Constraining NH_3 emissions using remote sensing and surface observations

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Why Study Ammonia?

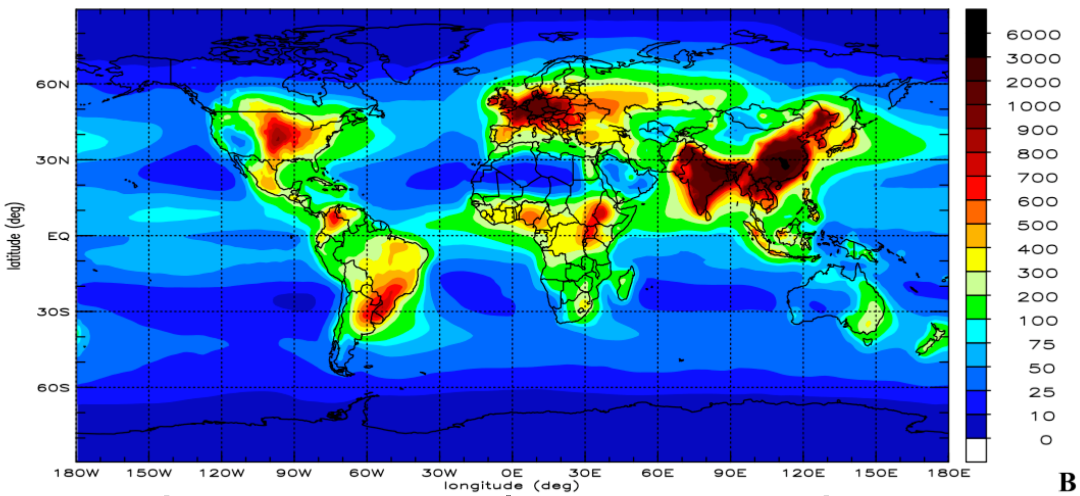
Inorganic aerosol: $(NH_4)_2SO_4$, NH_4NO_3

Health impacts

Importance for $PM_{2.5}$ control
(e.g., Pinder et al., 2007; Henze et al., 2009)

Climate impacts
(e.g, Abbat et al., 2006; Wang et al., 2008)

Deposition of reactive nitrogen



(Dentener et al., *GBC*, 2006)

Importance of NH_x transport
(Galloway, *Science* 2008)

NH_3 emissions to double by 2050.
(Denman et al., *IPCC*, 2007)

46 Tg gap in N budget?
(Schlesinger, *PNAS*, 2009)

NH_3 emissions very uncertain, often x2 or more

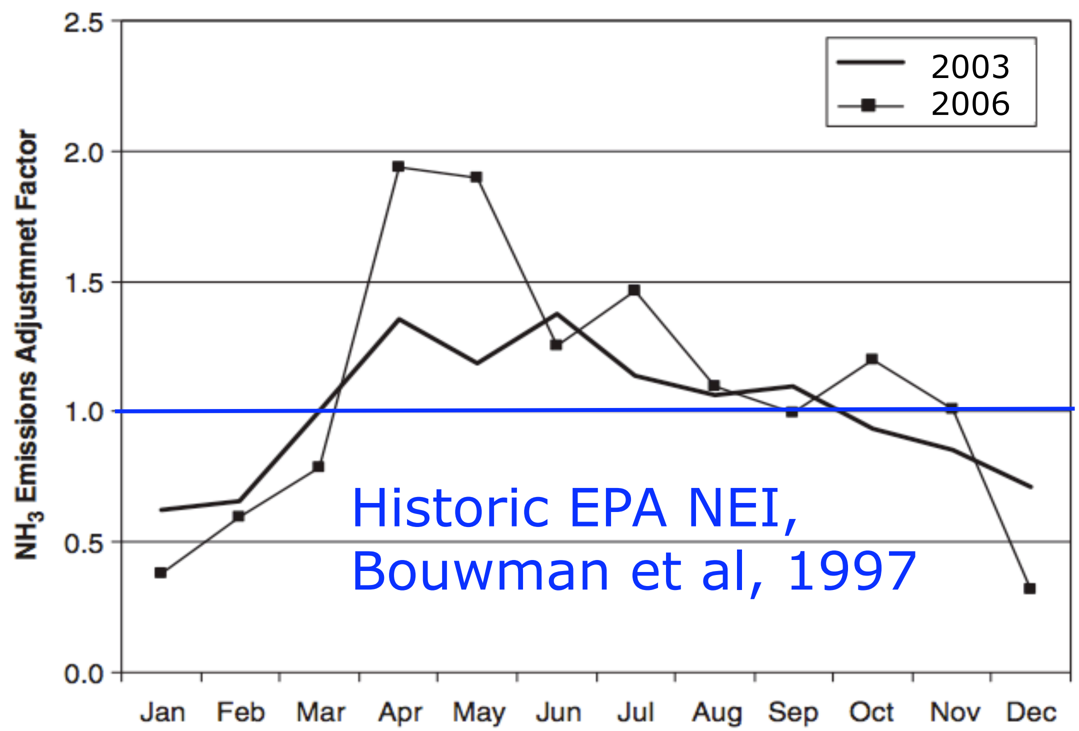
NH_3 inverse modeling: Gilliland et al.

Observations: wet NH_x = aerosol NH_4^+ + gas NH_3

Method: Kalman filter (BF) to adjust monthly nation-wide scale factors

Results:

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



NH_3 emissions variability and uncertainty: Beusen et al. (2008)

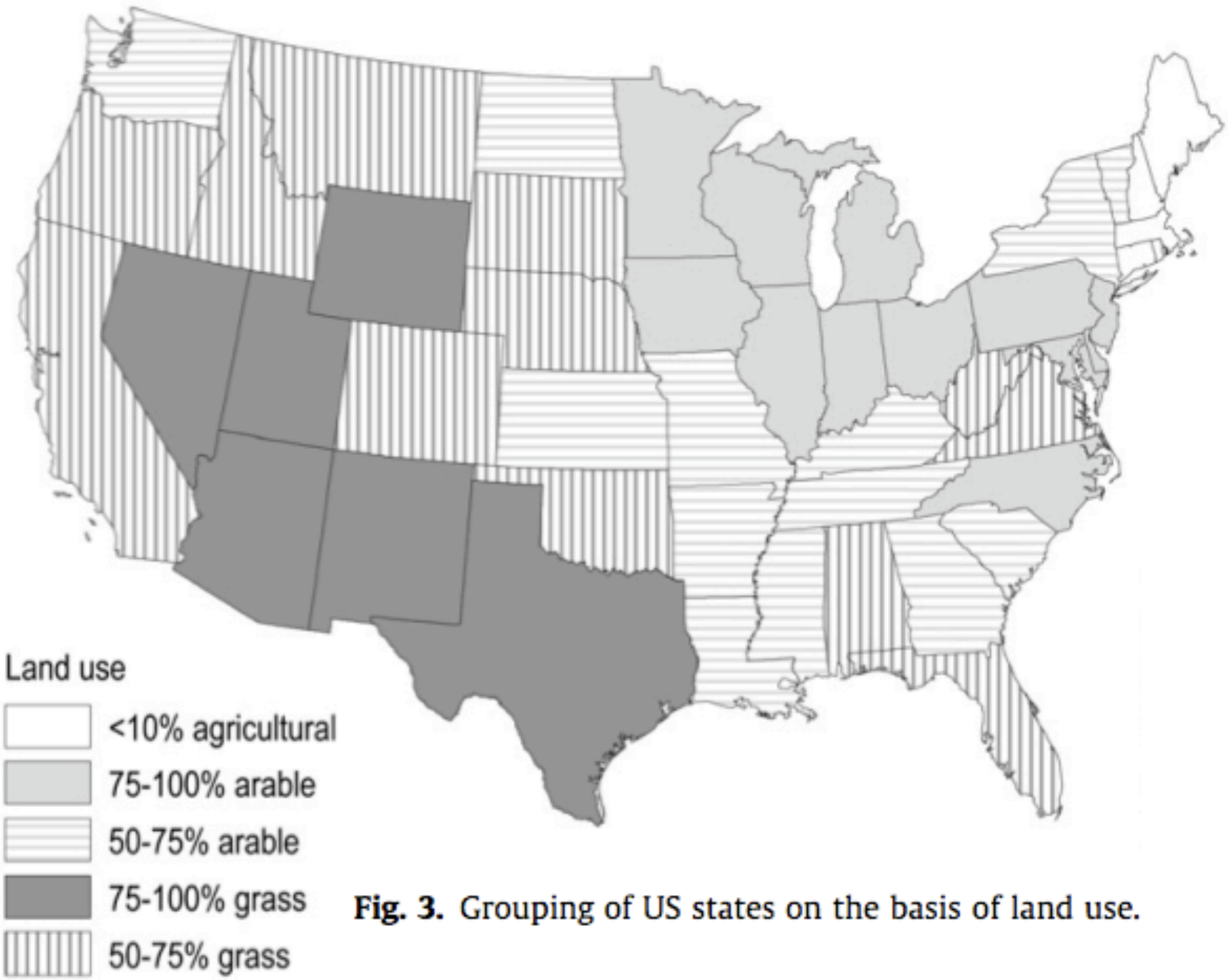
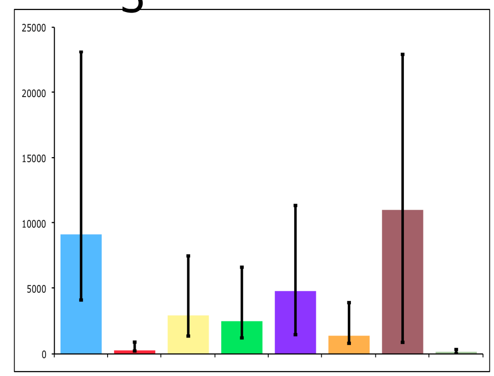


Fig. 3. Grouping of US states on the basis of land use.

Global animal NH_3 emissions

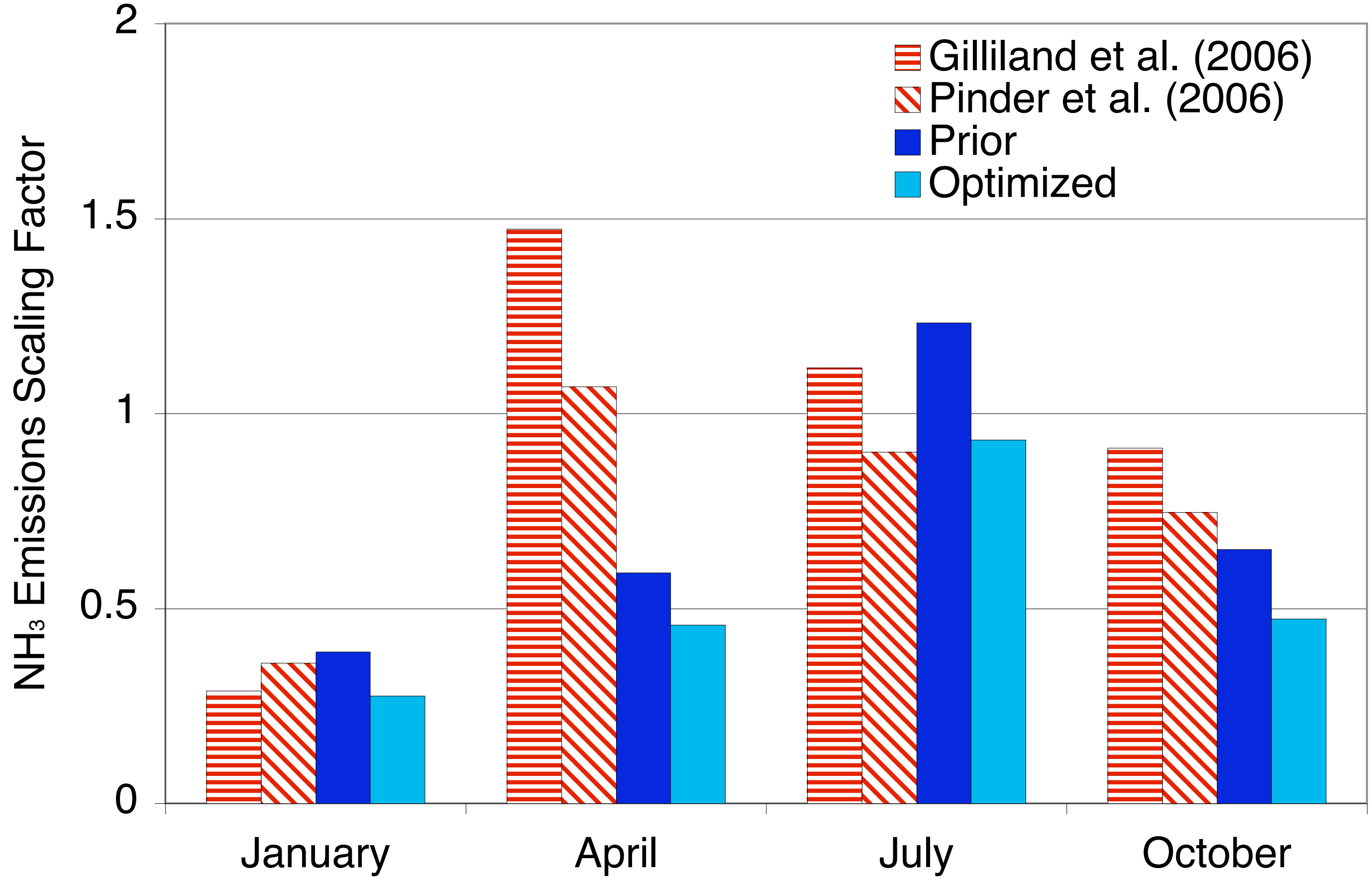


Source types

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

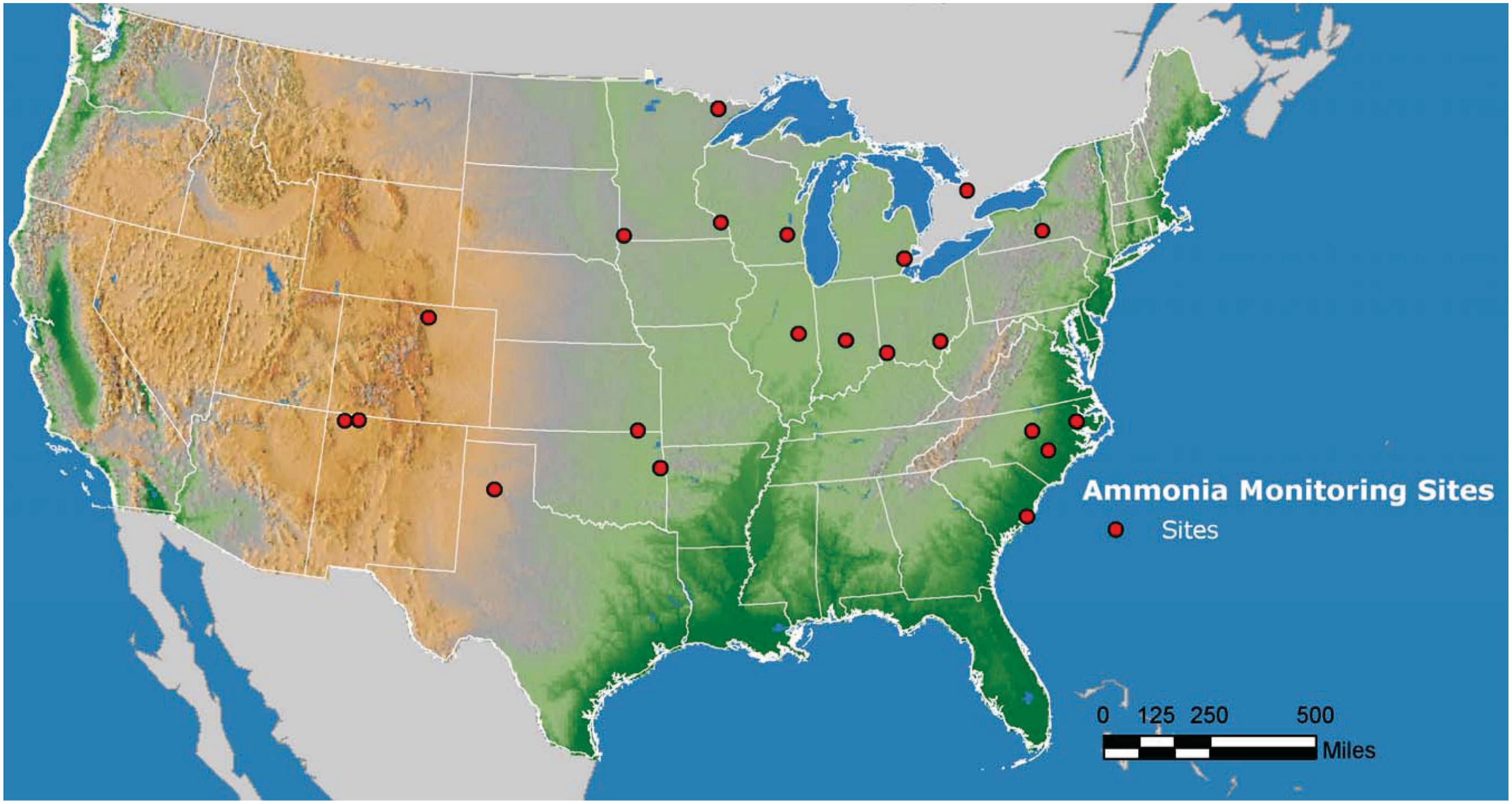
Inverse modeling: NH_3

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



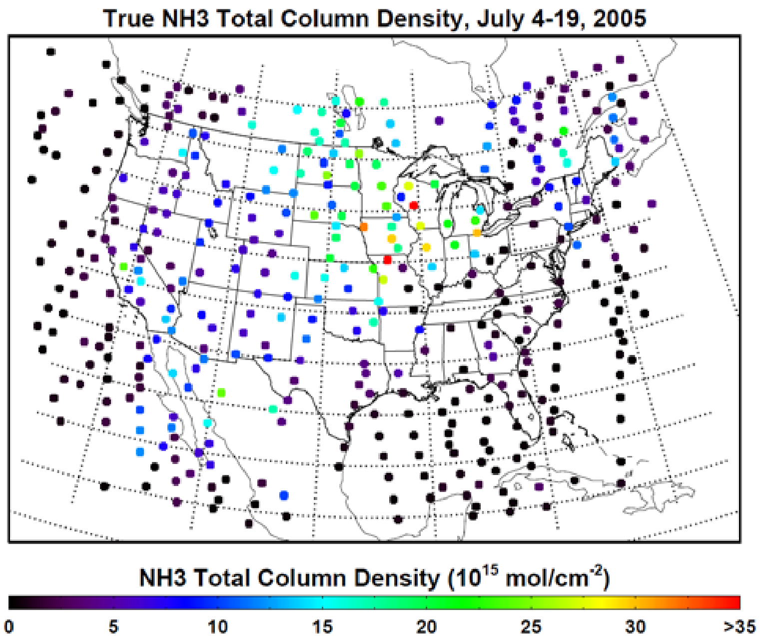
Seasonal peak in April or July?

NH_3 Monitoring Sites



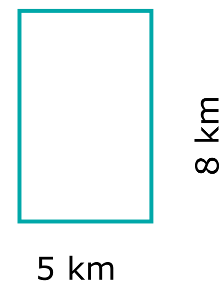
EPA's AMoN sites (Gary Lear). Also LADCO, SEARCH, CSU, ANARChE, etc., but no extensive network.

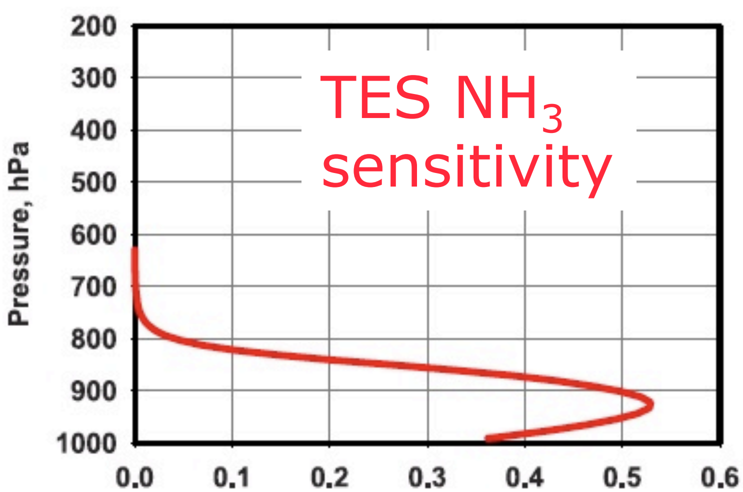
Remote sensing of NH_3: TES



"covers" globe in 16 days

Footprint size:





Properties

- Vertical "profile", ~ 1 DOF
- sensitive to BL
- more sparse, precise than IASI

Beer et al., 2008; Clarisse et al., 2010

Satellite validation challenges

Arriving at an NH_3 "observation" is an inverse problem itself.

- ill posed (multiple atmospheric states look alike)
- constraints required

Satellite products are a mix of measured and modeled quantities.

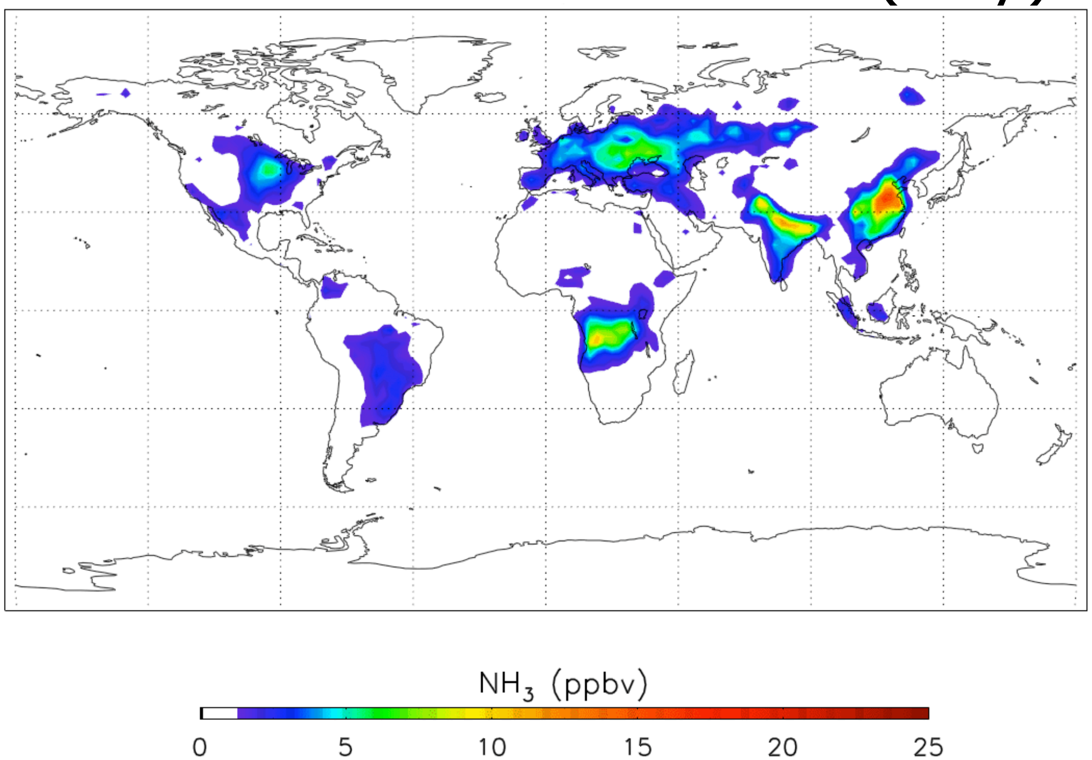
- model estimates used for initial profile
- profile scaled to match observations
- influence of model estimate can be removed

Need to address issues of collocation of observations

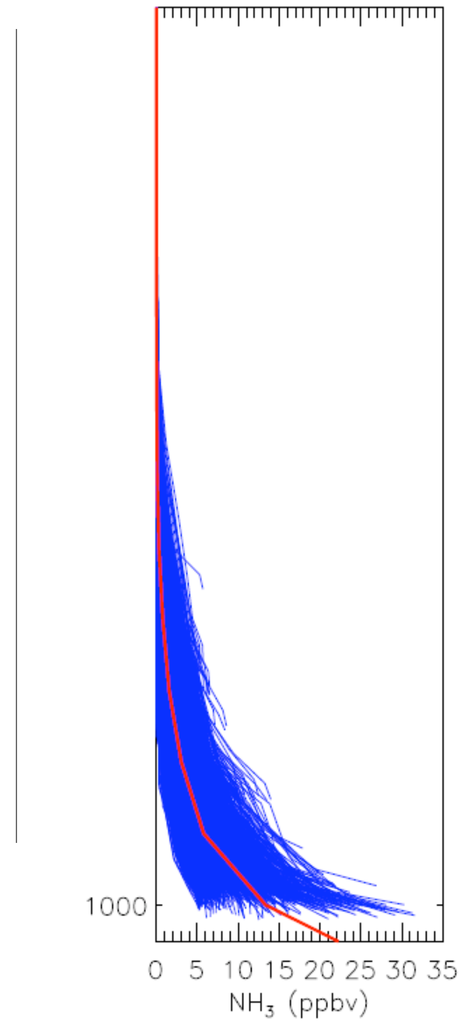
- horizontal
- vertical
- time

Global modeling support of NH_3 retrievals

GEOS-Chem surface level (July)

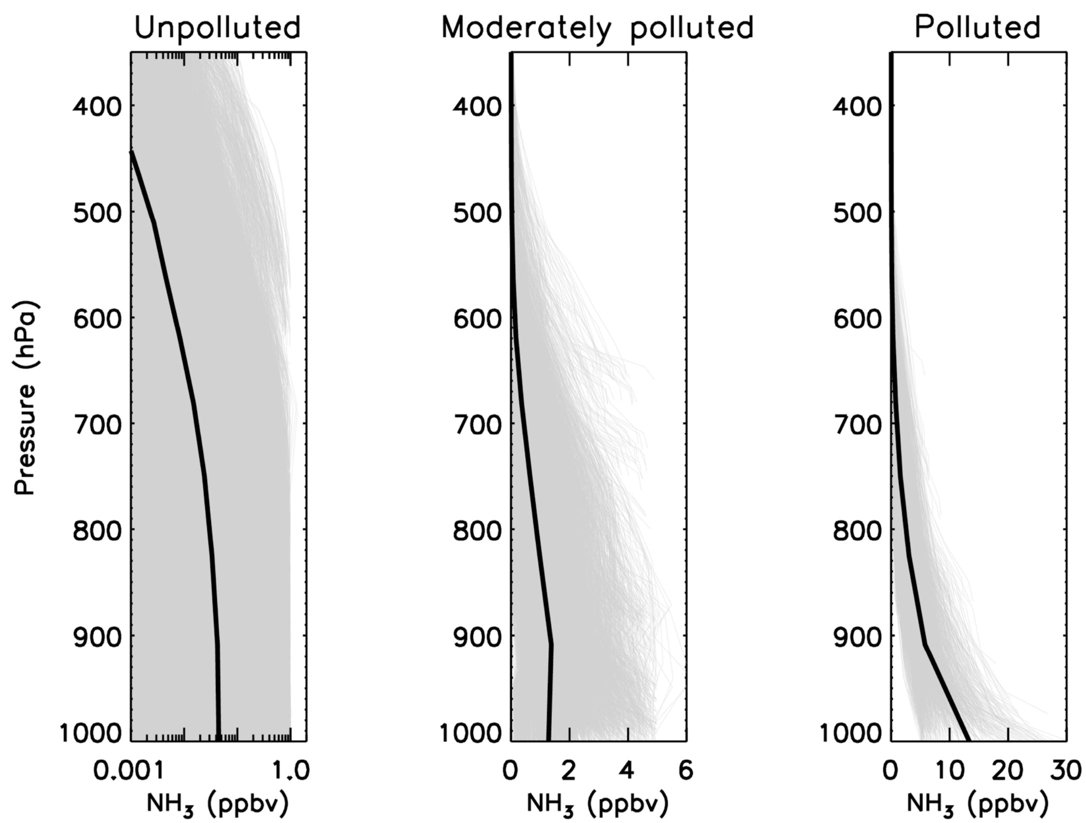


Vertical variance



Global modeling support of NH_3 retrievals

Classify model profiles based on surface values

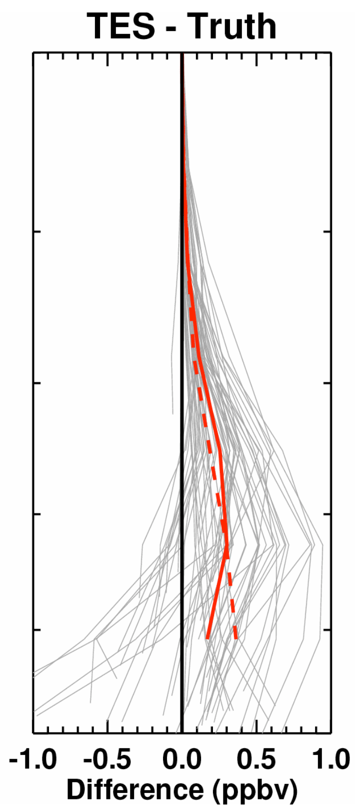


Consider three representative profiles

Cady-Pereira et al., *in prep*

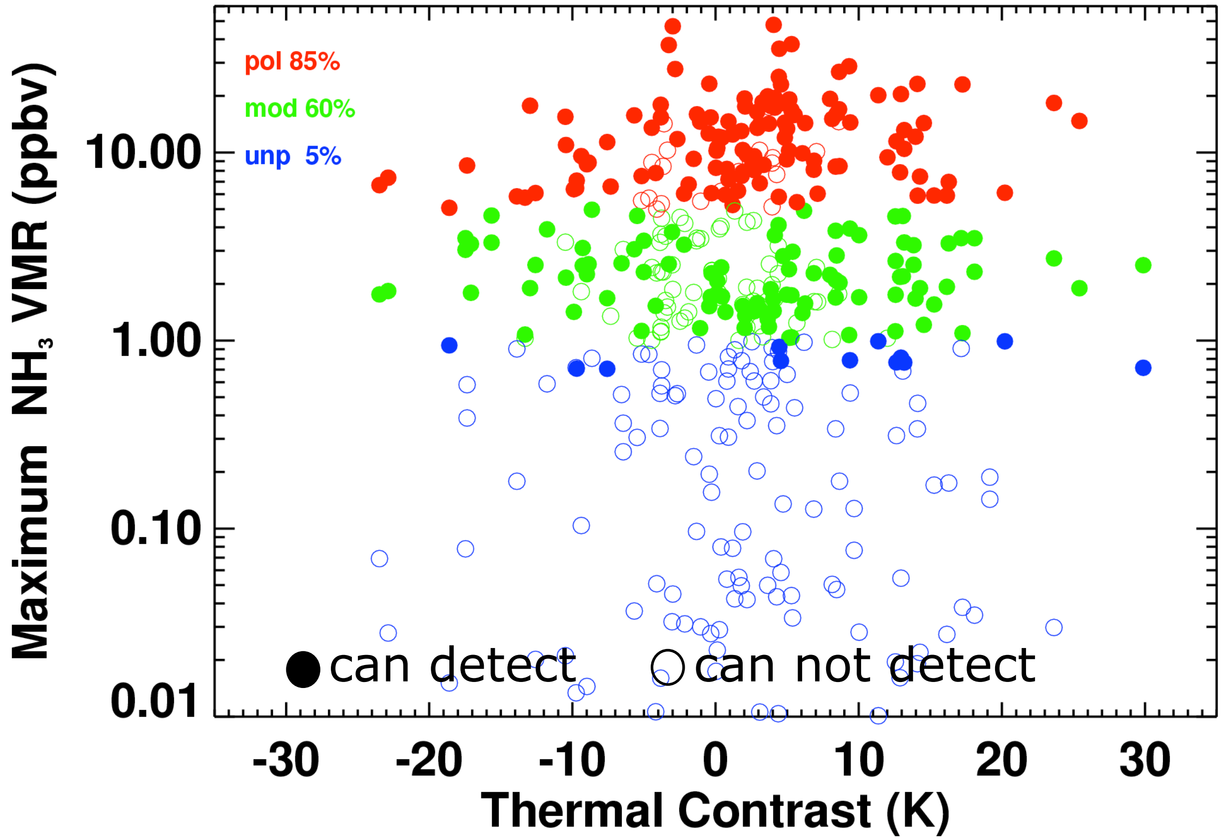
TES: measurement characterization

Bias Test



~ +0.5 ppb

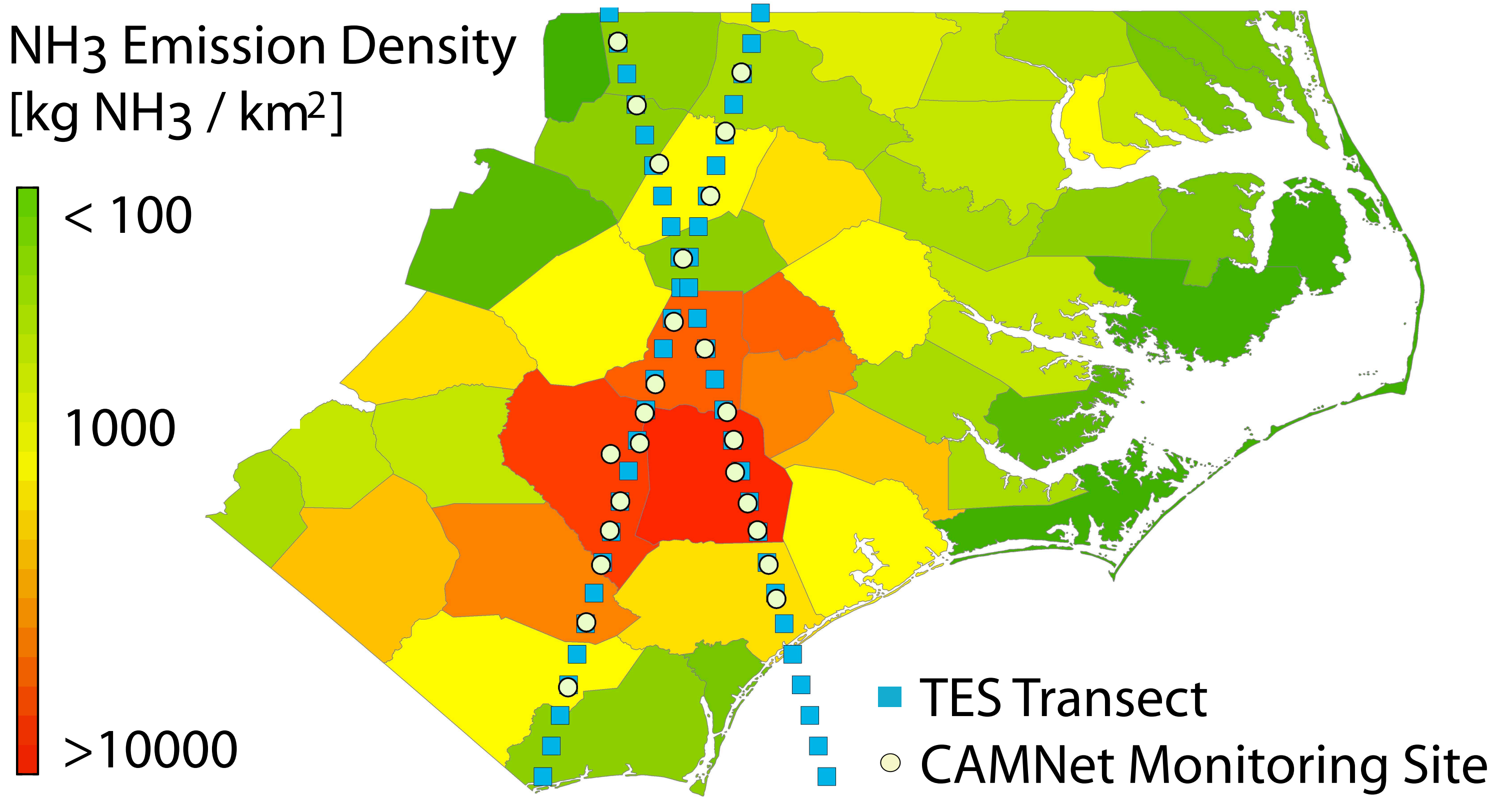
Detectability Test



> 1 pbb

Validating TES NH_3 with surface observations

Overlap with TES Transects for 2009



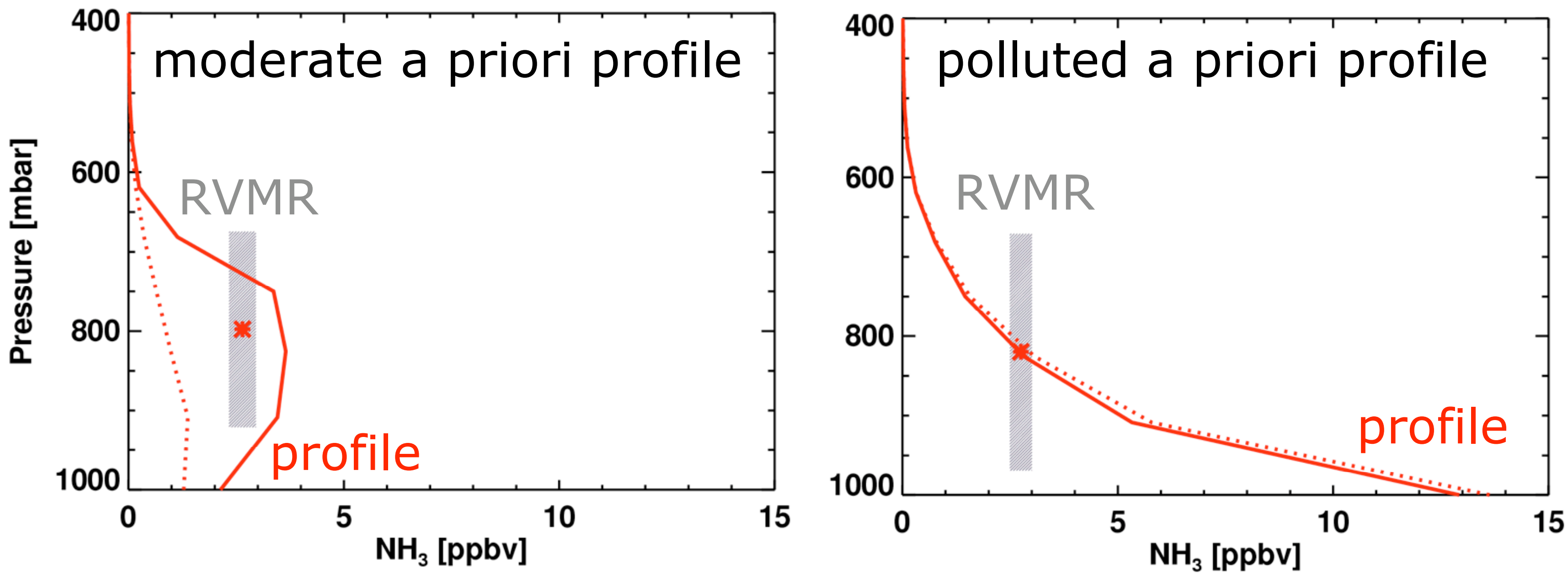
region selected for large values and large gradients

Pinder et al., *in prep*

Comparing retrievals (profiles) to surface concentrations

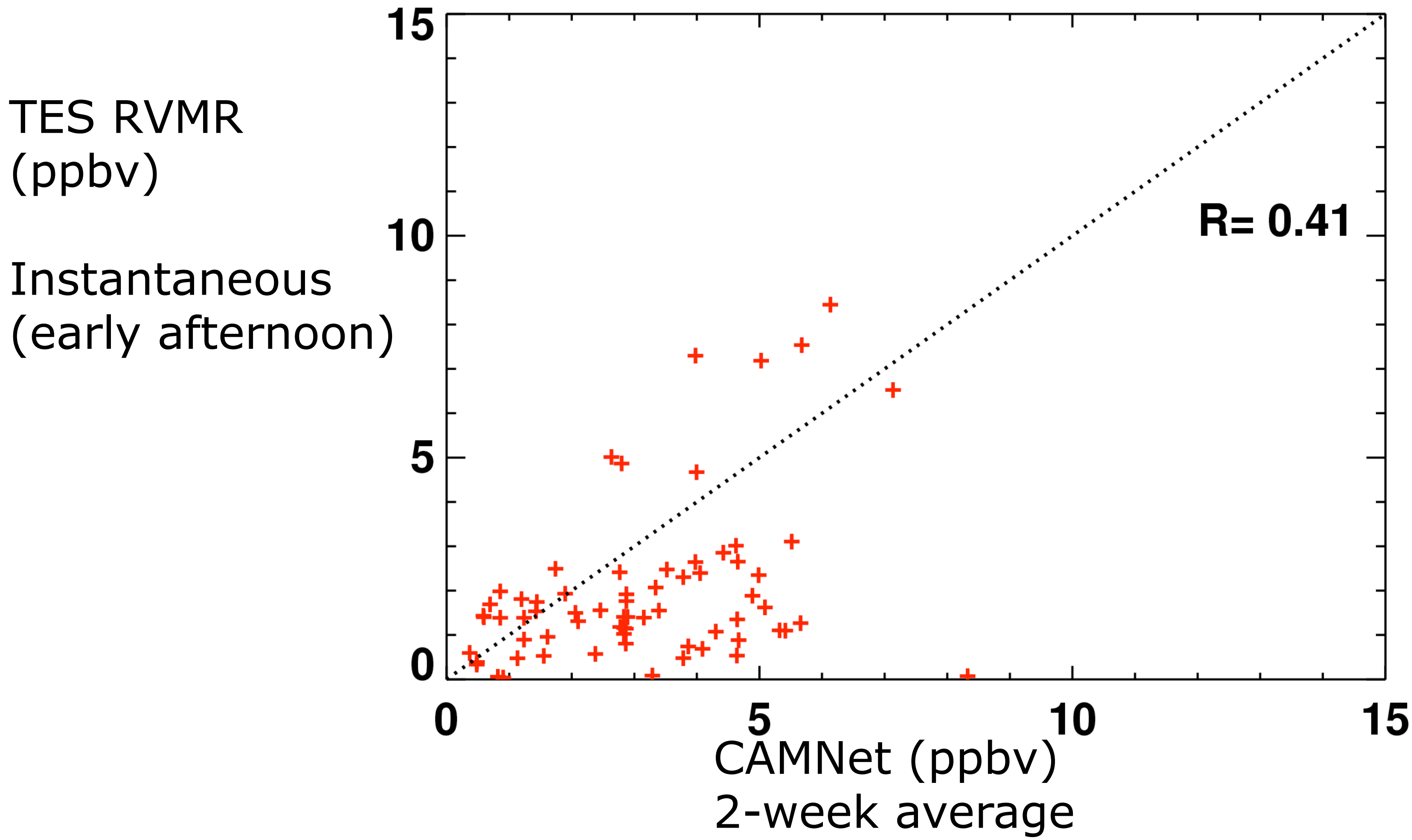
Comparison of surface measurements to satellite profile is influenced by choice of a priori

Cady-Pereira et al., *in prep*



RVMR: - representative volume mixing ratio (Payne et al., 2009)
- is more invariant than profile

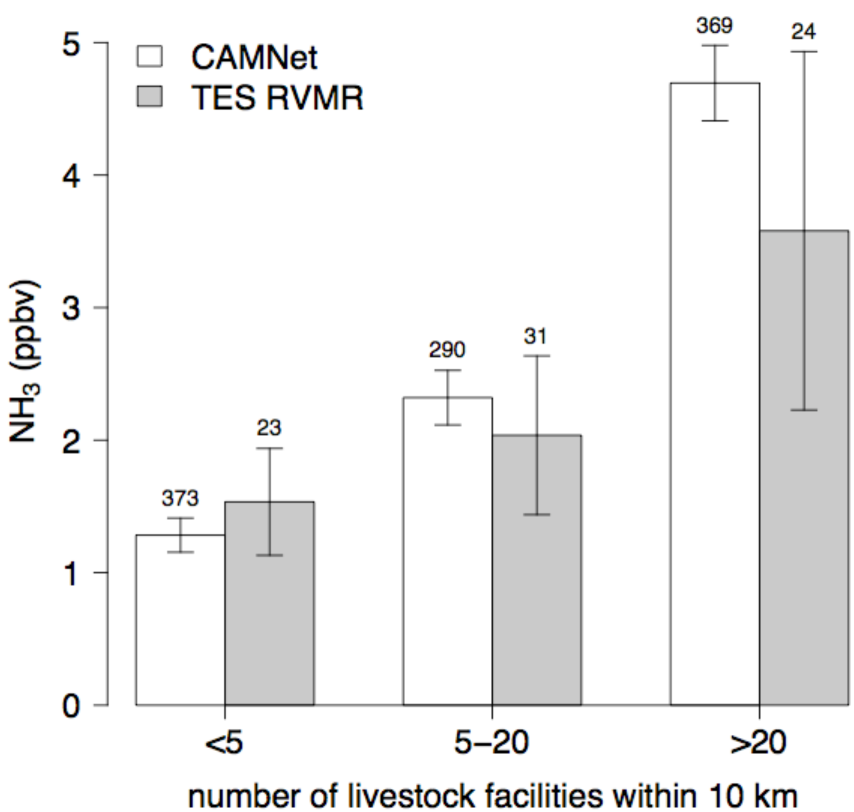
Comparing instantaneous values to 2-week averages



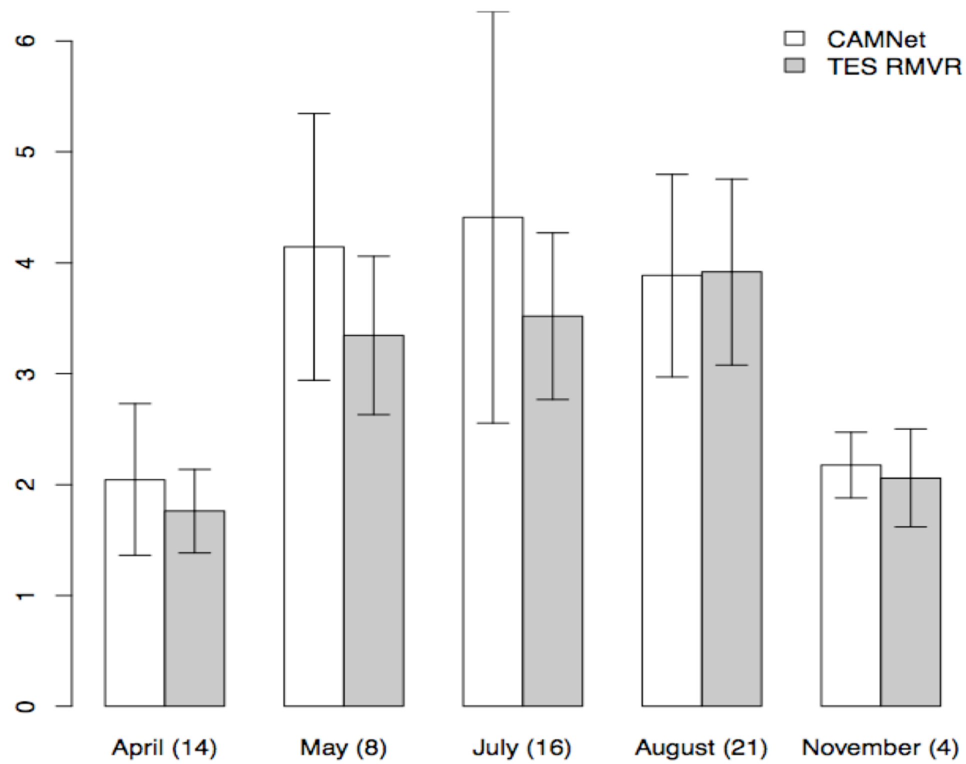
However, comparison of 2-week ave obs to instantaneous obs at TES overpass time has R=0.4.

TES vs surface obs: spatial and seasonal trends

spatial gradients



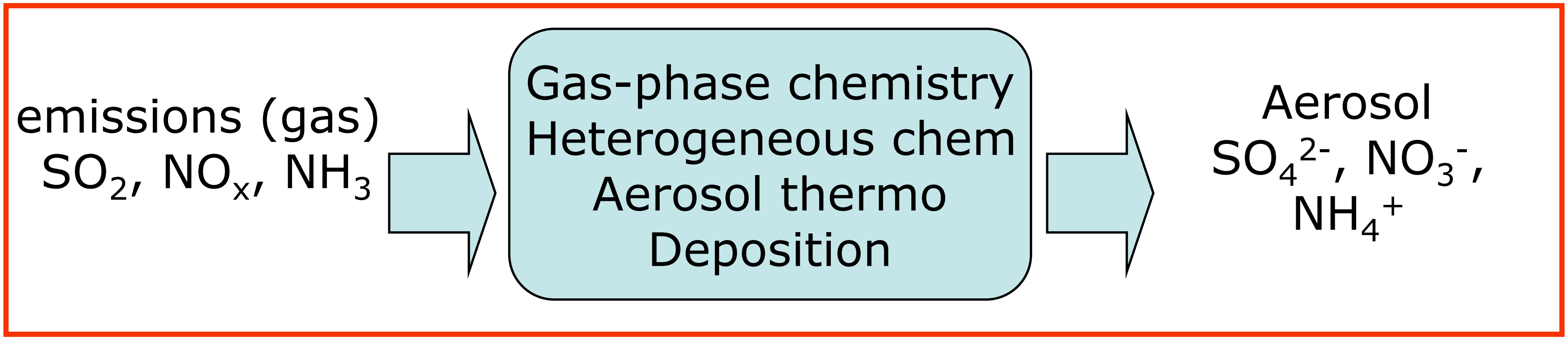
monthly variability



Pinder et al., *in prep*

4D-Var with GEOS-Chem

Forward model v8-02-03 (*Park et al.*, 2004)



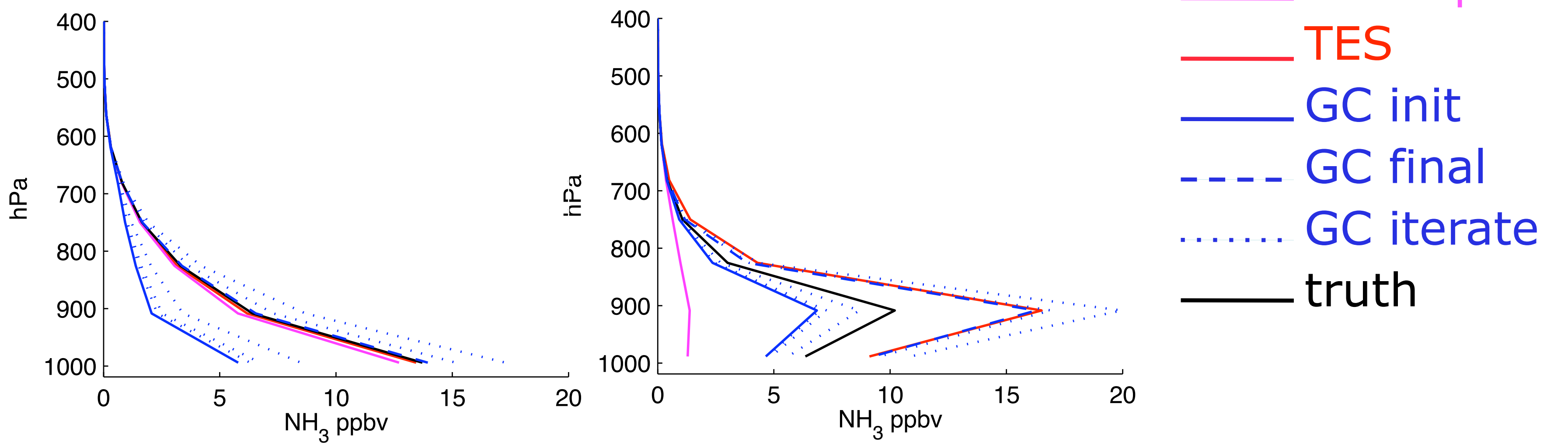
Adjoint model (*Henze et al.*, 2007; Kopacz et al., 2009)

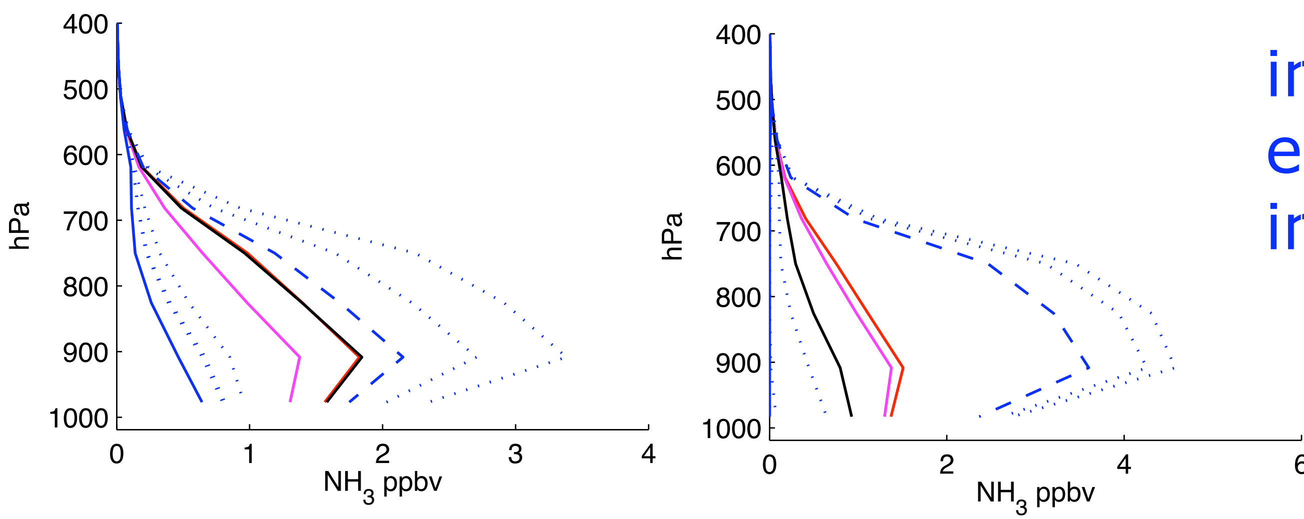
Test inversion with pseudo data:

- double GC emissions -> NH_3 → pseudo TES obs
- start with GC base
- invert using pseudo TES obs
- Converge to double GC emissions?

Inverse modeling: NH_3 profile

ideal retrievals | biased retrievals





impact on emissions inversions?

Starting from base case, opt to doubled

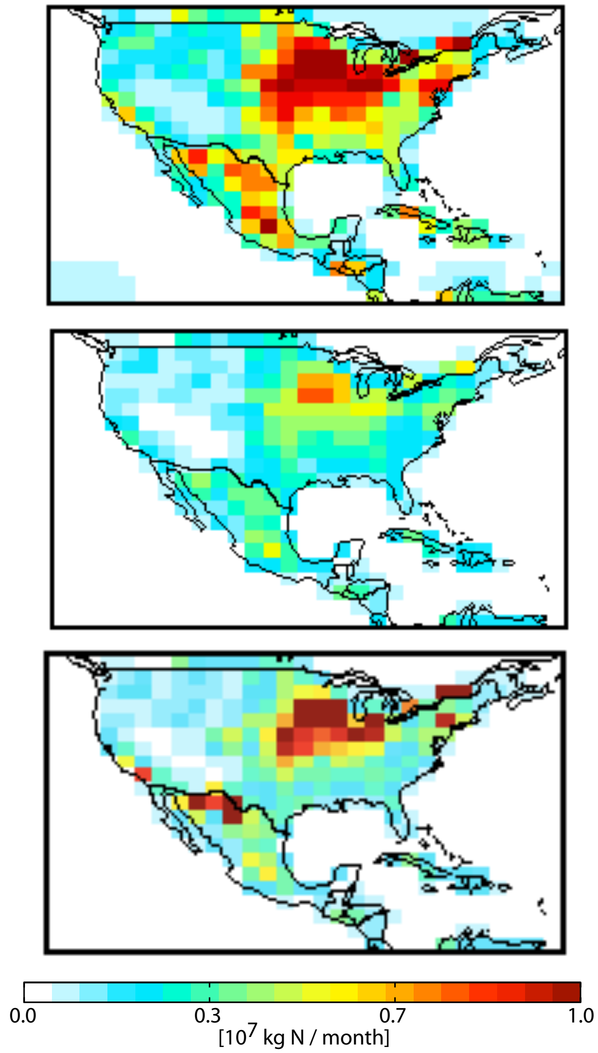
NH_3 emissions:

true:

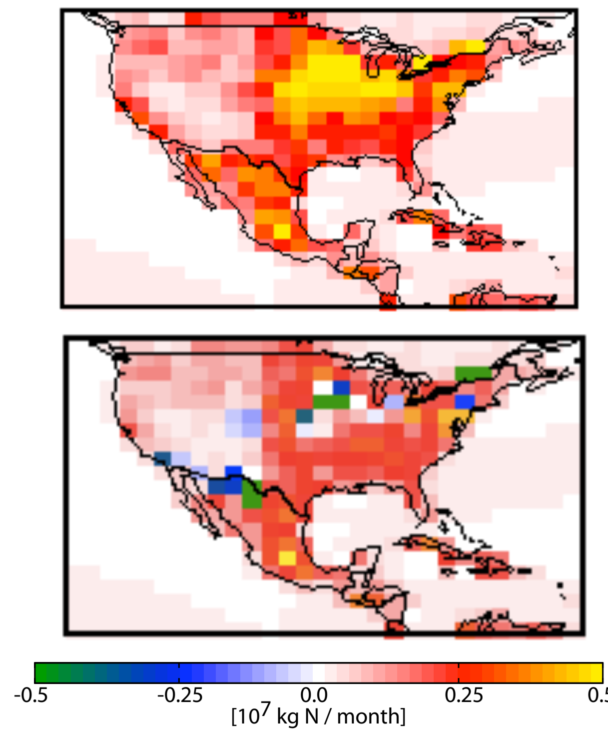
twice the initial

initial:

optimized:

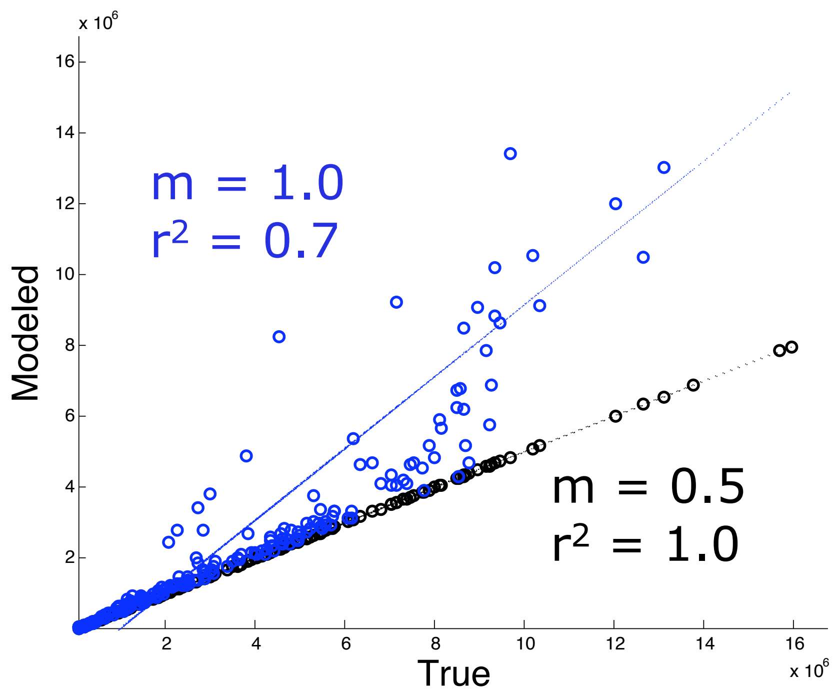


error: true - GC

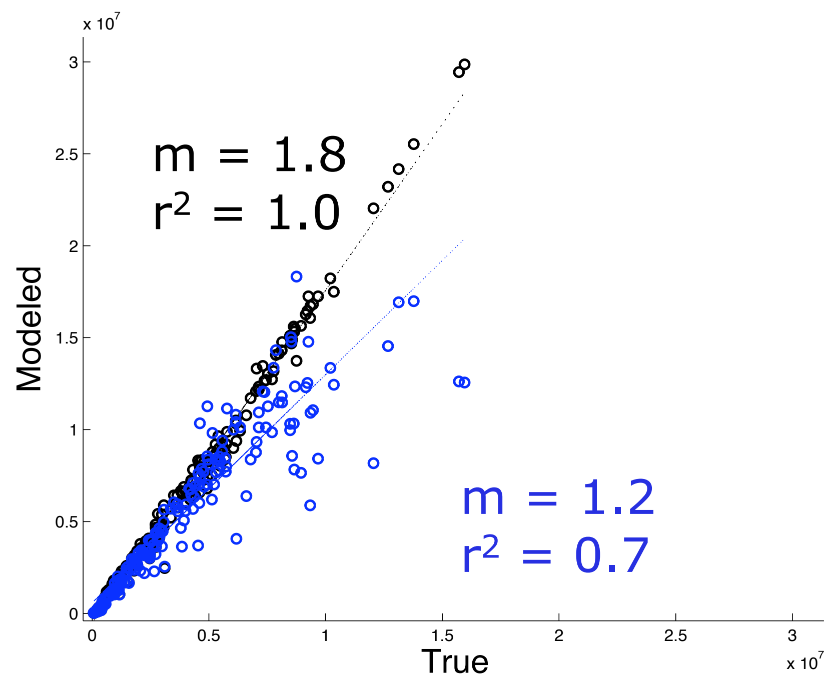


Pseudo tests: **before** and **after**

Starting consistently low



Starting high

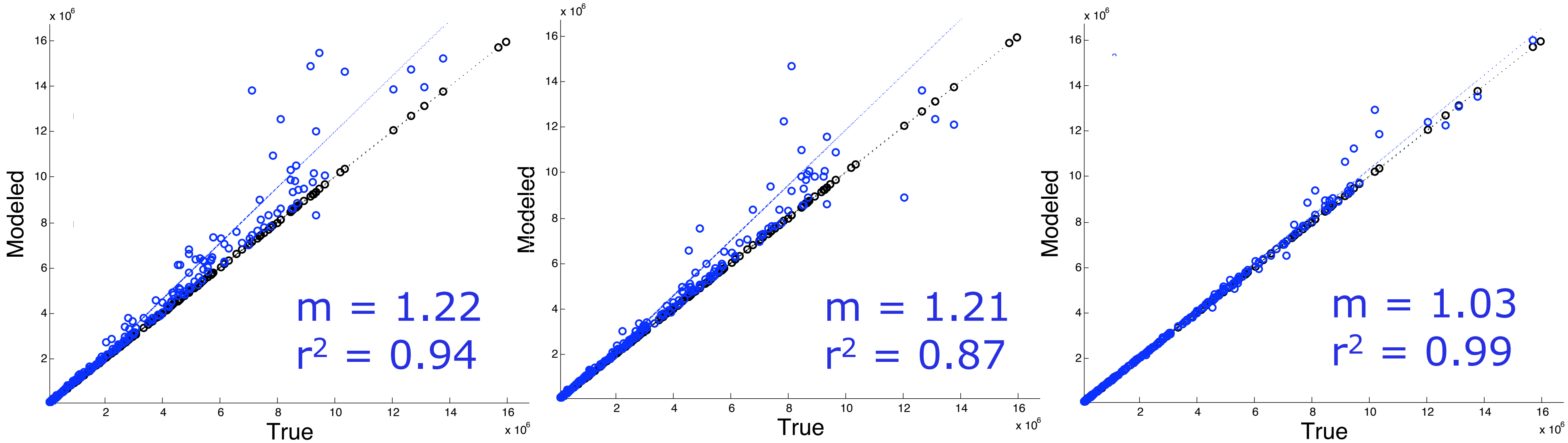


1. Can ideally capture 70% of source variability
2. Inversion has a high bias – what is the source?

Pseudo tests: **before** and **after**

Assimilating various pseudo data sets:

standard retrieval | modified retrieval | true profile



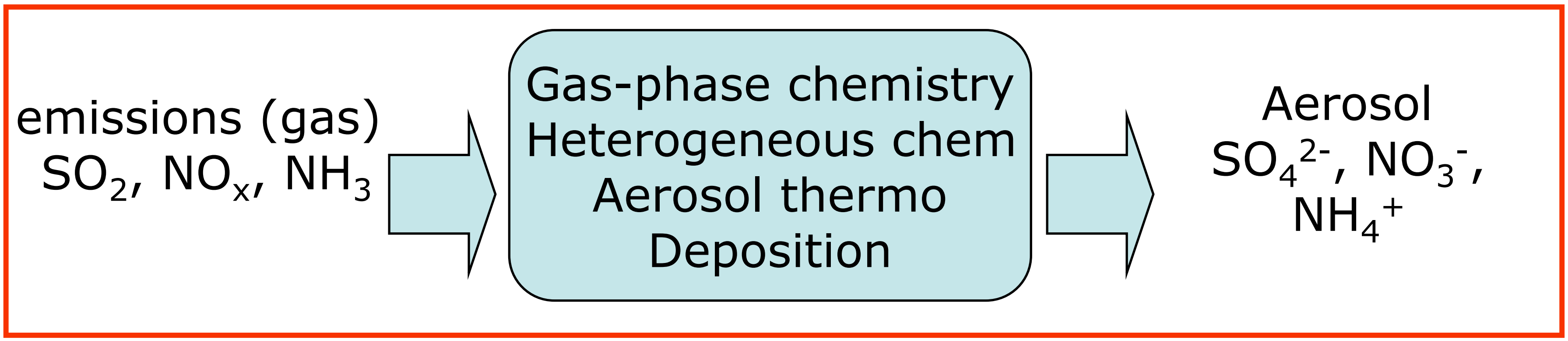
bias and noise

noise

Adjusted retrieval to reduce bias.
Problem is not severely ill-posed.

4D-Var with GEOS-Chem

Forward model v8-02-03 (*Park et al.*, 2004)



Adjoint model (*Henze et al.*, 2007; Kopacz et al., 2009)

Inversion with real data for several months in 2008.

$$J = \frac{1}{2} \sum_{\mathbf{c} \in \Omega} (H\mathbf{c} - \mathbf{c}_{obs})^T \mathbf{S}_{obs}^{-1} (H\mathbf{c} - \mathbf{c}_{obs}) + \frac{1}{2} \gamma_r (\boldsymbol{\sigma} - \boldsymbol{\sigma}_a)^T \mathbf{S}_{\sigma_a}^{-1} (\boldsymbol{\sigma} - \boldsymbol{\sigma}_a)$$

100%, 25% and 10% uncertainty in NH3, NOx, SO2 sources