

Inverse Modeling of Ammonia Emissions

Daven K. Henze

2/23/09

Mechanical Engineering
University of Colorado at Boulder

Collaborators

Thanks to:



Mark Shepard, Karen Cady-Pereria,



Ming Luo, Kevin Bowman, TES team



Rob Pinder, John Walker



NASA GSFC: NCCS
NASA JPL: SCC

Importance of studying NH₃ emissions

PM_{2.5}

- Itself leads to NH₄⁺, 10-20% of PM_{2.5} mass concentration
- Governs formation of NO₃⁻, which can be 20-30% in winter

PM_{2.5} NAAQS Regulations

- Not a presumptively regulated species, but can be very efficient (Pinder et al., 2007; Henze et al., 2008)
- Can be regulated in place of SO₂ or NO₂

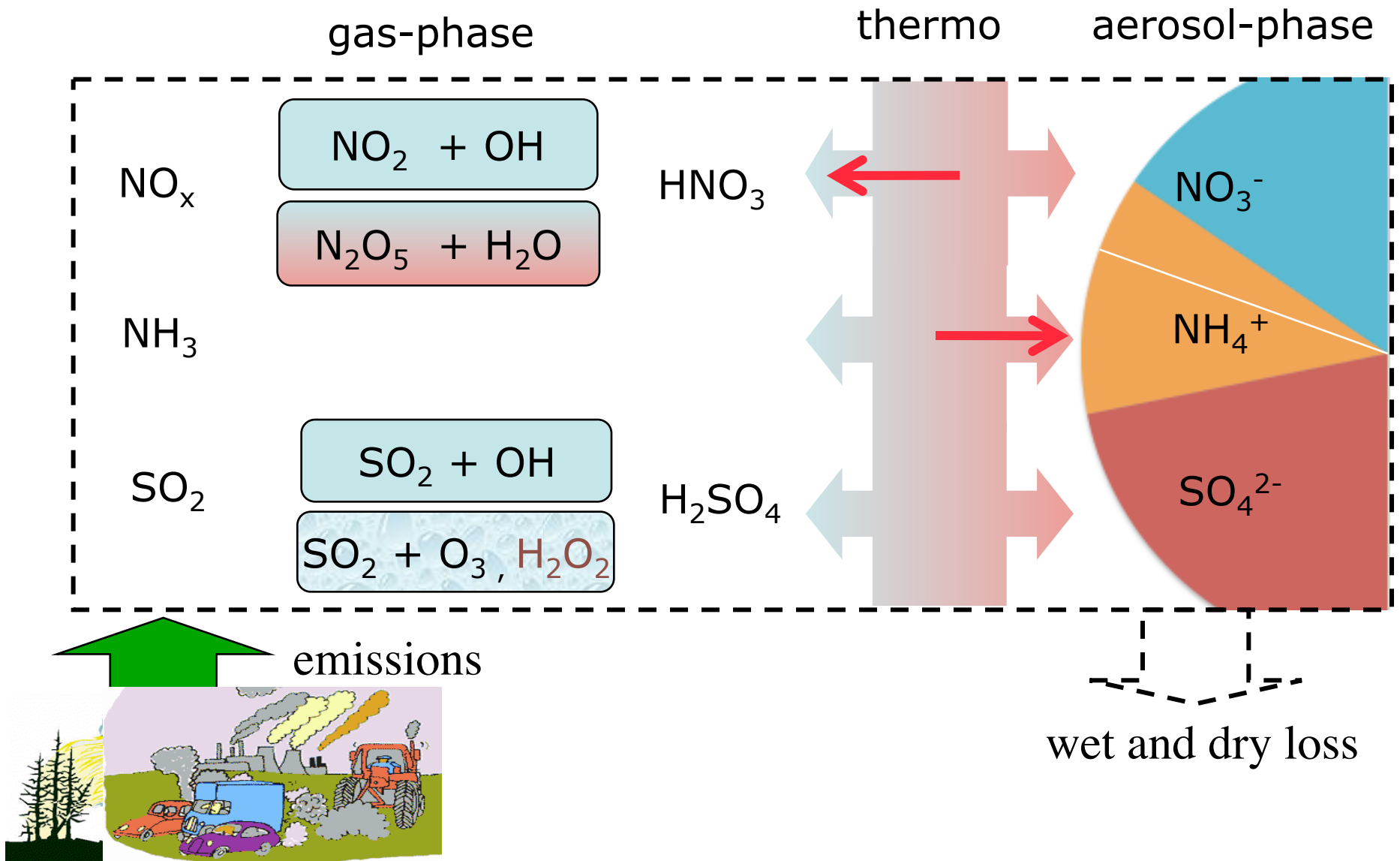
Ecosystem impacts

- 11% of worlds natural vegetation impacted by N dep (Dentener et al., 2006)
- N dep will increase 10-40% near NH₃ sources in U.S. by 2020 (Pinder et al., 2008)

Very large source of uncertainty

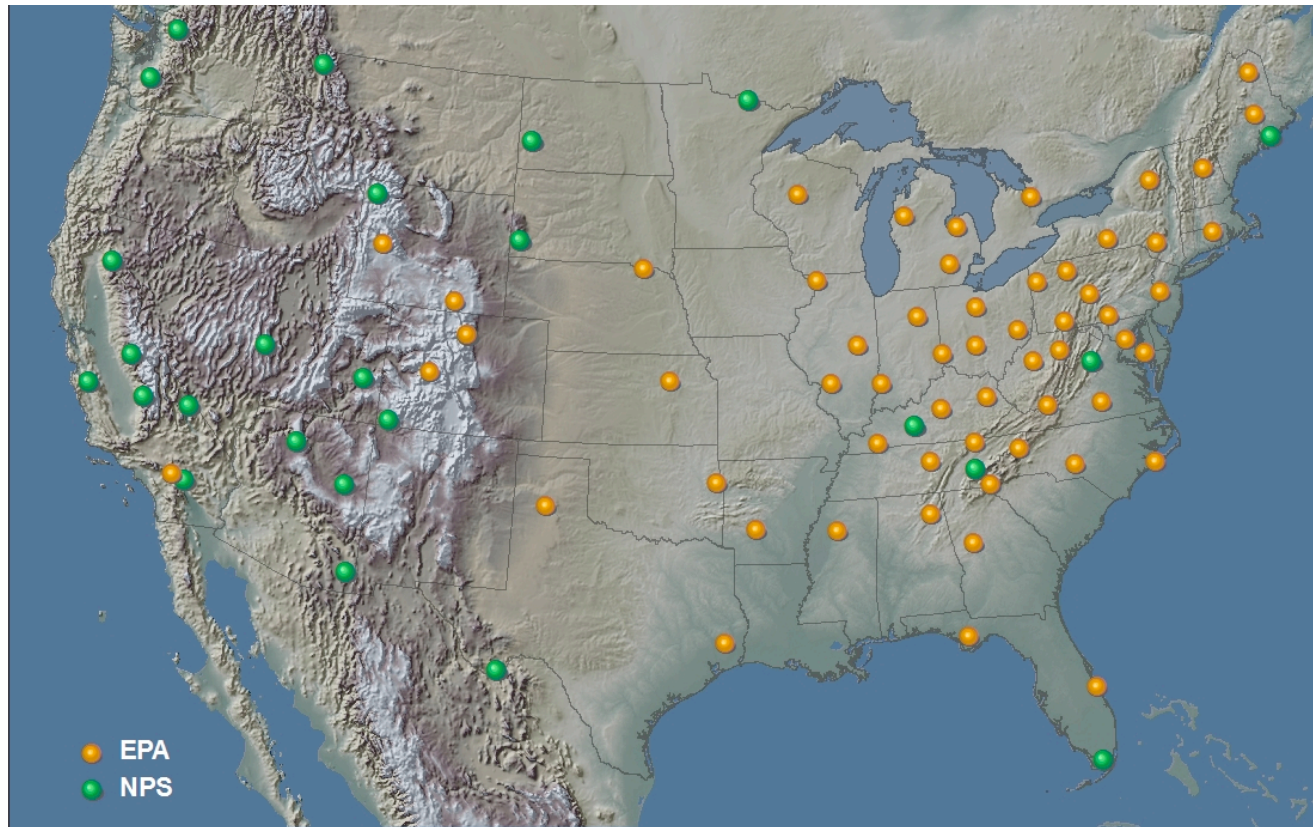
- estimating U.S. inorganic PM_{2.5} levels (Yu et al., 2005; Simon et al., 2008)
- global N dep. (Sutton et al., 2007)

Secondary inorganic aerosol formation



NH₄⁺ monitoring

CASTNet



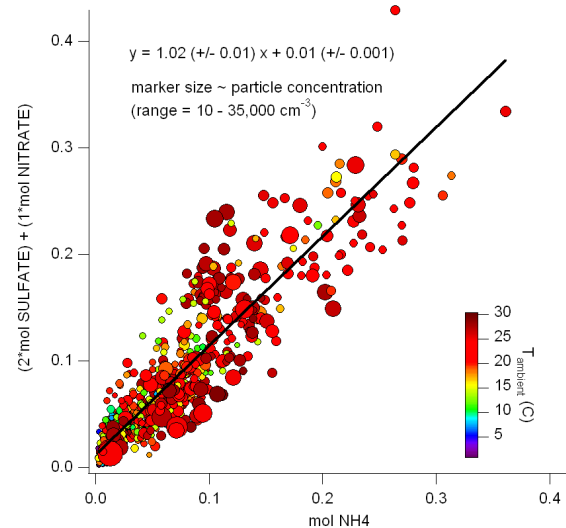
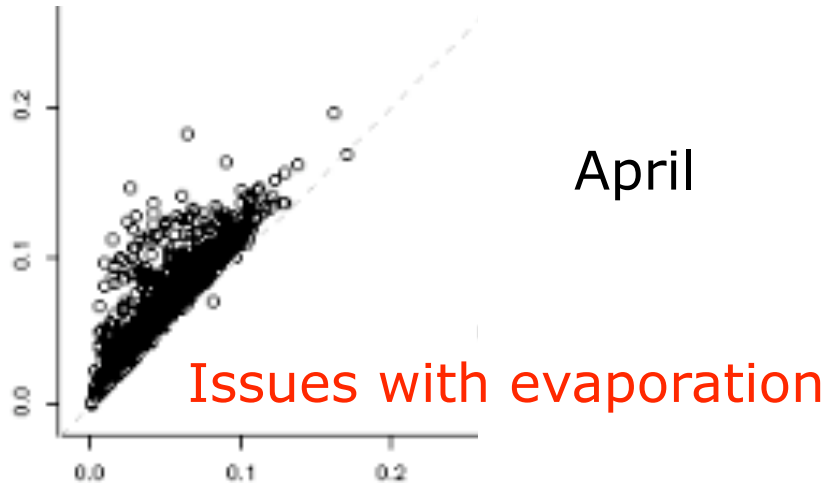
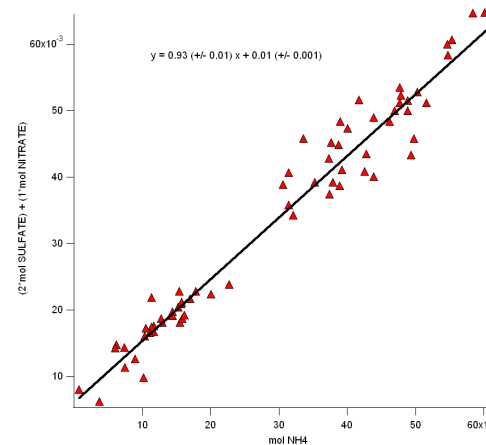
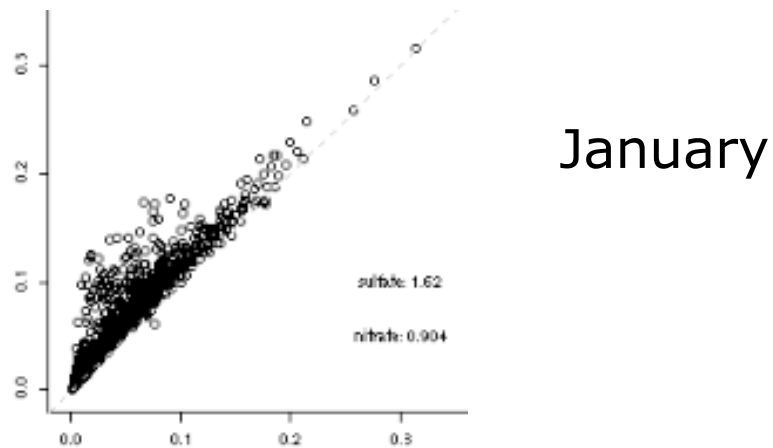
STN: another 200 sites

Checking ion balance:

$$n(\text{NH}_4^+) : 2n(\text{SO}_4^{2-}) + n(\text{NO}_3^-)$$

CASTNet, all sites,
2005-2006 (R. Pinder)

Field campaigns
(Sorooshian et al.)



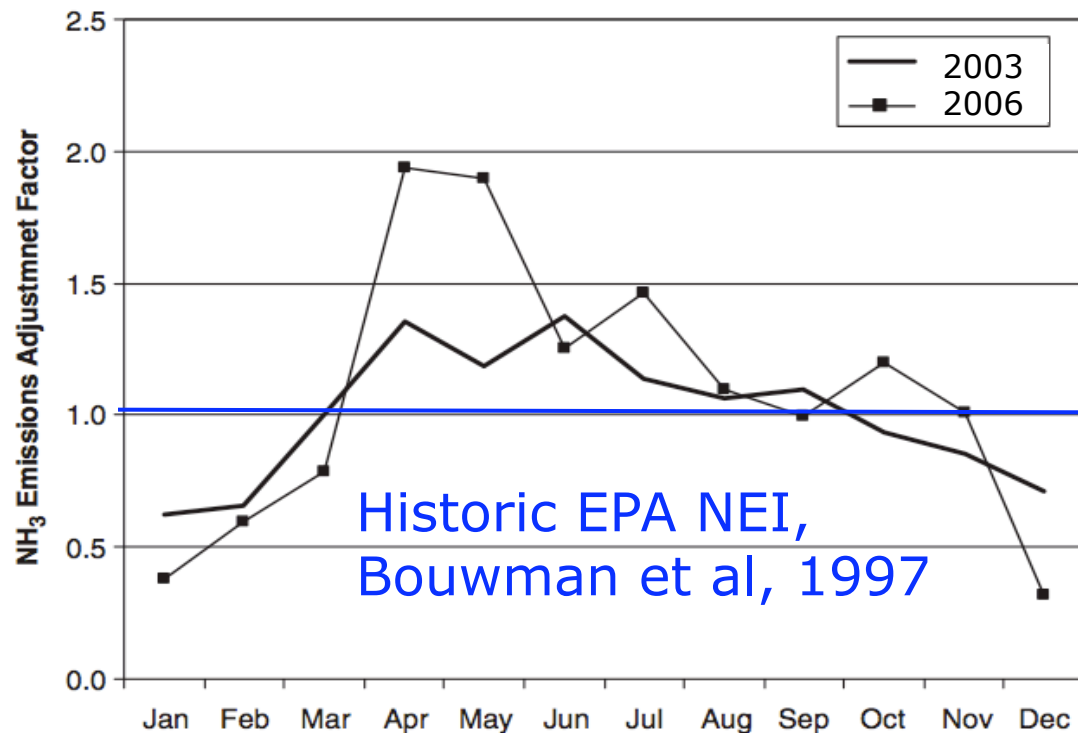
NH₃ inverse modeling: Gilliland et al.

Observations: wet NH_x = aerosol NH₄⁺ + gas NH₃

Method: Kalman filter (BF) to adjust monthly nationwide scale factors

Results:

Gilliland et al., 2003;
Gilliland et al., 2006



NH₃ emissions variability and uncertainty: Beusen et al. (2008)

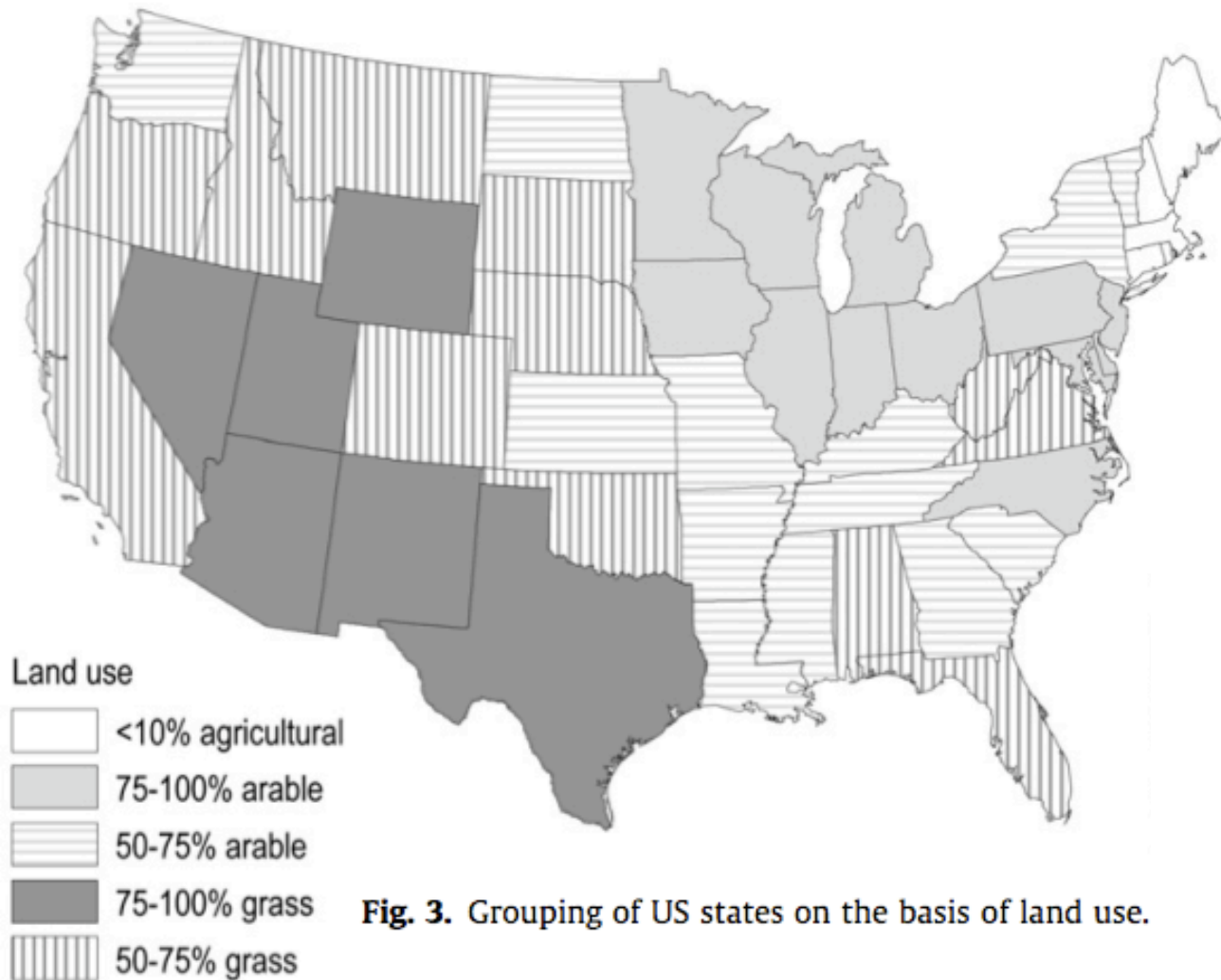
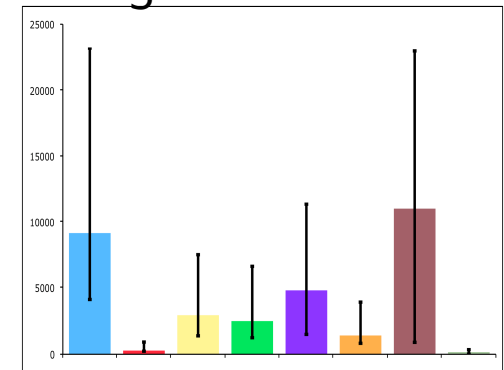


Fig. 3. Grouping of US states on the basis of land use.

Global animal NH₃ emissions

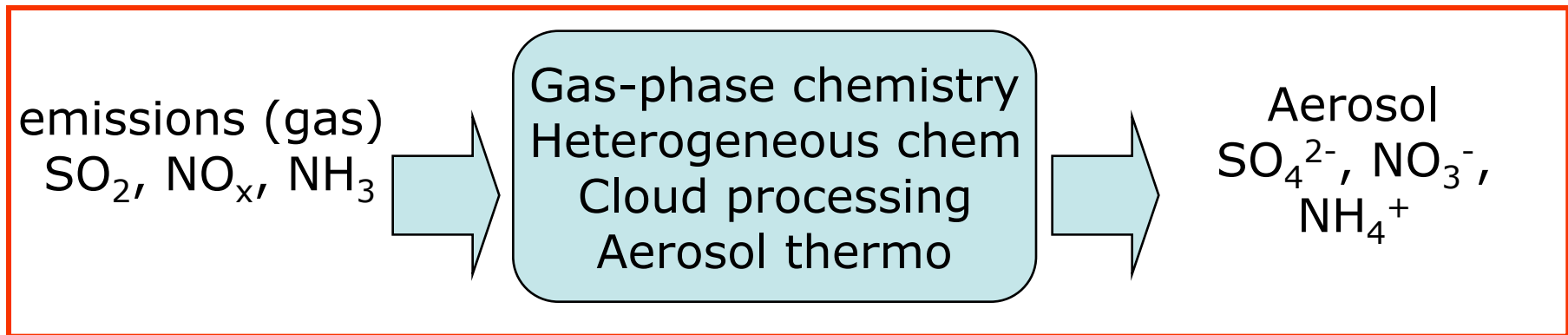


Source types

- housing mixed
- housing pastoral
- grazing mixed
- grazing pastoral
- spreading cropland
- spreading grassland
- fertilizer cropland
- fertilizer grassland

4D-Var with GEOS-Chem Adjoint Model

Forward model v6-02-05 (*Bey et al., 2001; Park et al., 2004*)

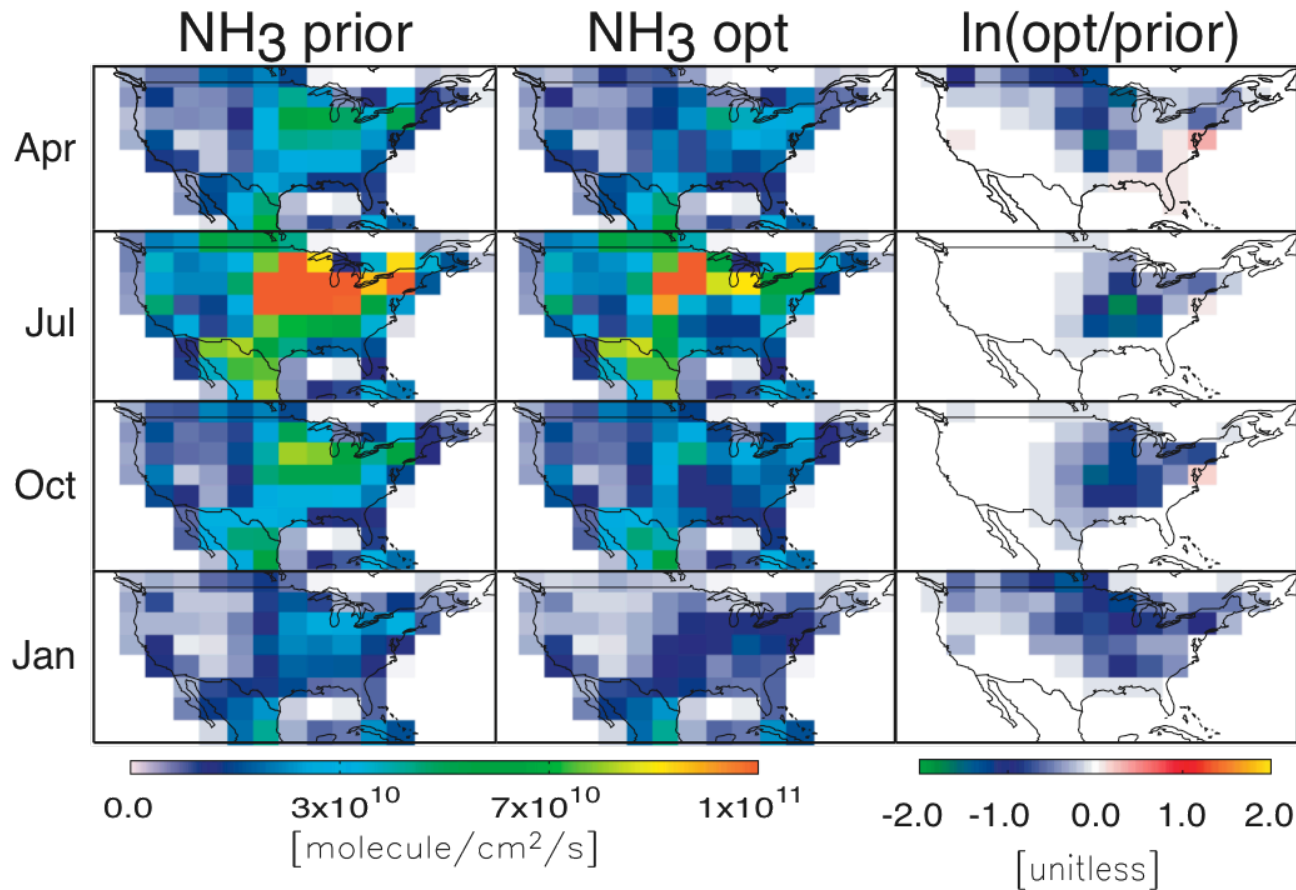


All included in adjoint (*Henze et al., 2007; Singh et al., 2009*)

Inverse modeling (*Henze et al., 2008*)

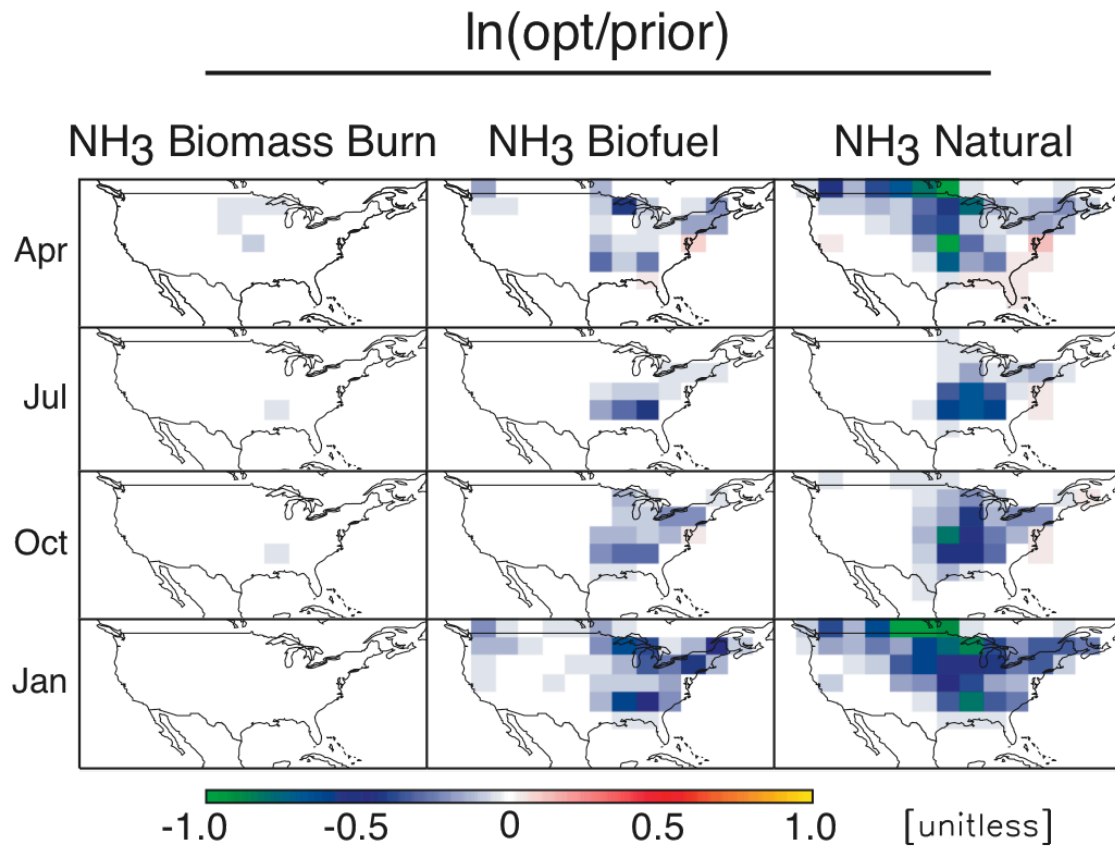
- Control parameters: all NH_3 , SO_2 , NO_x emissions
- Sulfate and nitrate from IMRPOVE network (24h, 1in3)
- Jan, April, July, October: 4 separate inversions

Inverse modeling: anthro NH_3 emissions



- scaling is spatially variable
- scaling generally reductions
- some increases
- Each month treated separately
- reduction in RMSE $\approx 40\%$

Inverse modeling: other NH_3 emissions



- scaling results from product of adjoints with prior emissions estimates

- reductions affect anthropogenic sources more than natural sources

- results across sectors are correlated

Can effectively distinguish between source sectors

Inverse modeling: assessing the solution

Dependence on inverse modeling assumptions:

- error covariance matrices
- regularization

Estimated uncertainty of solution

- approximate inverse hessian
- std error and correlations

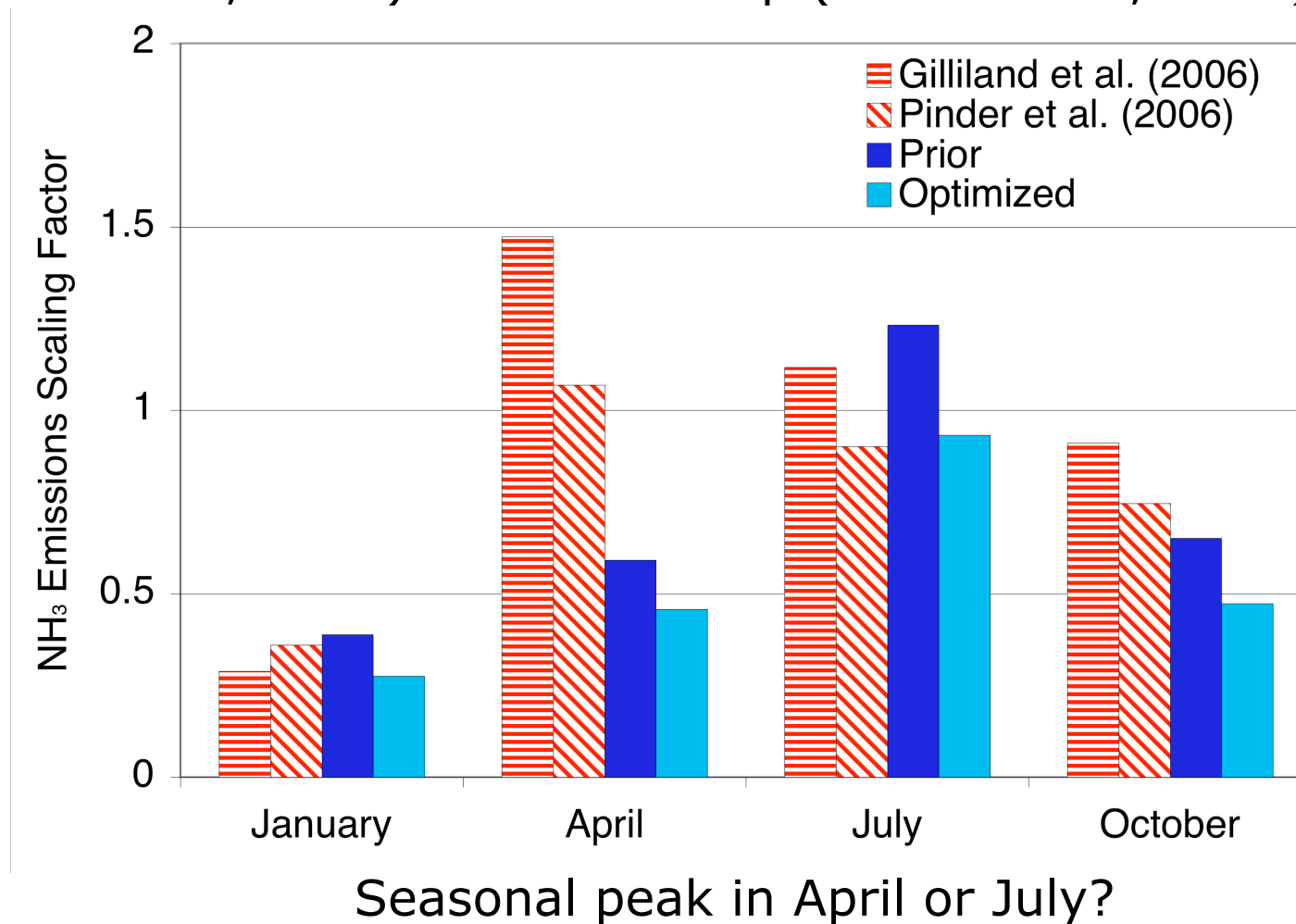
Compare to other studies

- inverse modeling
- bottom up inventories

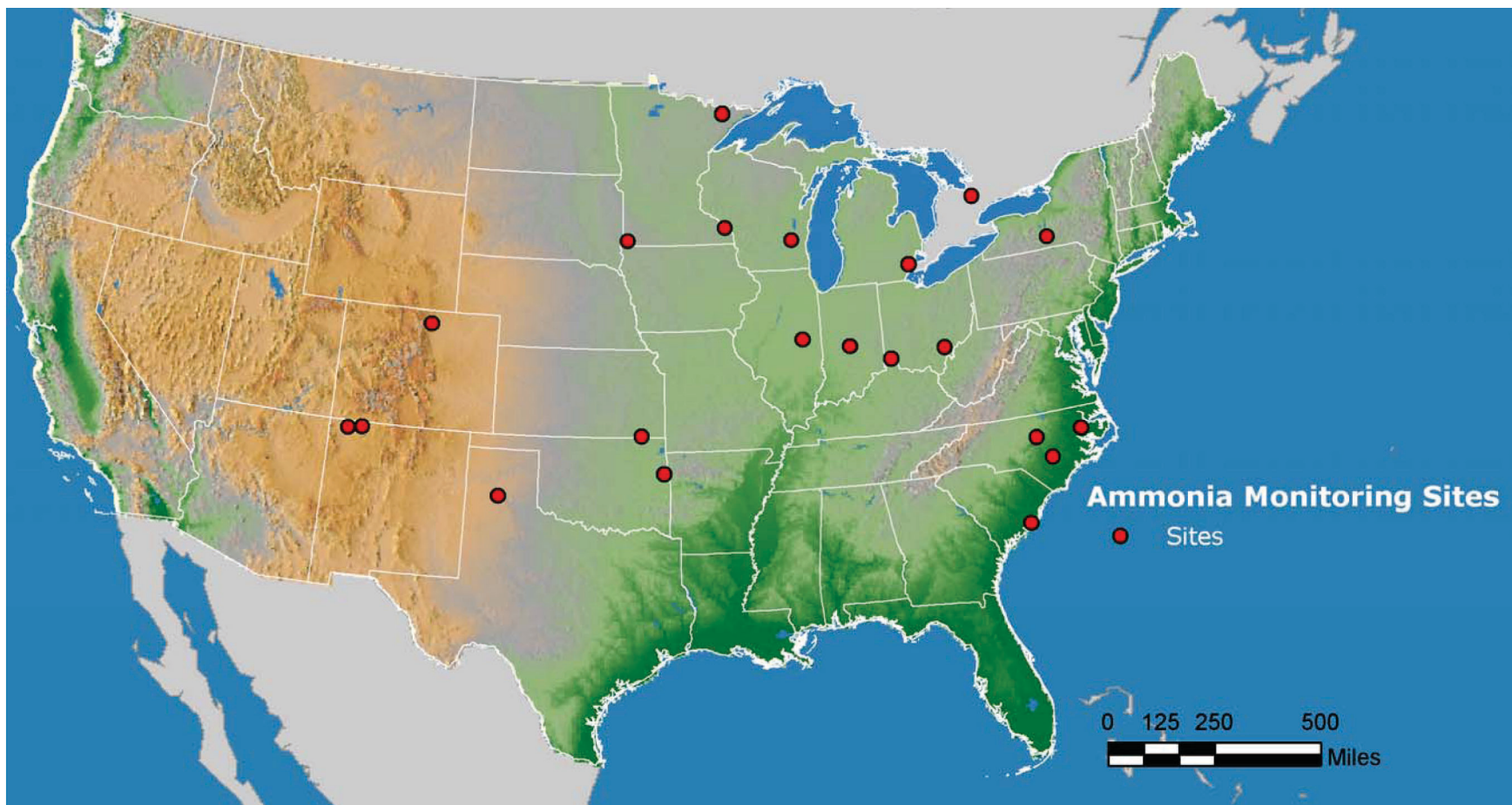
Compare to NH_3 observations

Inverse modeling: NH_3

Sum changes to NH_3 over U.S., compare to other inverse modeling (Gilliland et al., 2006) and bottom up (Pinder et al., 2006)



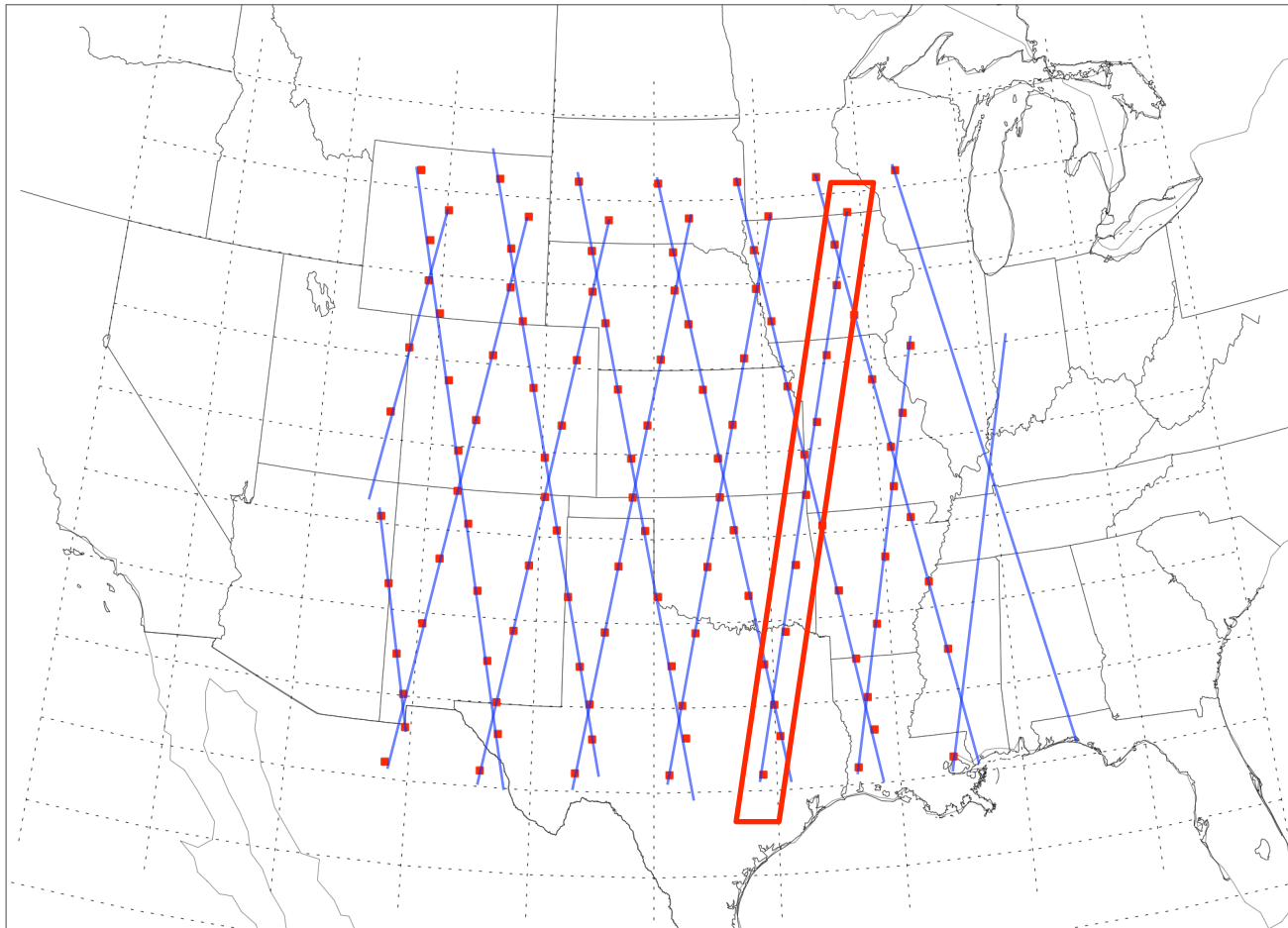
NH₃ Monitoring Sites



CAMD sites at CASTNet locations

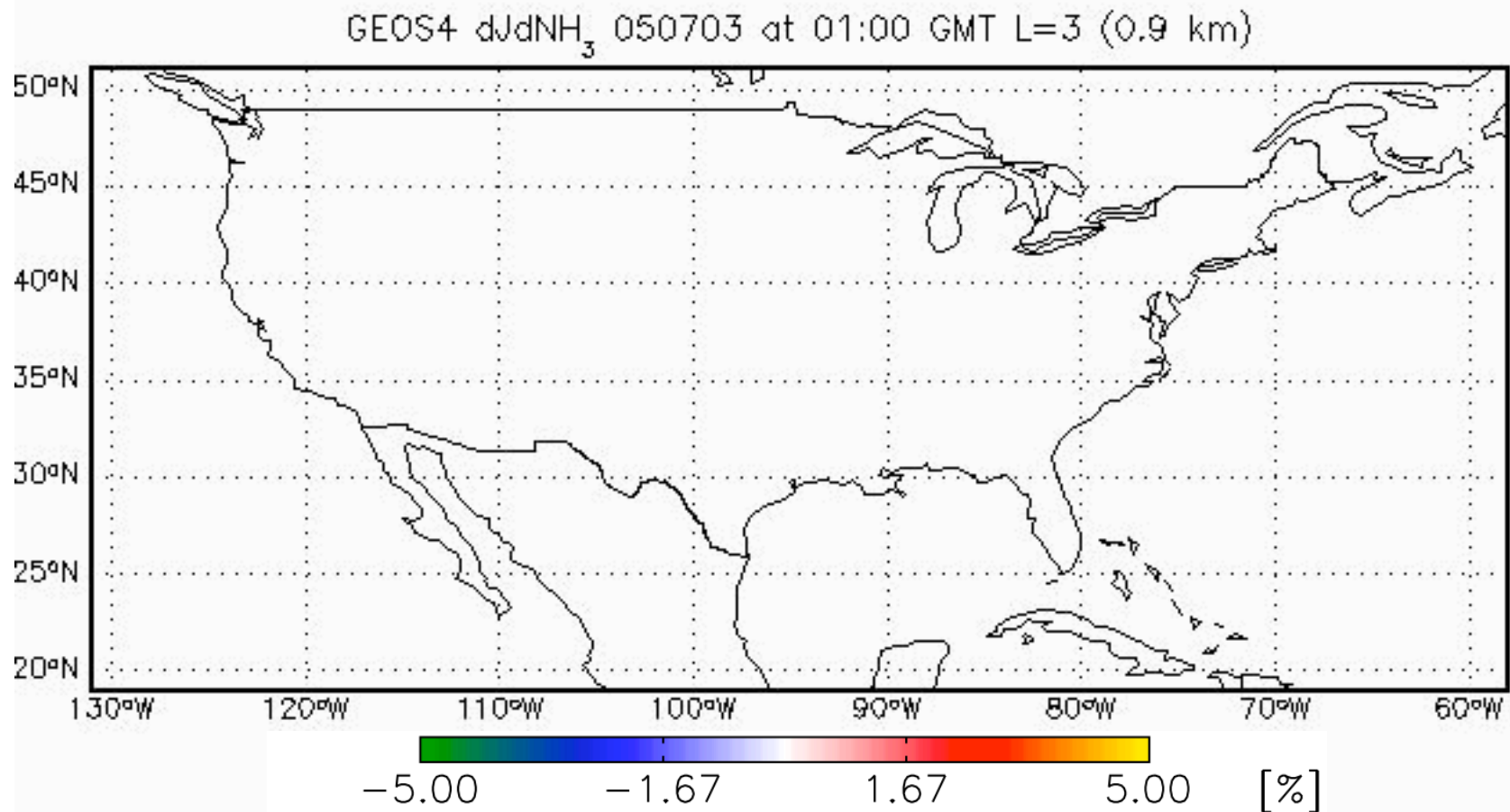
Potential for further constraints: TES coverage for 2 weeks in July 2005

TES GS Footprints, July 4-19, 2005



data selection: lat = [30.,45.], lon = [-110, -90.]; total number of profiles: 115

Adjoint sensitivities of modeled NH_3 retrievals



Sensitivities show the origins of the NH_3 that eventually will be “observed” by TES

Inverse Modeling: twin experiment

Parameter Estimate

$$\mathbf{p} = 2\mathbf{p}_a$$



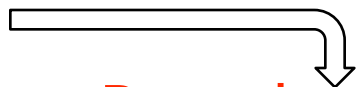
Forward Model

t_0 \longrightarrow t_f



Simulated NH_3 field

$$\mathbf{c}(x, t)$$



Pseudo retrievals, IMPROVE observations

Inverse Modeling using Adjoint Model

