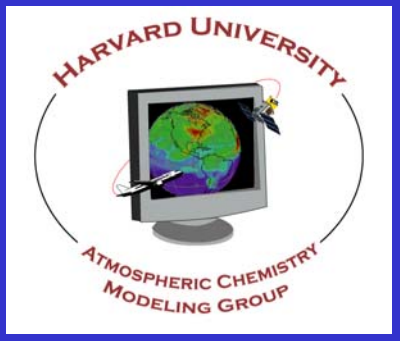
BAYESIAN AND ADJOINT INVERSE MODEL ANALYSES OF PM SOURCES IN THE UNITED STATES USING OBSERVATIONS FROM SURFACE, AIRCRAFT, AND SATELLITE PLATFORMS

Daniel J. Jacob, Harvard (P.I.) and John H. Seinfeld, Caltech (Co-I.)



- Park, R.J., D.J. Jacob, and J.A. Logan, Fire and biofuel contributions to annual mean aerosol concentrations in the United States, *AE (in press)*
- Fairlie, T.D., D.J. Jacob, and R.J. Park, The impact of transpacific transport on mineral dust in the United States, *AE, 41,* 1251-1266, 2007.
- Heald, C.L., D.J. Jacob, et al., Concentrations and sources of organic carbon aerosol in the free troposphere over North America, *JGR 111*, D23S47, 2006.
- Drury, E.E, D.J. Jacob, et al., Improved algorithm for MODIS satellite retrievals of aerosol optical depths over land, to be submitted, 2007
- CALTECH PUBLICATIONS TO BE ADDED!

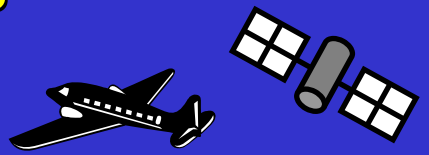
Supported by EPA-STAR

USING OBSERVED ATMOSPHERIC CONCENTRATIONS TO IMPROVE SOURCE INVENTORIES

A priori "bottom-up" source vector $\mathbf{x}_a$, error covariance matrix $\mathbf{S}_a$

Chemical transport model

$$\mathbf{y} = \mathbf{F}(\mathbf{x}) + \boldsymbol{\varepsilon}_m$$



Observation vector $\mathbf{y}$, error covariance matrix $\mathbf{S}_y$; diagnose $\mathbf{S}_y^{-1}(\mathbf{y}-\mathbf{F}(\mathbf{x}_a))$ (forward model analysis)

FORMAL INVERSE ANALYSIS: seek true source x by minimizing $J(\mathbf{x})$:

$$J(\mathbf{x}) = (\mathbf{y} - \mathbf{F}(\mathbf{x}))^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a)$$

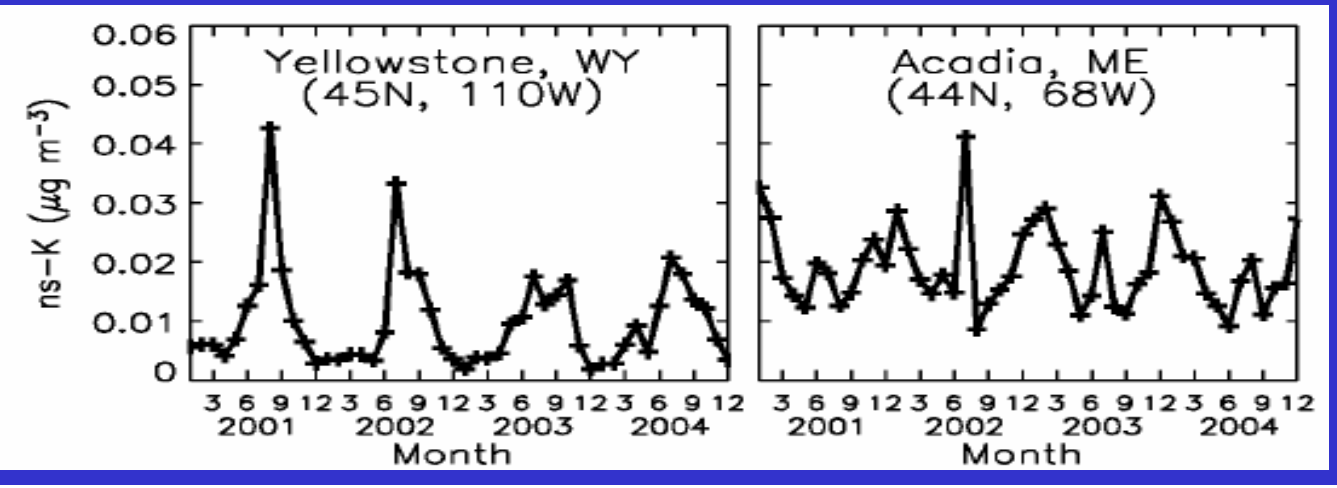
- **Analytical ("Bayesian") method:** for linear(ized) model $\mathbf{y} = \mathbf{K}\mathbf{x}$, solve analytically

$$\nabla_x J(\mathbf{x}) = 0 \;\Rightarrow\; \hat{\mathbf{x}} = \mathbf{x}_a + (\mathbf{K}^T \mathbf{S}_y^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} \mathbf{K}^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{K}\mathbf{x}_a)$$

- **Adjoint method:** solve numerically by steepest-descent algorithm based on successive calculations of $\nabla_x J(\mathbf{x})$:

$$\nabla_x J(\mathbf{x}) = -\underbrace{(\nabla_x \mathbf{F}(\mathbf{x}))^T}_{\text{adjoint of forward model}} \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x})) + \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a)$$

QUANTIFYING THE BIOMASS BURNING SOURCE OF PM2.5 FROM CORRELATION OF TOTAL CARBON (TC) WITH NON-SOIL K^+ AT IMPROVE SITES

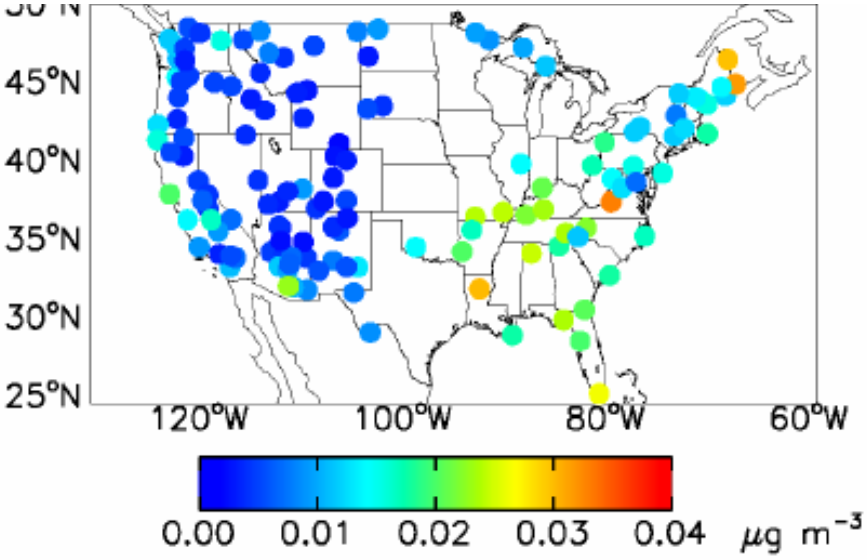


Non-soil K^+:

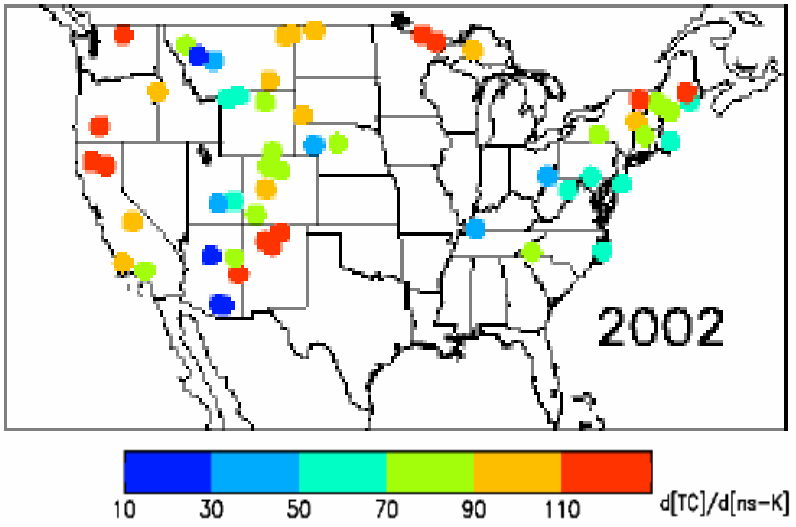
West: summer max, low background

East: winter and summer maxes, high background

Background ns-K^+ (traces industrial biofuel)



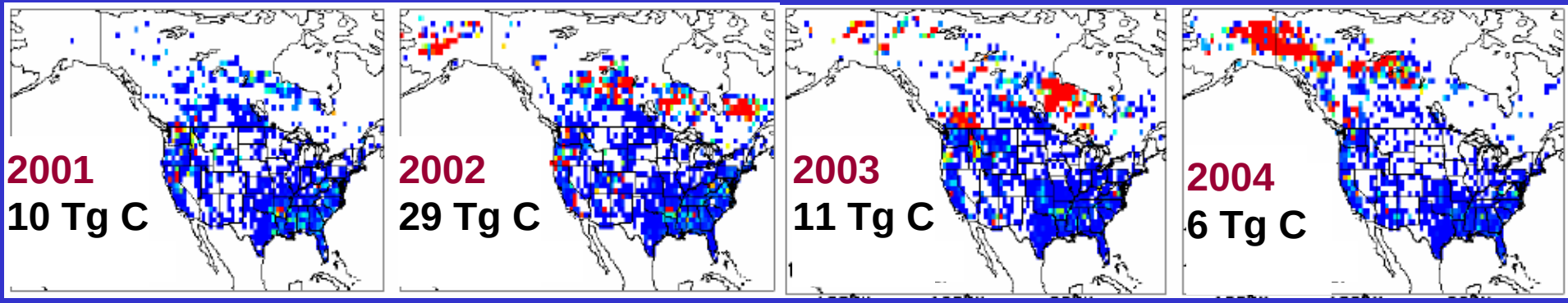
Attribute TC from open fires from RMA slope of TC vs. ns-K^+ linear regression

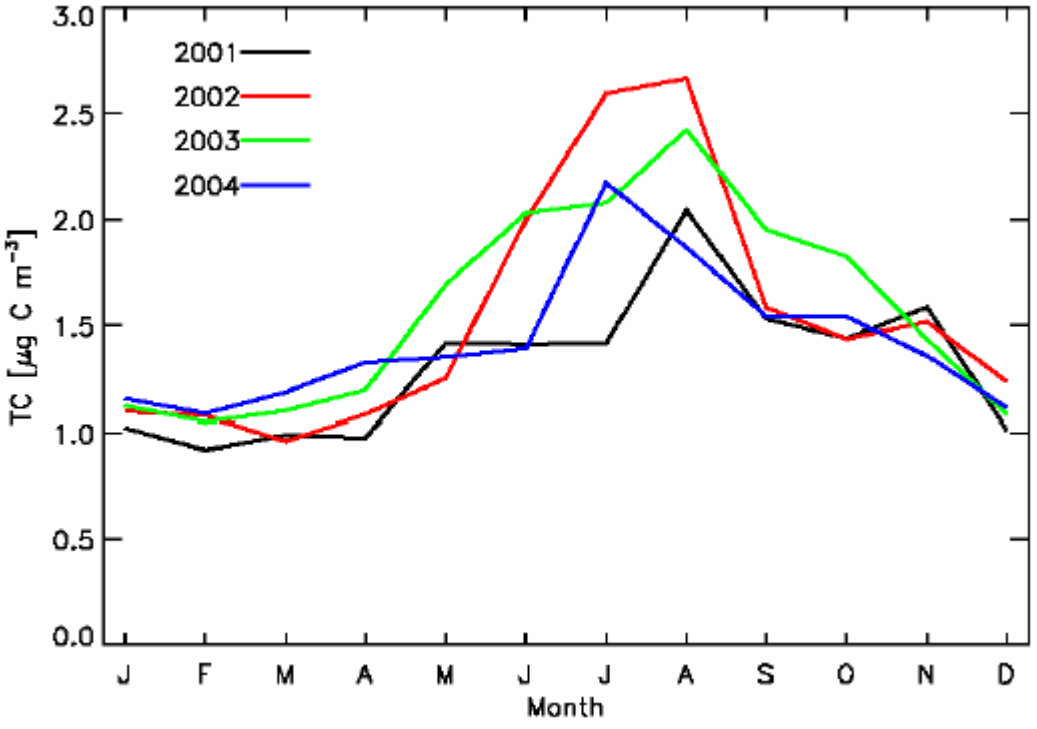


Park et al. [2007]

SEASONAL VARIATION OF TC IS DRIVEN BY SUMMER WILDFIRES

JJA fire emission inventory from MODIS data [van der Werf et al., 2006] and contiguous U.S. dry mass burned totals (1997-2004 mean 16 Tg C)



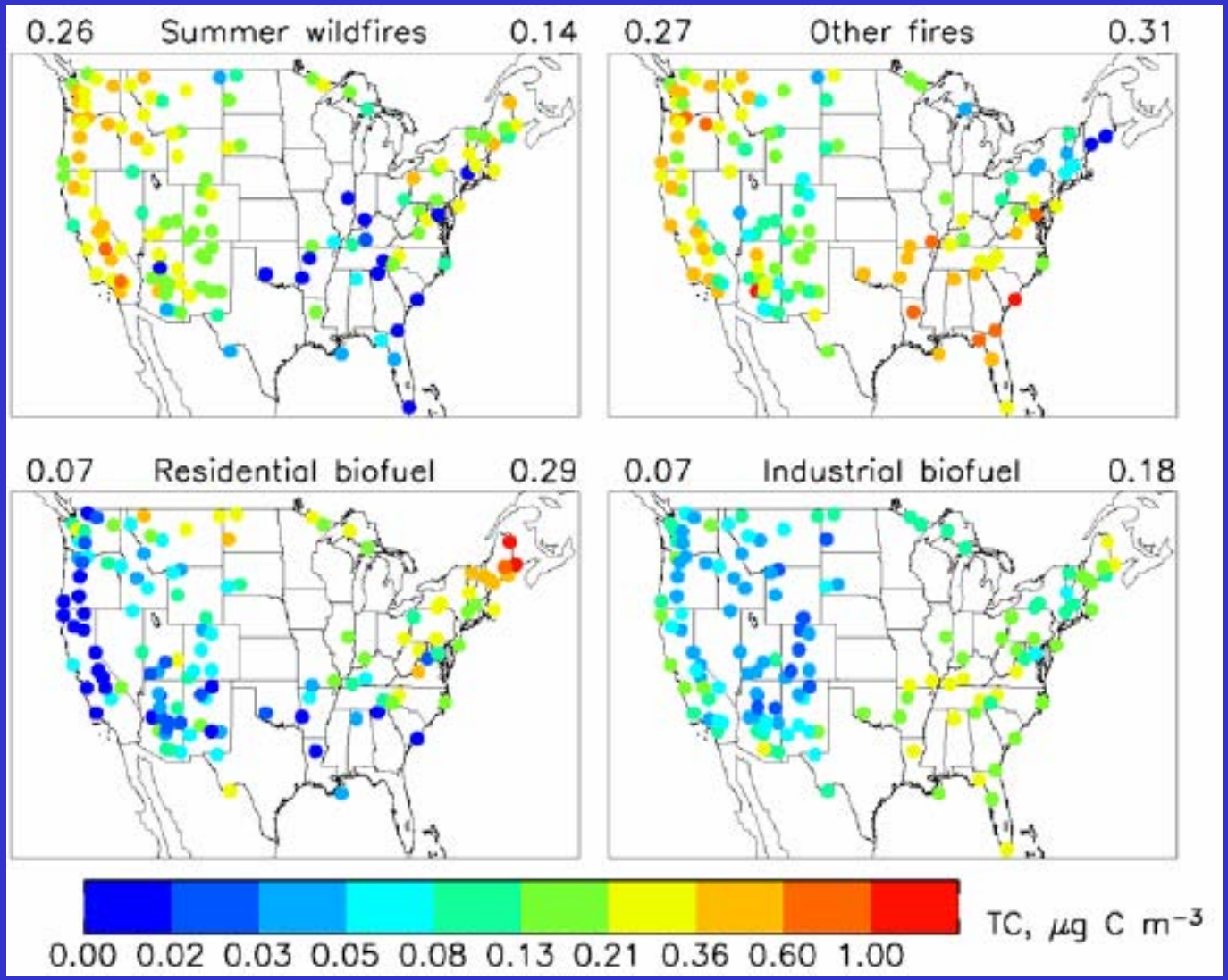


TC seasonal variation averaged over all U.S .IMPROVE sites

Park et al. [2007]

FIRE AND BIOFUEL CONTRIBUTIONS TO TC PM2.5

Annual mean C concentrations, 2001-2004 –also mean values in W and E



Park et al. [2007]

IMPLICATIONS FOR AQ STANDARDS AND EMISSION INVENTORIES

Source attribution at IMPROVE sites (annual means)

PM2.5, μg m-3 (x1.8 for TC)	West	East
Summer wildfires	0.47	0.25
Other fires	0.49	0.56
Residential biofuel	0.13	0.52
Industrial biofuel	0.13	0.32
Total fire+biofuel TC (this work)	1.2	1.6
Observed TC	2.2	3.4
Observed total PM2.5	3.8	8.2

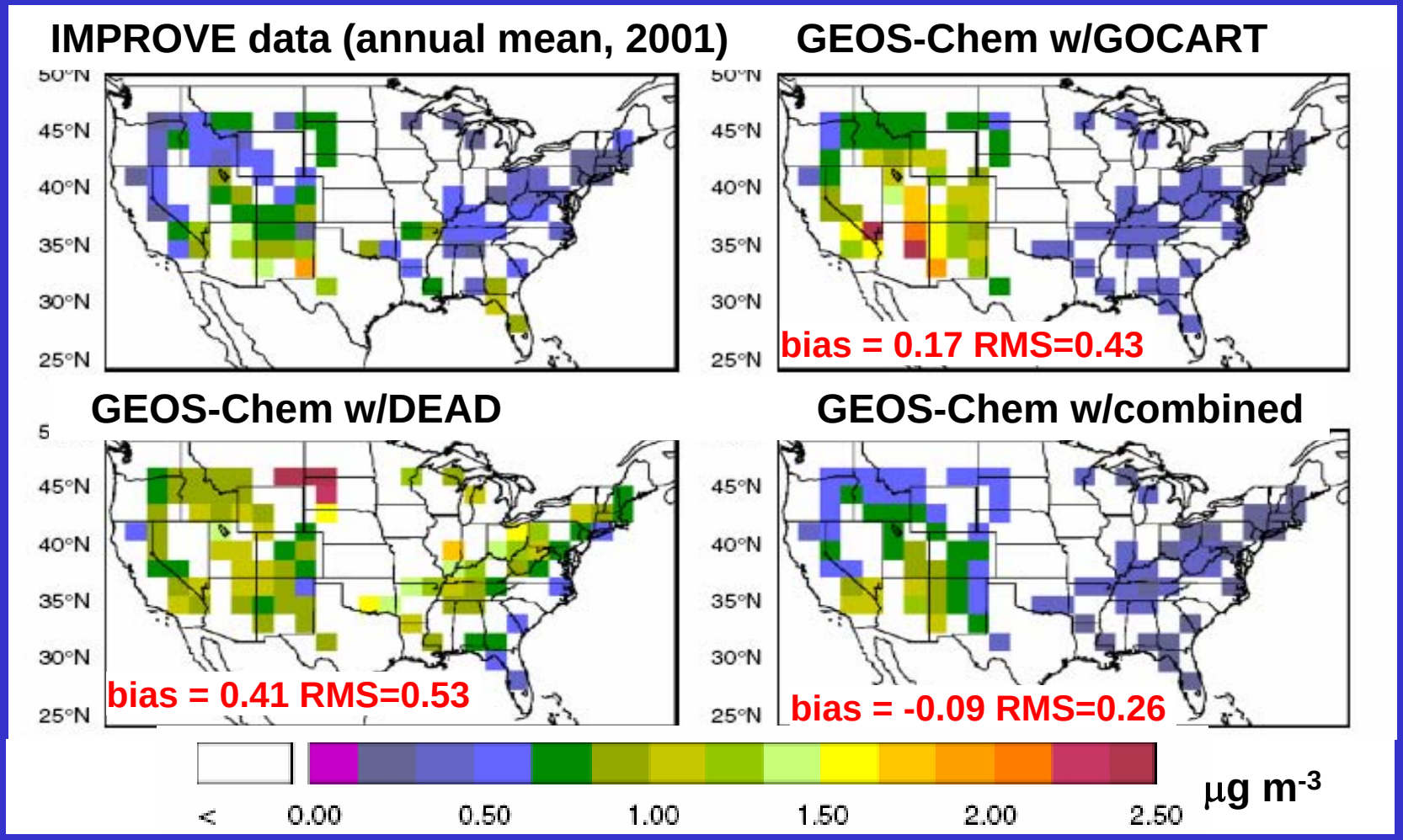
Emissions for contiguous U.S. (climatological fires)

	Tg C y^{-1}
Top-down	
This work	
Open fires	0.90
Biofuel	0.51 (40% ind.)
Park et al. [2003]	
Open fires	0.68
Biofuel	0.96 (80% ind.)
Bottom-up	
Bond et al. [2004]	
Open fires	0.89
Biofuel	0.42 (<10% ind.)
NEI99	
Biofuel	0.23 (<10% ind.)

Park et al. [2007]

TESTING DUST EMISSION SCHEMES WITH IMPROVE DATA

GOCART [Ginoux et al., 2004] , DEAD [Zender et al.,2003] and combined schemes
Implemented in GEOS-Chem global simulation



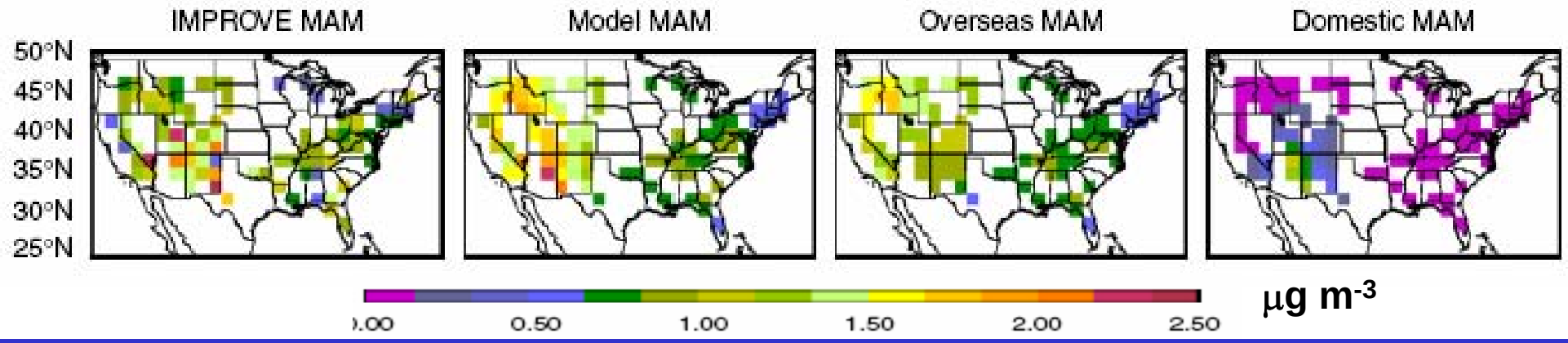
GOCART has two low wind threshold
DEAD has insufficient areal restriction

} combination of two gives significant improvement

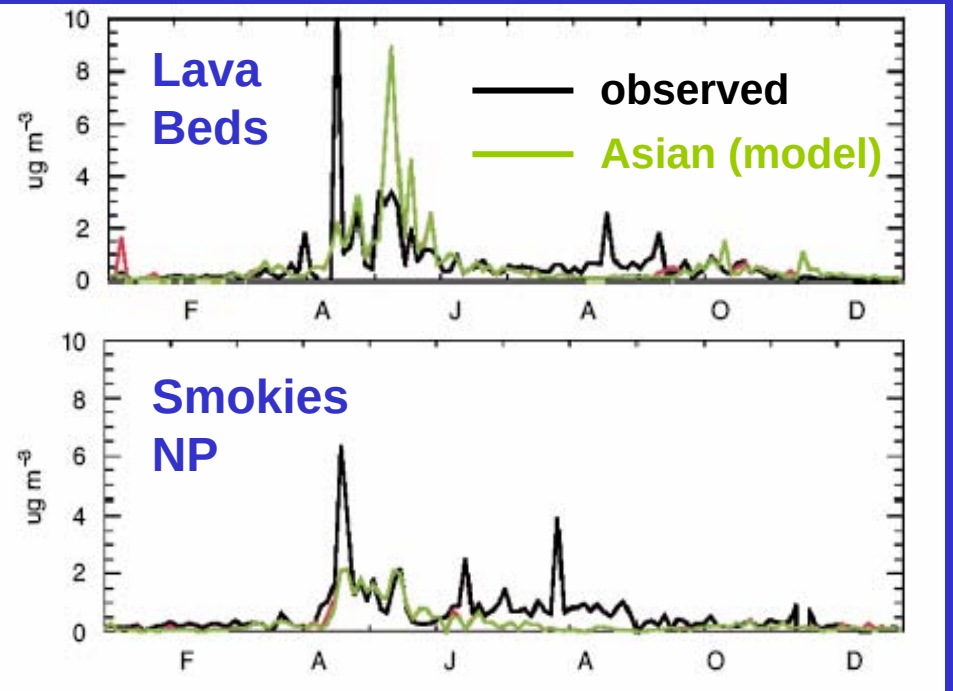
Fairlie et al. [2007]

ASIAN CONTRIBUTION TO U.S. DUST (2001)

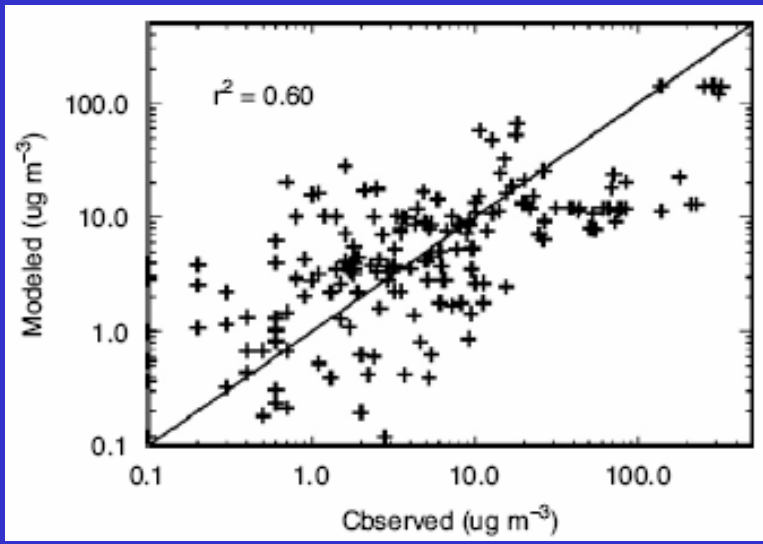
Spring is max dust season in most of U.S., and source is mainly from Asia



Annual time series; Asian dust is combination of events + seasonal background



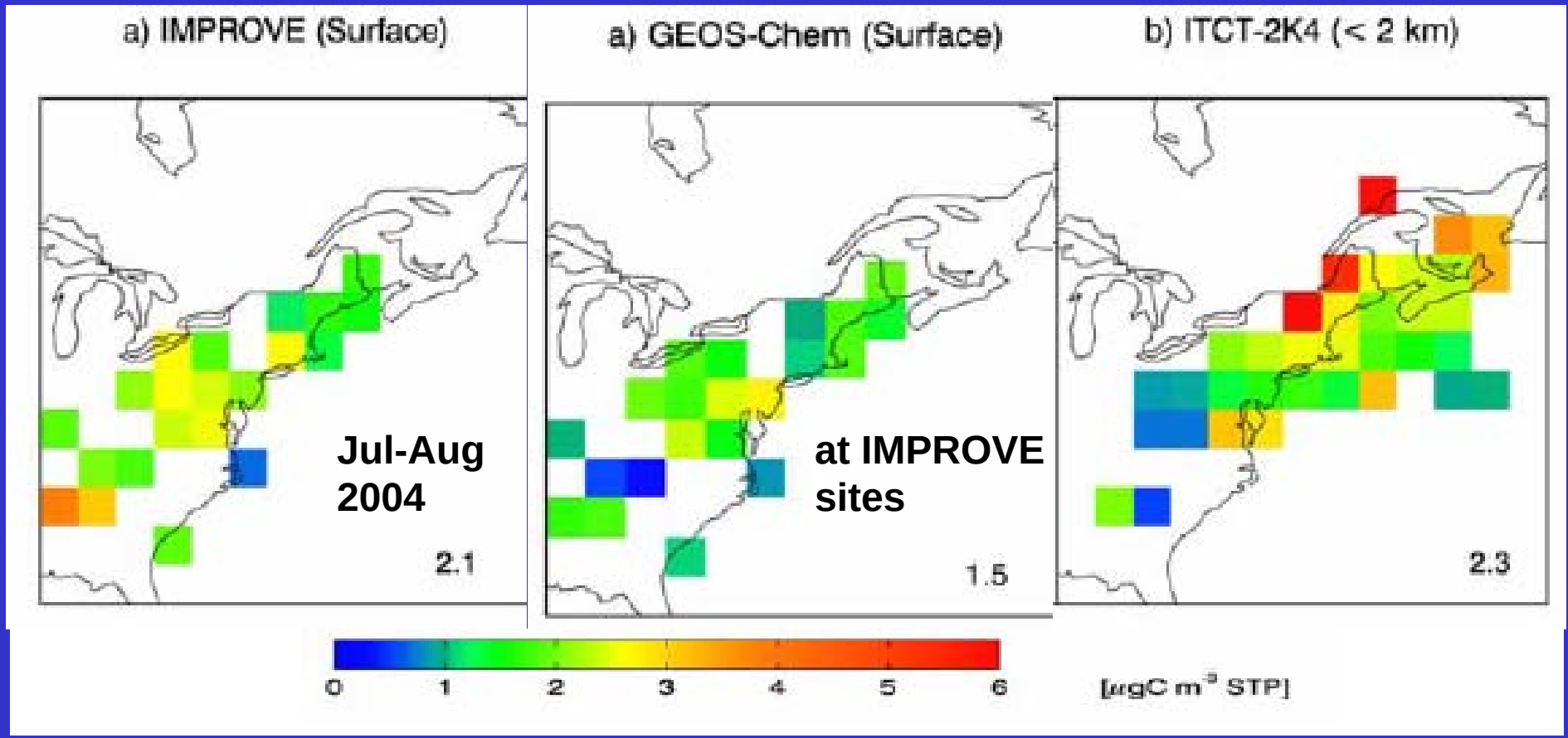
Model simulation of spring 2001 Asian outflow (TRACE-P aircraft data)



Fairlie et al. [2007]

TESTING OC SOURCES WITH ITCT-2K4 AIRCRAFT DATA

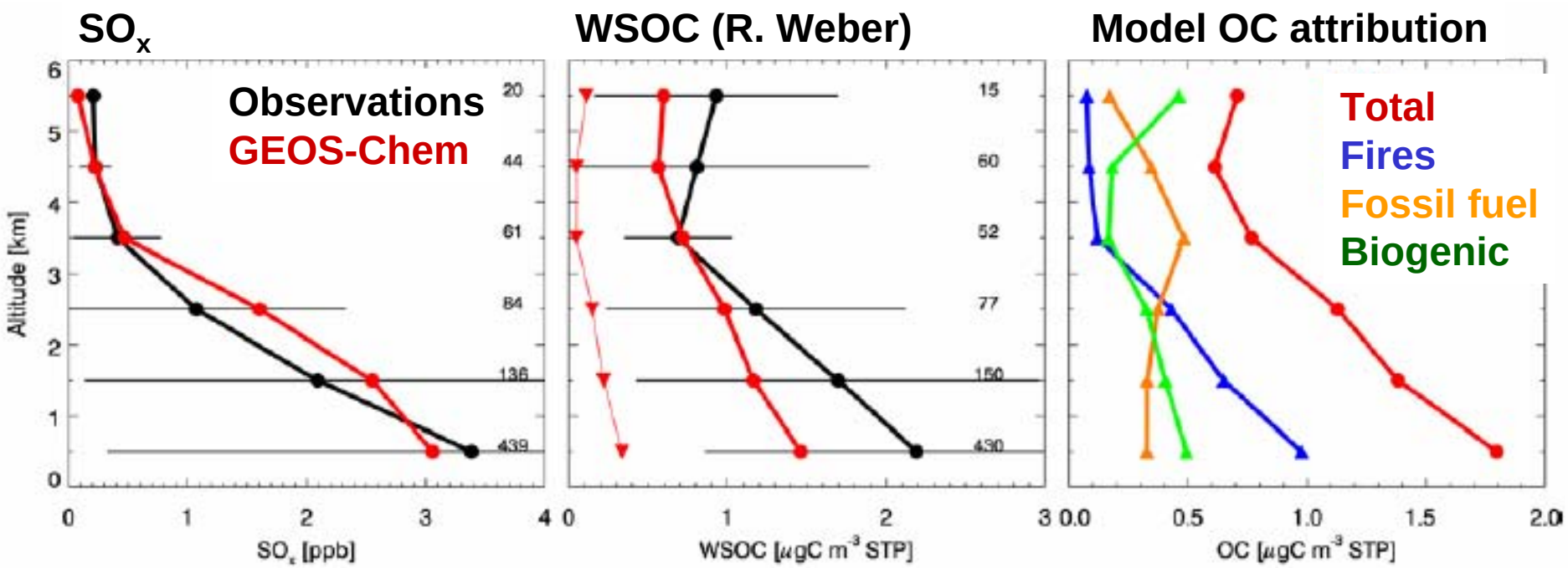
Jul-Aug 2004 out of Portsmouth, NH;
water-soluble OC (WSOC) measurements by R. Weber (Georgia Tech)



- **IMPROVE and ITCT-2K4 consistent; high aircraft values in N. New England due to Canadian fires**
- **Model low by 30%, weak correlation w/aircraft data (R^2 = 0.10)**
- **0-2 km source attribution in model: 49% anthropogenic, 27% fires, 8% biogenic (isoprene), 16% biogenic (other)**

Heald et al. [2006]

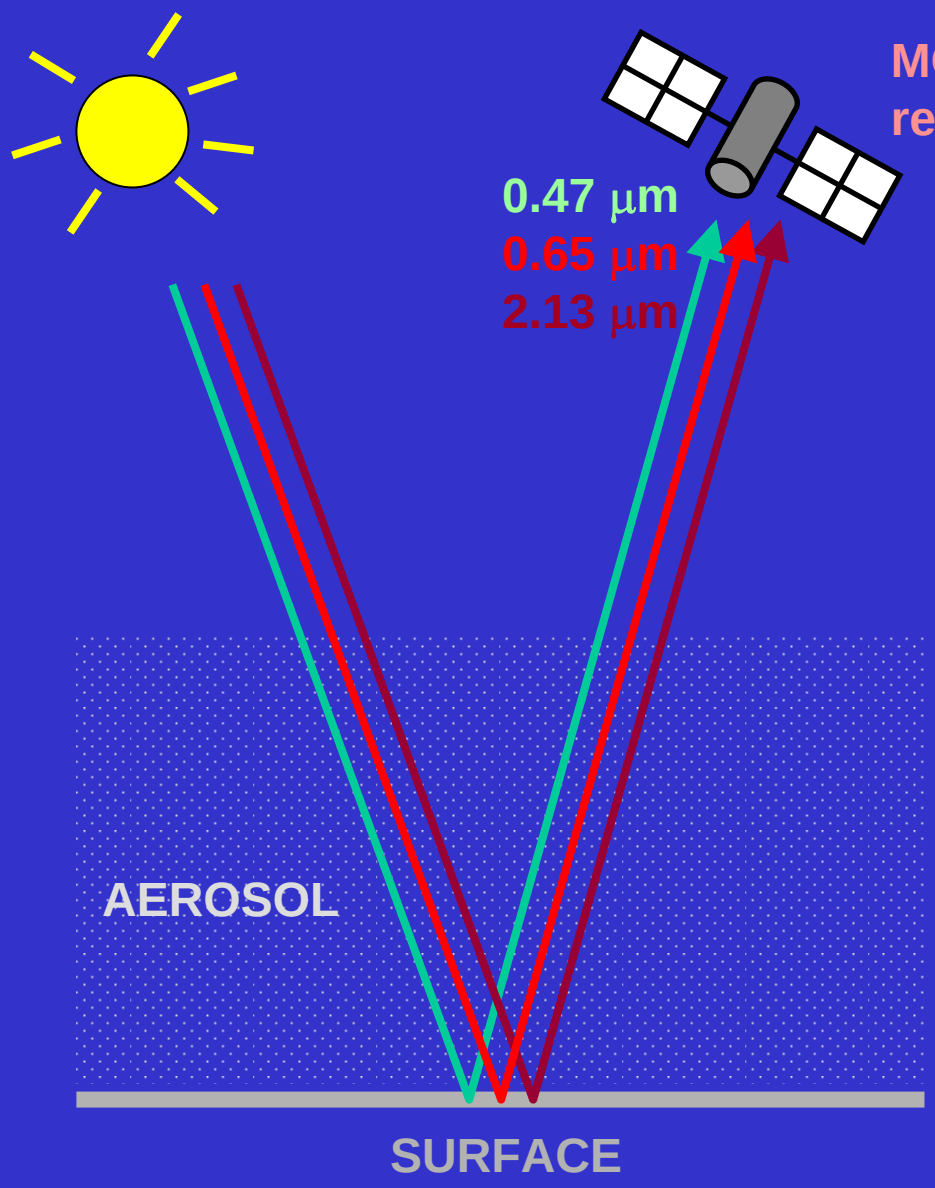
WSOC AEROSOL OVER EASTERN U.S. IN ITCT-2K4



- Observed free tropospheric concentrations are lower than in ACE-Asia; model is only 25% too low in the mean…
- …but is incapable of reproducing observed variability in boundary layer (r^2 = 0.11) or free troposphere (R^2 = 0.05)
- Observed free tropospheric WSOC has significant correlation (R^2 ~ 0.3) with methanol combined with sulfate, nitrate, or toluene; suggests cloud production mechanism and combined bio-anthro source

Heald et al. [2006]

IMPROVING MODIS SATELLITE RETRIEVALS OF AEROSOL OPTICAL DEPTHS OVER LAND



Drury et al. [2007]

MODIS measures top-of-atmosphere (TOA) reflectance in several wavelength channels

- **Interpretation of TOA reflectance in terms of AOD requires assumptions on surface reflectance, aerosol optical properties**

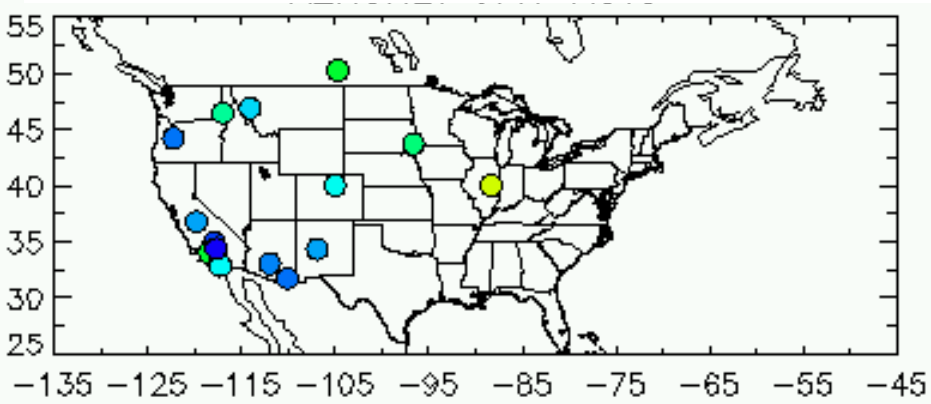
- **Use TOA reflectance at 2.13 μm (transparent atmosphere) to derive surface reflectance**

- **MODIS operational algorithm relies on general assumptions for 0.47/2.13 surface reflectance ratios and aerosol optical properties; we improve by deriving those locally using GEOS-Chem model information**

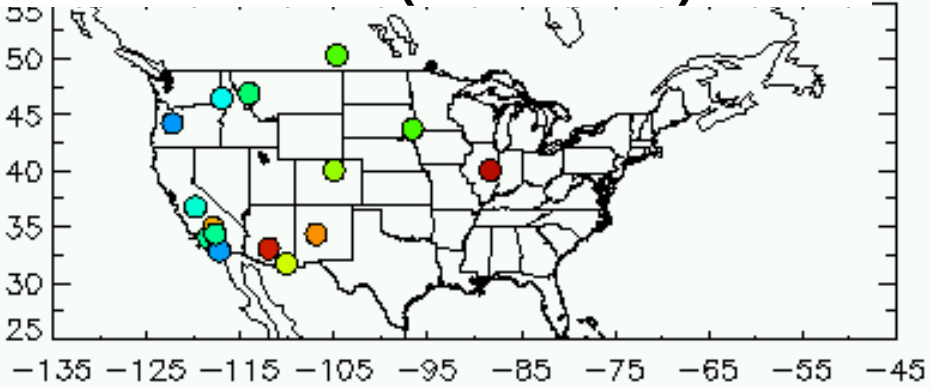
- **The resulting retrieval (or model reflectance) is suited for quantitative interpretation including inverse analyses**

COMPARISON vs. AERONET 0.47 μm AODs (Jul-Aug 2004)

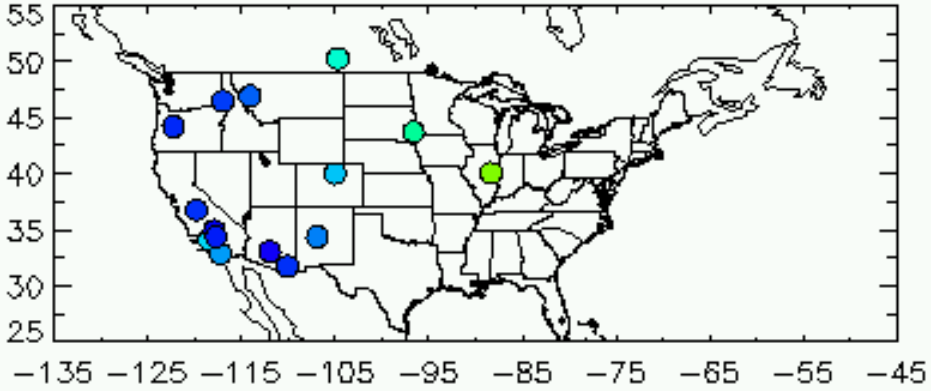
AERONET



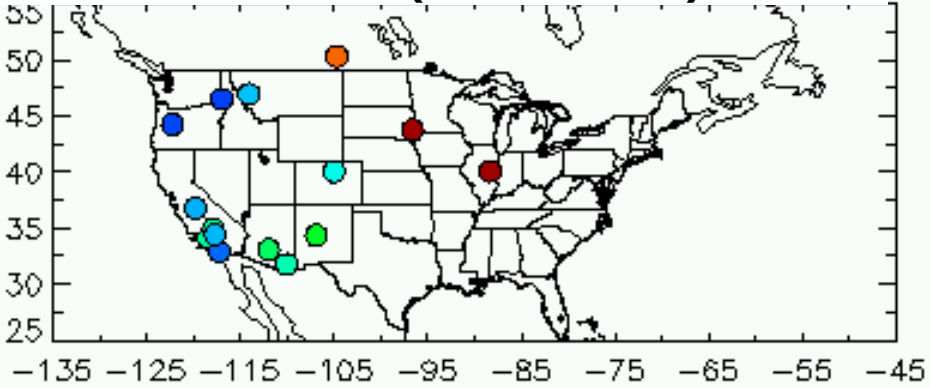
MODIS (collection 5)

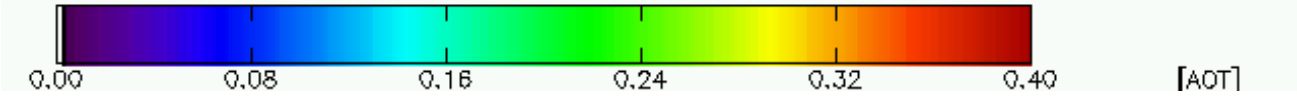


MODIS (this work)



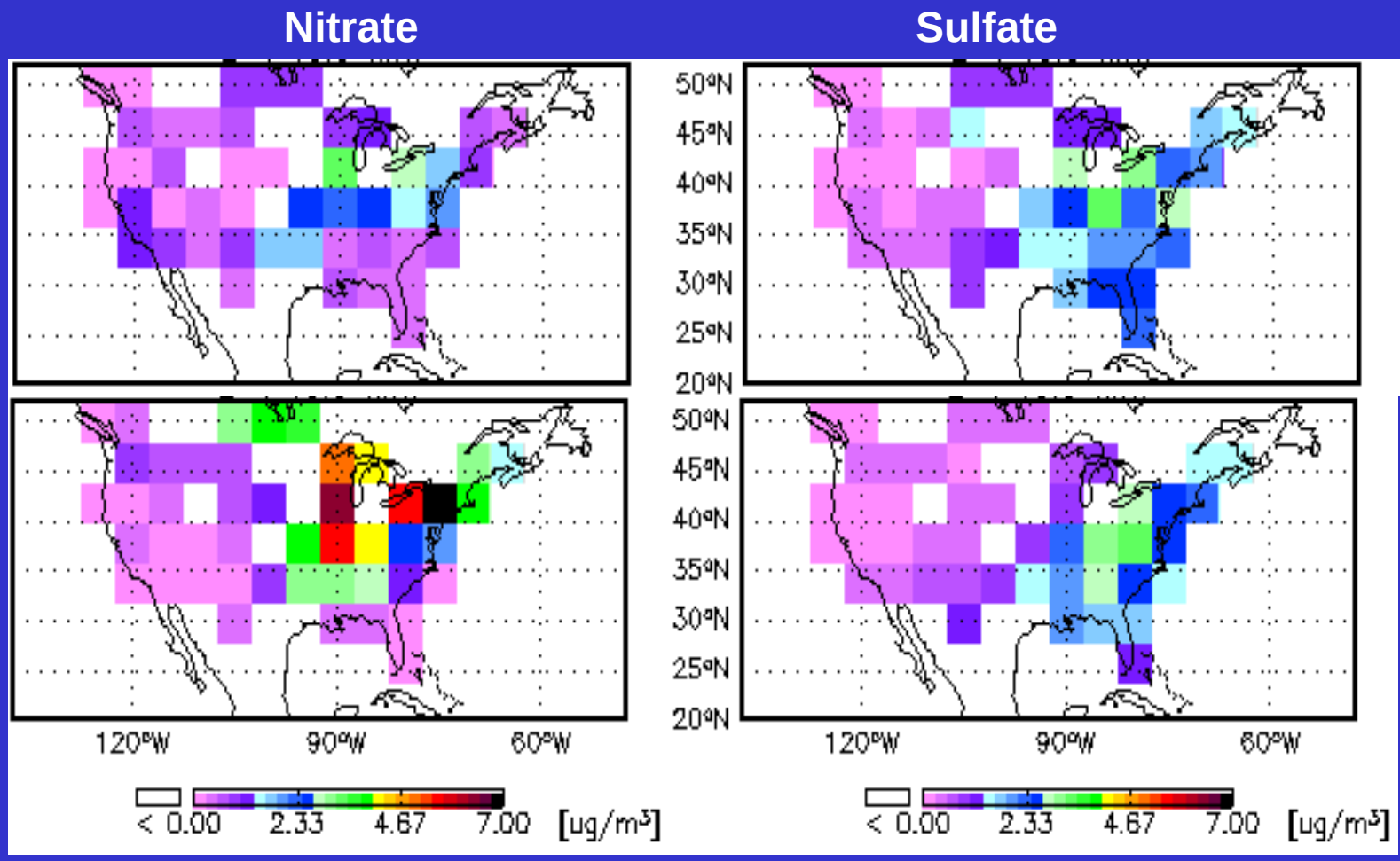
MODIS (collection 4)





Drury et al. [2007]

ADJOINT MODEL APPROACH TO IMPROVE PM NITRATE SIMULATION

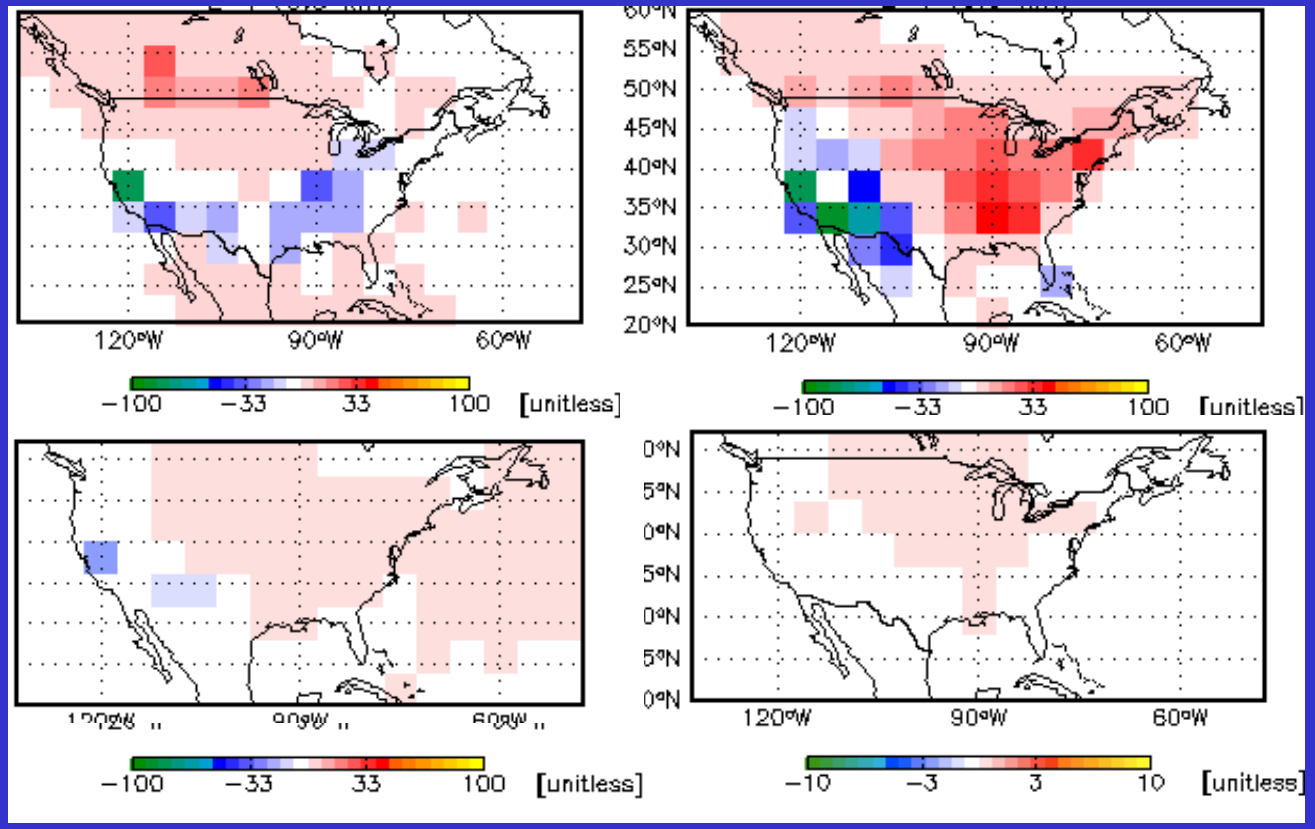


Use LSQ cost function *J* to fit $[NO_3^-]$ and $[SO_4^{2-}]$ as function of model parameters

Henze et al. [2007]

SENSITIVITY OF COST FUNCTION TO MODEL PARAMETERS

NO_x emissions



Anthro. NH_3 emissions

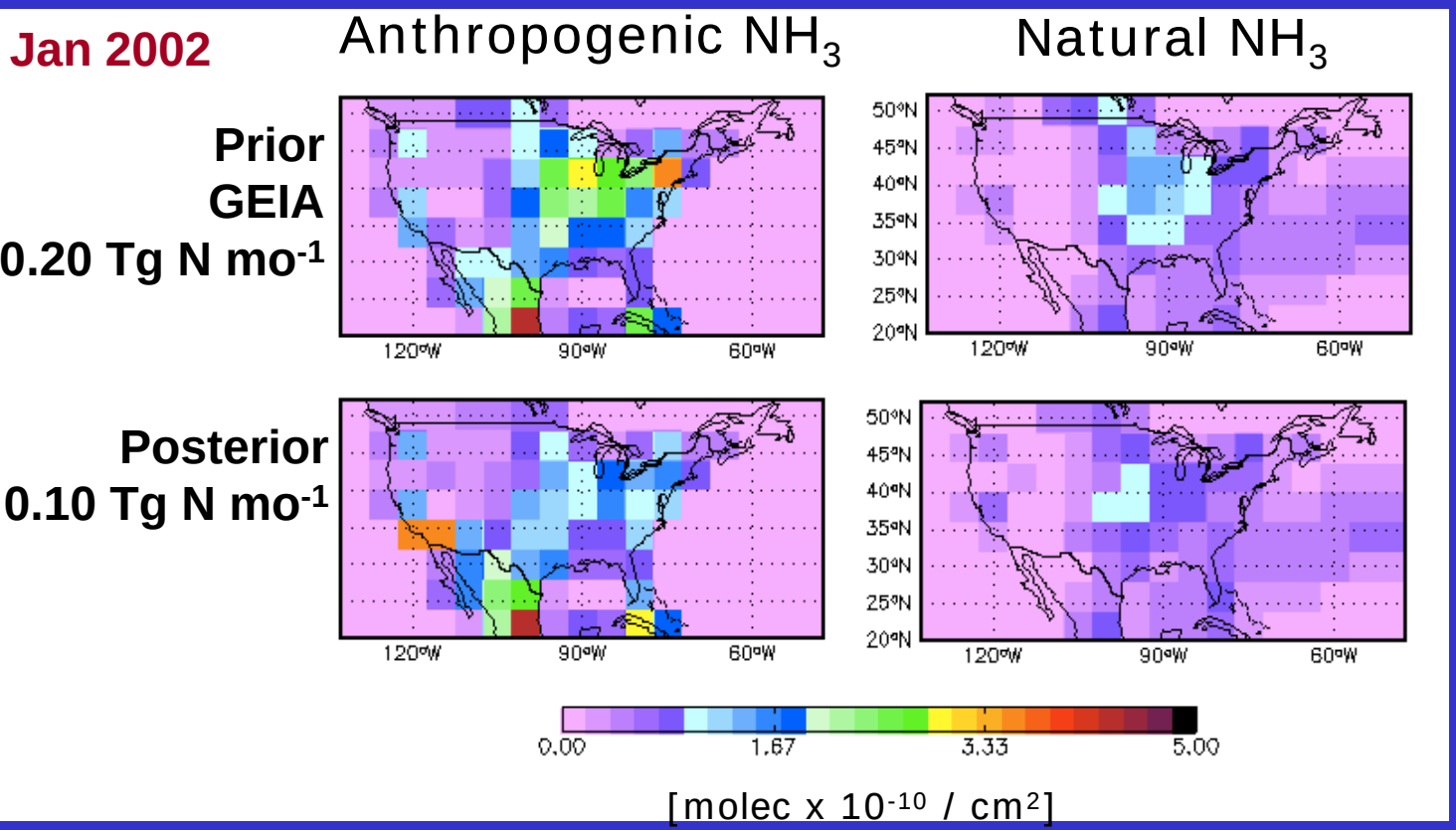
γ_{N2O5}

Initial conditions

Henze et al. [2007]

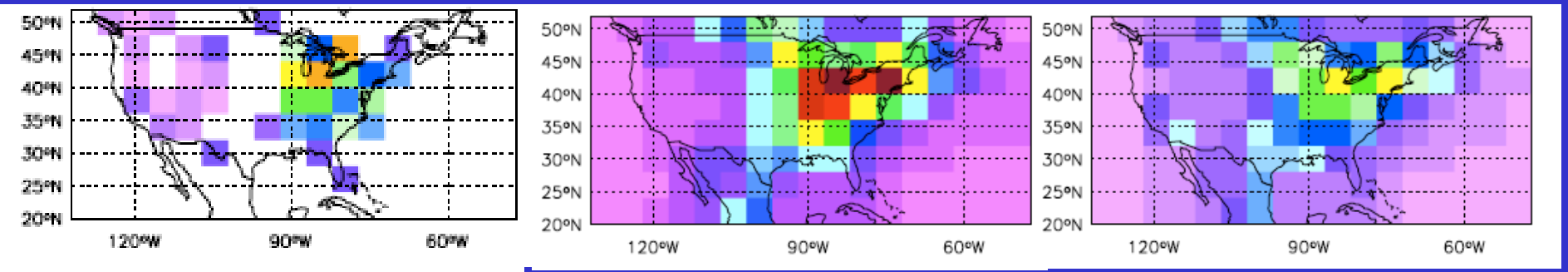
OPTIMIZATION OF NH_3 EMISSIONS USING NO_3^- AND SO_4^{2-} DATA

Jan 2002



Other recent estimates are in range 0.08-0.12 Tg N mo^{-1} [Gilliland et al., 2003, 2006; Pinder et al., 2003, 2006; Park et al., 2004]

NH_4^+, observed | **NH_4^+, model (*prior*)** | **NH_4^+, model (*posterior*)**



Henze et al. [2007]