Towards Refining Estimates of Ammonia Emissions: Modeling Framework Preparations

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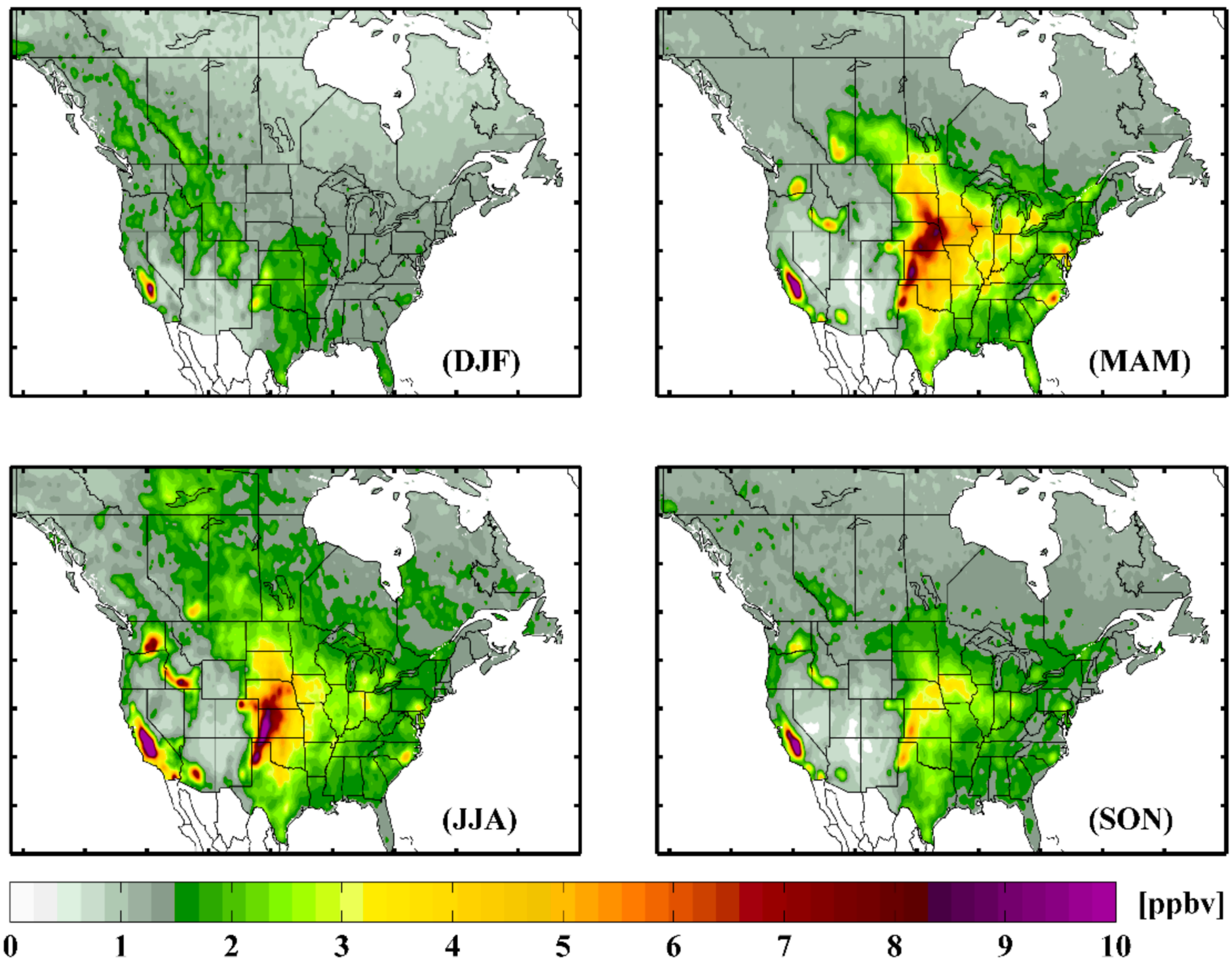
CMAS 2019

Ammonia in the Atmosphere

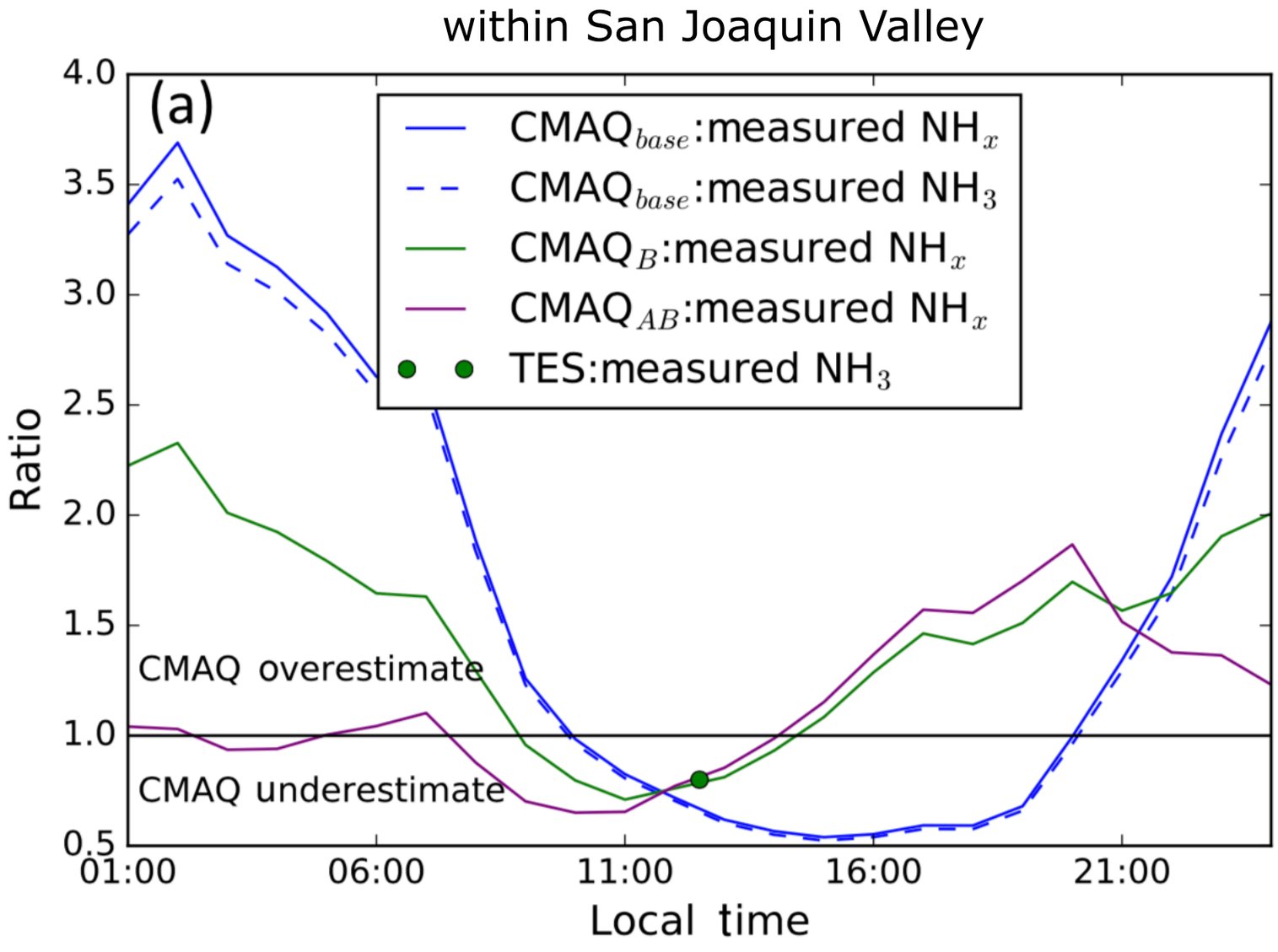
5-year mean of surface NH_3 from CrIS (2013-2017)



(Shephard et al., ACPD, 2019)

Challenges Modeling Ammonia

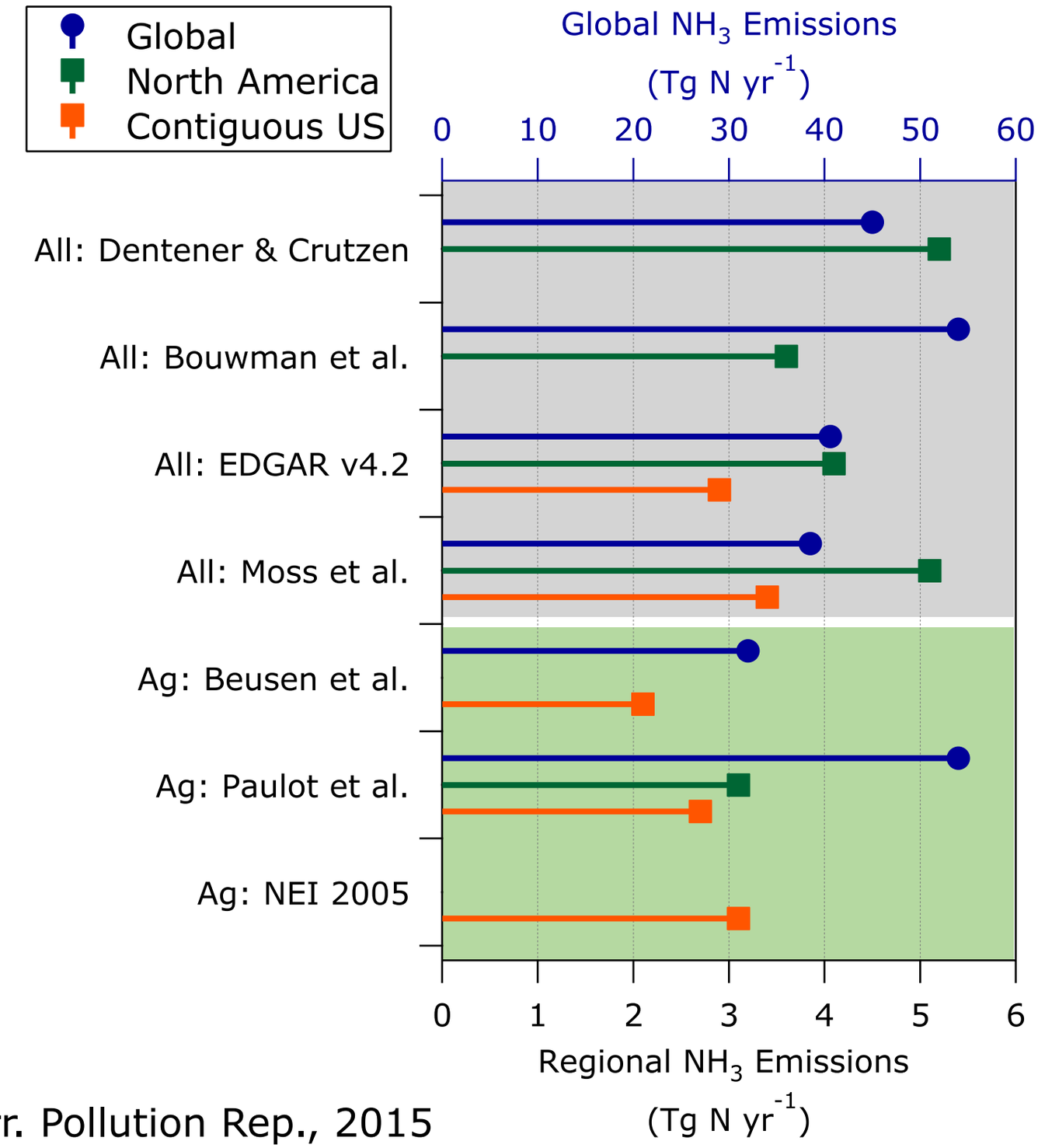
- Emissions estimation from variable sources
- Volatility of gas
- Potential for bidirectional flux



Lonsdale et al., ACP, 2017

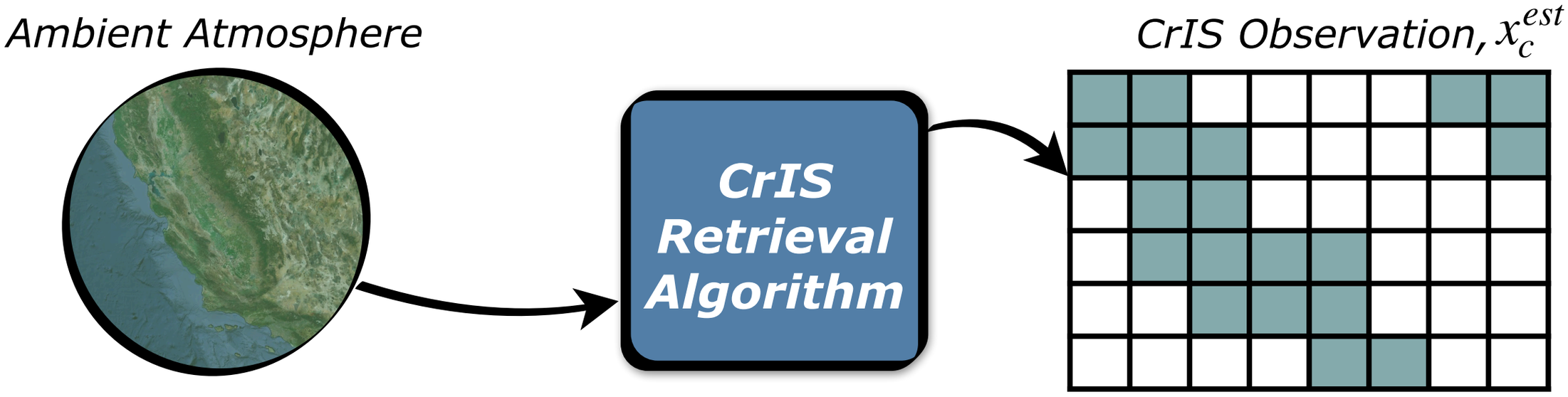
Uncertainty in Ammonia Emissions

- Between 25% and 50% spread in emissions estimates exists across inventories.
- Select agricultural contributions are equivalent to other estimates of total emissions.



Zhu et al., Curr. Pollution Rep., 2015

Assimilating CrIS Observations

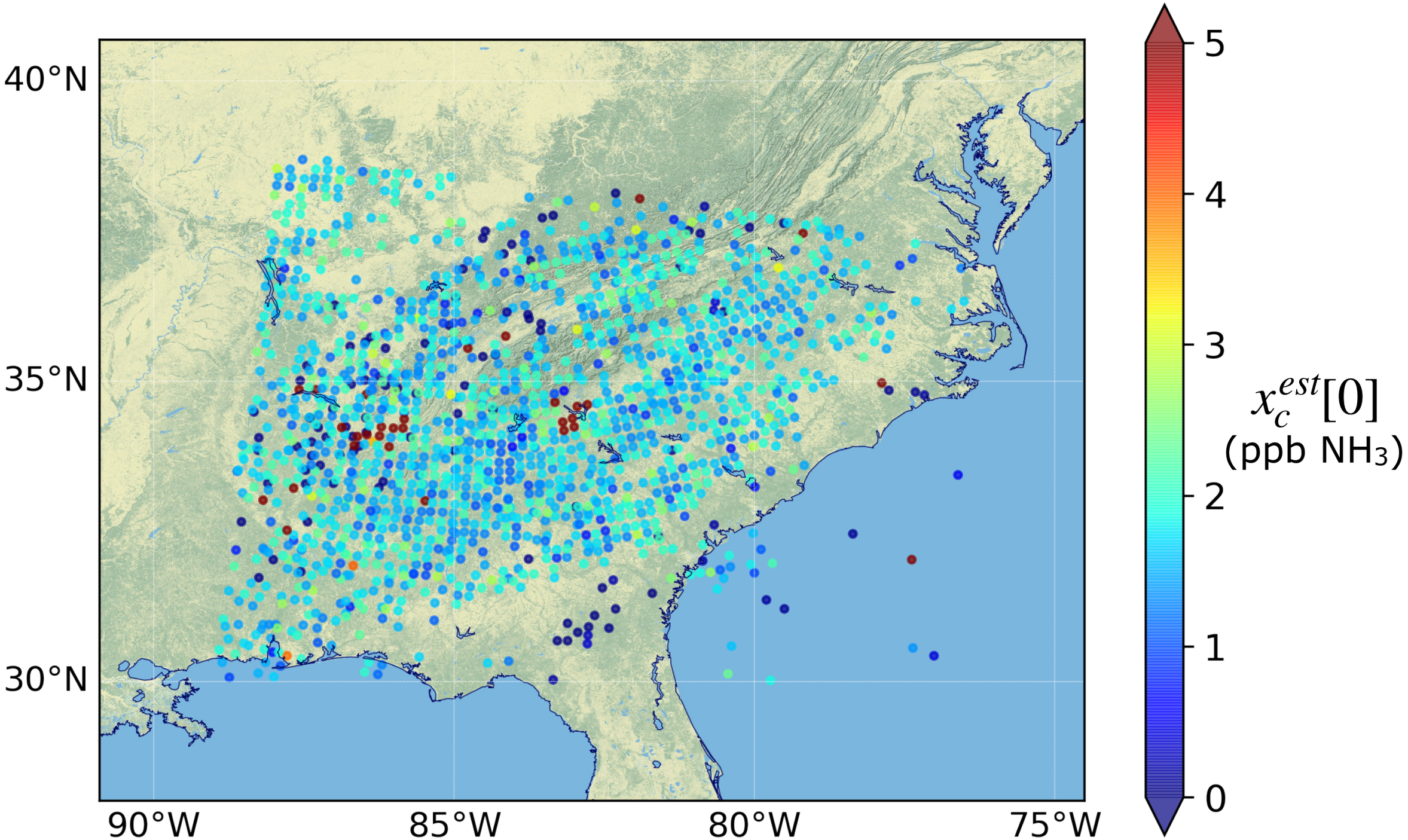


$$x_c^{est} = x_a + A(x_c^{mapped} - x_a)$$

where x_a is a profile based on
clean, moderate, or polluted conditions

Shephard and Cady-Pereira, Atmos. Meas. Tech., 2015

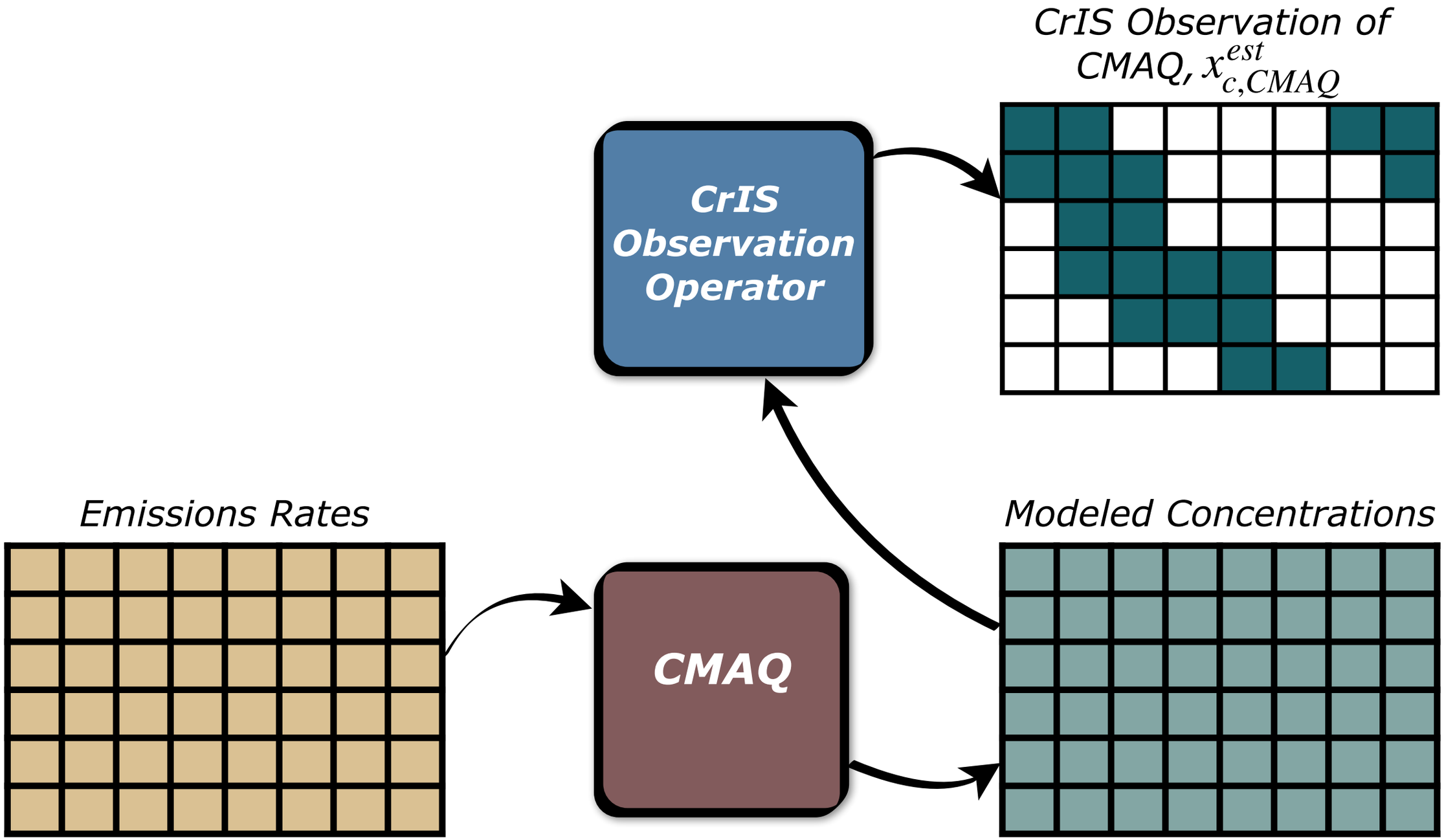
Sample CrIS Retrievals



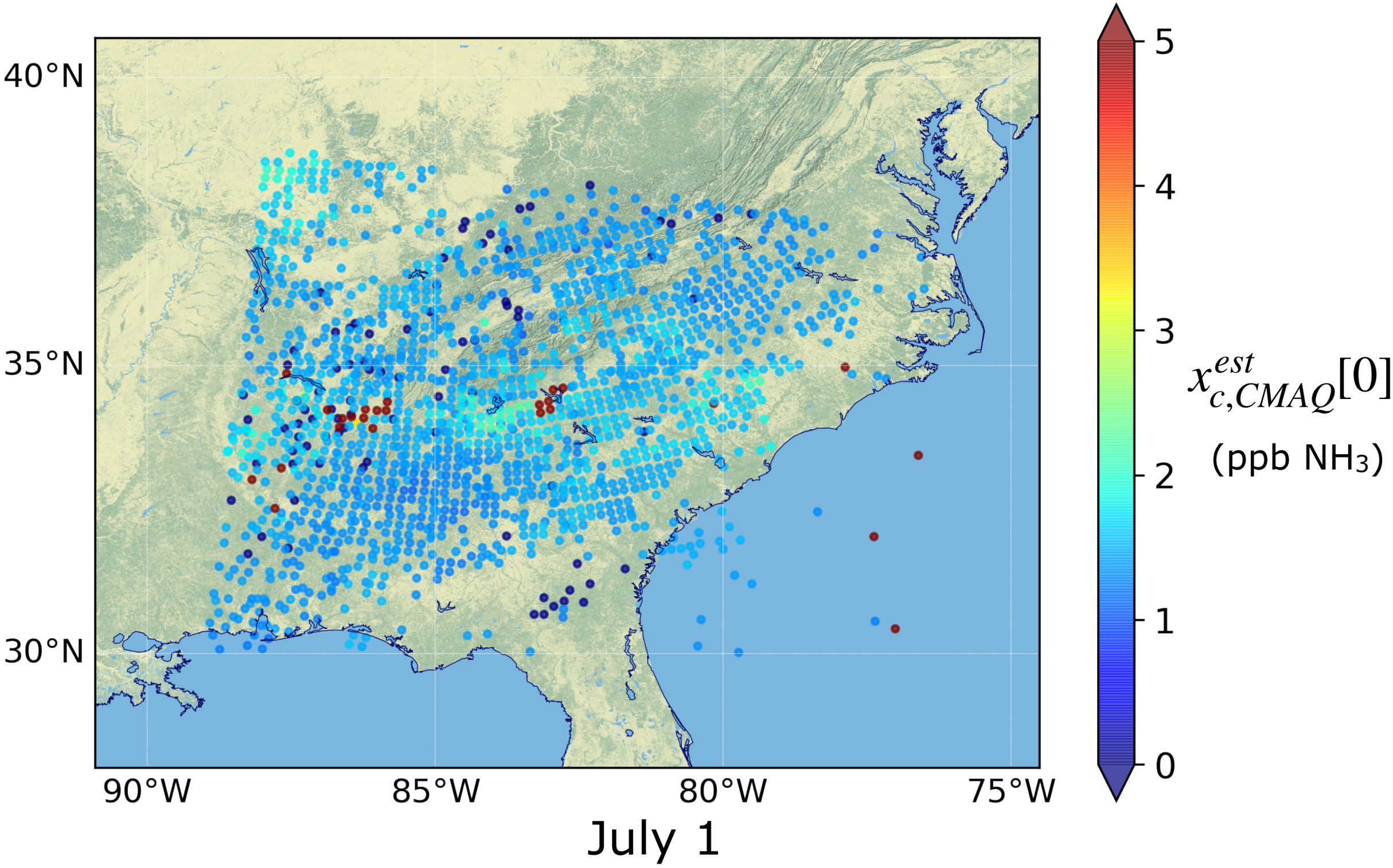
CrIS v.1.3, July 1

courtesy of Mark Shephard

Simulating CrIS Observations

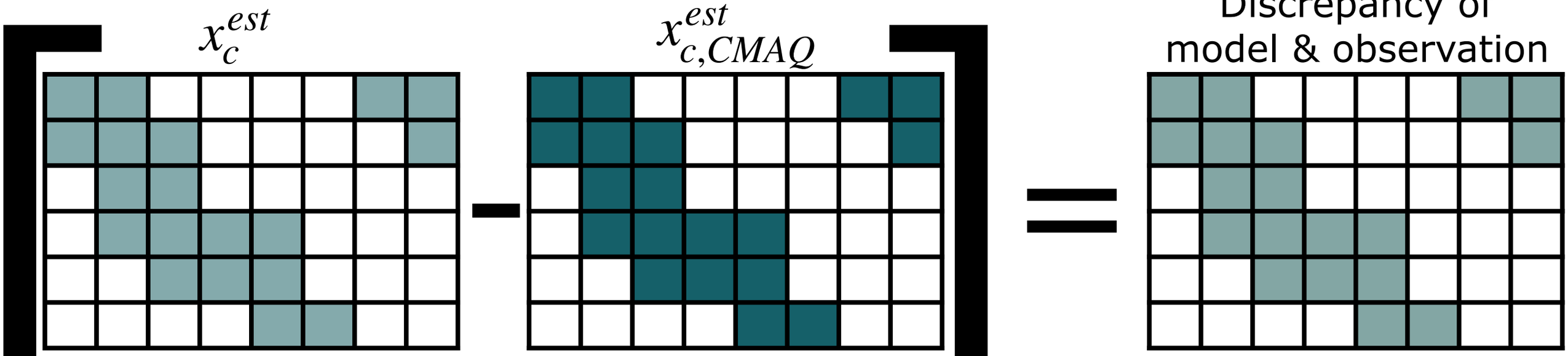


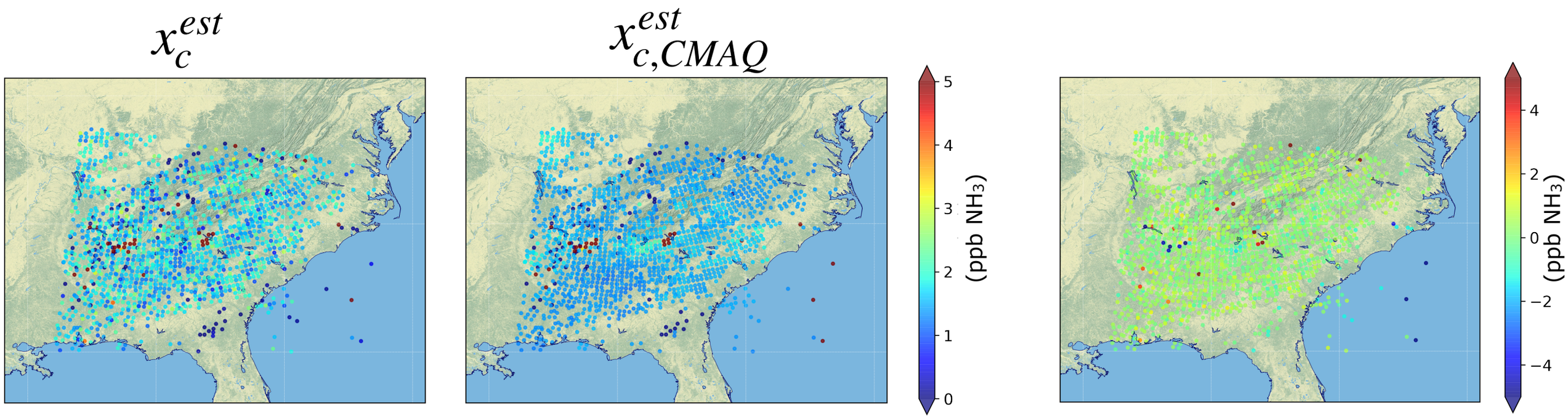
CrIS Retrieval Applied to CMAQ



Comparing CrIS Observations of Ambient & CMAQ Ammonia

$$J = (x_{c,CMAQ}^{est} - x_c^{est}) S_{obs}^{-1} (x_{c,CMAQ}^{est} - x_c^{est})$$





Attribute Difference to Emissions with CMAQ adjoint

$$\frac{\partial(\text{concentrations})}{\partial(\textbf{emissions})}$$

∂(emissions)

∂(SO_2 Power Plant Emissions)

∂(NH_3 Agricultural Emissions)

∂(NO_x Refinery Emissions)

∂(NO_x Vehicle Emissions)

∂(Seasalt Emissions)

∂(concentration)

$$\frac{\partial J}{\partial NH_3}$$

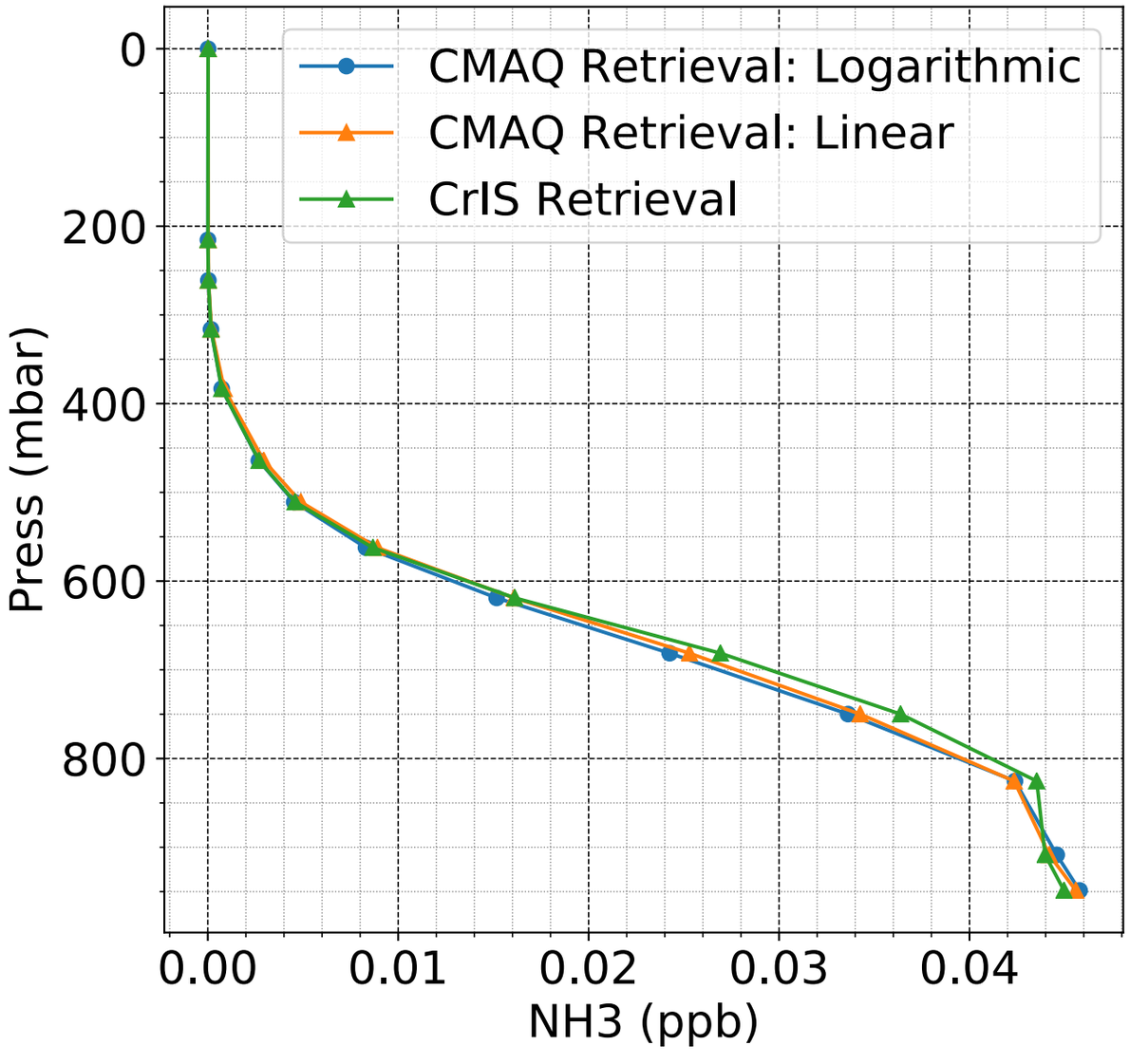
$$\partial\vec{y} = F'^{T}\partial x$$

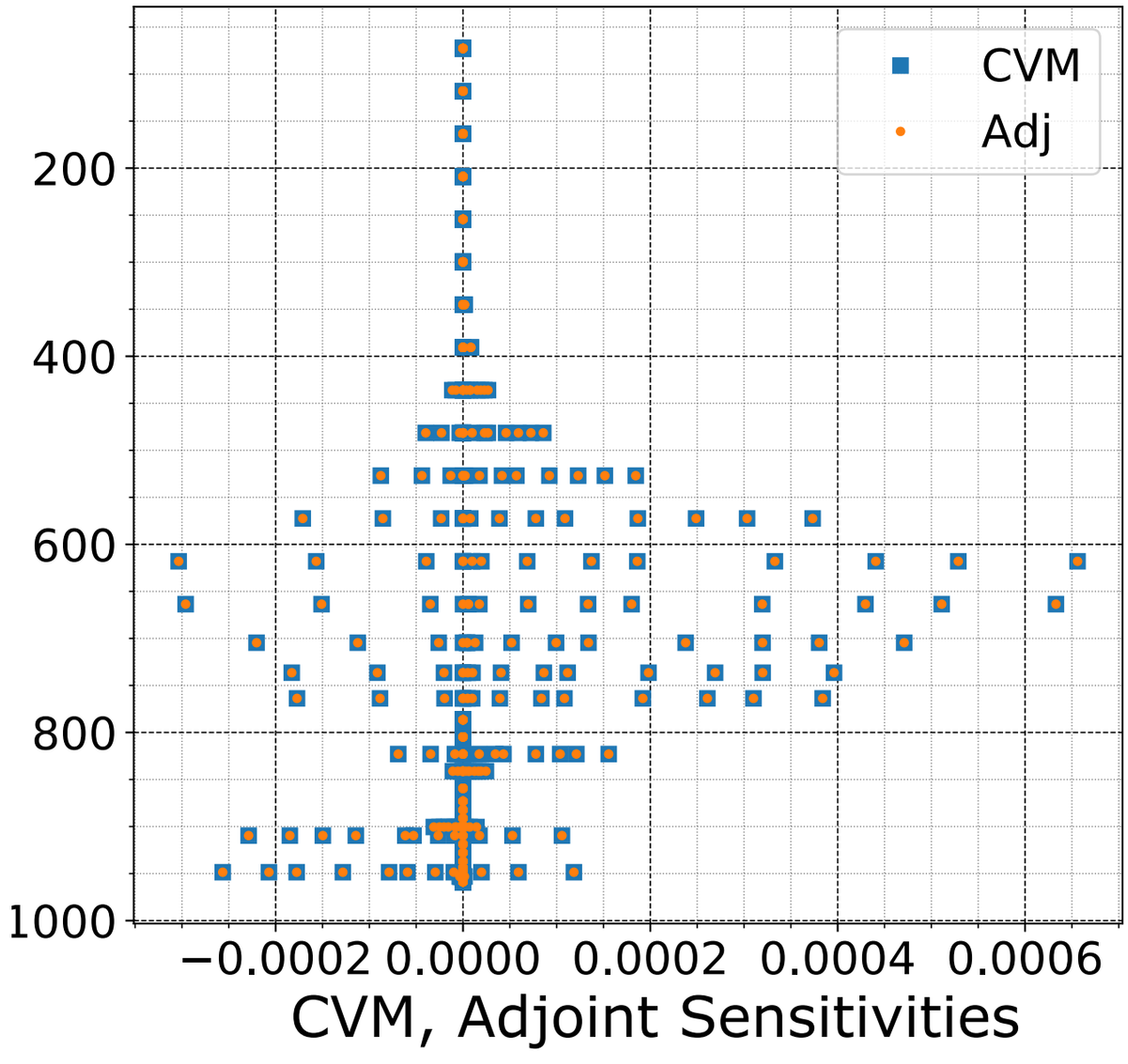
Menut et al., 2000; Hakami et al., 2006; Henze et al., 2007

Attribute Difference to Emissions with CMAQ adjoint

Adjoint of CrIS Observation Operator

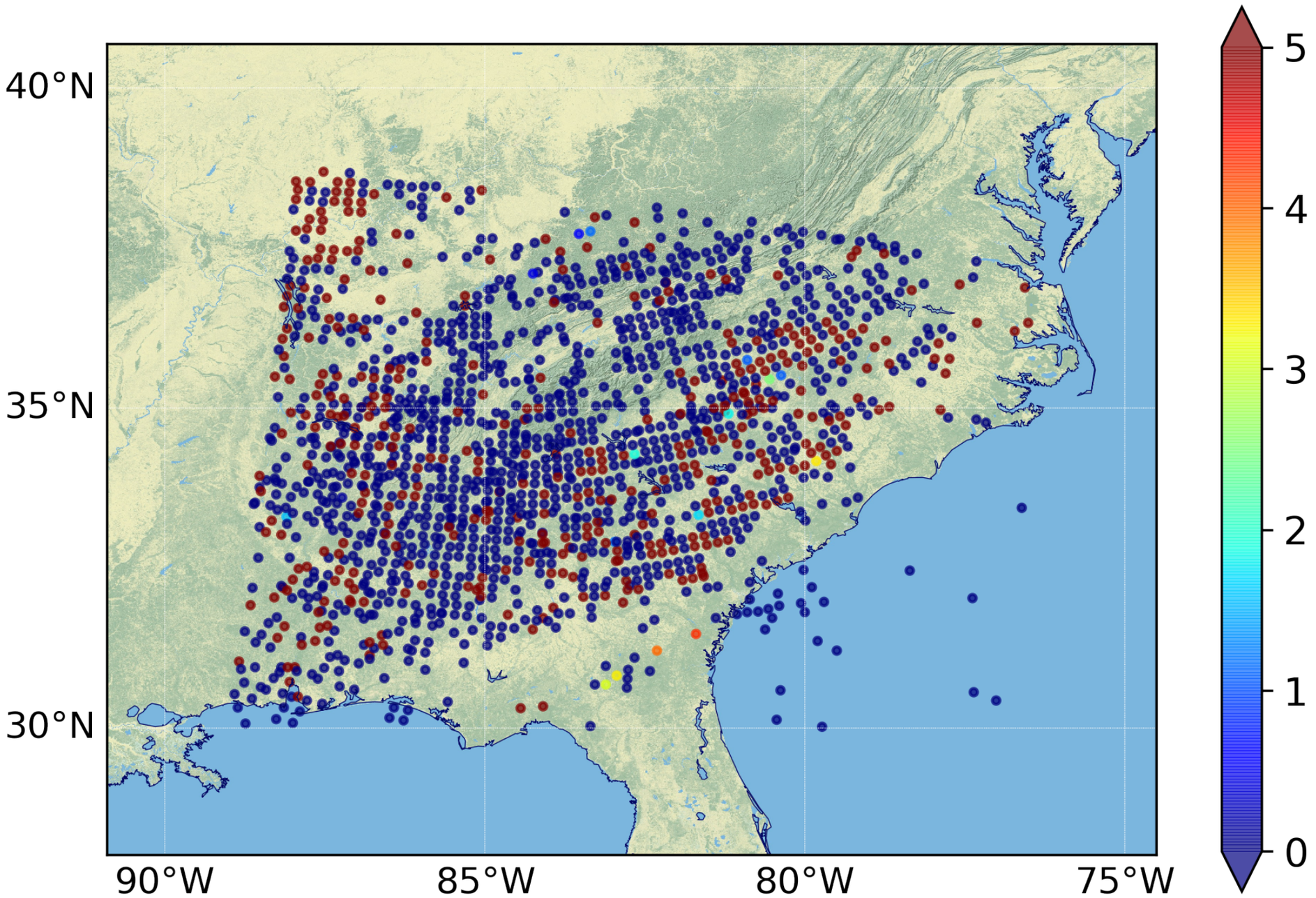
The Jacobian of the observation operator compares well between the adjoint and the complex variable method (CVM).





Sample Adjoint Forcing

$$\frac{\partial J}{\partial [NH_3]} = S_{obs}^{-1}(x_{c,CMAQ}^{est} - x_c^{est})$$



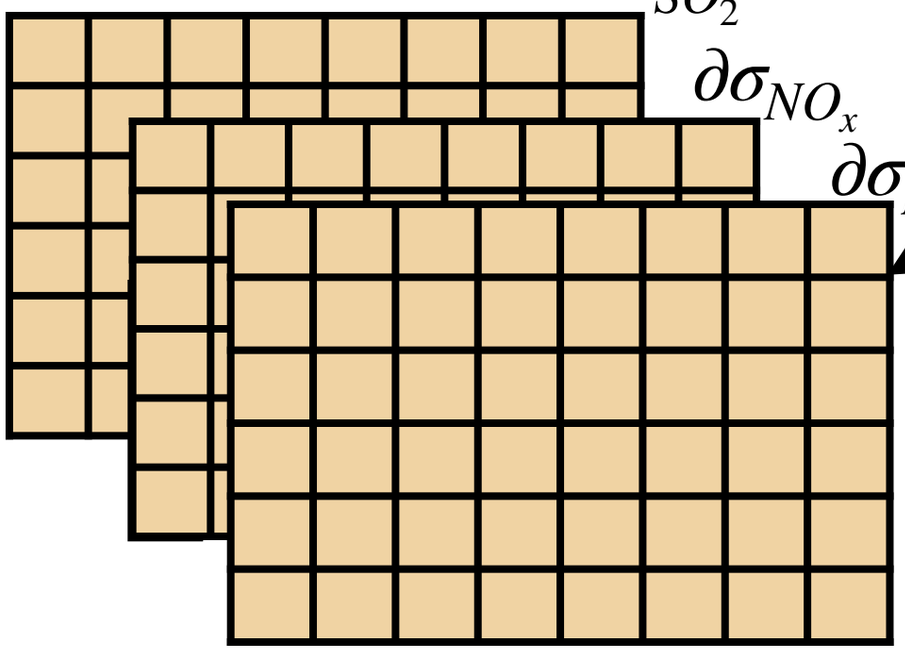
Refining Emissions with CrIS Observations

Emissions Scaling Factors

$\partial\sigma_{SO_2}$

$\partial\sigma_{NO_x}$

$\partial\sigma_{NH_3}$

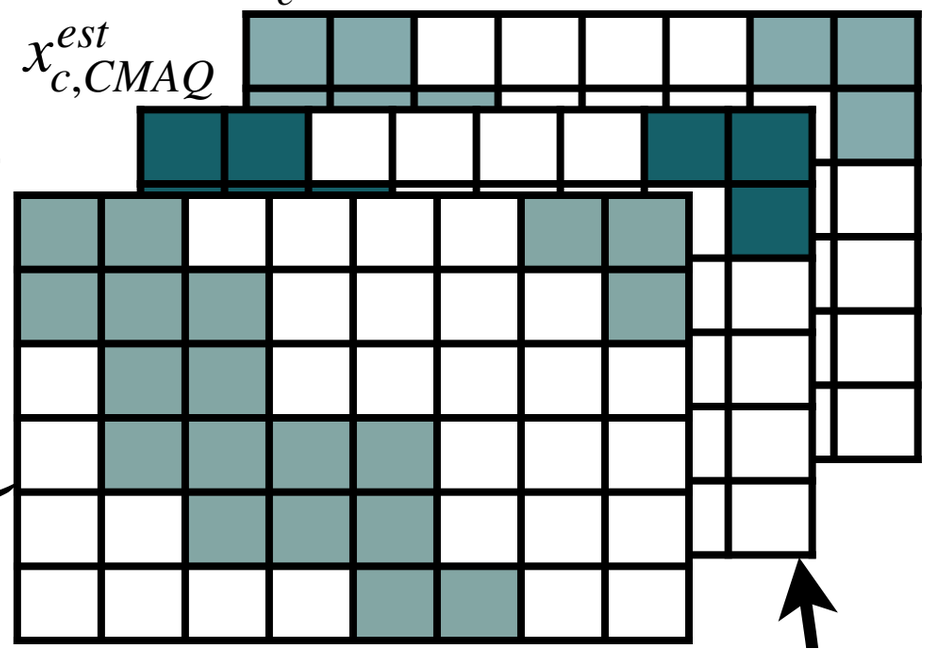


CMAQ adjoint

$\frac{\partial J}{\partial NH_3}$

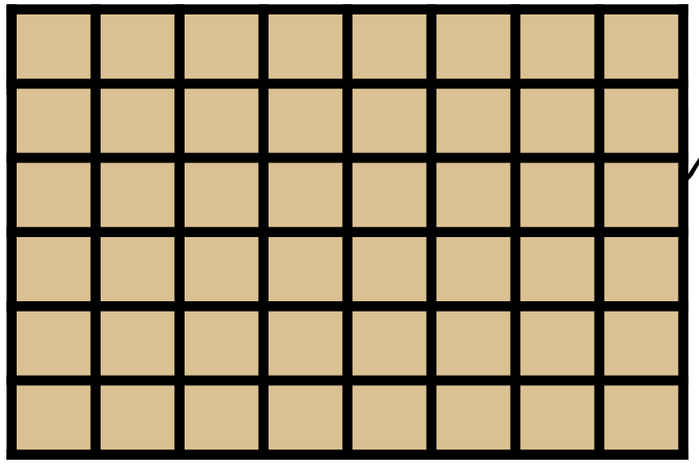
x_c^{est}

$x_{c,CMAQ}^{est}$



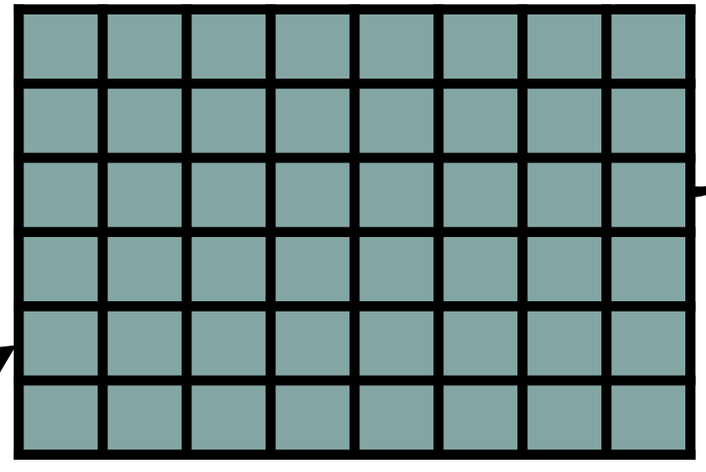
CrIS Observation Operator

Emissions Rates

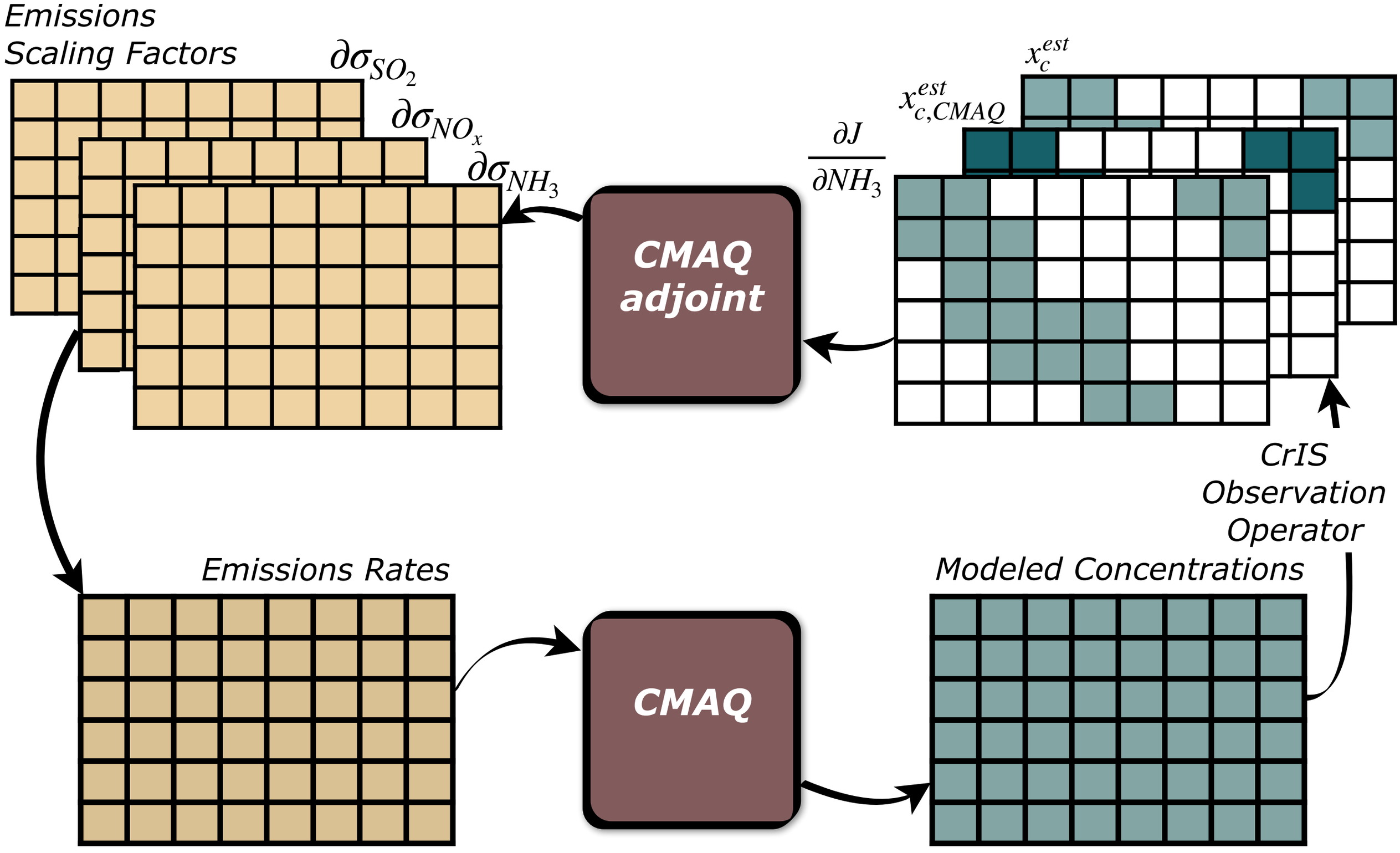


CMAQ

Modeled Concentrations

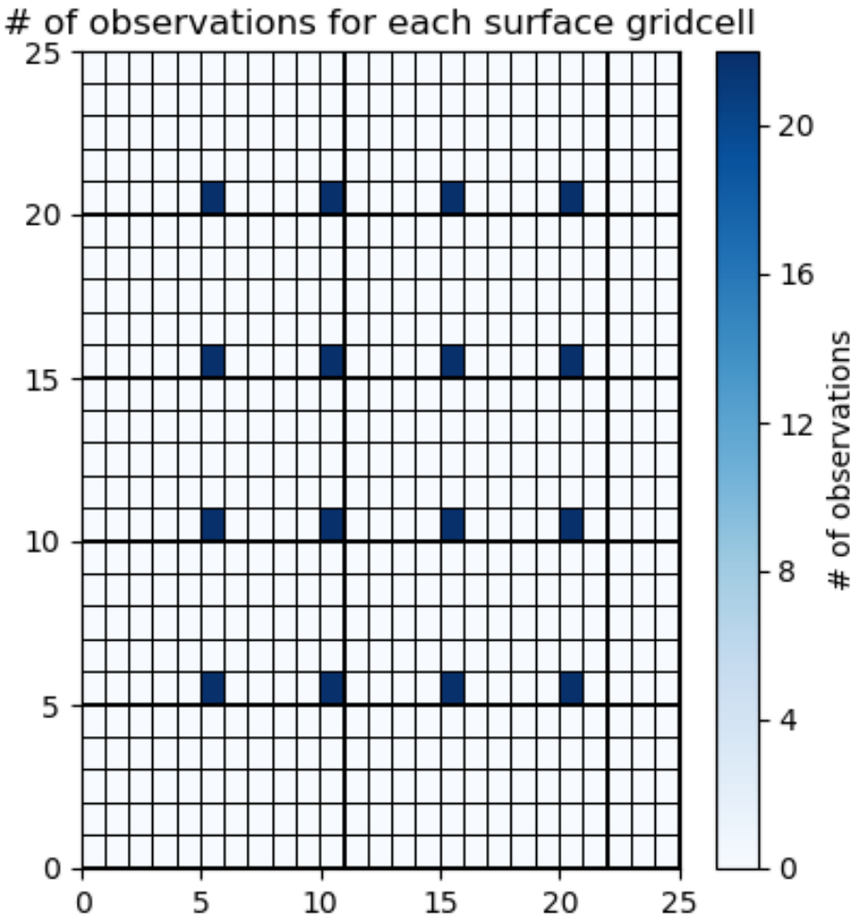


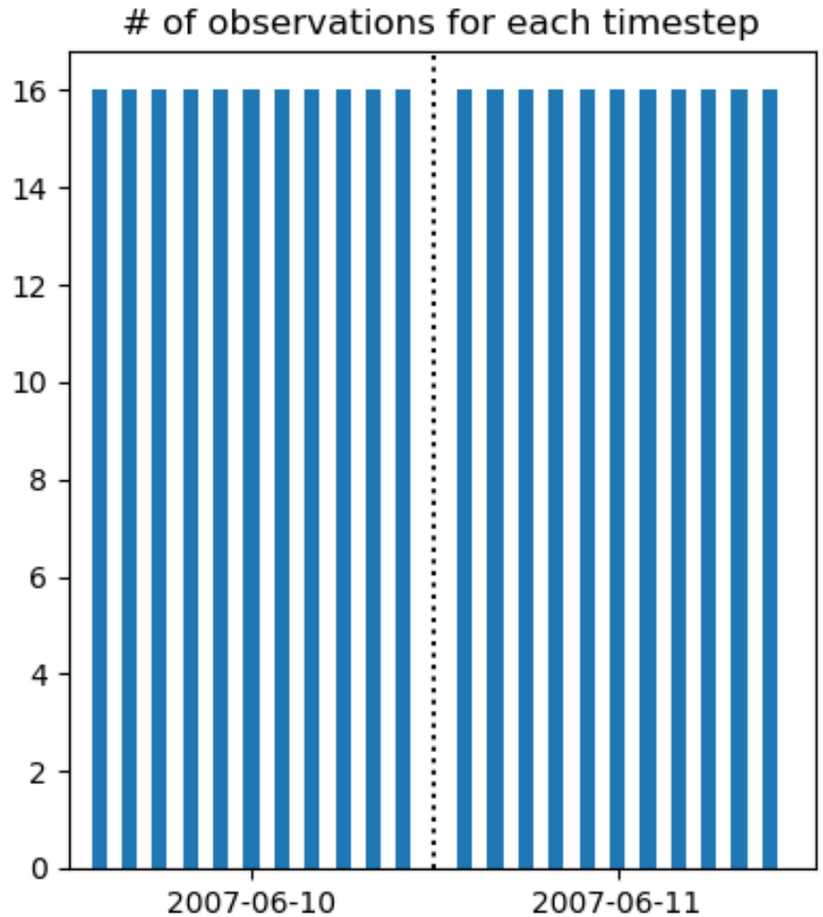
Refining Emissions with CrIS Observations



Evaluating a python-based 4-D Variational Framework

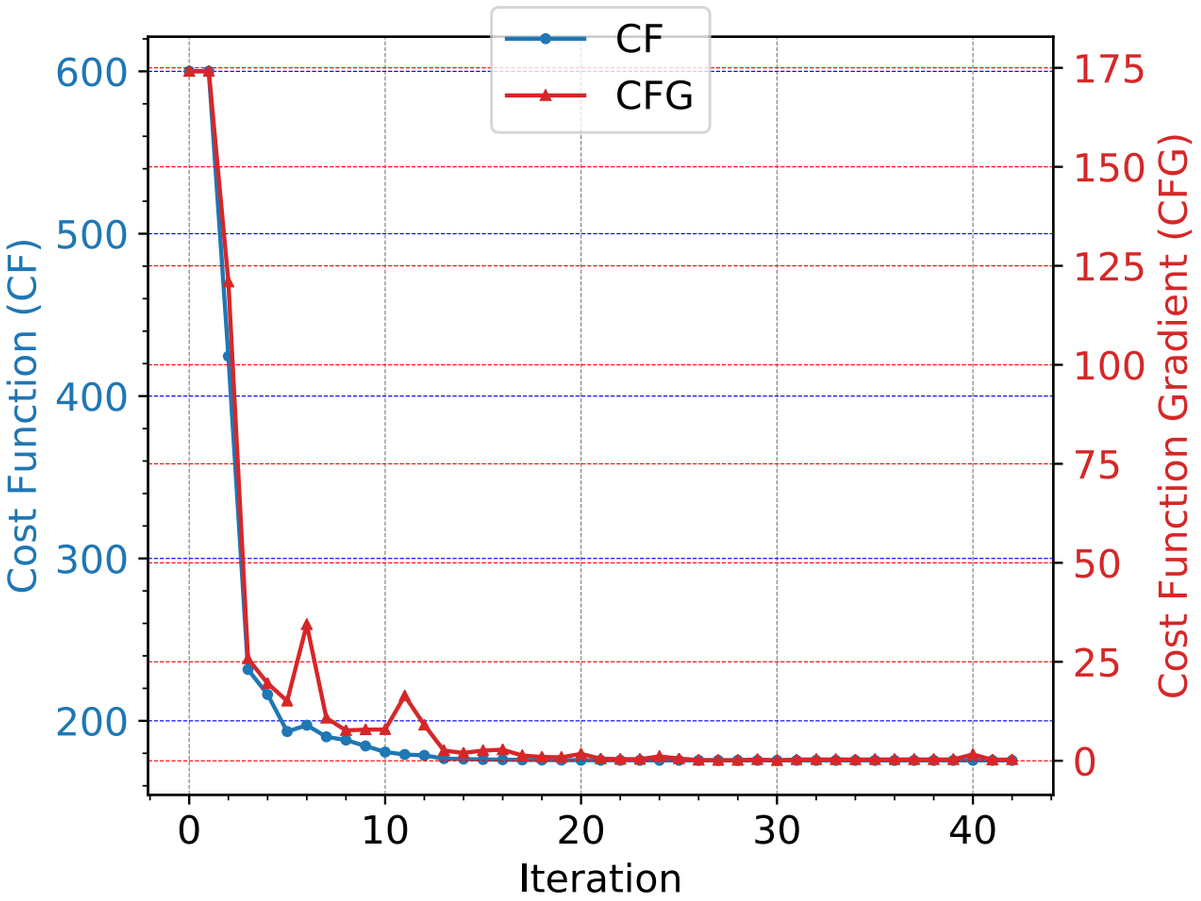
$$J(\frac{\boldsymbol{ef}}{u_{emis}}) = \frac{1}{2}(\boldsymbol{efu}_n - \frac{\boldsymbol{ef}_b}{u_{emis}})^T(\boldsymbol{efu}_n - \frac{\boldsymbol{ef}_b}{u_{emis}}) + \frac{1}{2}\sum_{k=1}^{K}(H[M[\boldsymbol{efu}_n]] - \boldsymbol{c}_{obs}^k)^T(\frac{H[M[\boldsymbol{efu}_n]] - \boldsymbol{c}_{obs}^k}{u_{obs}^2})$$

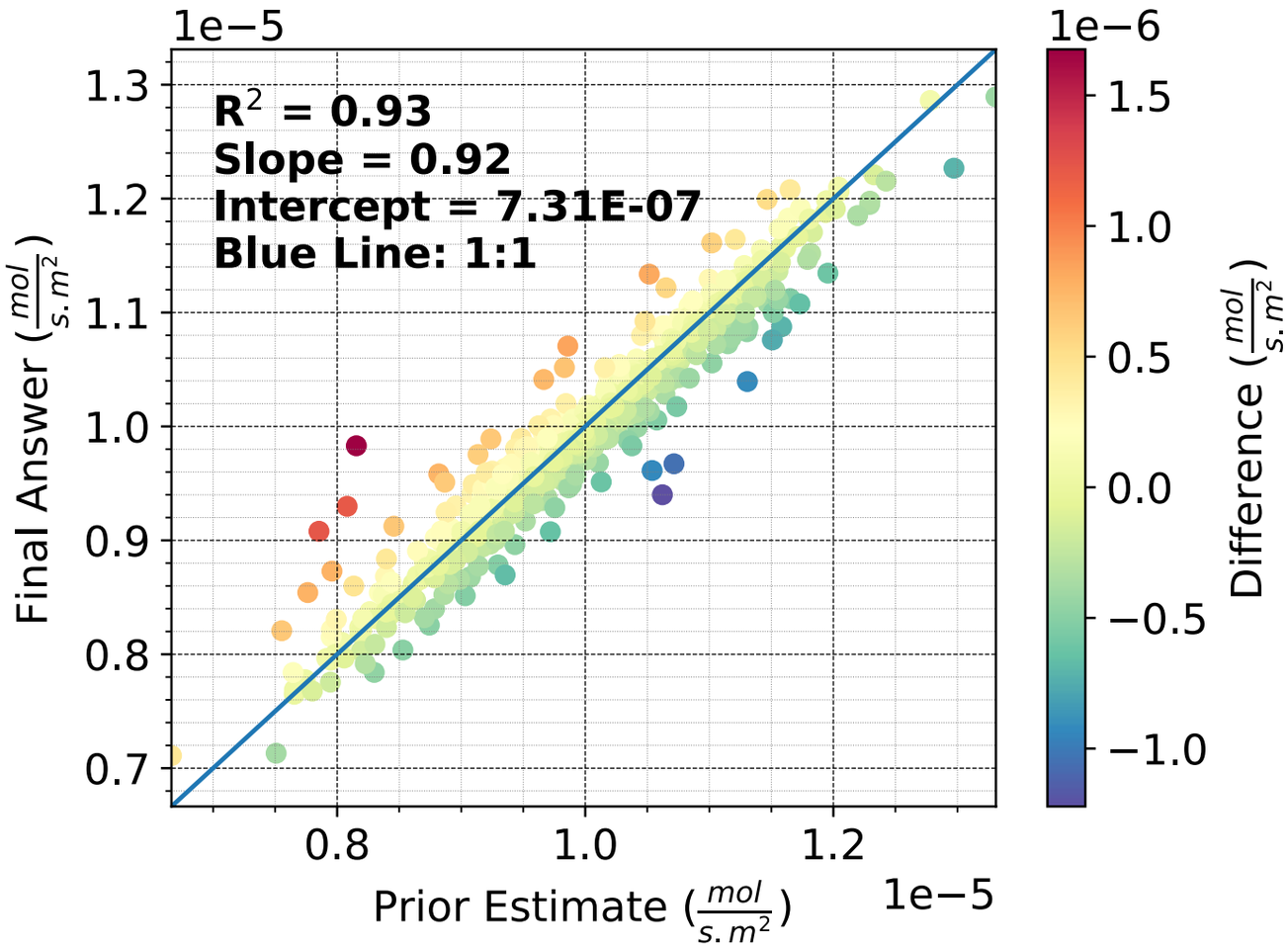




Random noise is applied to each background prior estimate and observation to assess ability to recover original prior.

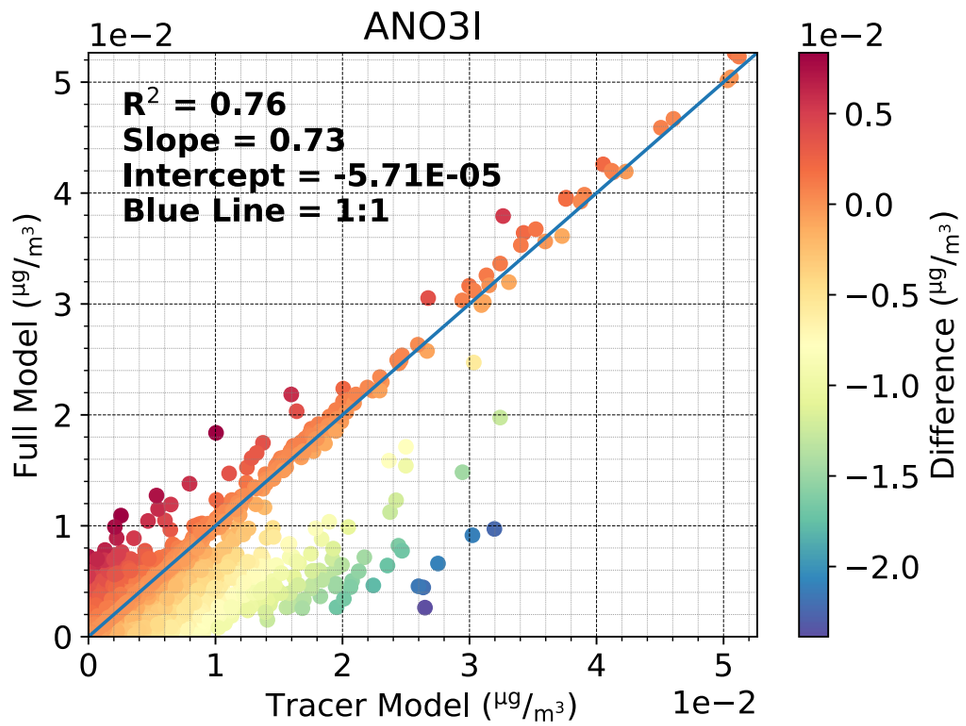
Evaluating a Python-based 4-D Variational Framework

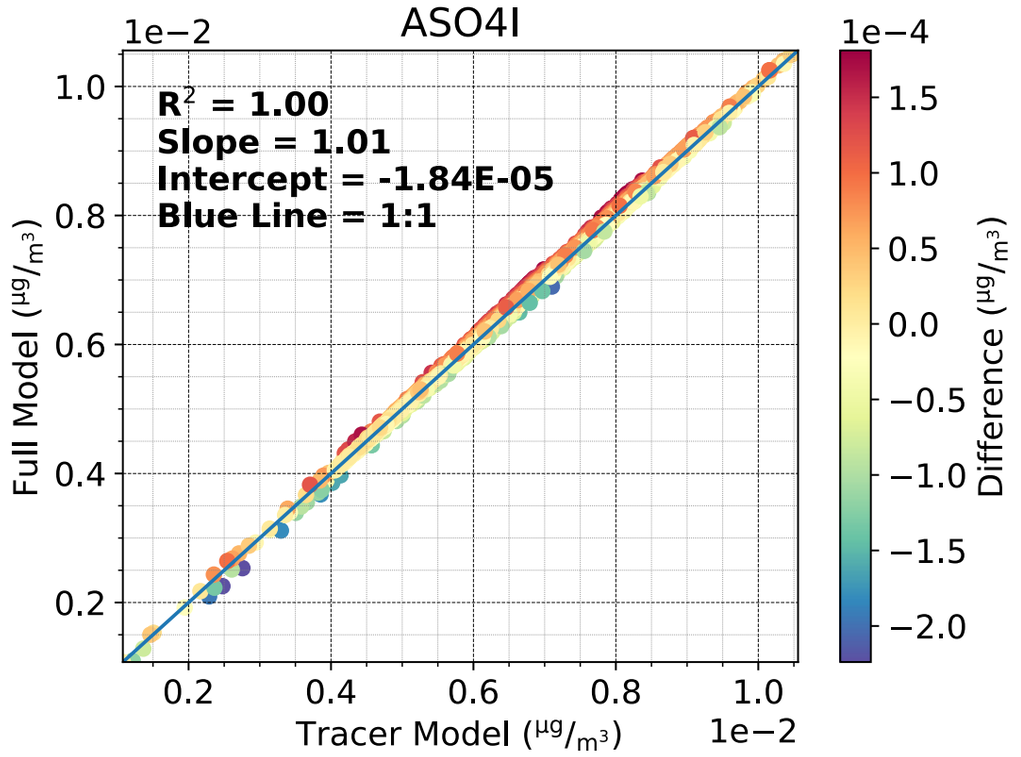


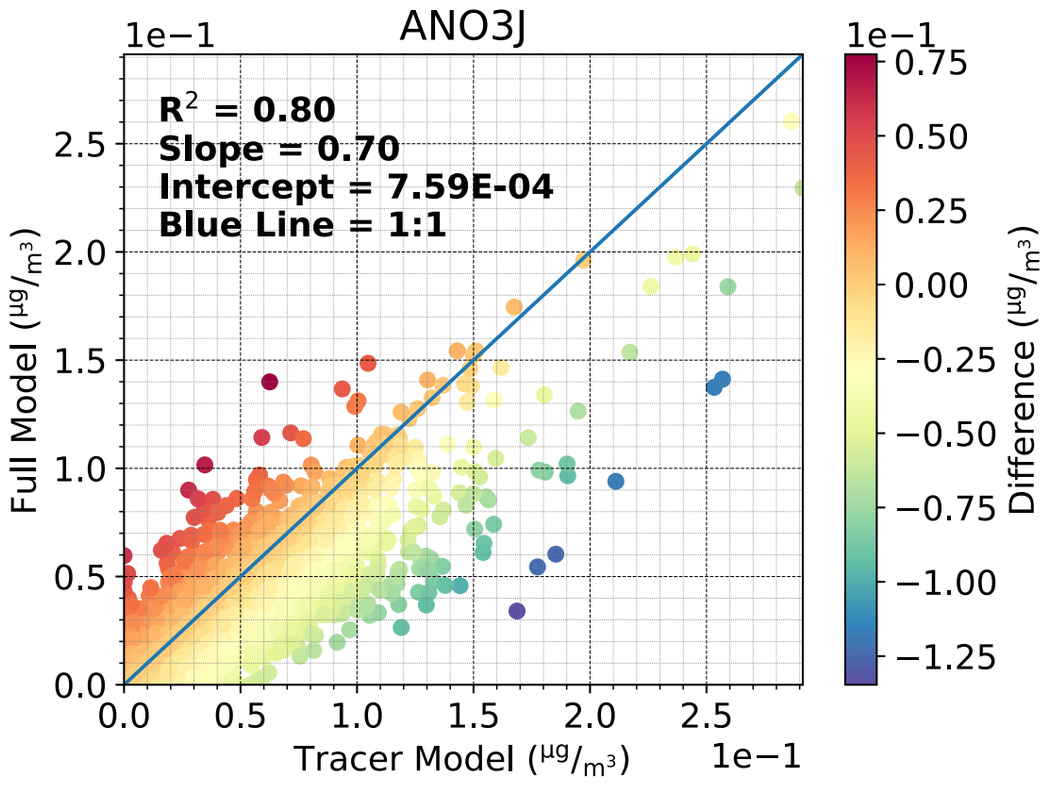


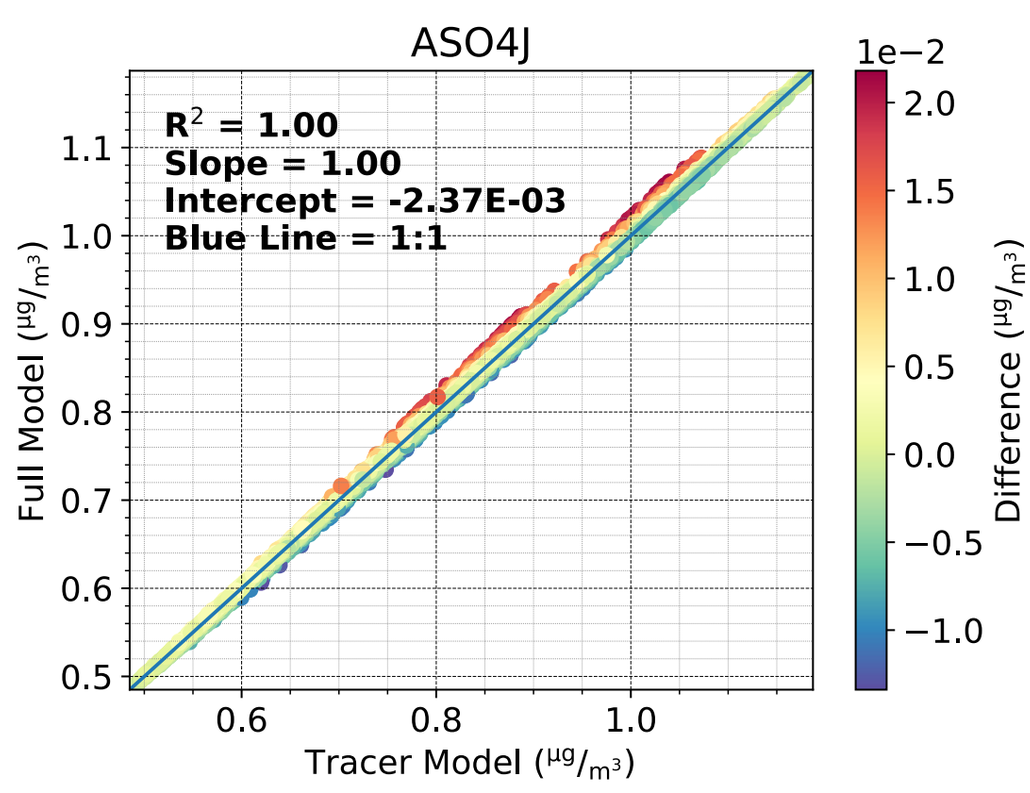
Reasonable recovery of the prior in this pseudo-observational tracer experiment provides confidence in the 4-D Var python framework.

Development of an Offline Ammonia Model & Adjoint for CMAQ

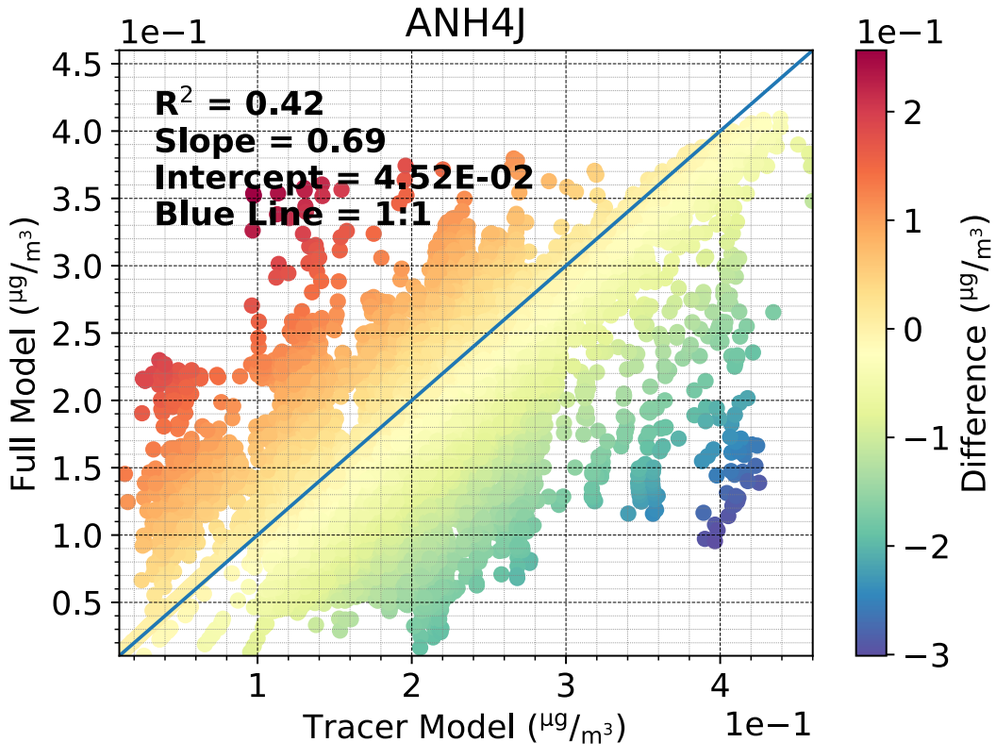


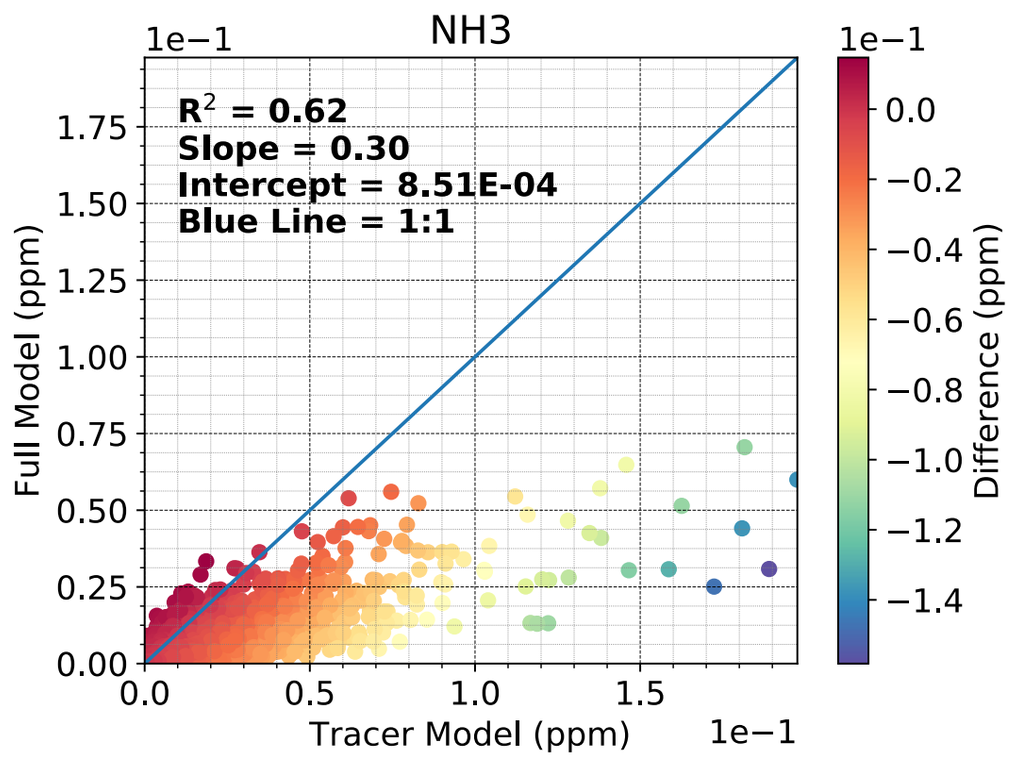


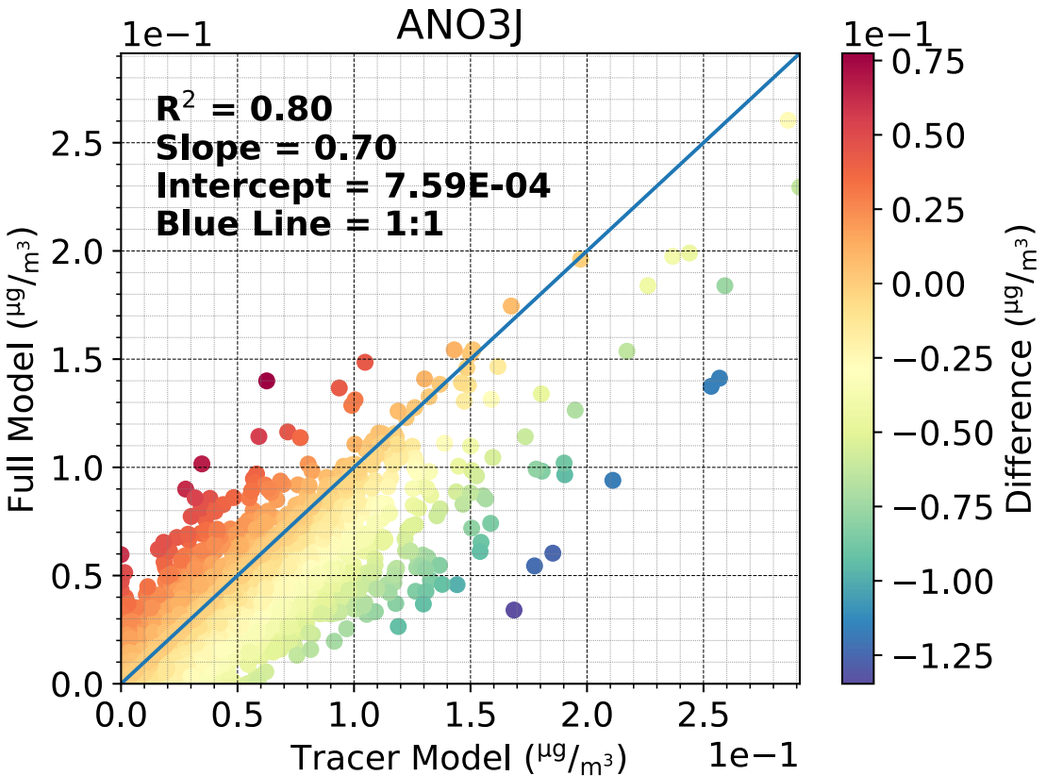


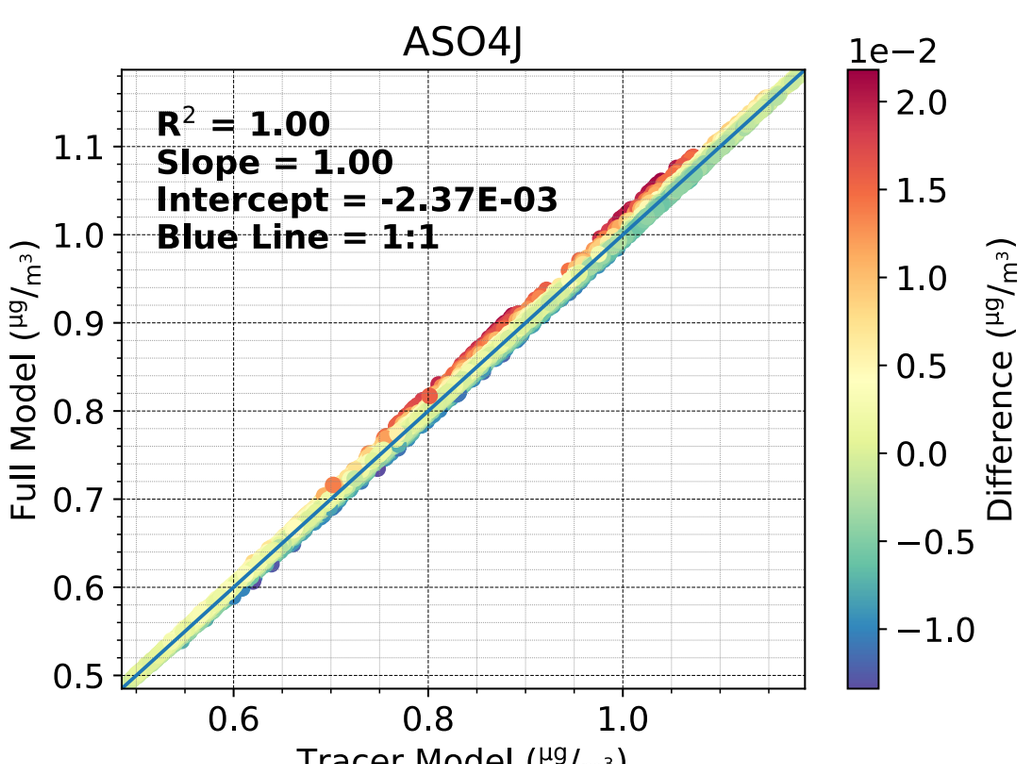


Development of an Offline Ammonia Model & Adjoint for CMAQ









Summary & Next Steps

- Resolving differences in NH_3 observations from CrIS and *simulated CrIS observations of CMAQ fields* will allow revision of NH_3 emissions with spatial specificity.

- A python-based 4-DVar framework with the CMAQ-adjoint has been shown to *recover emissions with generic pseudo-observations*.

- *Transport, inorganic thermodynamics, and depositional processes* will be included in the revision of emissions.

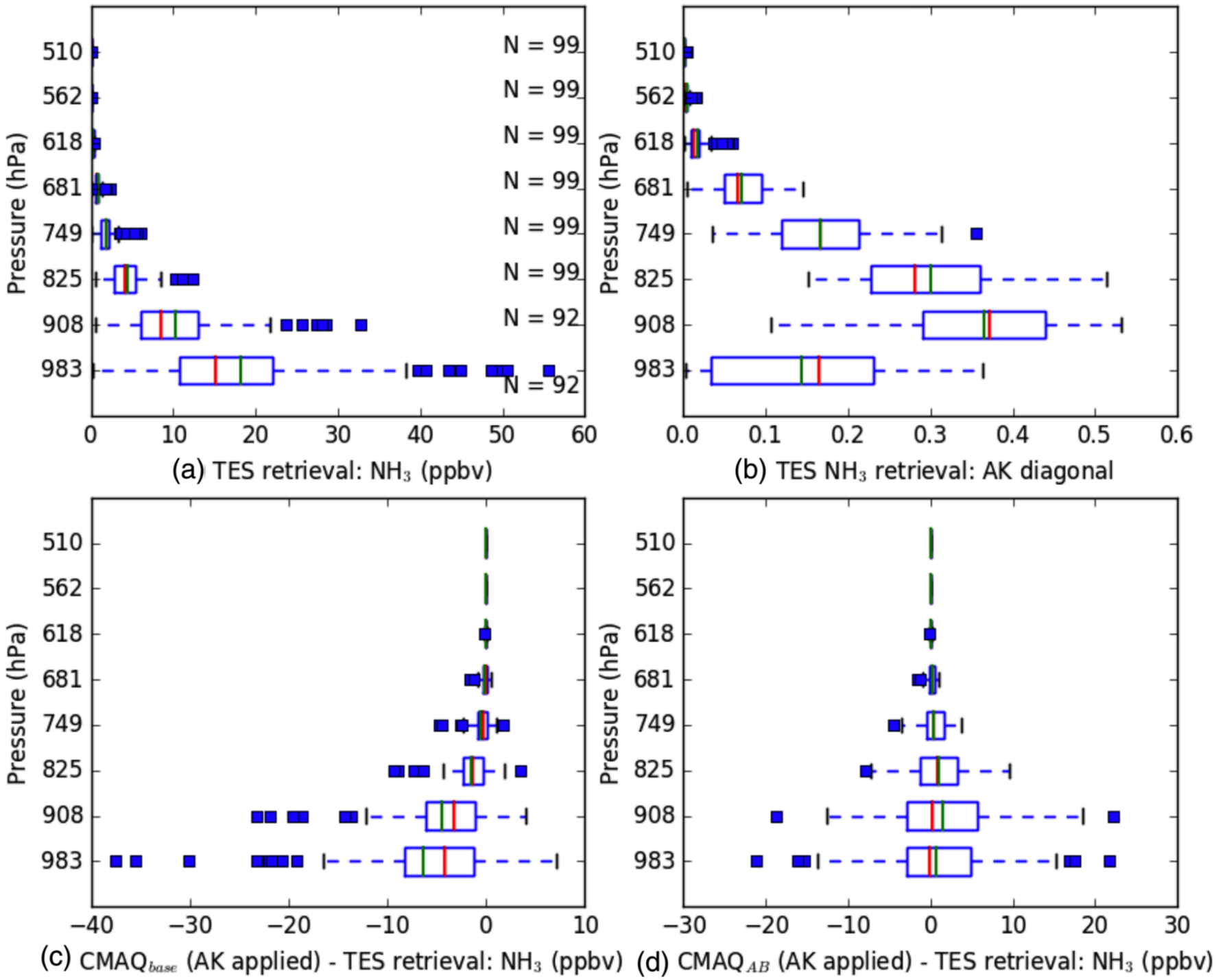
Acknowledgements

- Mark Shephard of Environment and Climate Change Canada for CrIS observations

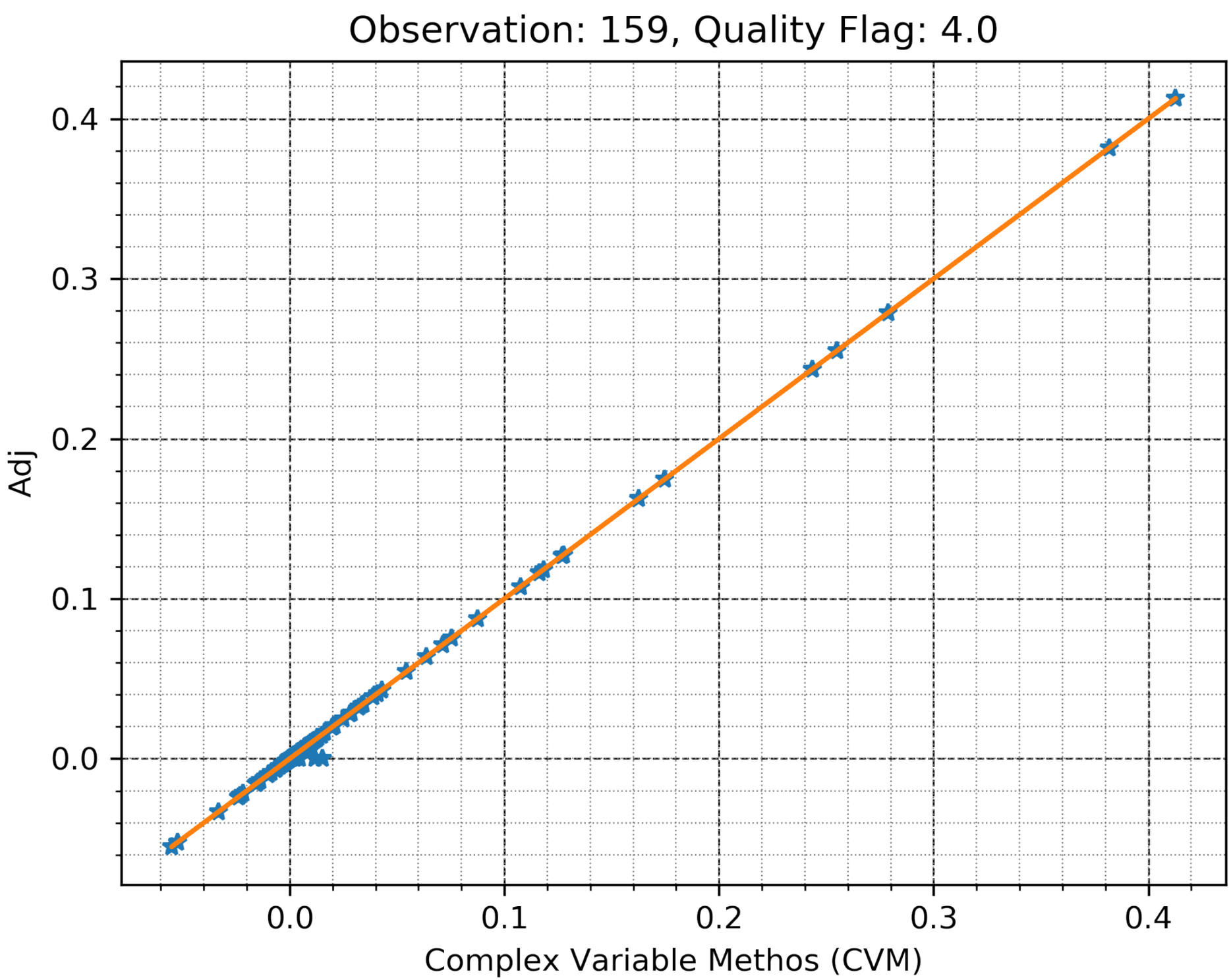
- NASA New Investigator Program Award #80NSSC18K1302

Questions?

Satellite-based Observations



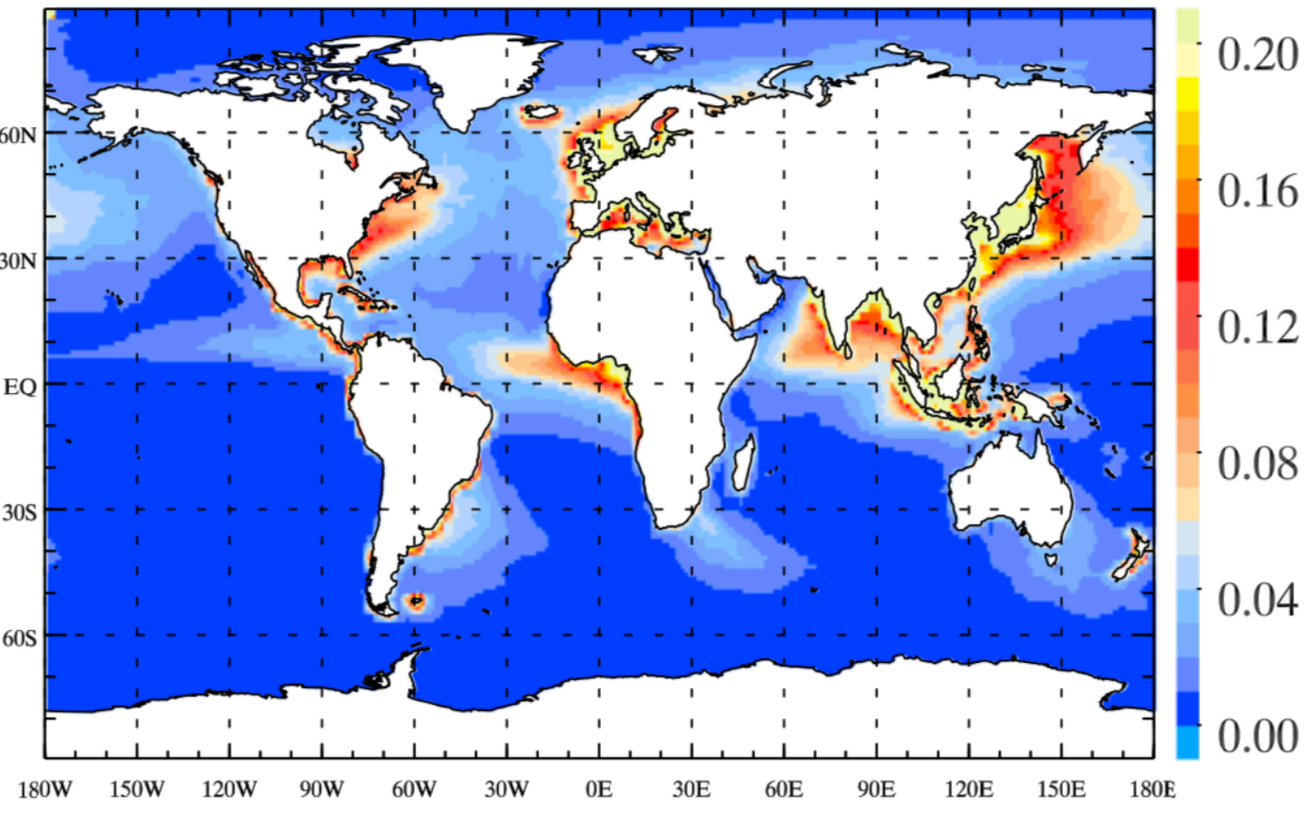
Lonsdale et al., ACP, 2017


Observation: 159, Quality Flag: 4.0
Adj
0.4
0.3
0.2
0.1
0.0
0.0
0.1
0.2
0.3
0.4
Complex Variable Methos (CVM)

Impacts: Nitrogen Deposition

Deposition of ammonia, ammonium, and associated nitrate aerosol affects ecosystems.

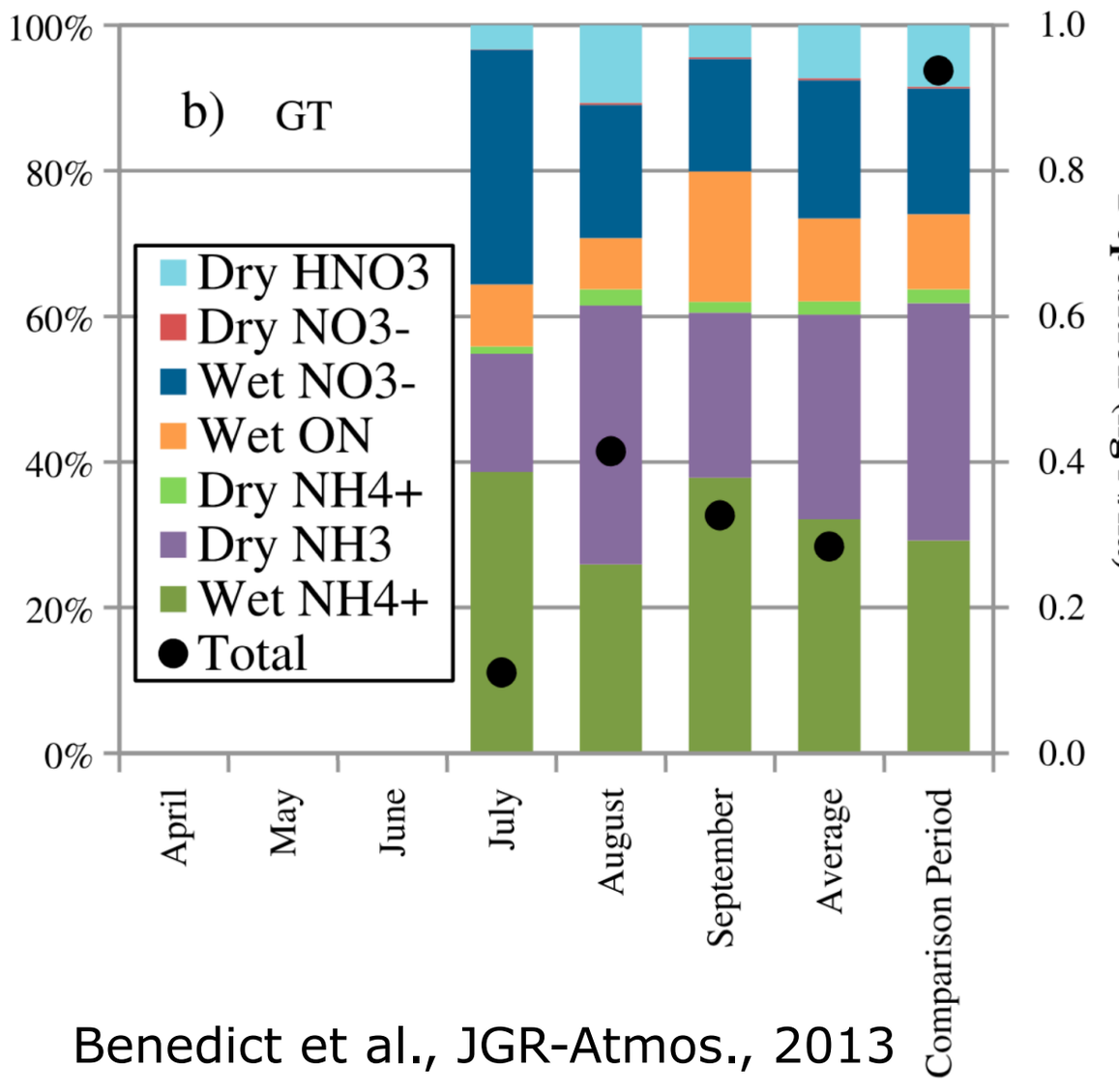
Atmospheric deposition of NH_x to oceans



(g N m^{-2} y^{-1})

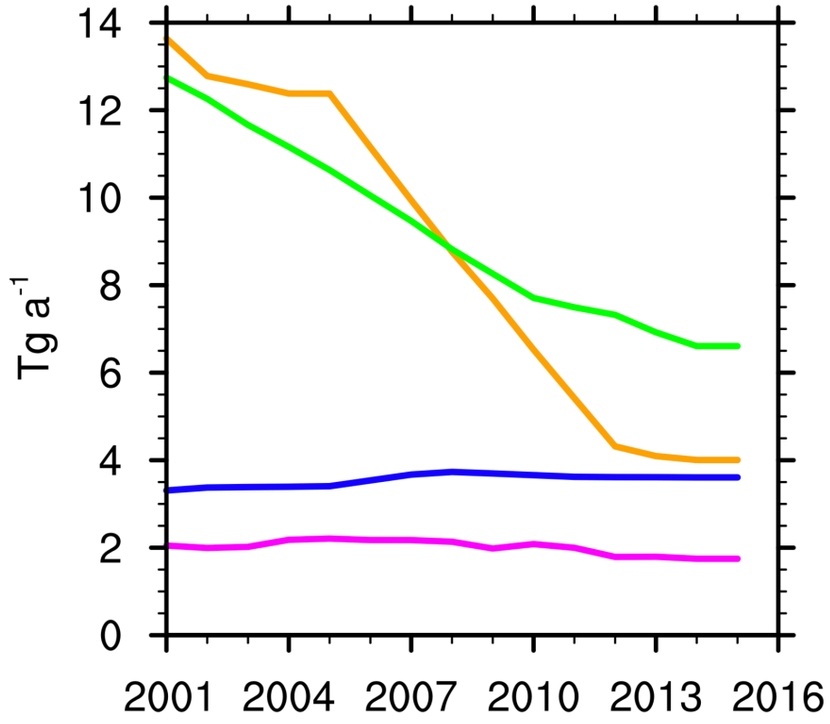
Jickels et al., Global Biogeochem. Cycles, 2018

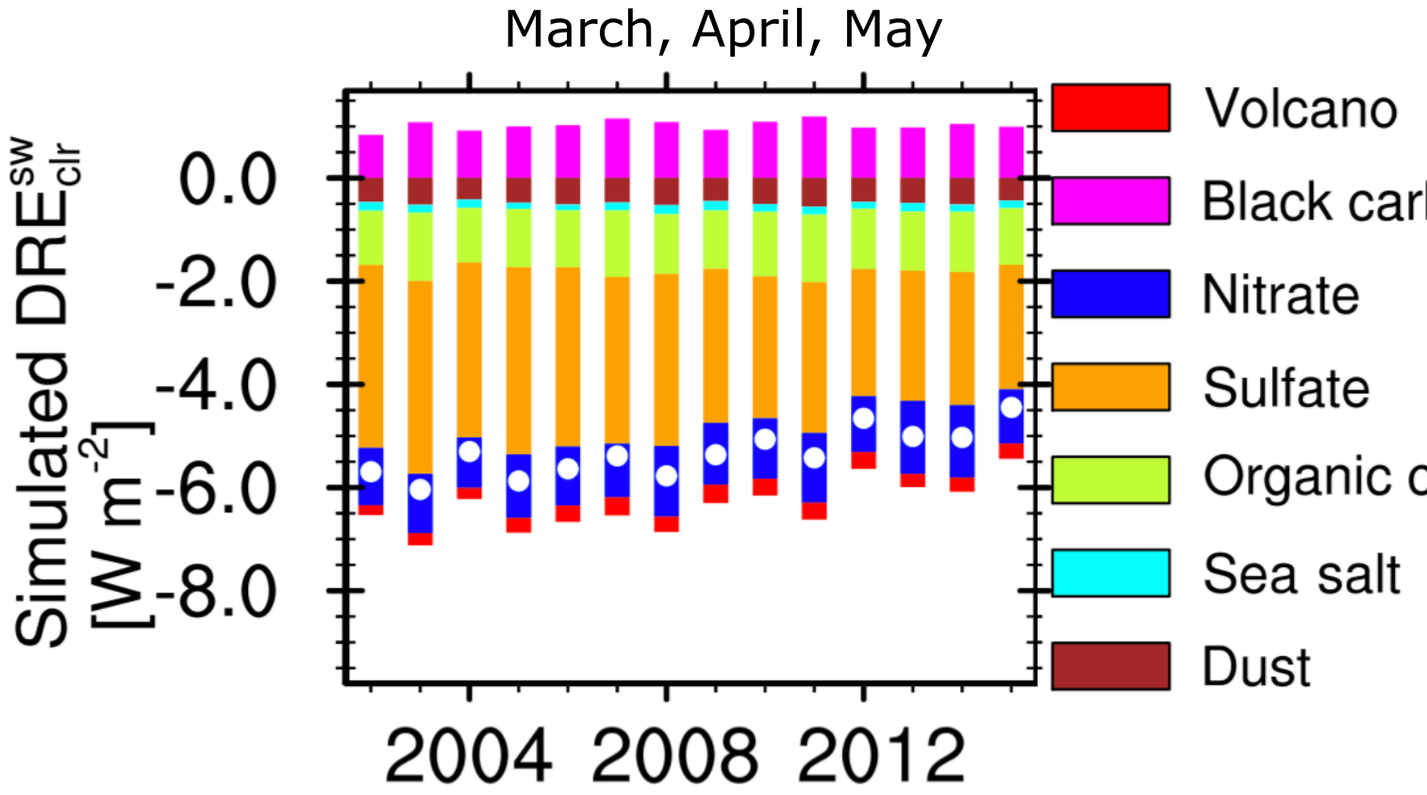
Deposition near Grand Tetons National Park



Benedict et al., JGR-Atmos., 2013

Impacts: Radiative Forcing





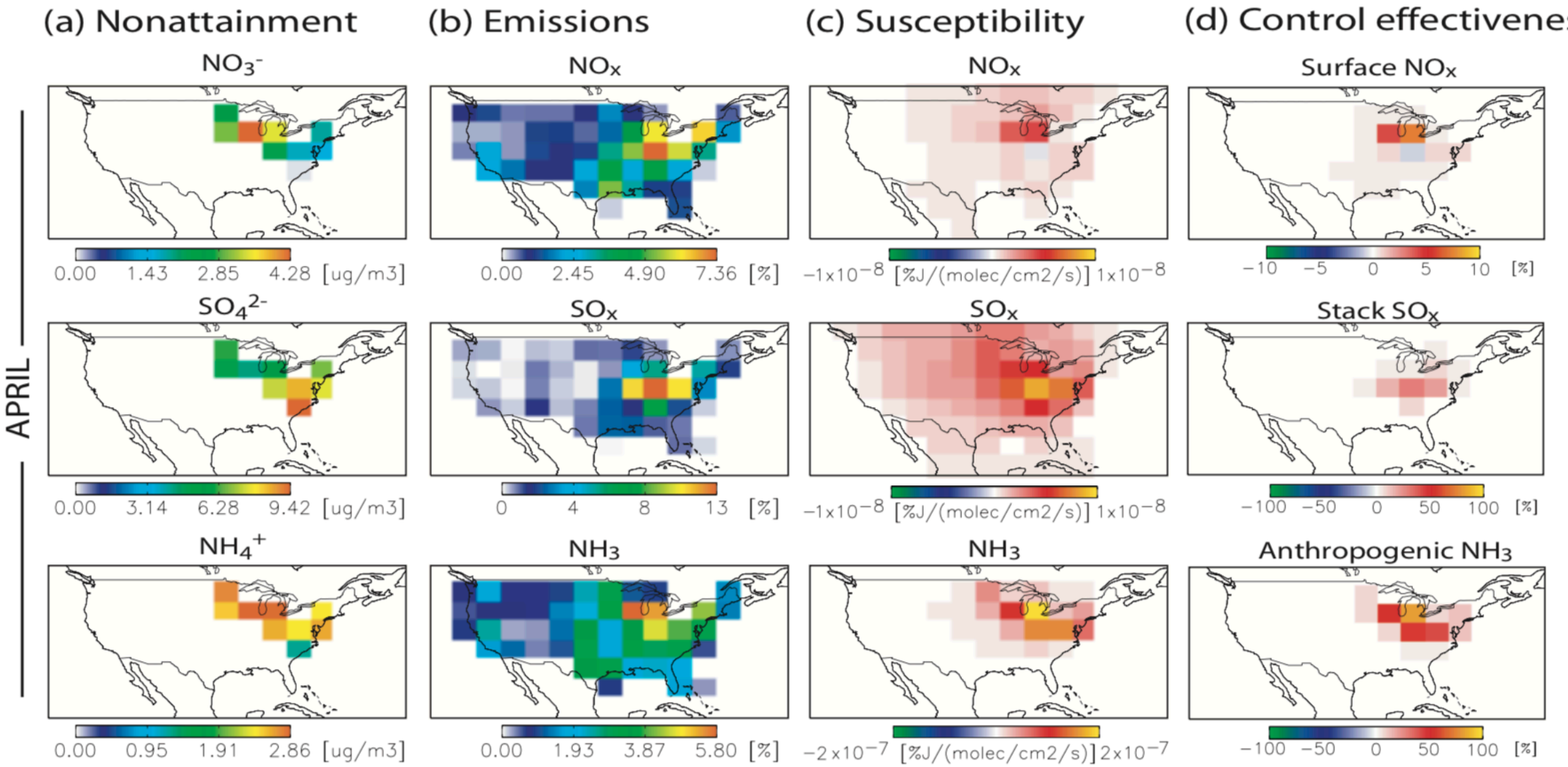
Paulot et al. found small increases in nitrogen-related emissions had a disproportionate impact on the clear sky, direct radiative effect.

Paulot et al., Atmos. Chem. Phys., 2018

Impacts: $PM_{2.5}$ Contribution

Ammonia neutralizes sulfate and nitrate; sulfate decreases leave more ammonium for neutralizing nitrate.

At certain times and places, controlling ammonia emissions would be the most efficient way to avoid a $PM_{2.5}$ exceedance.



Henze et al., Atmos. Chem. Phys., 2009