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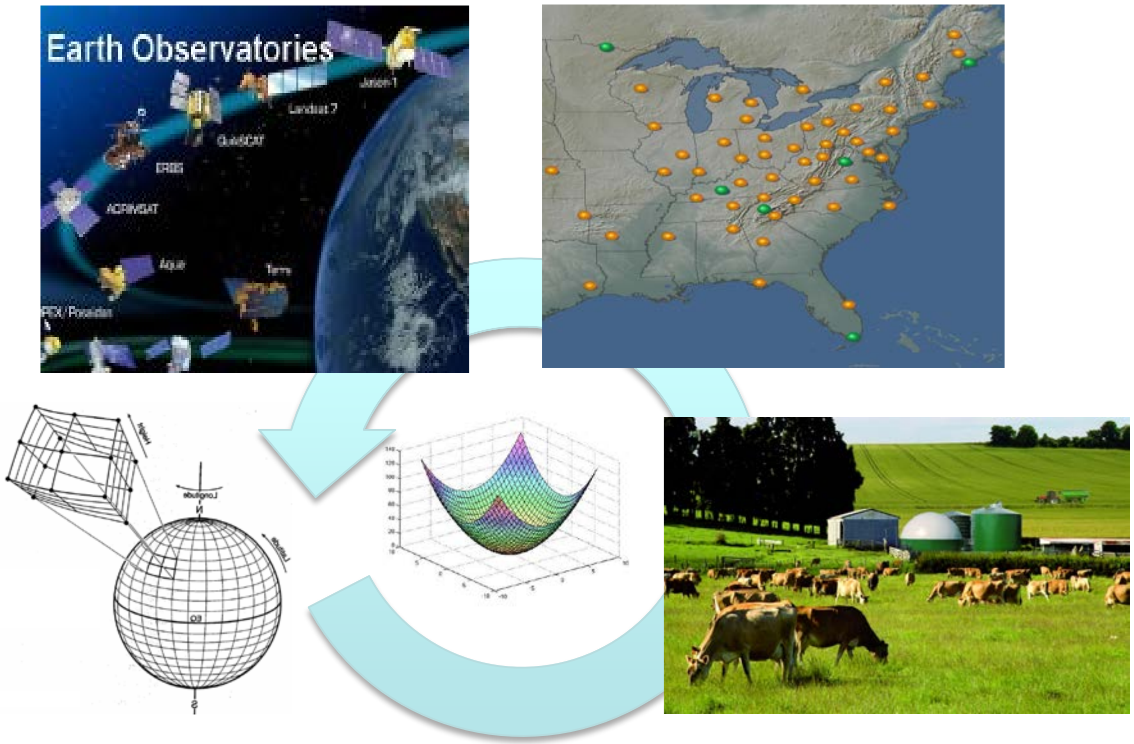
Top-down constraints on NH_3 emissions



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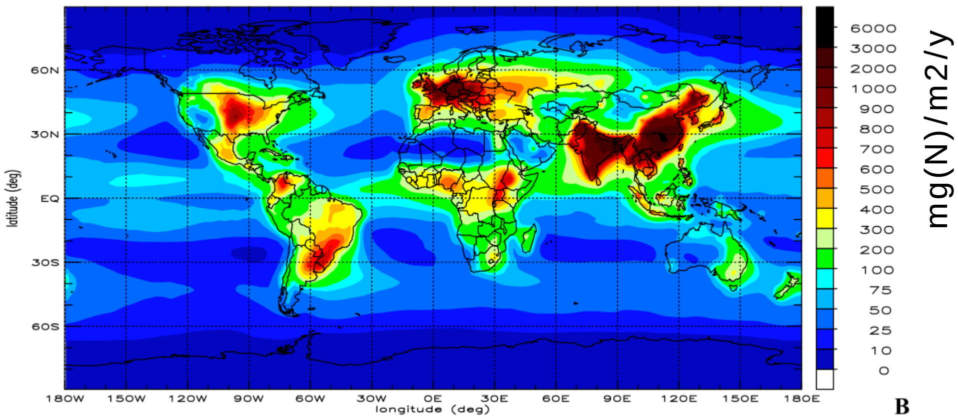
EPA STAR RD834559. This work does not reflect official agency views, policies.

Impacts of NH_3

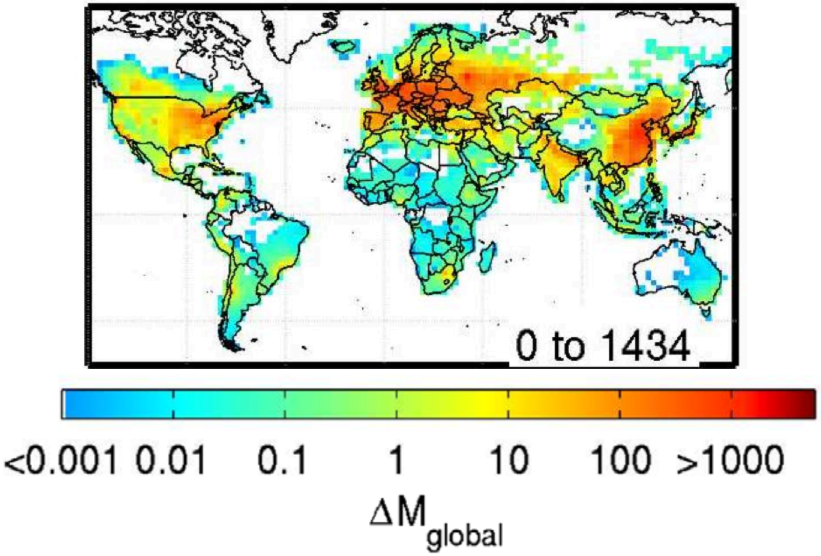
Deposition

Estimated N deposition from NHx
(Dentener et al., 2006)



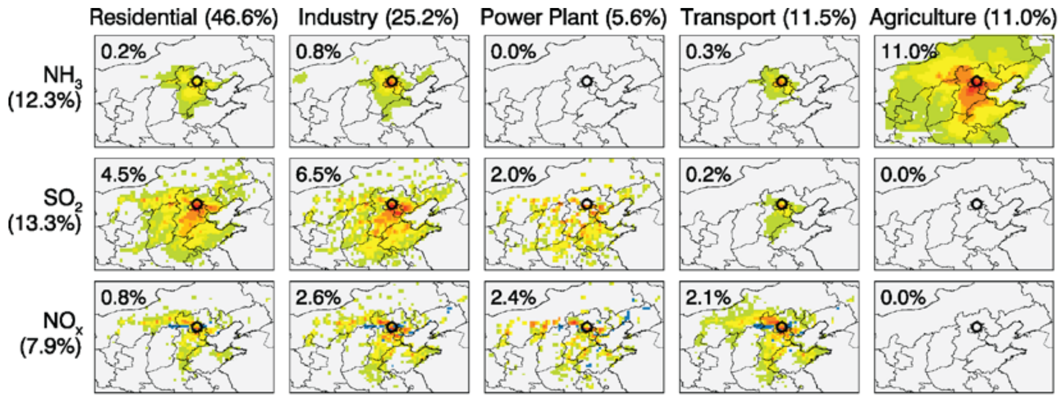
Health

Impacts of 10% Δemissions
(Lee et al., ES&T, 2015)



Air Quality

Source attribution of Jan. PM2.5 event
(Zhang et al., ERL, 2015)



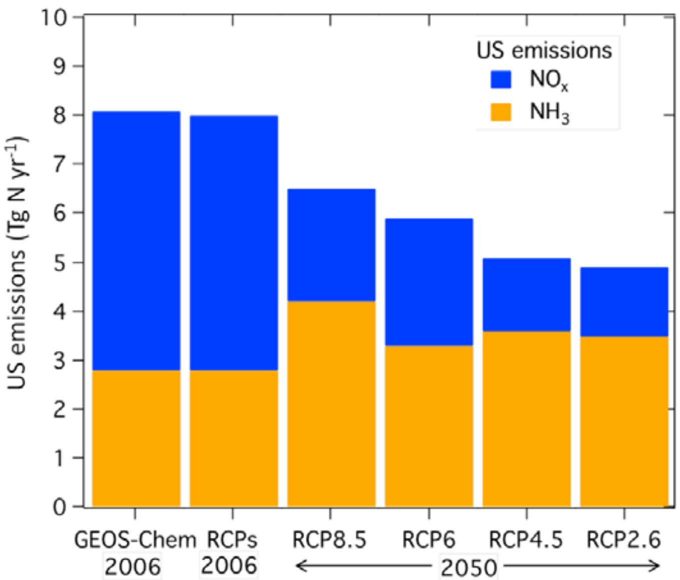
- Agricultural emissions lead to 20% of global premature deaths from ambient air pollution (Lelieveld et al., Nature, 2015) – largely the impact of NH_3 emissions on $PM_{2.5}$.

NH_3 is a growing concern

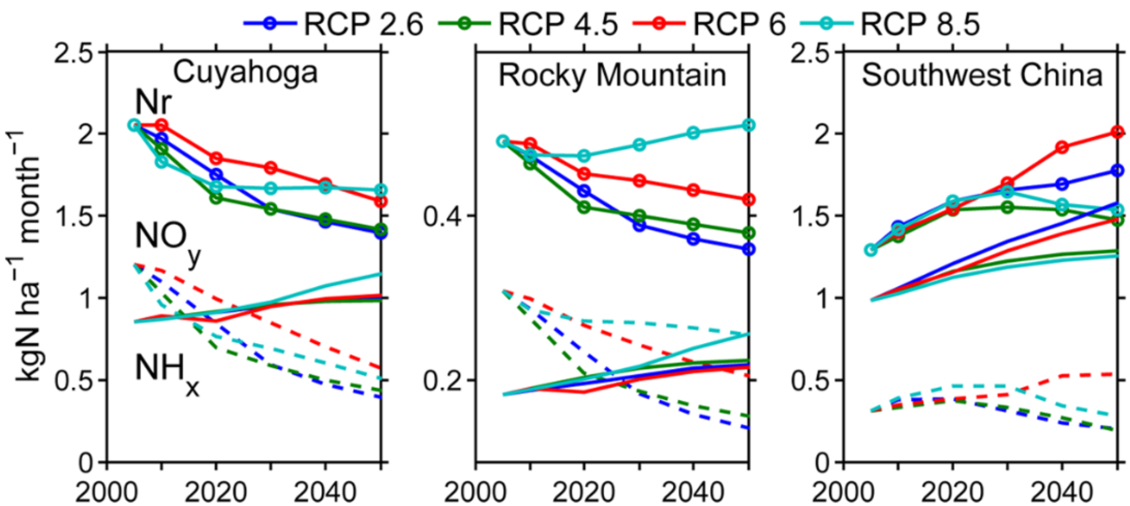
Denman et al. (2007), *IPCC*: NH_3 emissions have increased by x2-x5 since preindustrial times and are estimated to double by 2050.

NH_3 projected to soon overtake NO_x as the driver of Nr deposition:

Emissions (Ellis et al., 2013)



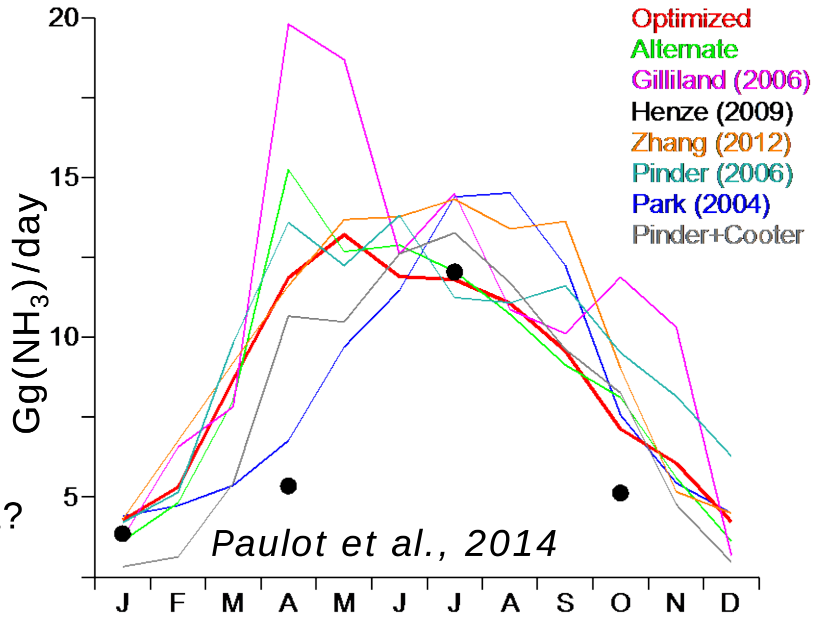
Deposition (Paulot et al., 2013)



Transition may have occurred already in the US
(Li et a., PNAS 2016; Sun et al., PNAS, 2016; Liu et al., PNAS, 2016)

Uncertainties in NH_3 emissions

Substantial variability in estimates of total US NH_3 emissions →



Larger uncertainties at regional scales
(e.g., Novak et al., 2012; Walker et al., 2012)

Global inventories also uncertain
(e.g., Beuson et al., 2008)
- Schlesinger (PNAS, 2009): a 46 Tg gap in N budget?

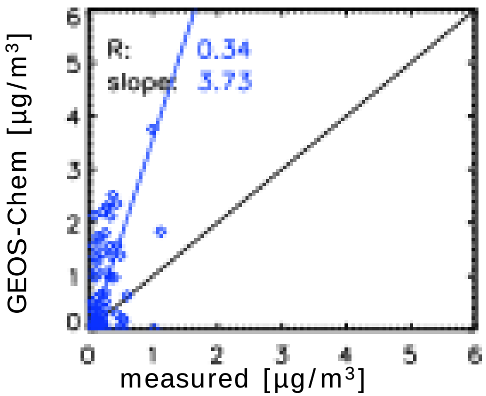
Why so uncertain?
- lack of direct source measurements (hard, expensive)

- difficulty in relating associated species to NH_3 sources
  - constraints from observations of [NH_4^+] or [NH_x] complicated by model/measurement error, precipitation
  - observations of [NH_3] less prevalent

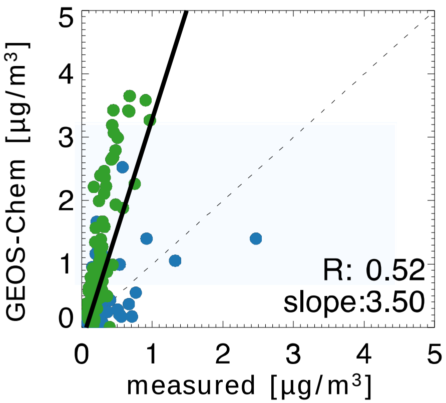
Uncertainties in NH_3 emissions: Implications for air quality and environment

- contribute to errors in assessing $PM_{2.5}$

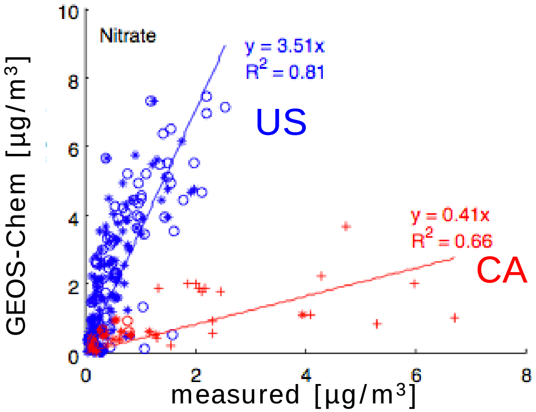
Ex: GEOS-Chem overestimates nitrate at IMPROVE / CASTNET (July)



Zhu et al., 2013



Heald et al., 2012

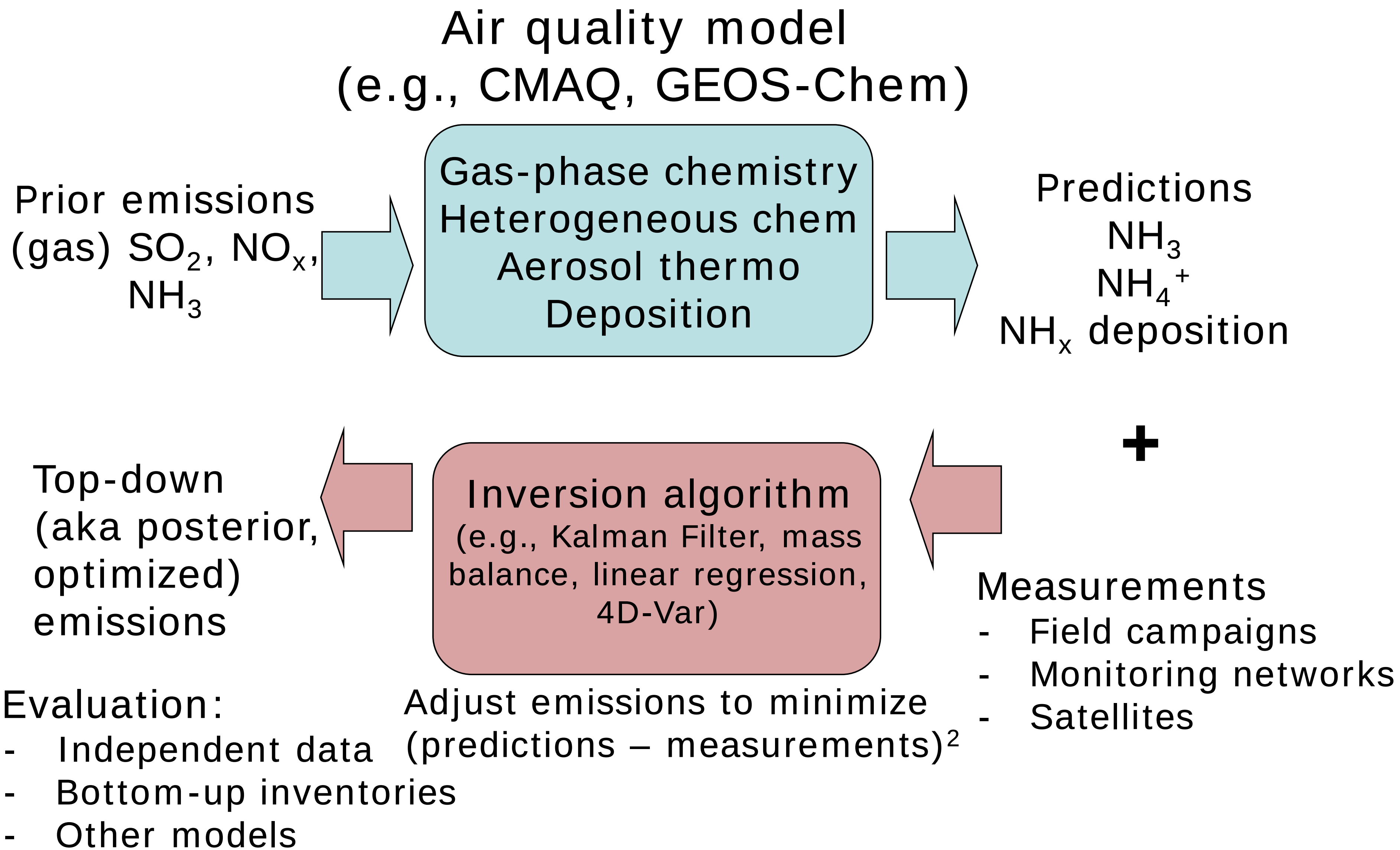


Walker et al., 2012

(also Liao et al., 2007; Henze et al., 2009; Zhang et al., 2012)

- undermine regulatory capabilities for secondary standards on SO_x, NO_x to control N_r dep (e.g., Koo et al., 2012)

Top-down constraints



Constraints on NH_x deposition from inverse modeling

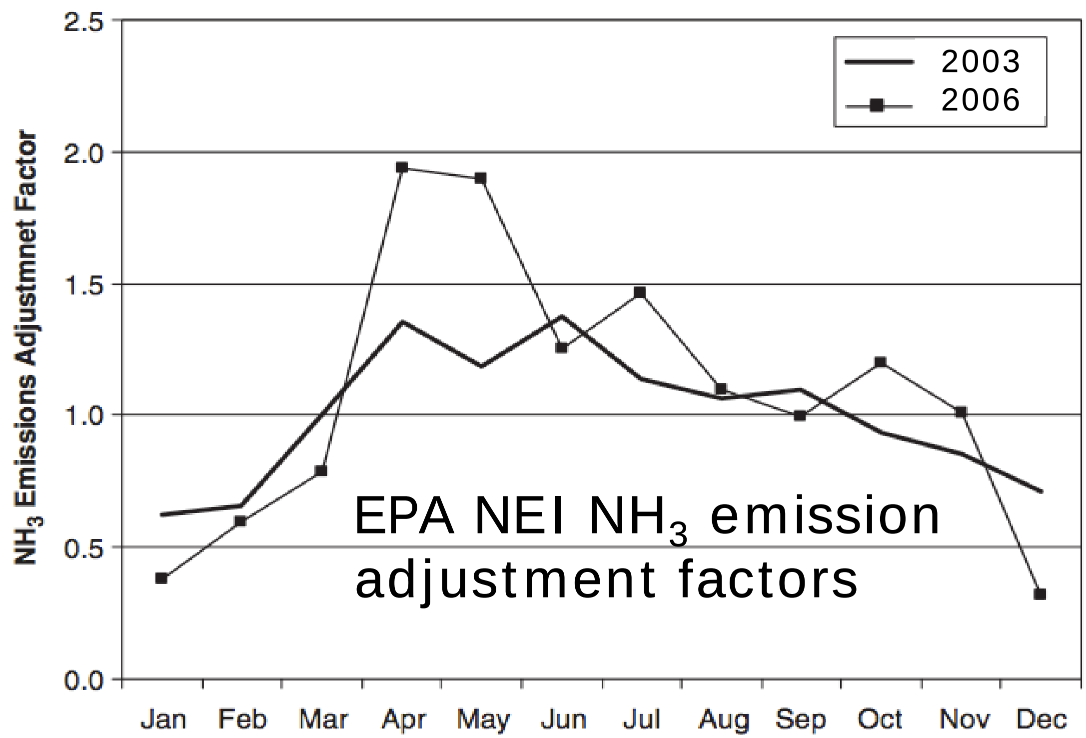
Many US air quality models get NHx deposition correct via assimilation.

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

Method: adjust (w/Kalman Filter) monthly nation-wide scale factors

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

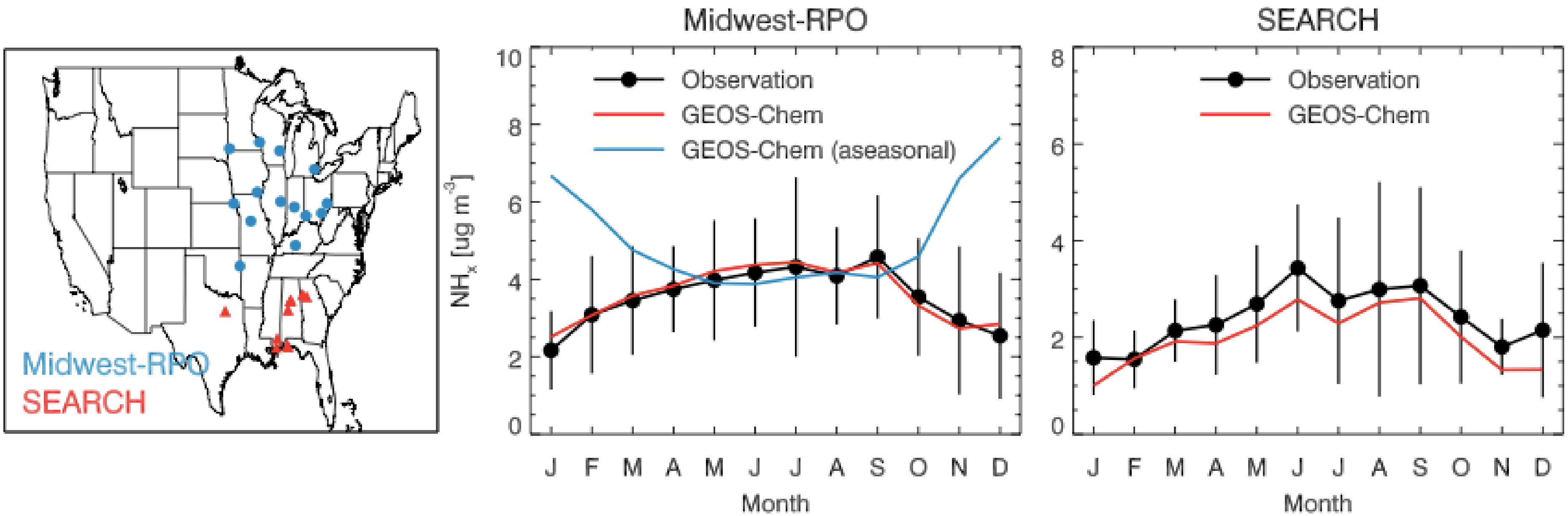
Assumptions:
- uniform seasonality throughout broad regions of US



Top-down constraints based on NH_x

Mendoza-Dominguez and Russell, 2001: constraints on NH_3 sources in the SE

Zhang et al., 2012: Seasonality of NH_3 sources adjusted so that Modeled matched RPO and SEARCH NHx measurements



- Resulting annual NHx and NO3 deposition unbiased.
- Enforces a spatially uniform seasonality / correction factor across the US.

Potential for making new inroads on this problem: ambient measurements of NH_3

Remote sensing with TES (Beer et al., 2008):

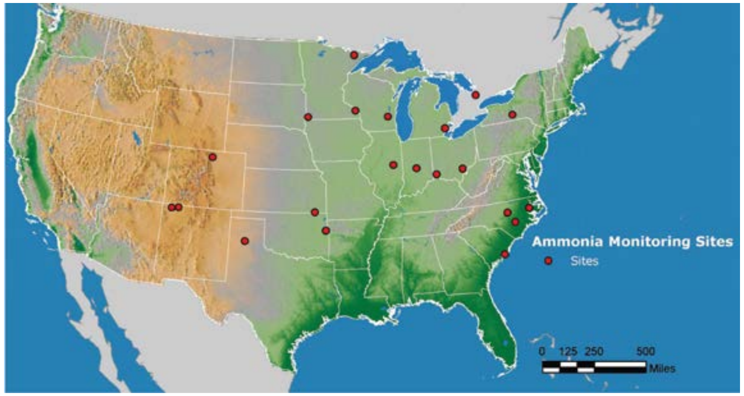
- 5 km x 8 km footprint
- sensitive to boundary layer NH_3
- detection limit of ~ 1 ppb
- bias of +0.5 ppb

July, 2005

Passive surface measurements:

EPA's AMoN sites (>2007)
(Puchalski et al., 2011)
+LADCO, SEARCH, CSU, ANARChE

2009 AMoN Sites



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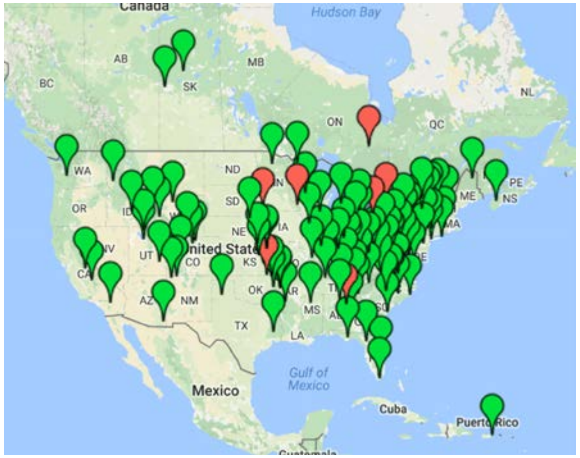
Now: AMoN

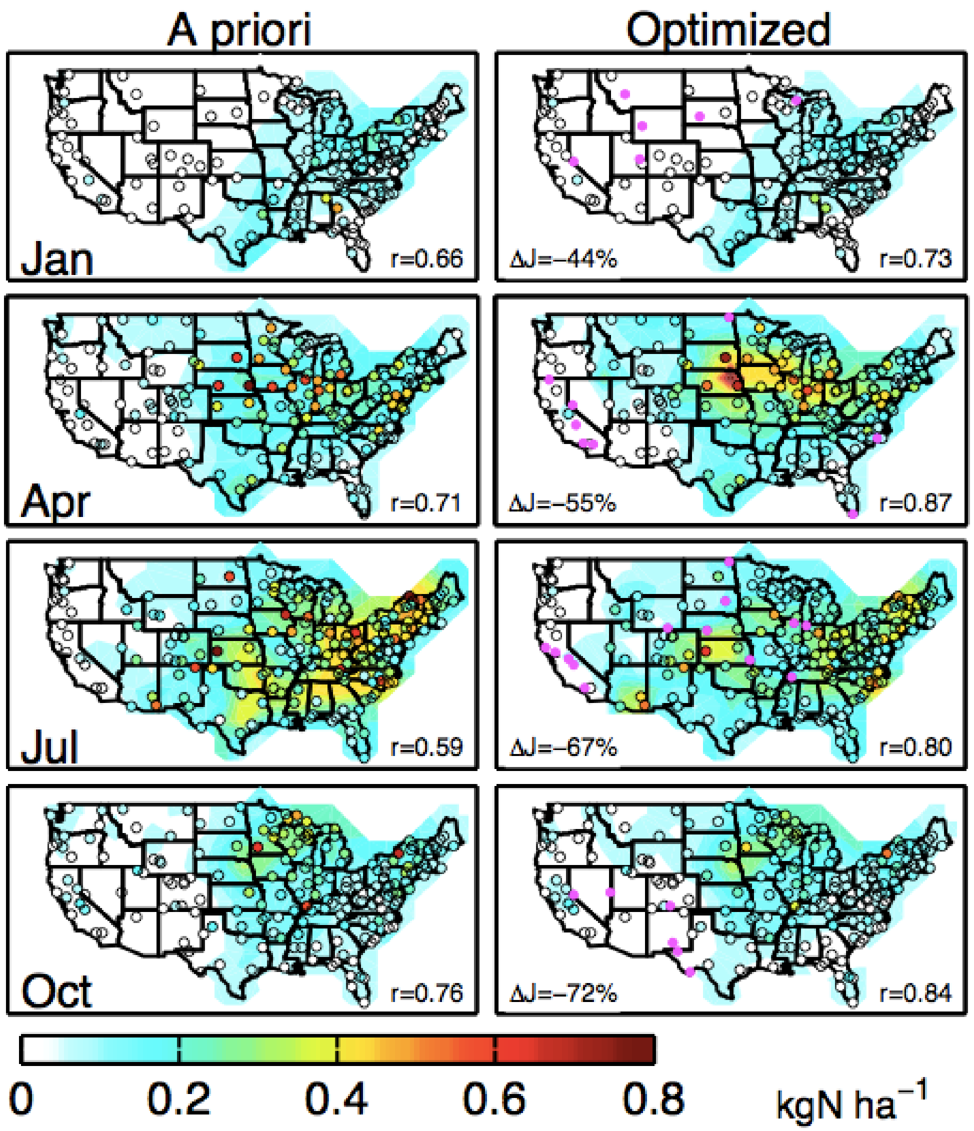


Passive surface measurements:

EPA's AMoN sites (>2007)
(Puchalski et al., 2011)
+LADCO, SEARCH, CSU, ANARChE

Now: aircraft (e.g. DISCOVER-AQ) and mobile surface (e.g., M. Zondlo, R. Volkamer)

Constraints from NH_x deposition, and an alternate bottom up inventory



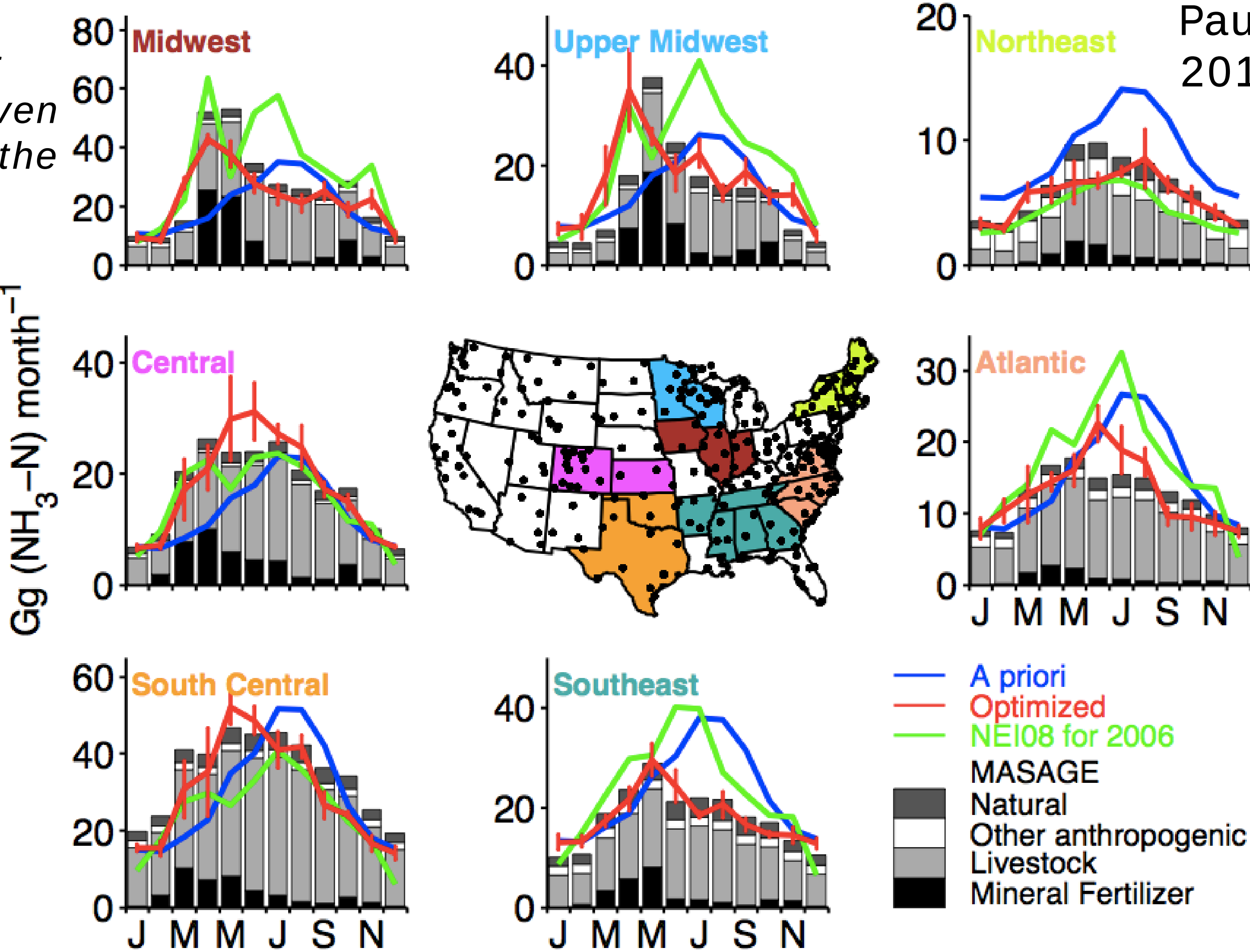
Paulot et al., 2014

- GEOS-Chem 4D-Var (Henze et al., 2007)
- Global 2x2.5
- Assimilate NTN, EMEP, ...

Constraints from NH_x deposition, and an alternate bottom up inventory

Paulot et al., 2014

Evidence for fertilizer-driven Emission in the Midwest?

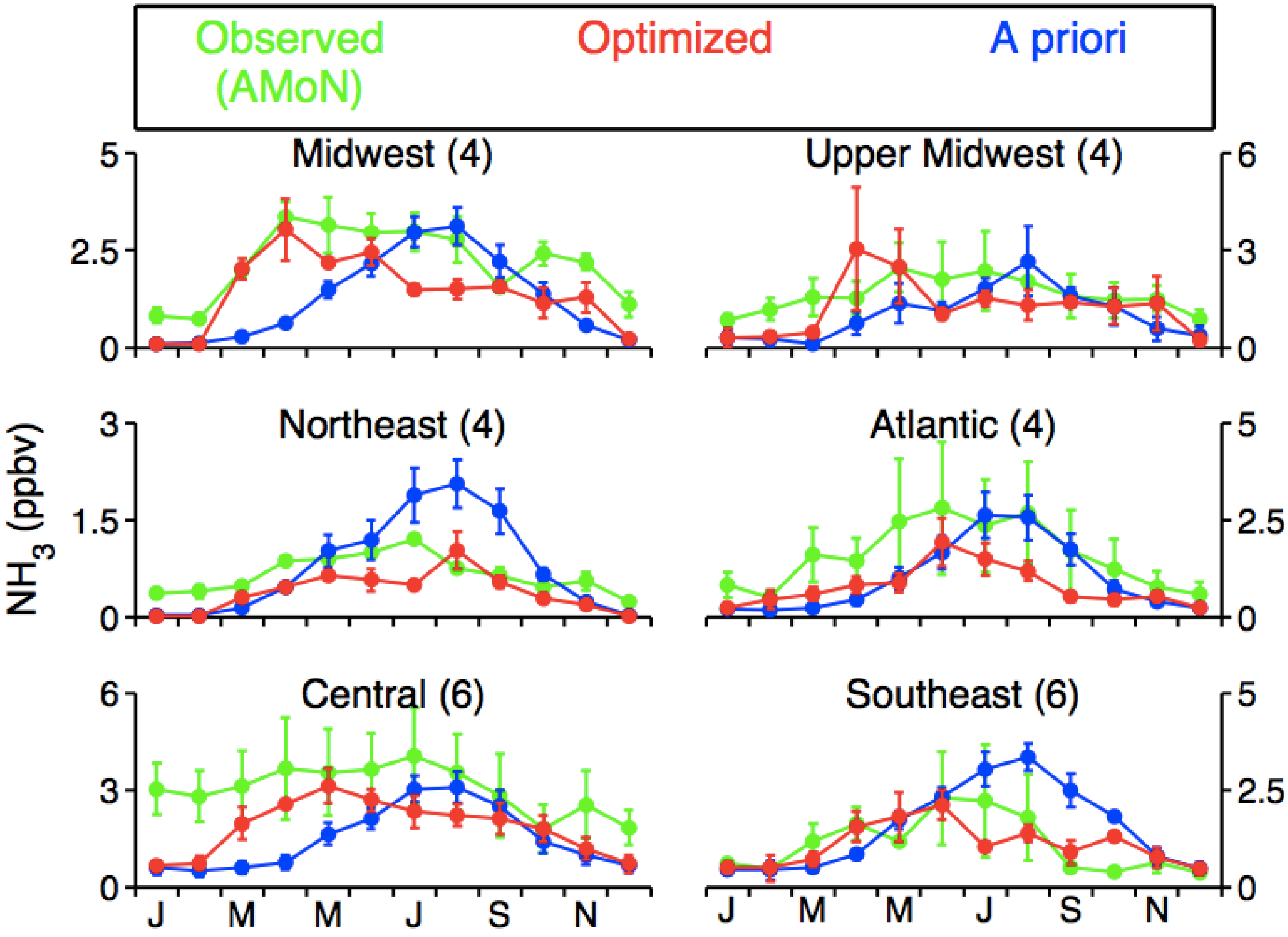


Alternate bottom-up inventory

No support for homogeneous seasonality in the US. Alternate bottom-up inventory has some success reproducing patterns of optimized emissions.

Constraints from NH_x deposition, and an alternate bottom up inventory

Comparison to surface NH3 measurements (Puchaski et al., 2011) before and after assimilation:

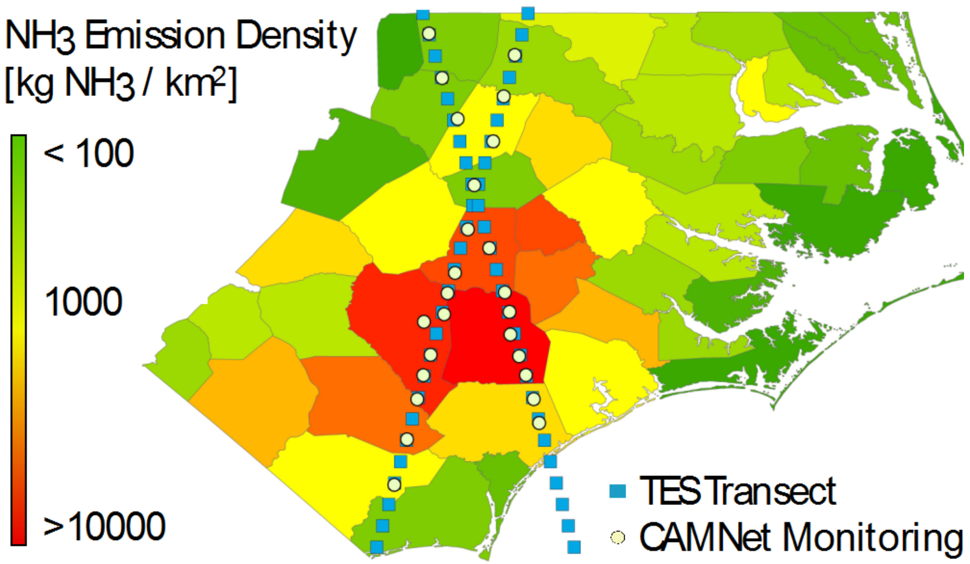


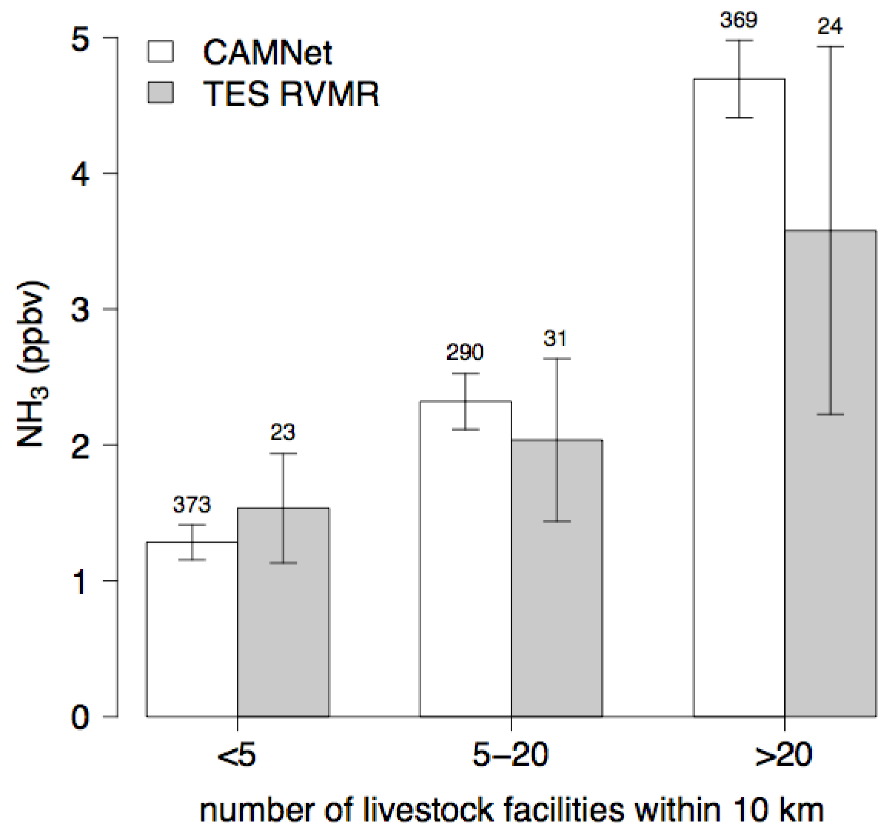
Paulot et al., 2014

TES NH_3 visualization

Detection of NH_3 gradients with TES

Overlap surface obs with TES Transects for 2009:



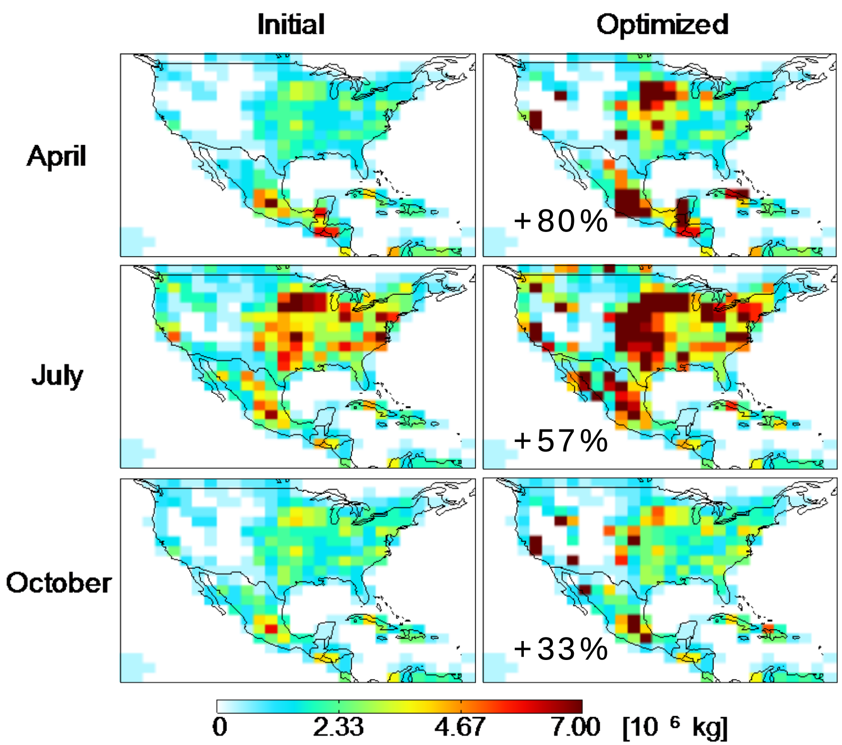


TES reflects real-world spatial gradients and seasonal trends

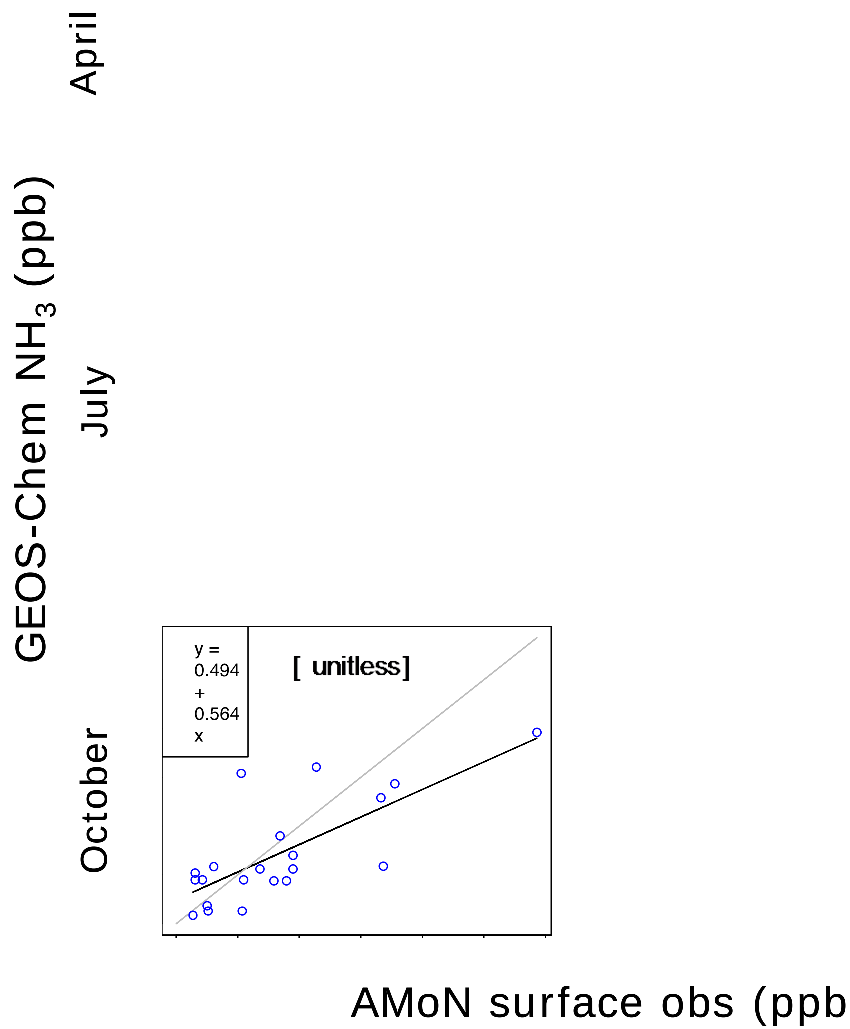
Pinder et al., GRL, 2011

Constraining emissions of NH_3 in GEOS-Chem using 4D-Var technique (Zhu et al., 2013)

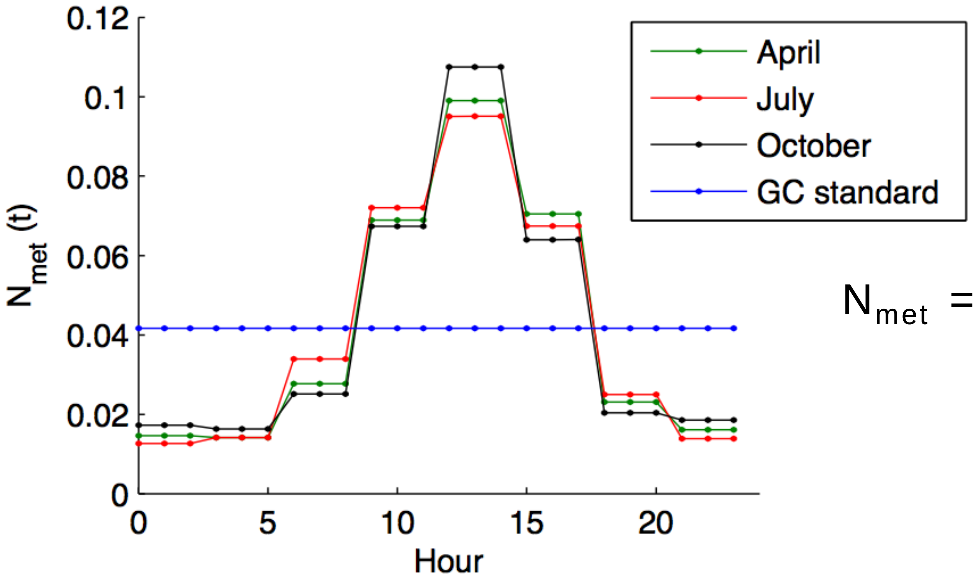
NH_3 emissions in GEOS-Chem



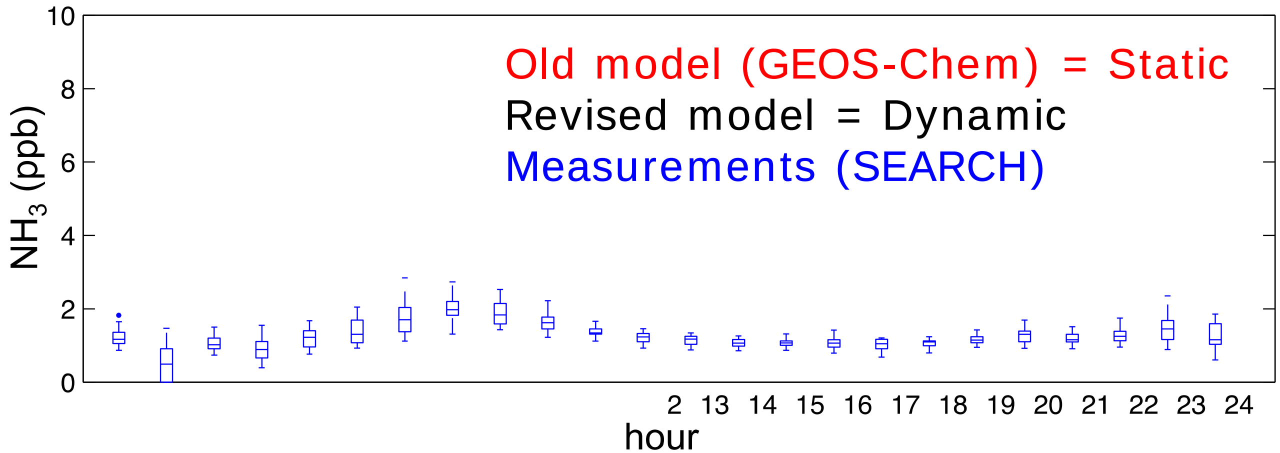
Agrees with constraints using NH_x deposition & new bottom up inventory from Paulot in April (+/- 20%) but not in July



Revised diurnal variability of NH_3 emissions

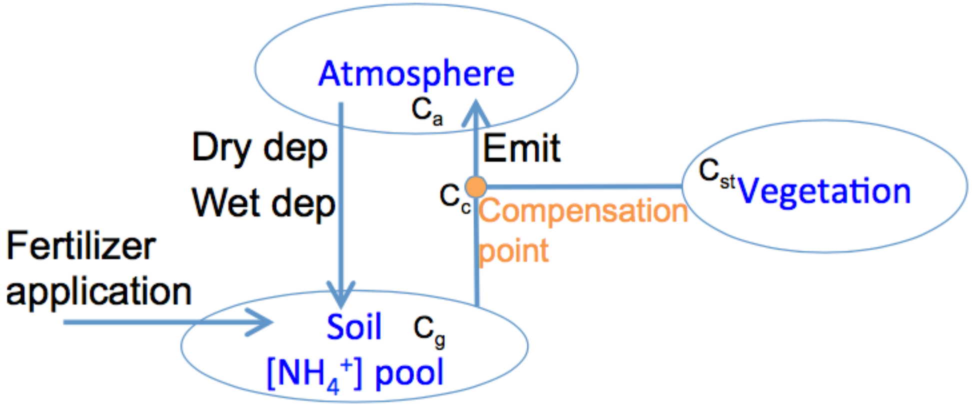


N_{met} = fraction of daily NH_3 emission



Zhu et al., 2014

NH_3 bidirectional exchange

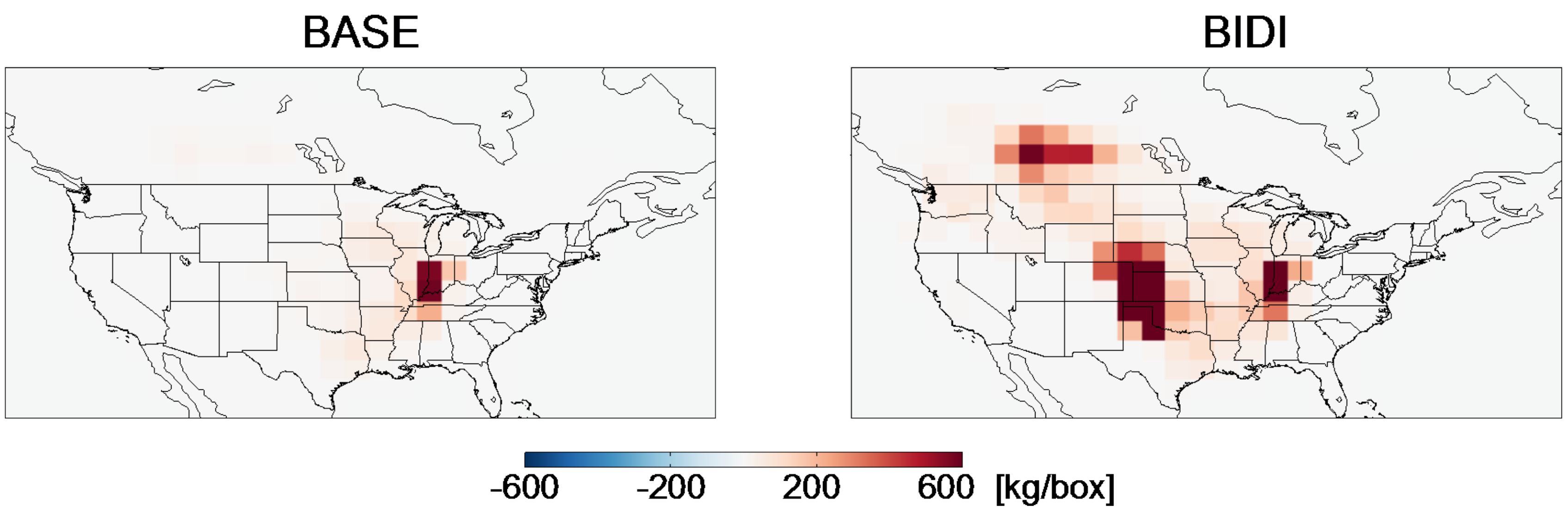


Implemented for the 1st time in a global model (Zhu et al., 2014)

Based on scheme developed for CMAQ (Bash et al., 2013)

Bidi-exchange increases the "lifetime" of NH_3:

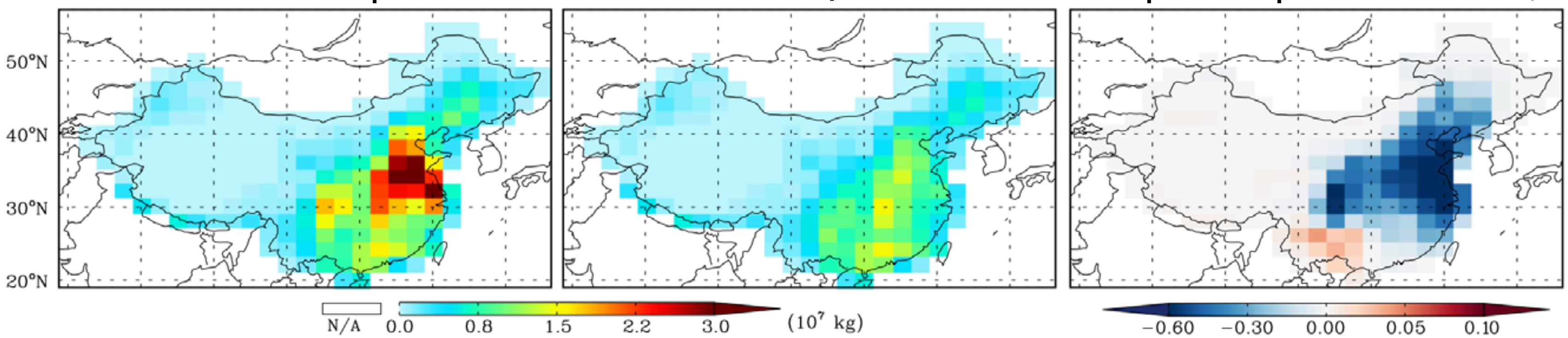
$$\frac{\partial J(NH_3)}{\partial \sigma_{ENH_3}}$$



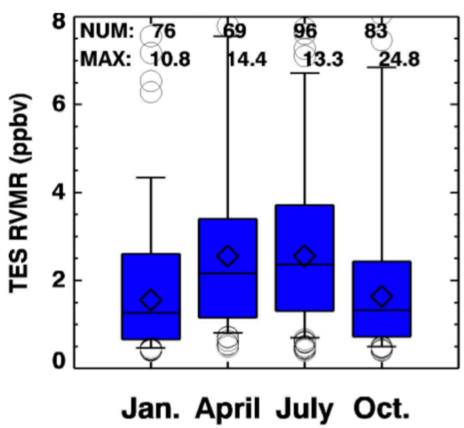
Constraining speciated aerosol sources using MODIS AOD

GEOS-Chem prior | GEOS-Chem posterior | post / prior Xu et al., 2013

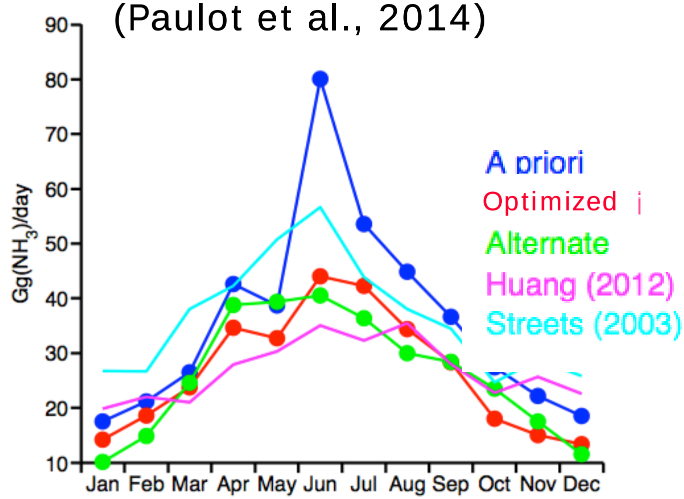
NH_3
April



TES NH_3 observations
(Shephard et al., 2011)



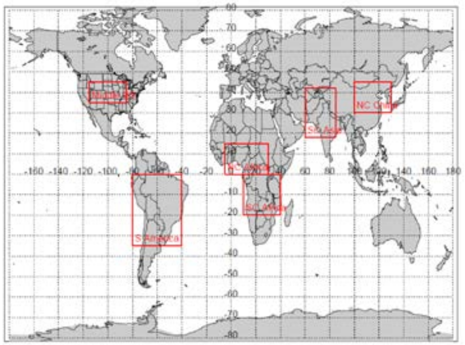
Inversion using wet NH_x deposition
(Paulot et al., 2014)



Top-down constraints agree with recent bottom up inventories: Huang (2012) and Alternate.

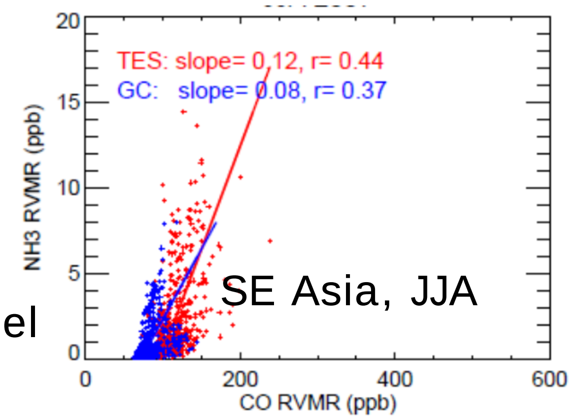
Constraints on NH_3 from AOD-based inversion consistent with satellit
NH_3 and NHx deposition inversion.

Evaluation of NH_3/CO ratios

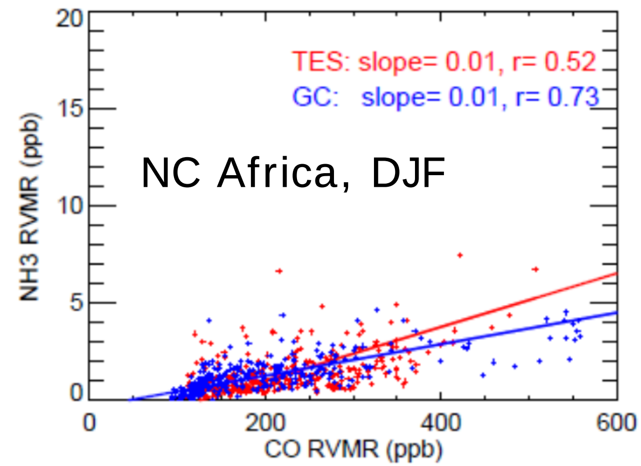


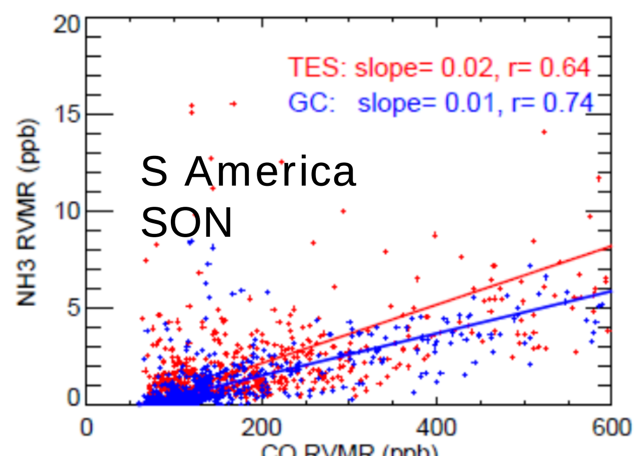
TES = satellite
GC = GEOS-Chem model

Non-biomass burning impacted region/season



Biomass burning impacted region/season



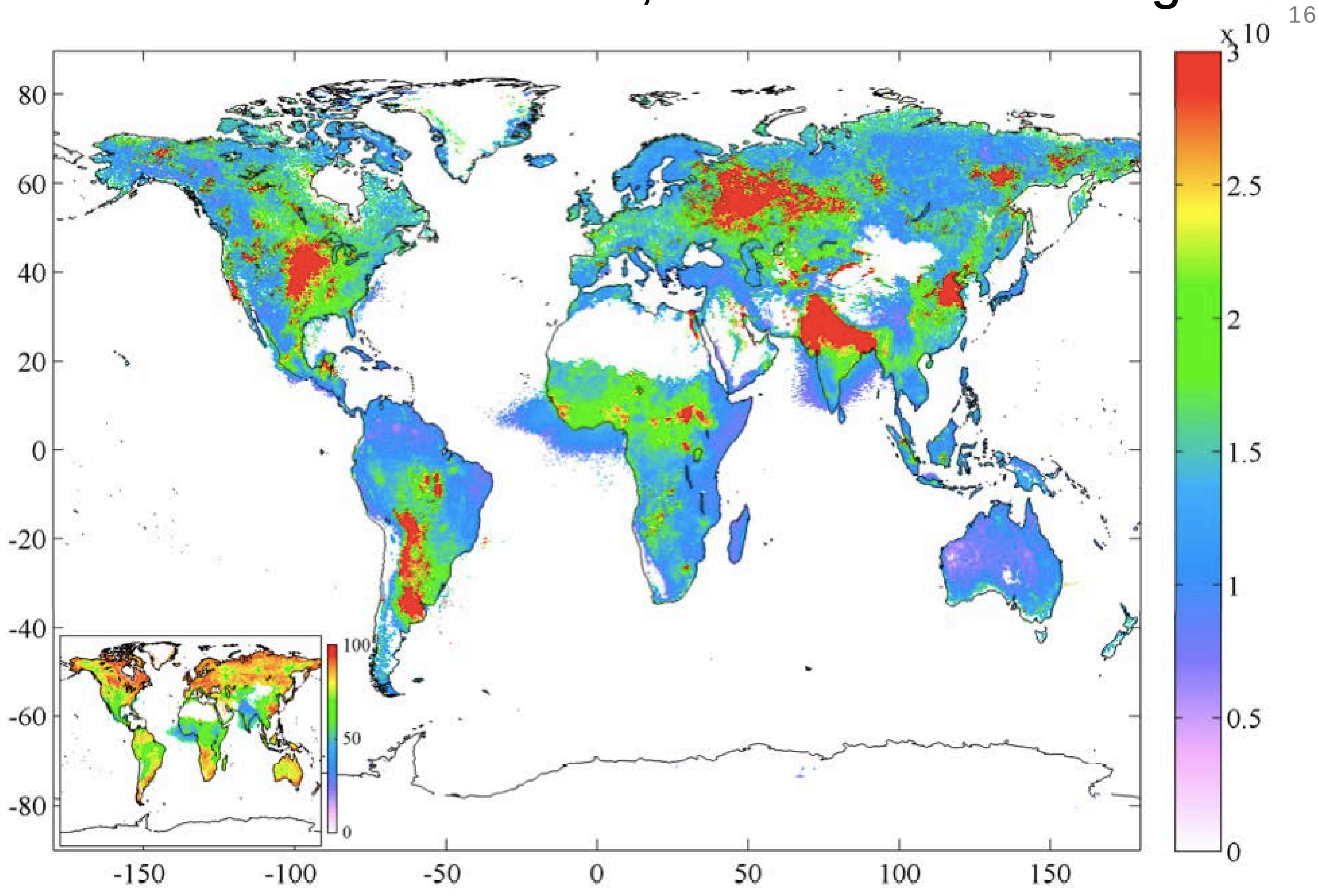


- Evaluate model emissions factors
- Higher slopes indicative of biomass burning sources

Luo et al., 2014

Remote sensing of NH_3: IASI

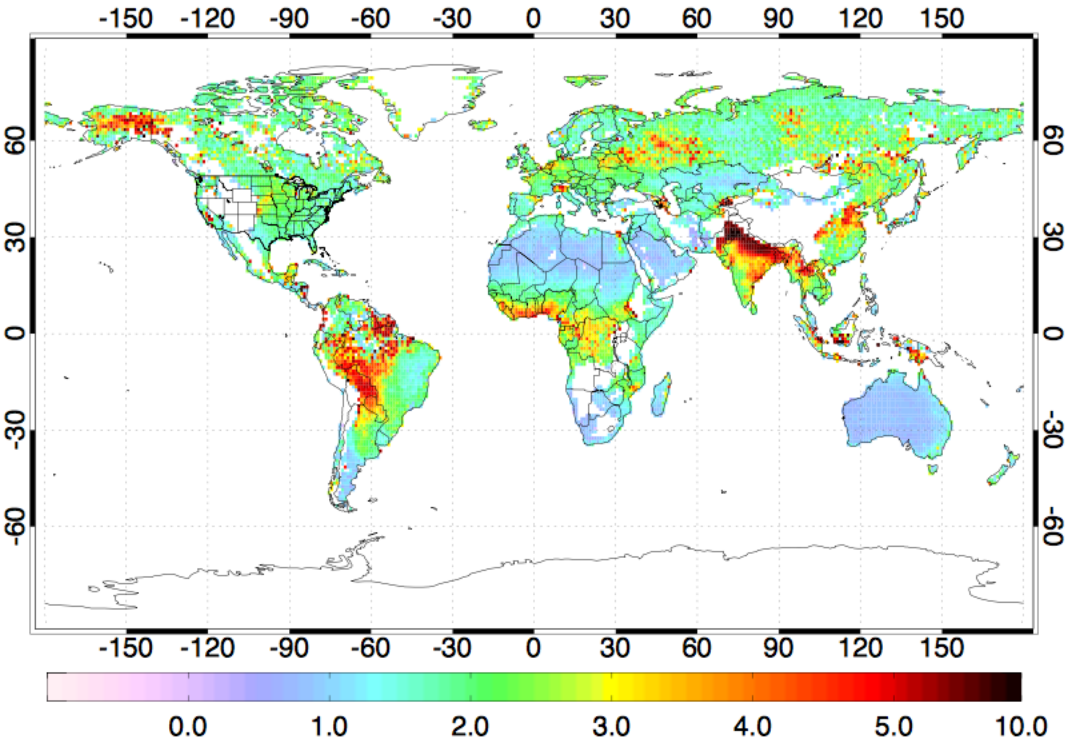
NH3 total columns, 2007-2012average



Van Damme et al., ACP, 2014

Remote sensing of NH_3: AIRS

NH3 VMRs at 918 hPa, 2002-2015 average



Warner et al., ACP, 2016

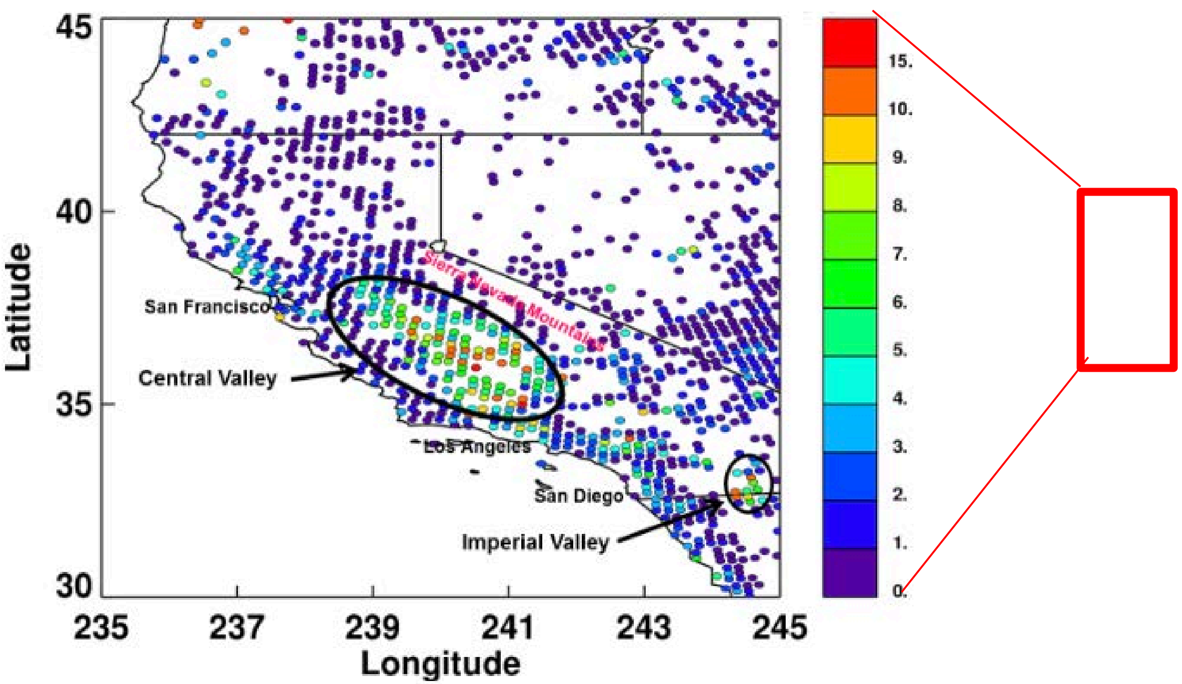
Remote sensing of NH_3: CrIS

Shephard and Cady-Pereira, AMT, 2015:

- New retrievals from CrIS (aboard Suomi-NPP)
- Will be produced operationally by end of 2017
- Much greater spatial density (x100) and sensitivity (x4) than TES
- evaluated with in situ and aircraft data

(a) CrIS: June 13, 2012

(b) TES: July 4 – 19, 2005



Final summary

NH_3 emissions pose a range of concerns on regional to global scales.

In situ measurements providing increased constraints for top-down NH_3 emissions estimates

Inverse modeling shows regionally variable seasonality throughout the US. Also guided other AQ model improvements (diurnal variability, bidi-exchange).

More data is available now (networks, mobile measurements, satellites) to revisit these questions and further evaluate both bottom-up and top down inventories.

Questions?

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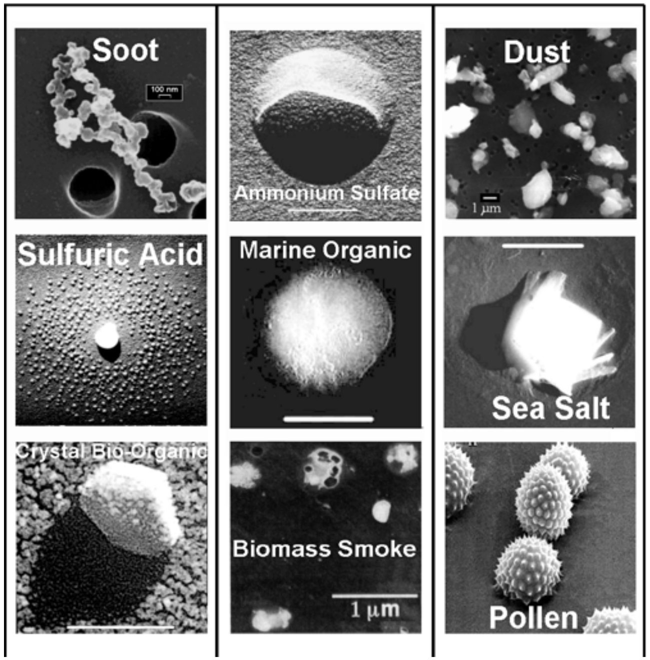
Atmospheric aerosols

Lifetime of 3 – 10 days

Significant impacts on
- air pollution
- visibility
- climate and meteorology

From emissions of
- dust, sea-salt, BC, OC (solid)
- SO_2, NH_3, NO_x, VOCs (gas-phase)



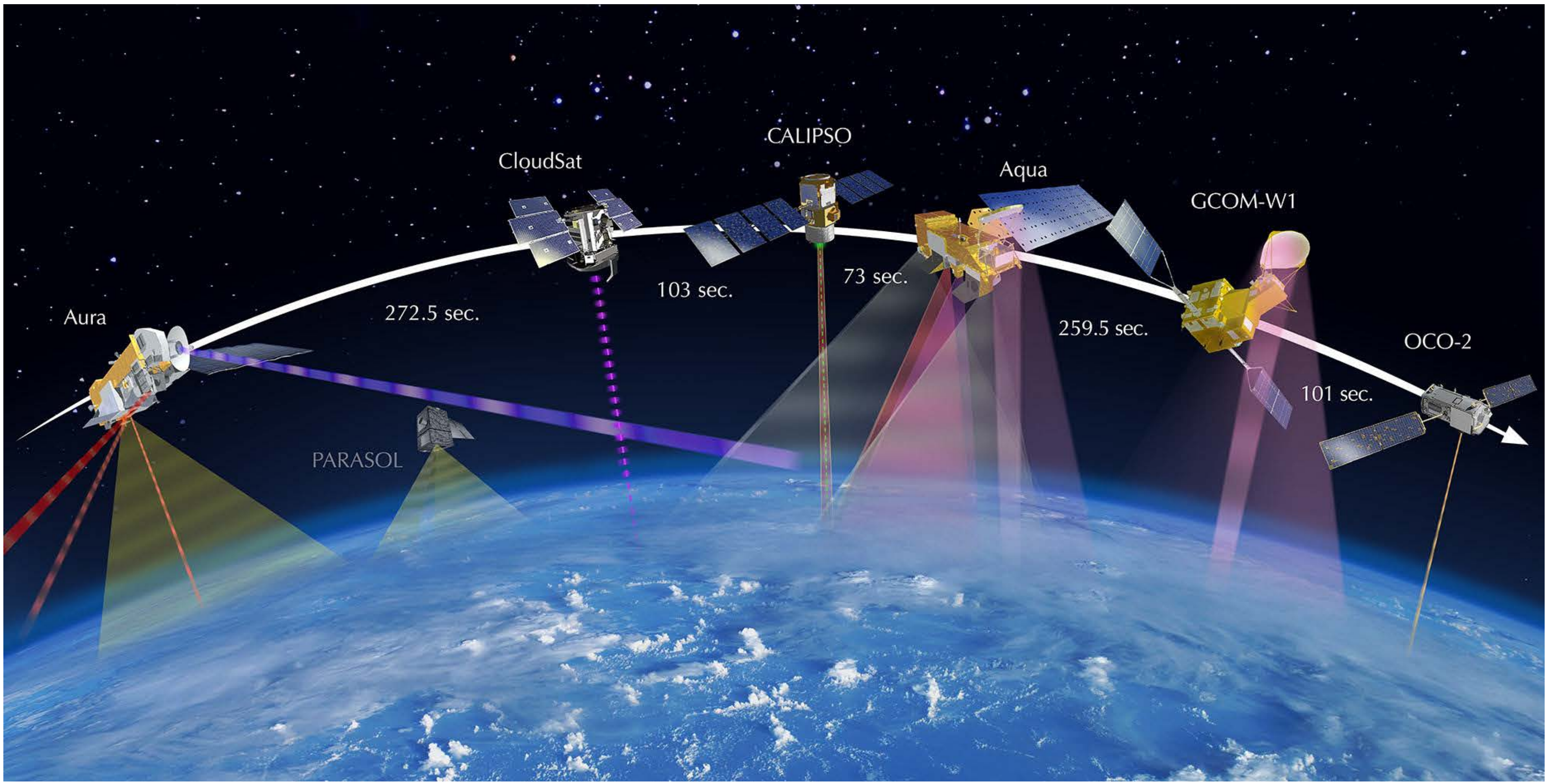
Peter Buseck, Arizona State

By a mix of anthropogenic and natural sources: transportation, energy generation, fires, industry, agriculture, residential heating and cooking, ...

- 4.2 (3.7-4.8) million annual premature deaths in 2015, #5 death risk factor (Cohen et al., Lancet, 2017).

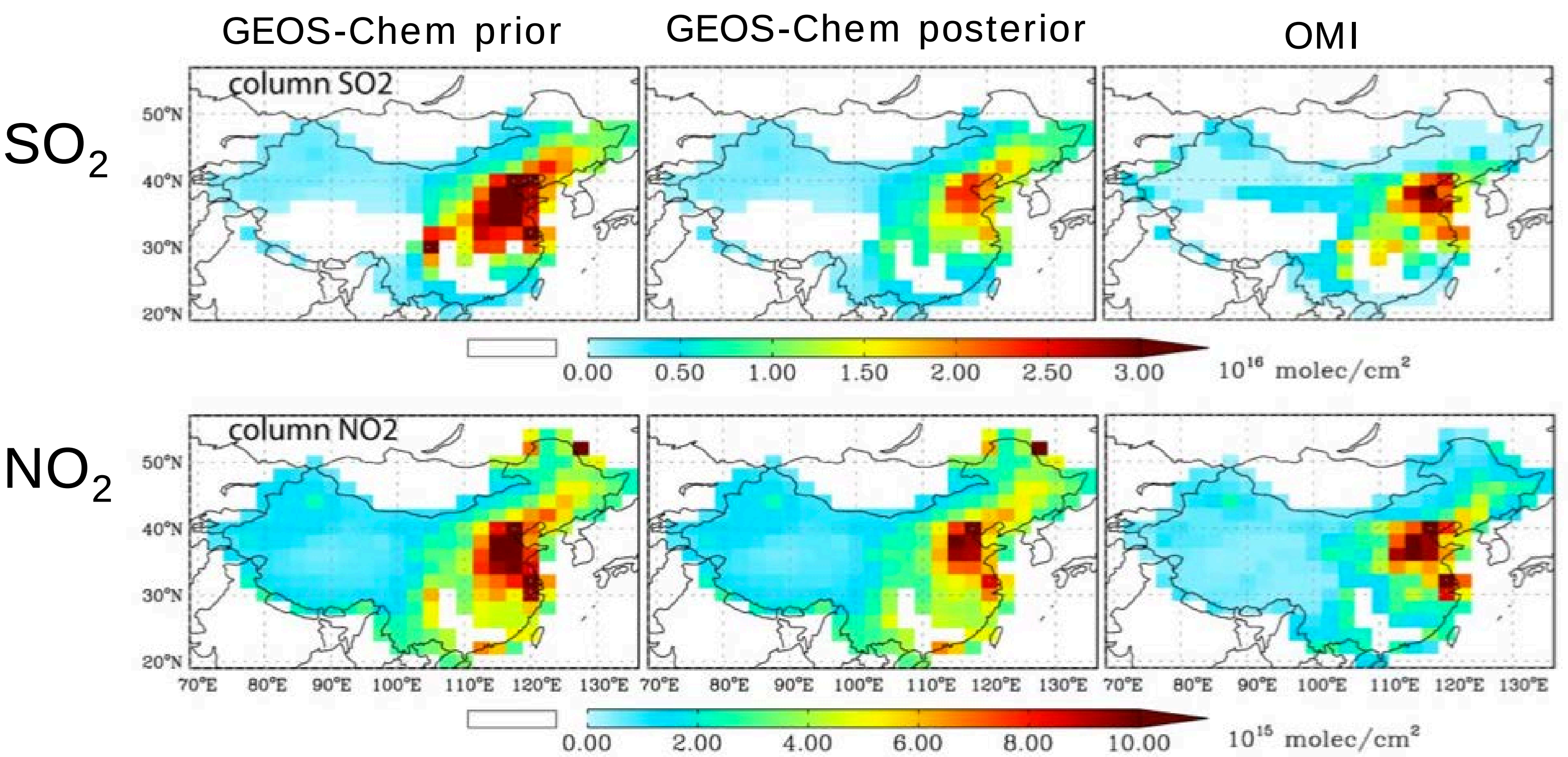
Current remote sensing of tropospheric composition

A-TRAIN (NASA)
Additional measurements from NOAA (VIIRS, CrIS), ESA (IASI), Korea (GOCI)



Constraining speciated aerosol sources using MODIS AOD

- constrain multiple aerosol precursor emissions with AOD
- evaluate constraints with gas-phase remote sensing



Xu et al., 2013

Questions?