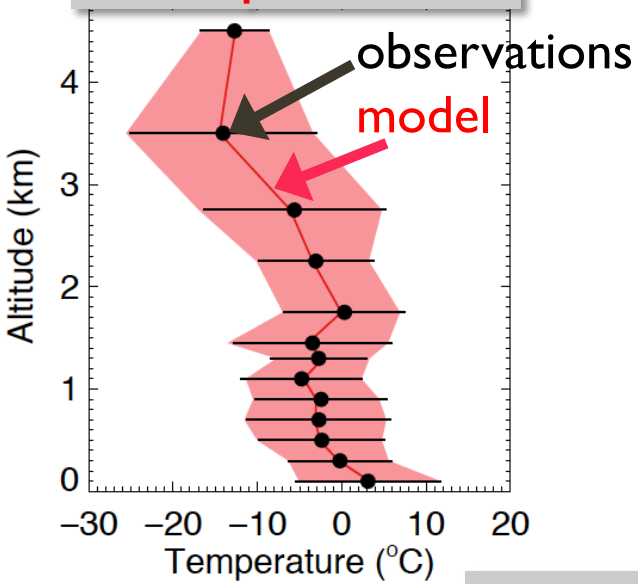


★Note: the current merges include the Near-Real-Time GC simulation (v9-02, NEI2008x0.5, low PBLH)

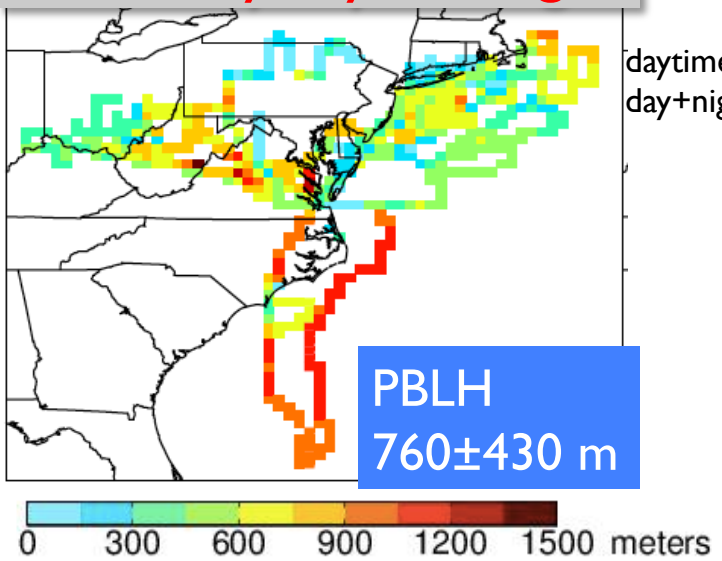
★The next version of the merges will include the updated GC simulation

Meteorological conditions

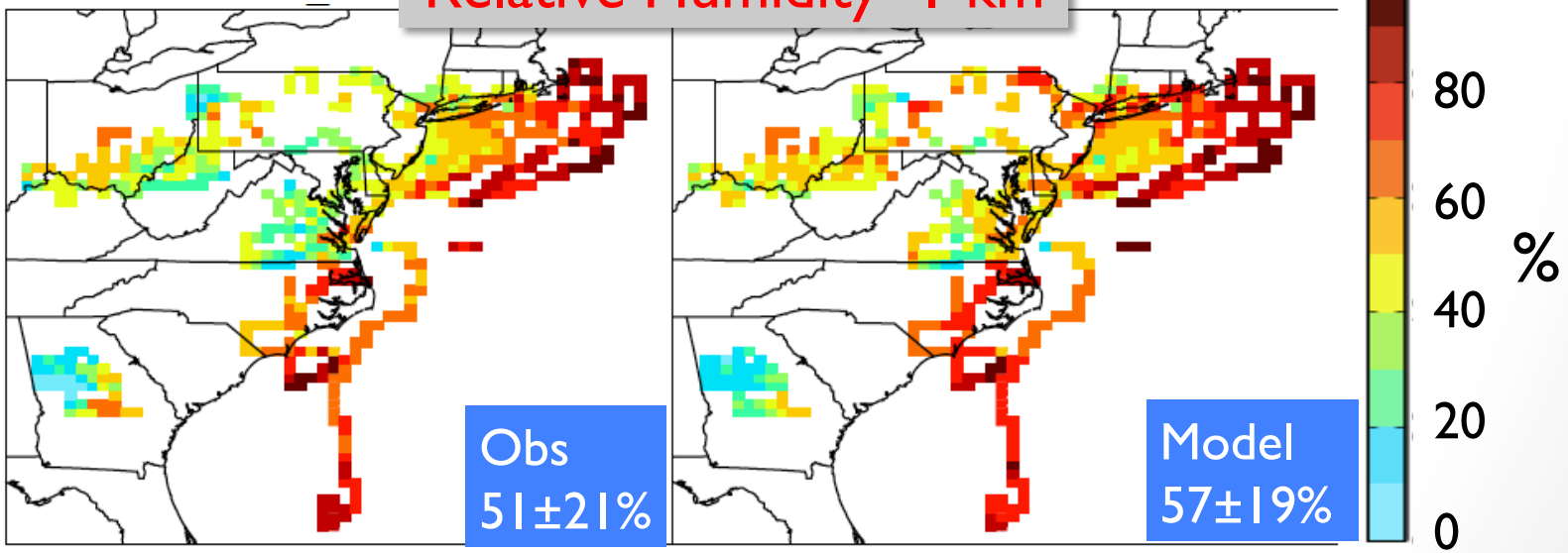
Temperature



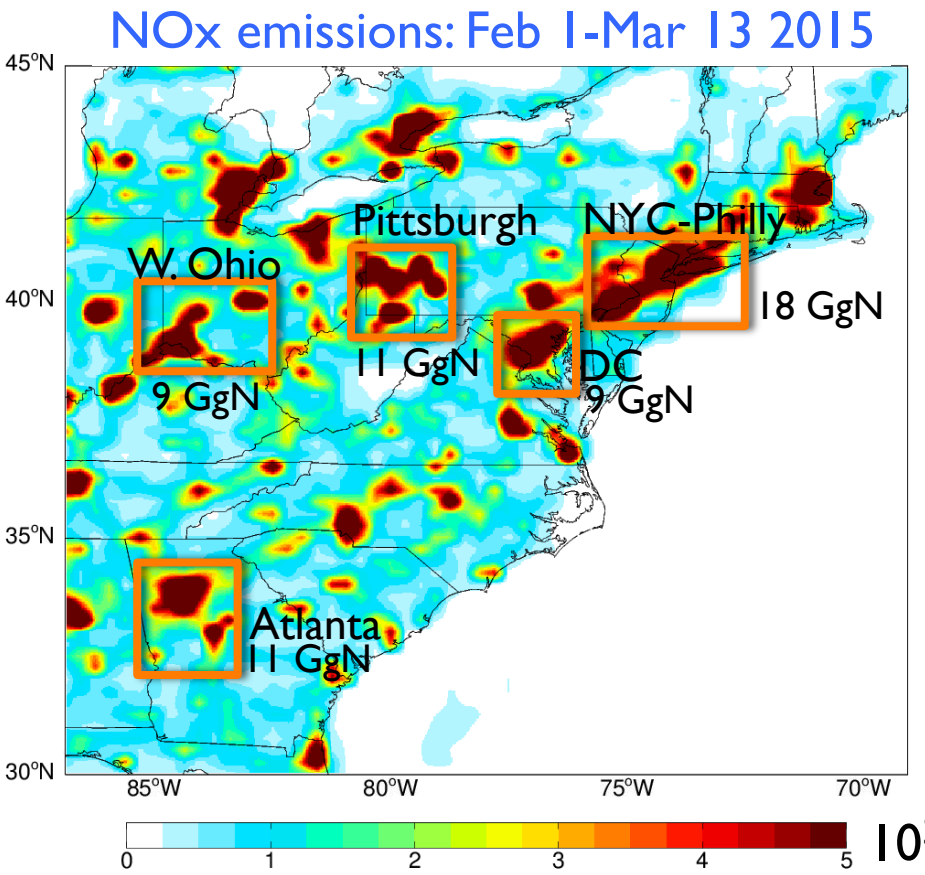
Boundary Layer Height



Relative Humidity<1 km



NOx emissions: Feb 1-March 13 2015



Base inventory: EPA NEI 2011

Scaled down by 20%

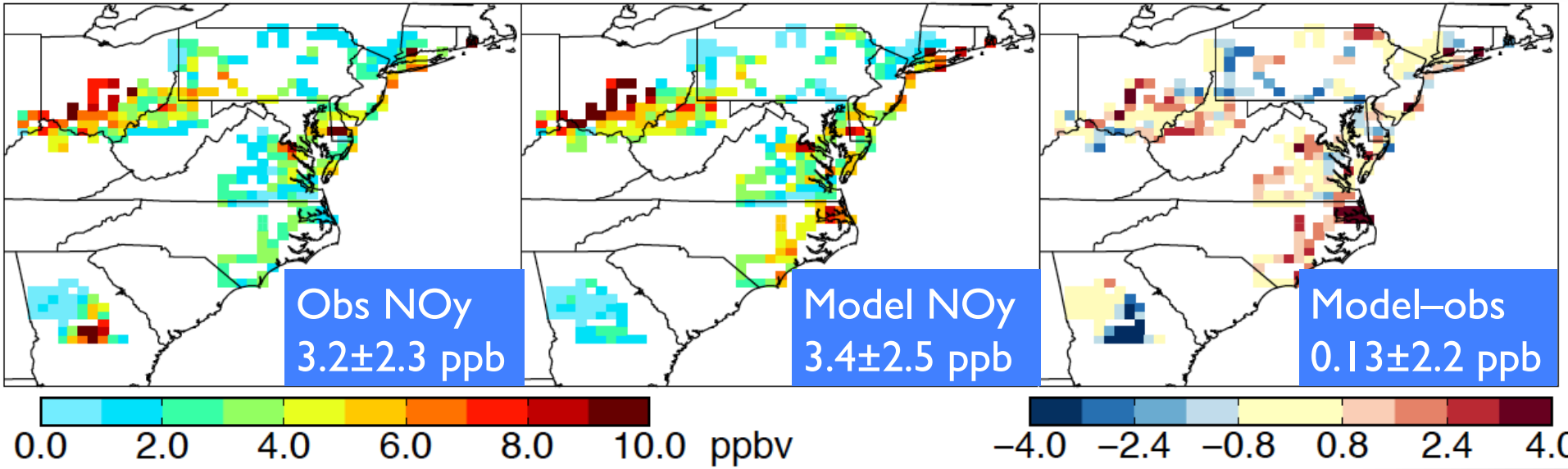
- based on 2015 Q1+Q2 CEMS: 25% lower NOx emissions from power plants relative to 2011
- Projected transportation emission decrease (7%/year)

Feb 1-Mar 13 2015, WINTER region:
Total NOx emissions = 155 GgN

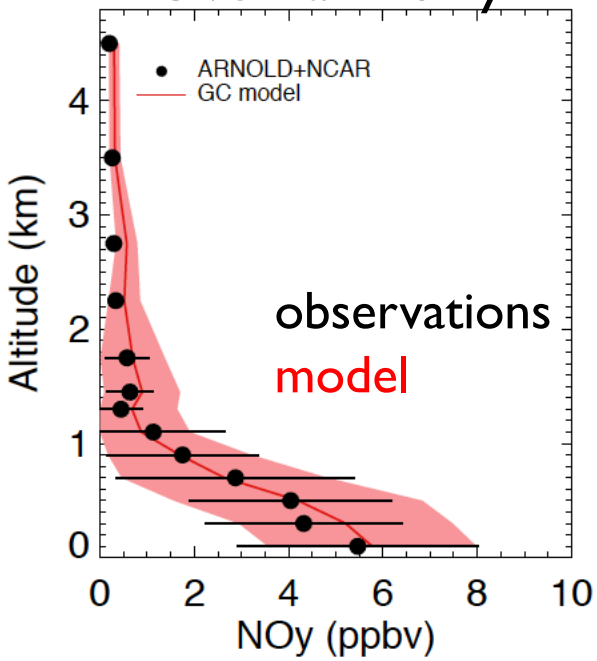
Entire U.S.:

- 53% transportation (onroad, offroad, aircraft)
- 25% stationary fuel combustion
- 11% industrial and other processes
- 3% soils
- 8% lightning

Comparison: NOy < 1 km altitude



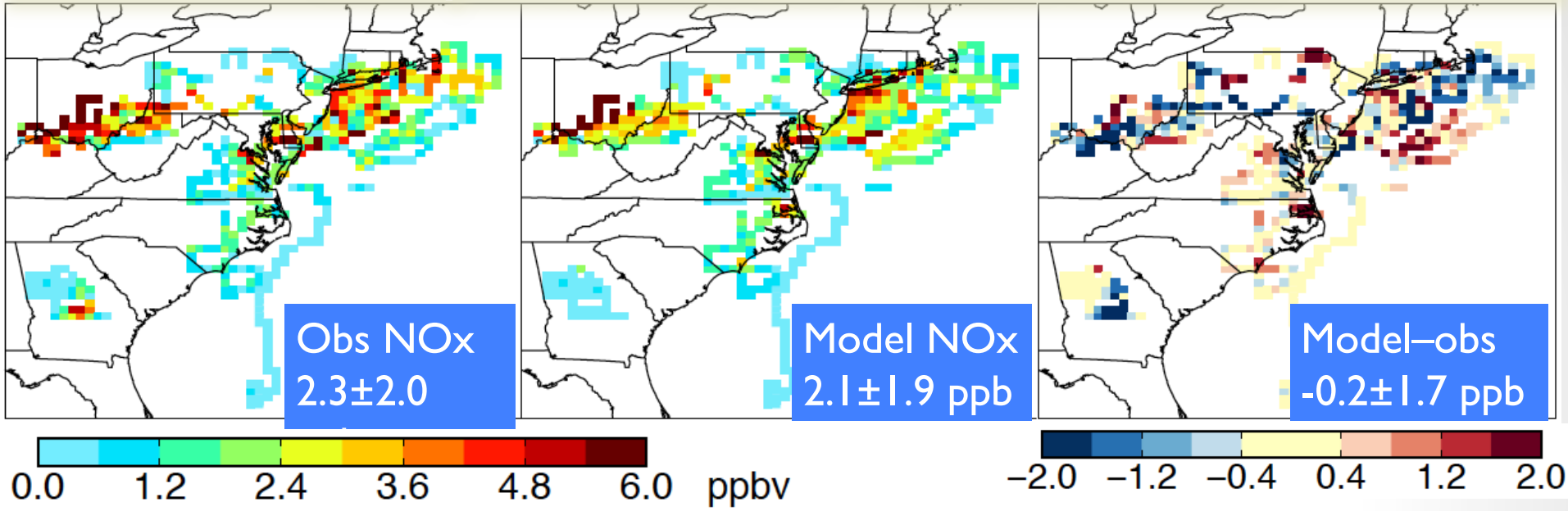
Over land only

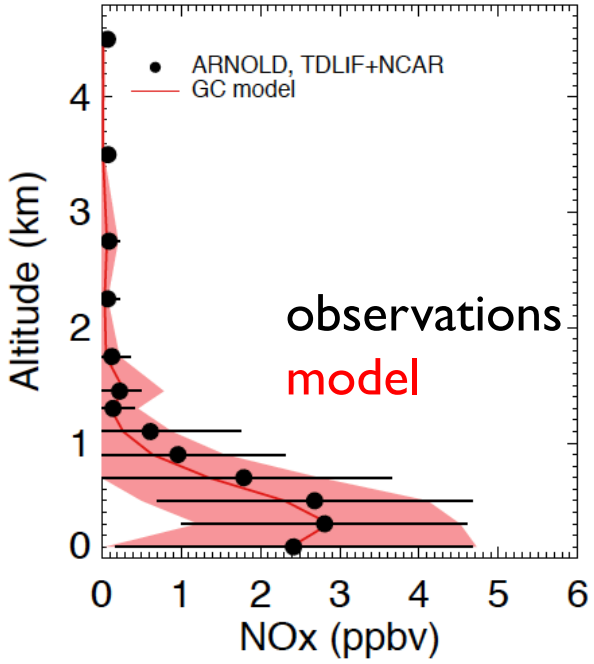


- Very good agreement for vertical distribution of NOy
- Below 1 km over land modeled NOy is ~10% higher than observations on average.

NO_y: ARNOLD + CL

Comparison: NOx < 1 km altitude

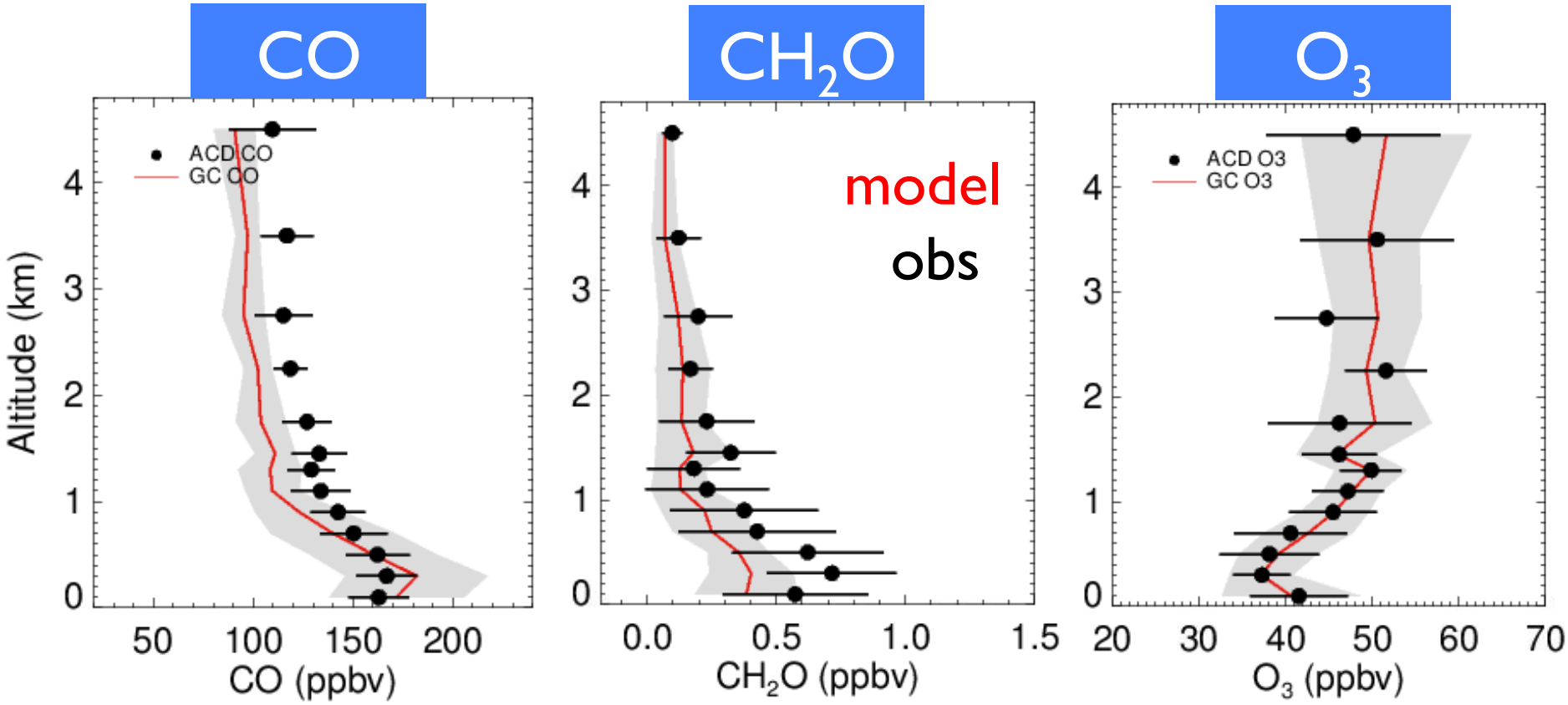




- Very good agreement for vertical distribution of NOx
- Below 1 km over land modeled NOx is ~10% lower than observations on average.

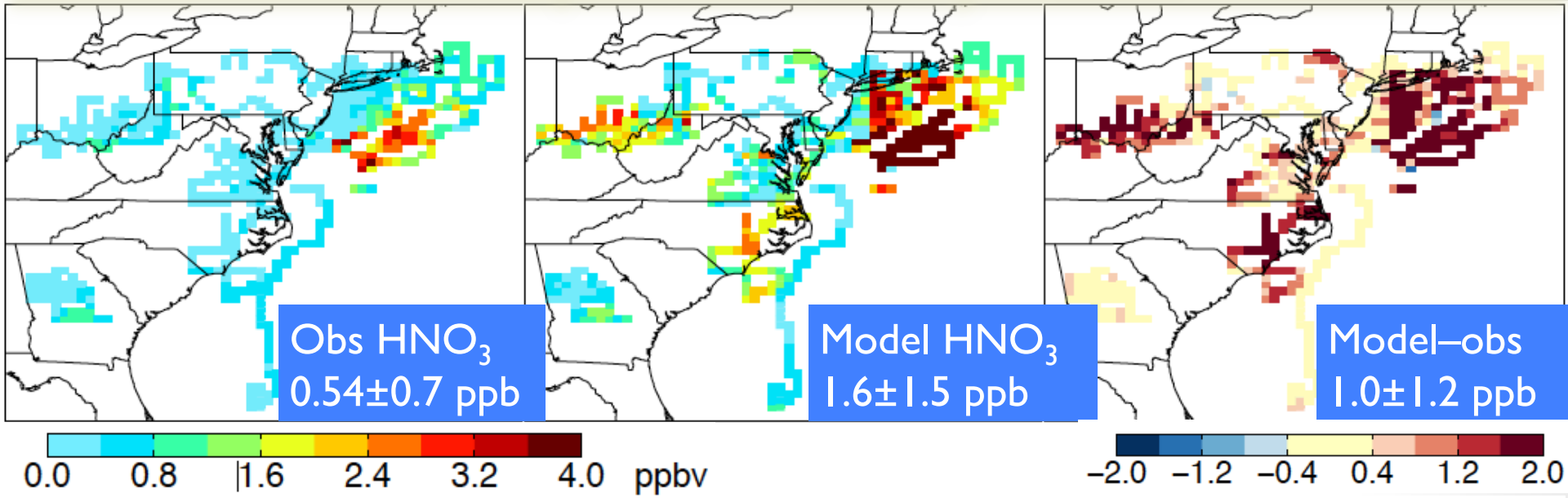
NO_x: ARNOLD + (TDLIF+CL)

Vertical profiles



CO & O_3: NCAR; CH_2O: ISAF

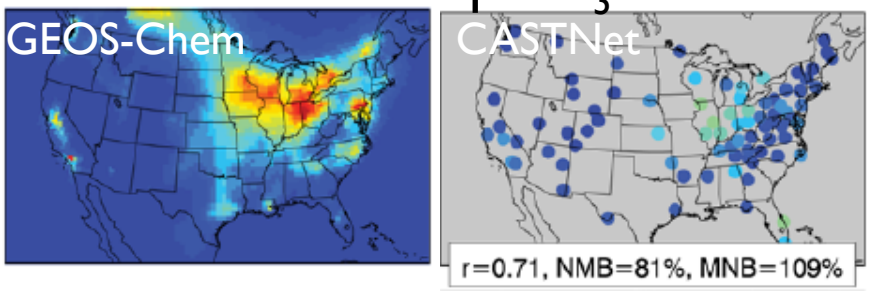
Comparison: HNO_3 < 1km



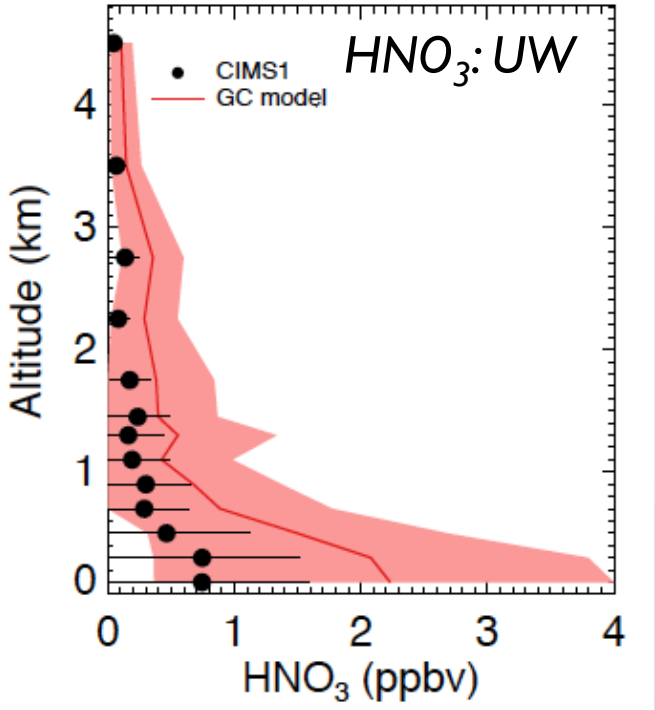
- Model HNO_3 is 2-3 times too high!
- Same for pNO_3 (see Viral's presentation)

→ *overestimate in HNO_3 production?*

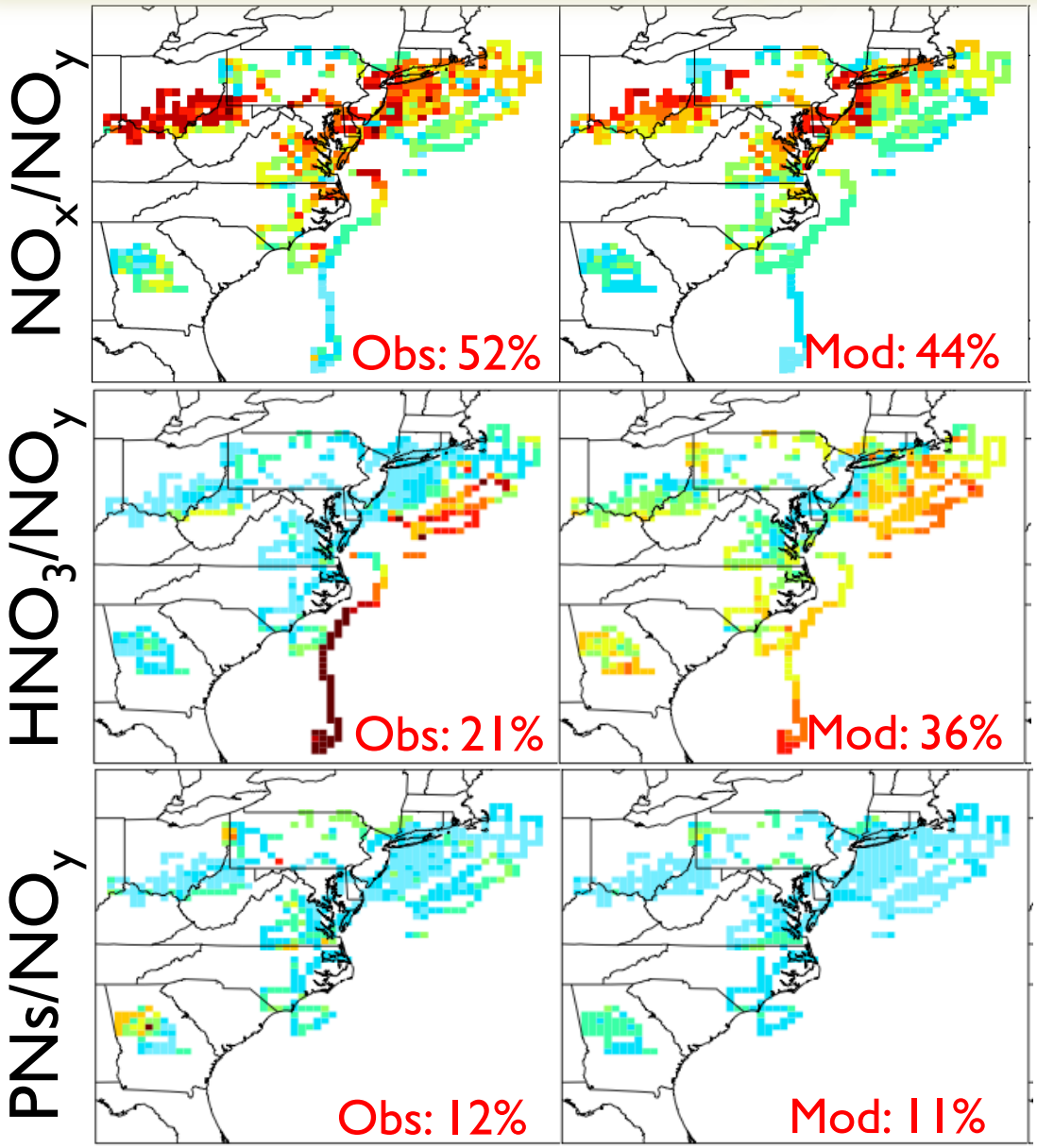
Surface pNO_3



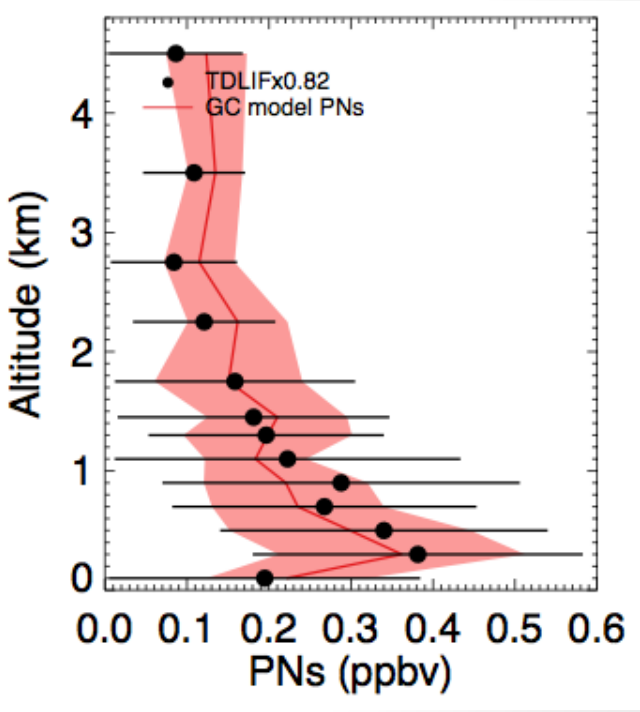
Zhang et al. (2012)



NOy partitioning

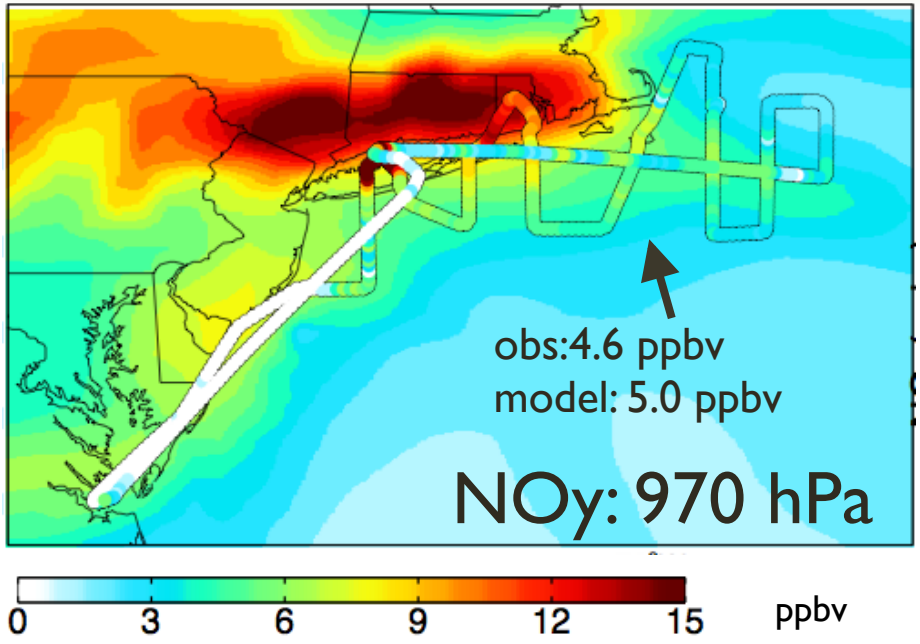


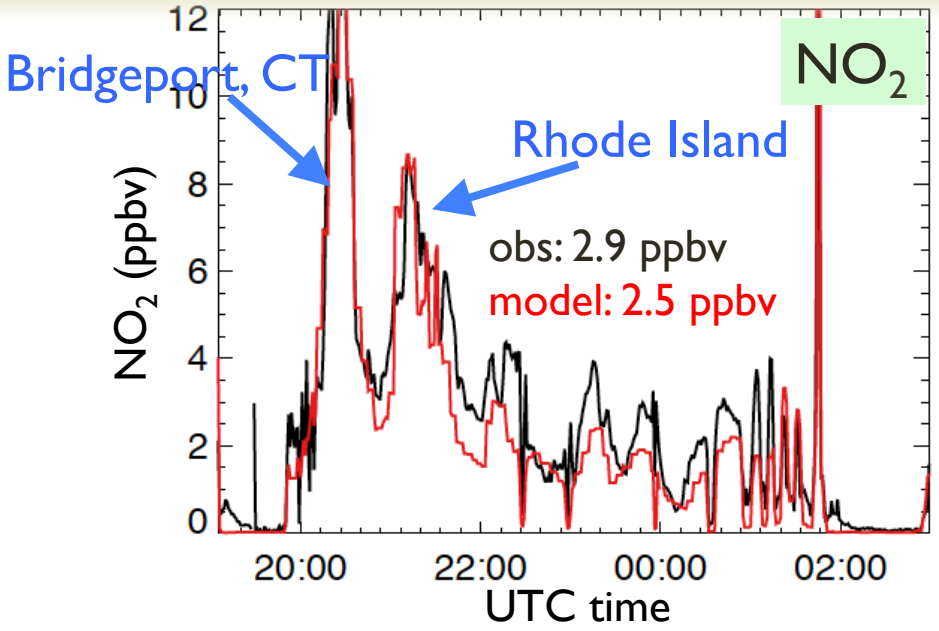
- NOx/NOy ranged from 90% to 10%, with an average of 52%. Model too low by 15%
- PNs: good agreement
- Too much HNO_3!

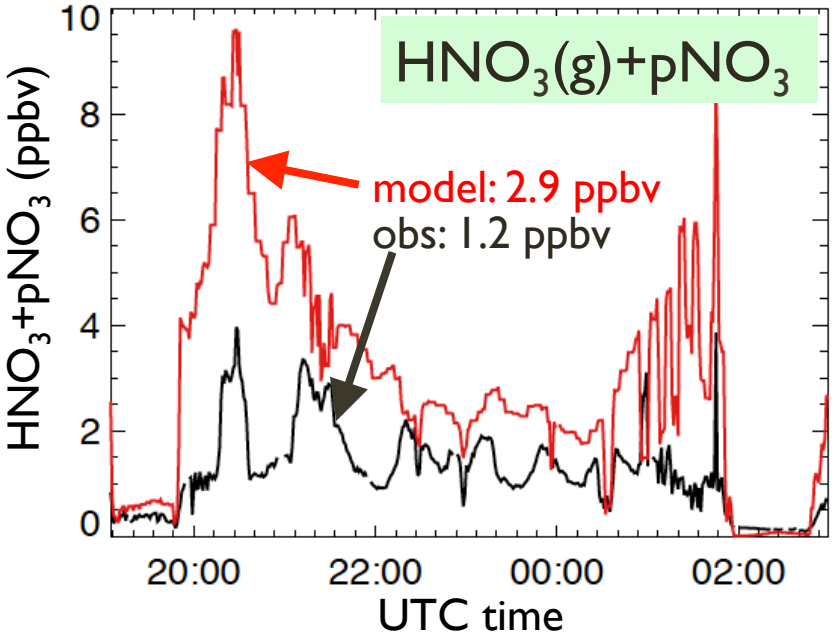


NO_y: ARNOLD+CL, HNO3: UW, PNs: TD-LIF

RF03 - Pollution outflow: Feb 7, 2015





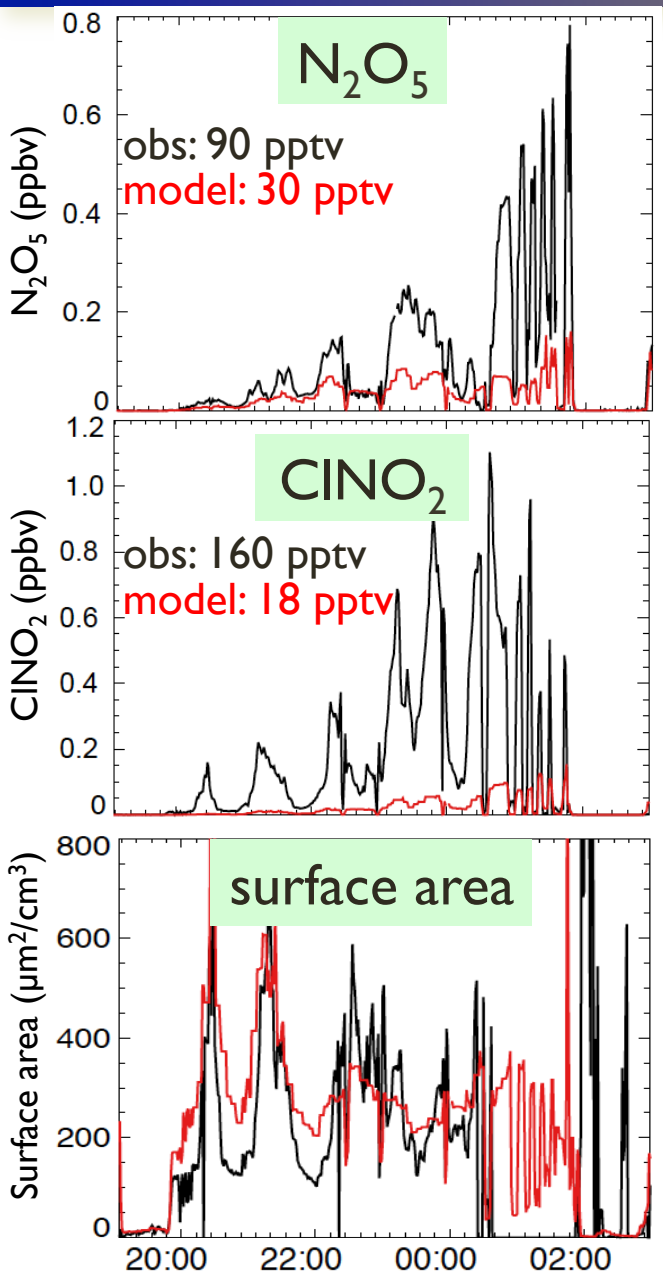


- Good agreement for NOx and NOy
- Model HNO_3+pNO_3 too high by x2

→ *overestimate in HNO_3 production?*

NOx, NOy: ARNOLD; HNO_3: UW, pNO_3: AMS

Nighttime NOx chemistry: Feb 3, 2015



Nighttime chemistry:

$NO_2 + O_3 \rightarrow NO_3$

$NO_3 + NO_2 \rightleftarrows N_2O_5$

N_2O_5 + aerosol $\rightarrow (2-\phi)HNO_3 + \phi ClNO_2$

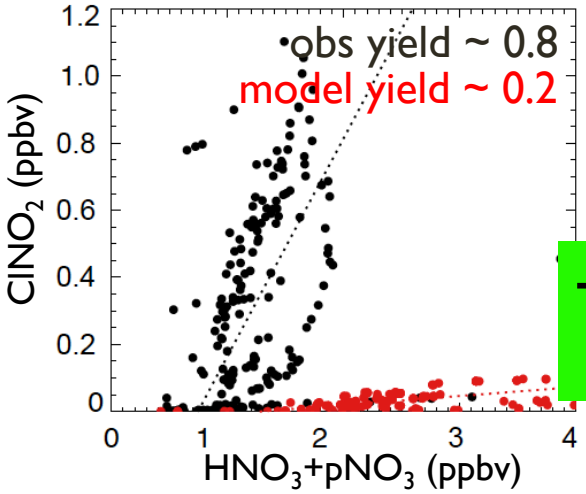
In GEOS-Chem:

$\gamma(N_2O_5)$ = f(RH, aerosol type, T) ~ 0.02 for RF03

Evans & Jacob (2005), Zhang et al. (2012)

ϕ = 1 on sea salt aerosol and 0.2 on other aerosols

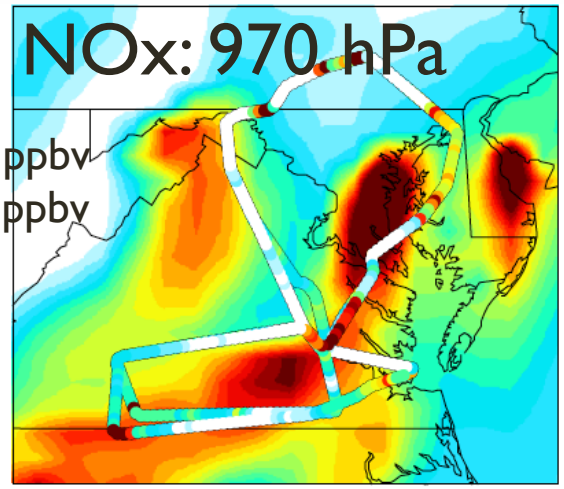
→ Model γ (N_2O_5) too high by a factor of 2-3

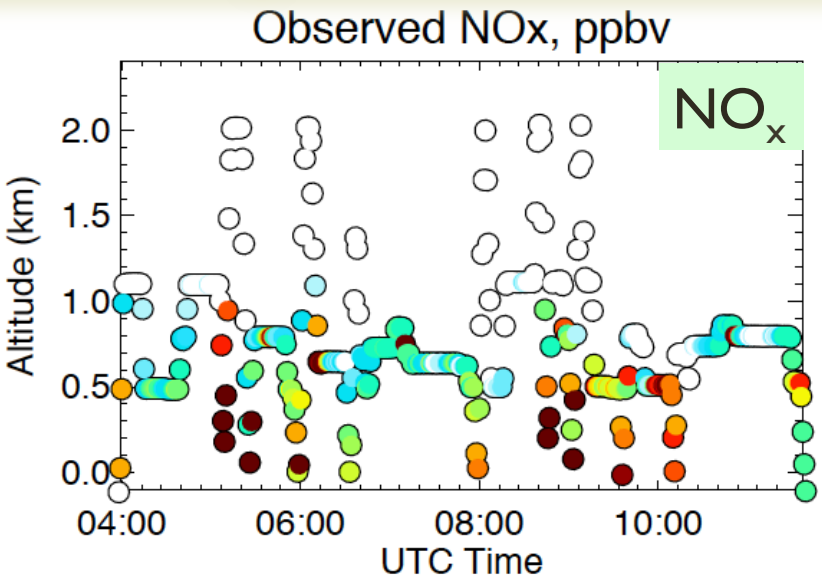


→ Model $\phi(ClNO_2)$ too low by a factor of 5

N_2O_5:ARNOLD+UW, $ClNO_2$:UW, pNO_3:CU, SA: RAF

RF07 – Virginia/Maryland night: Feb 24, 2015

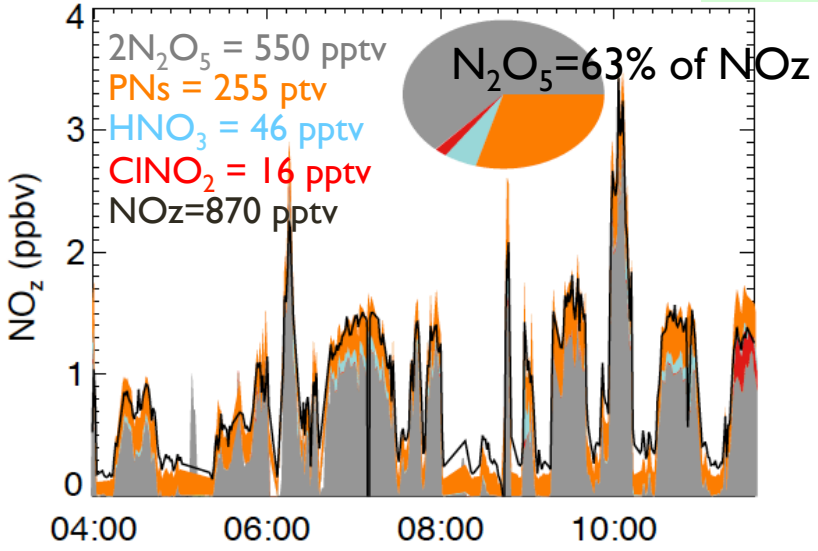


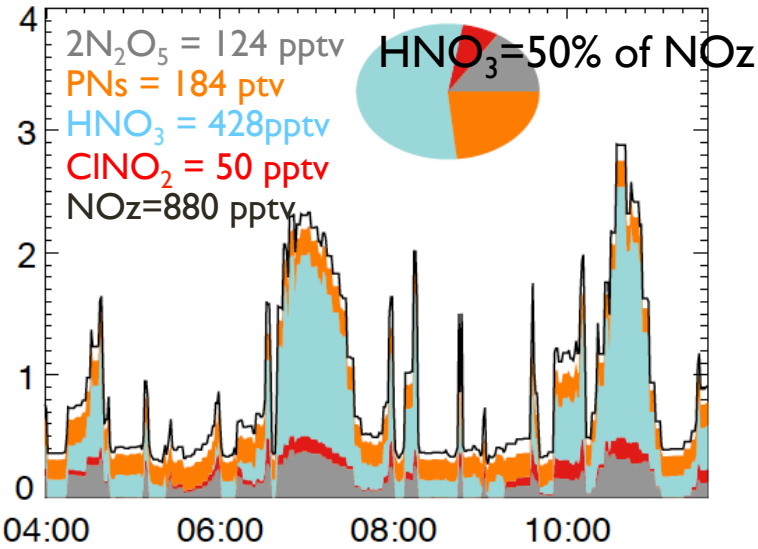


Observations

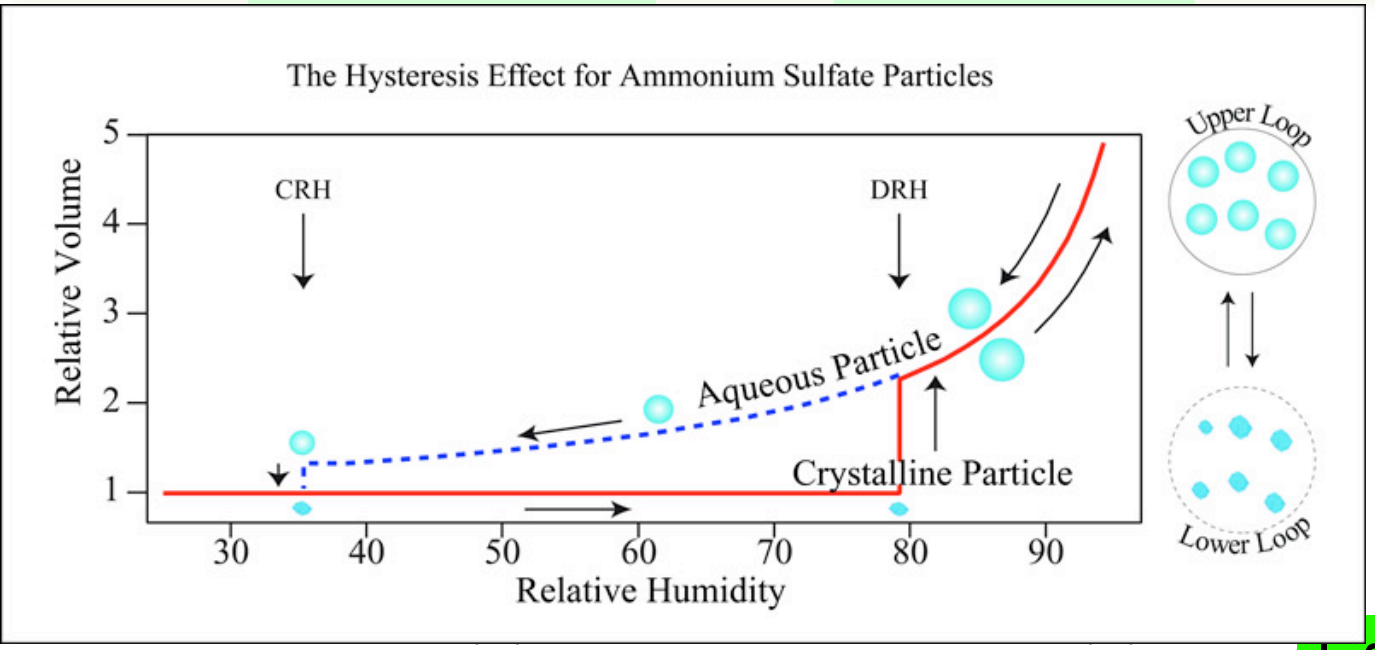
$NOz = NO_y - NO_x$

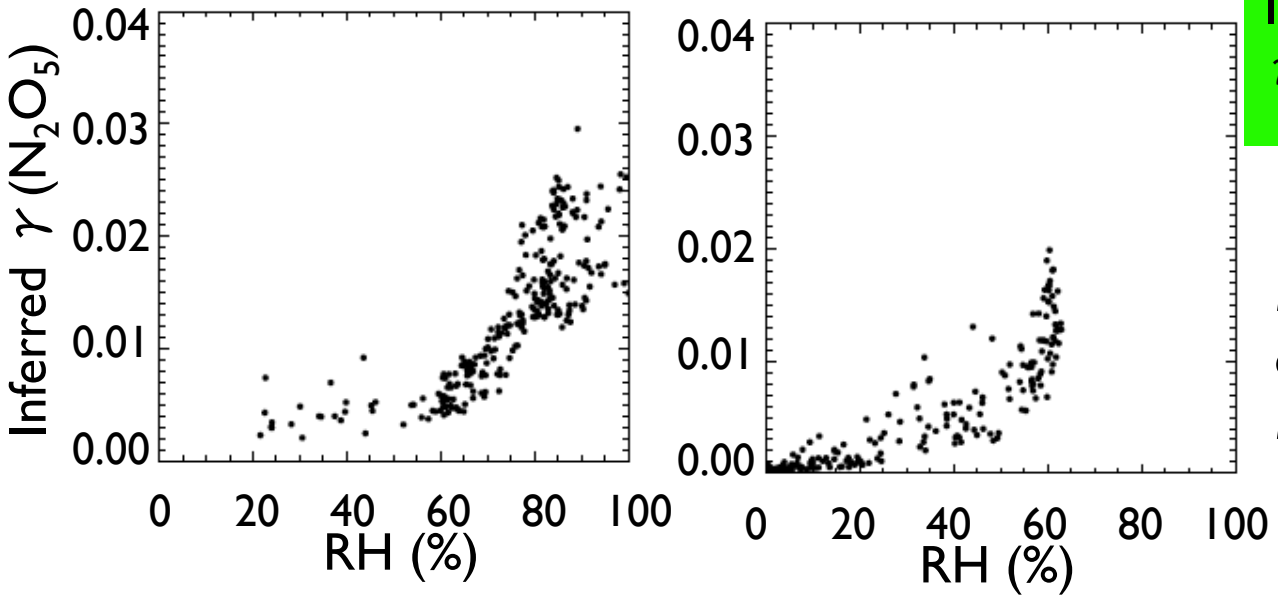
GEOS-Chem





Why is HNO_3 production slow?





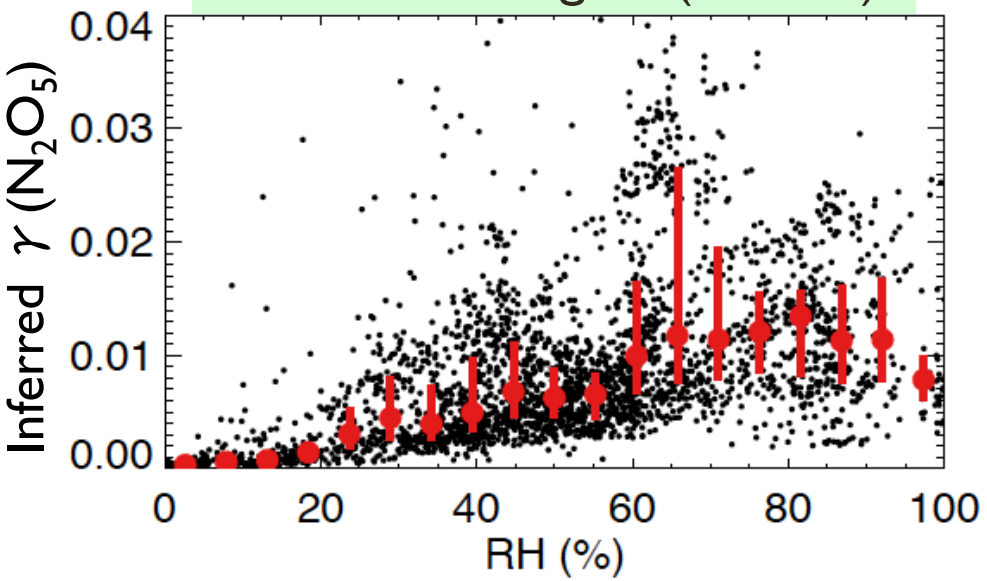
Infer γ (N_2O_5)=

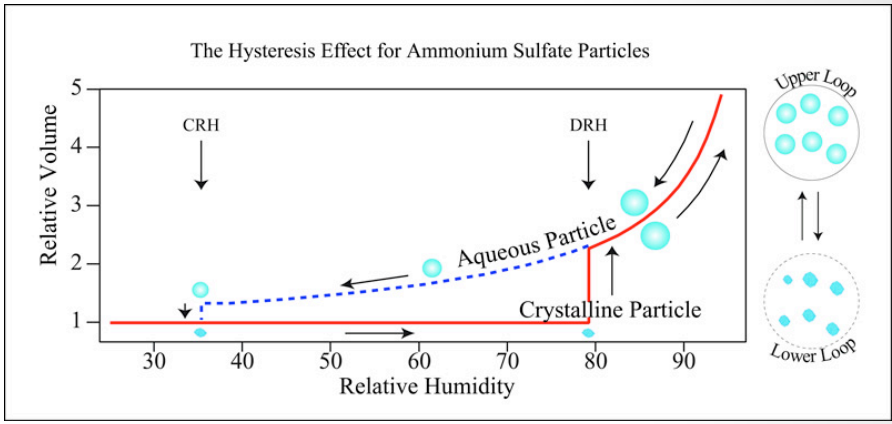
γ_{model} x obs (HNO_3+$ClNO_2$+pNO_3)
model(HNO_3+$ClNO_2$+pNO_3)

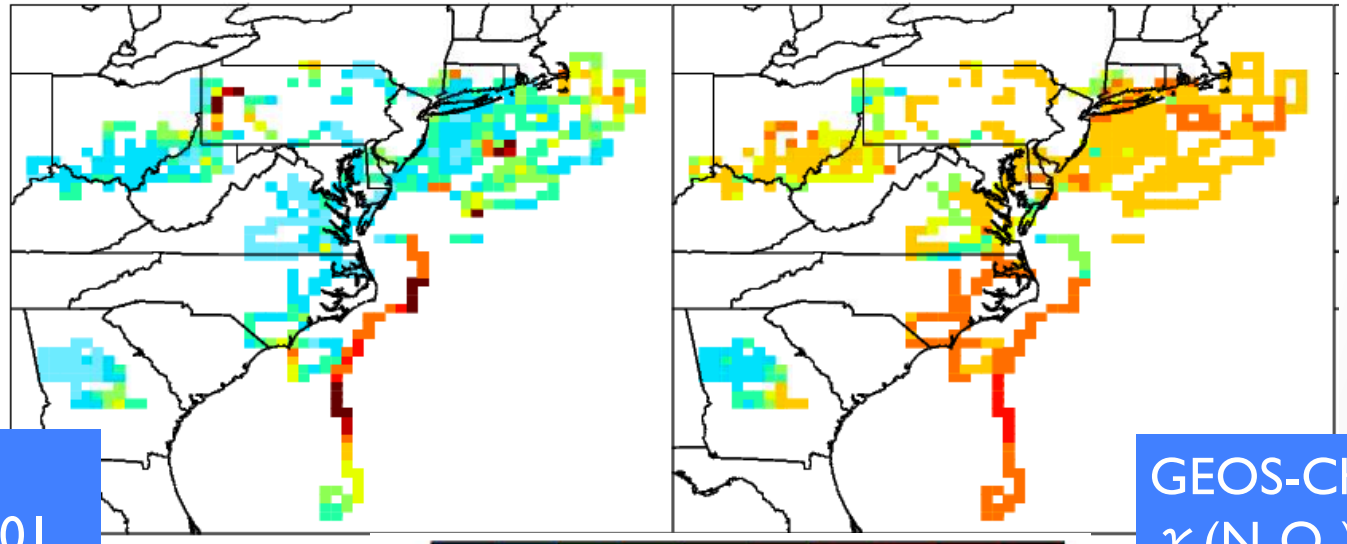
N_2O_5 uptake suppressed on crystalline particles at low RH?

An attempt at inferring γ (N_2O_5) during all flights

All WINTER flights (< 1 km)







Inferred γ (N_2O_5)=0.01

GEOS-Chem γ (N_2O_5)=0.02

Next steps

- Why is N_2O_5 aerosol uptake so slow? (Jessica Haskins)
- What are the implications of a lower γ (N_2O_5) on NOy lifetime and export?
- What is the yield of $ClNO_2$? How does it affect the NOx lifetime, O_3 and VOCs?
- How to the aircraft observations relate to surface observations and satellite observations?