Inverse Modeling and Attainment Analysis for Improved Decision Support of $PM_{2.5}$ Air Quality Regulations

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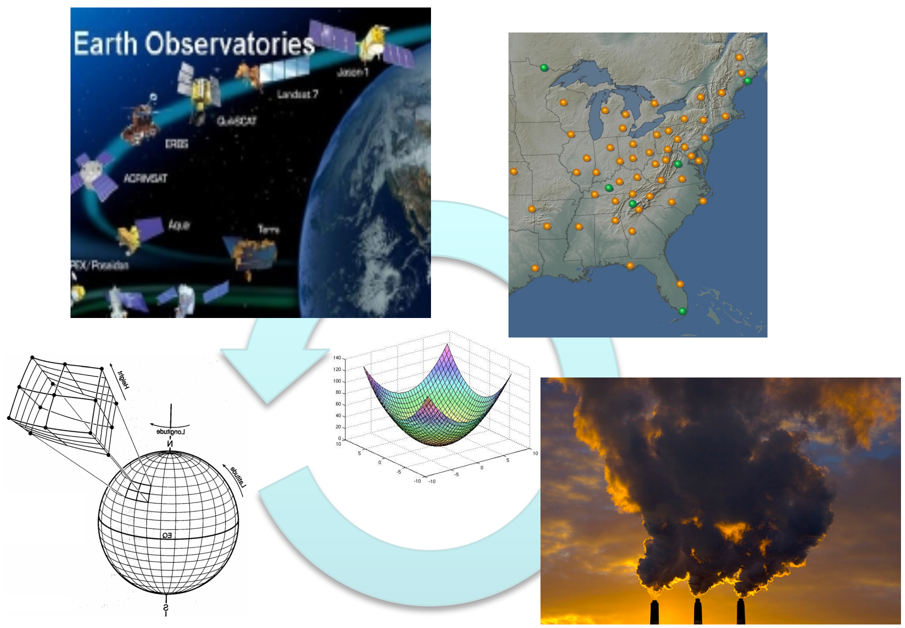
Patrick L. Kinney, Darby Jack, Ying Li, Columbia University
Loretta Mickley, Daniel Jacob, Harvard University
Drew Shindell, NASA GISS
Randall Martin, Dalhousie University
Amir Hakami, Carleton University



Partners at US EPA:

AMAD
- Sergey Napelenok
- Robert Pinder
- Alice Gilliland
- S.T. Rao

Emissions Group
- Doug Solomon
- Sally Dombrowski

OAQPS
- Bryan Hubbell

Project summary

- Motivation: ~130,000 deaths owing to $PM_{2.5}$ exposure in the U.S. annually (Fann et al., 2012). How can we AQ improve models that relate mortalities to specific precursor sources?

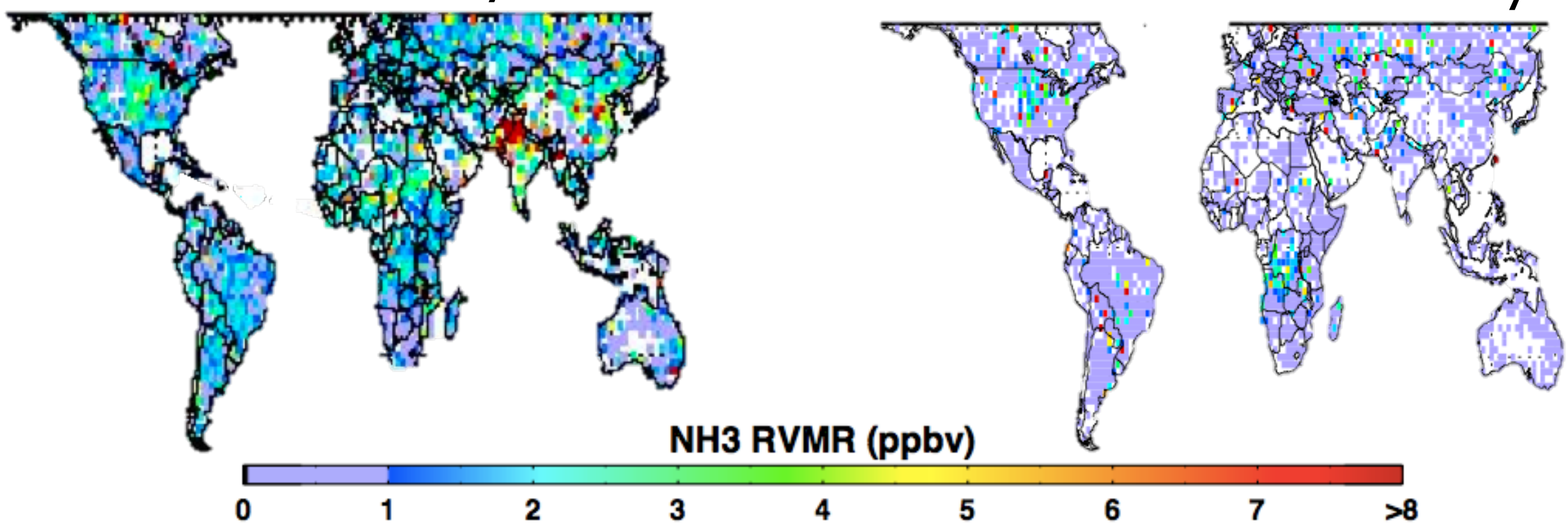
- Remote sensing is used to inform US EPA National Emissions Inventories, and adjoint modeling is used to relate these emissions to attainment of $PM_{2.5}$ exposure and ambient air quality standards.

- Earth observations and models applied:
  - TES NH3, OMI and SCIAMACHY NO2
  - IMPROVE, CASTNet, NDP, AMoN
  - GEOS-Chem, GISS Model E, CMAQ

Remote sensing of $PM_{2.5}$ precursors

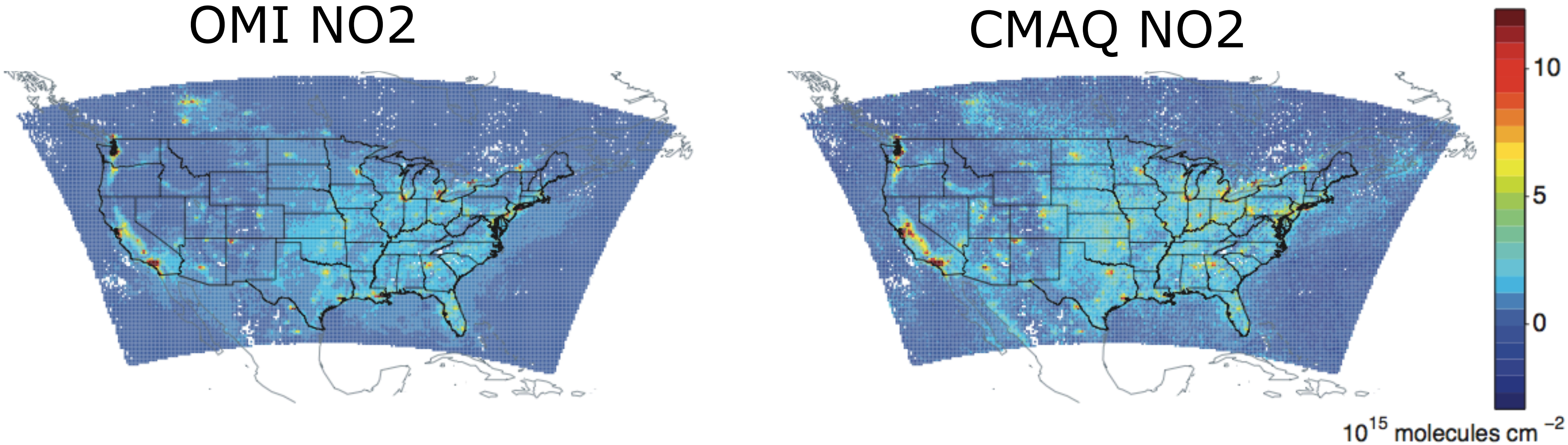
TES NH3 July

GEOS-Chem NH3 July



OMI NO2

CMAQ NO2

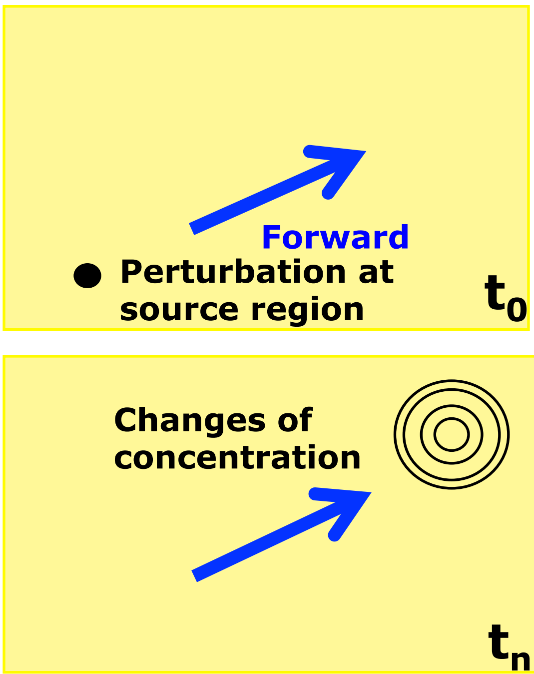


How to use remote sensing to constrain emissions?

Adjoint modeling for source-receptor analysis:

Forward Model (source-oriented)

Sensitivity of all model concentrations to one model source



Adjoint modeling for source-receptor analysis:

Forward Model (source-oriented)

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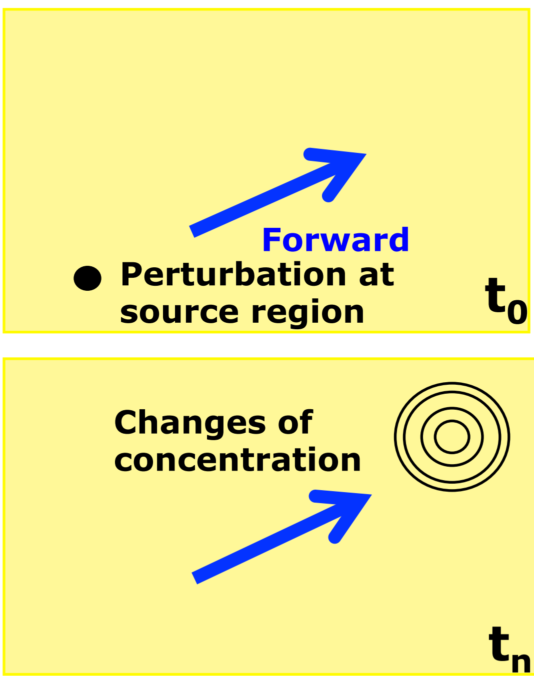
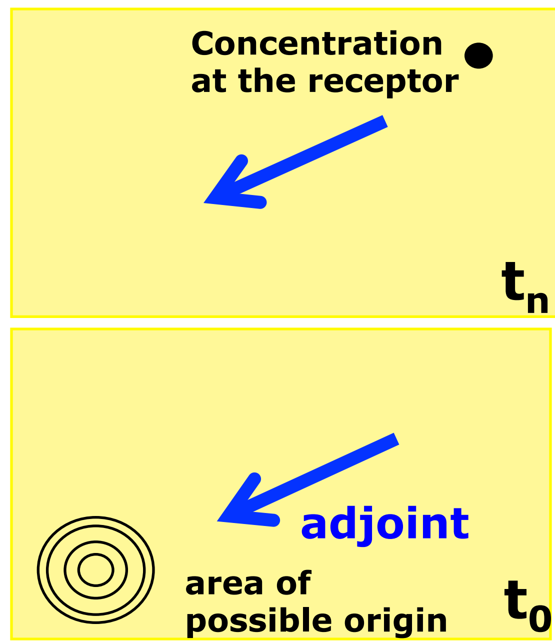


Adjoint Model (receptor-oriented)

Sensitivity of model concentration in specific location to many model sources



Adjoint modeling for source-receptor analysis:

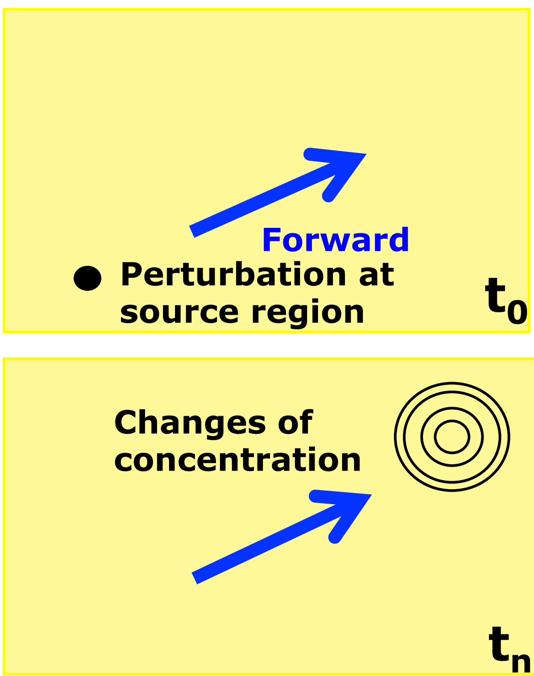
Forward Model (source-oriented)

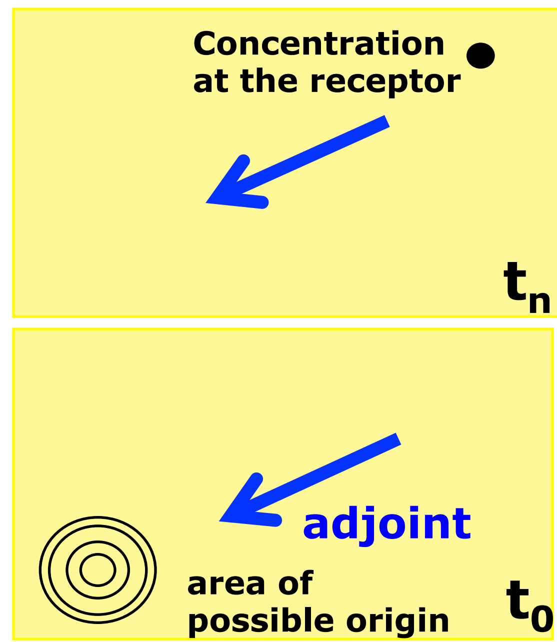
Sensitivity of all model concentrations to one model source

Adjoint Model (receptor-oriented)

Sensitivity of model *response over any region* to many model sources

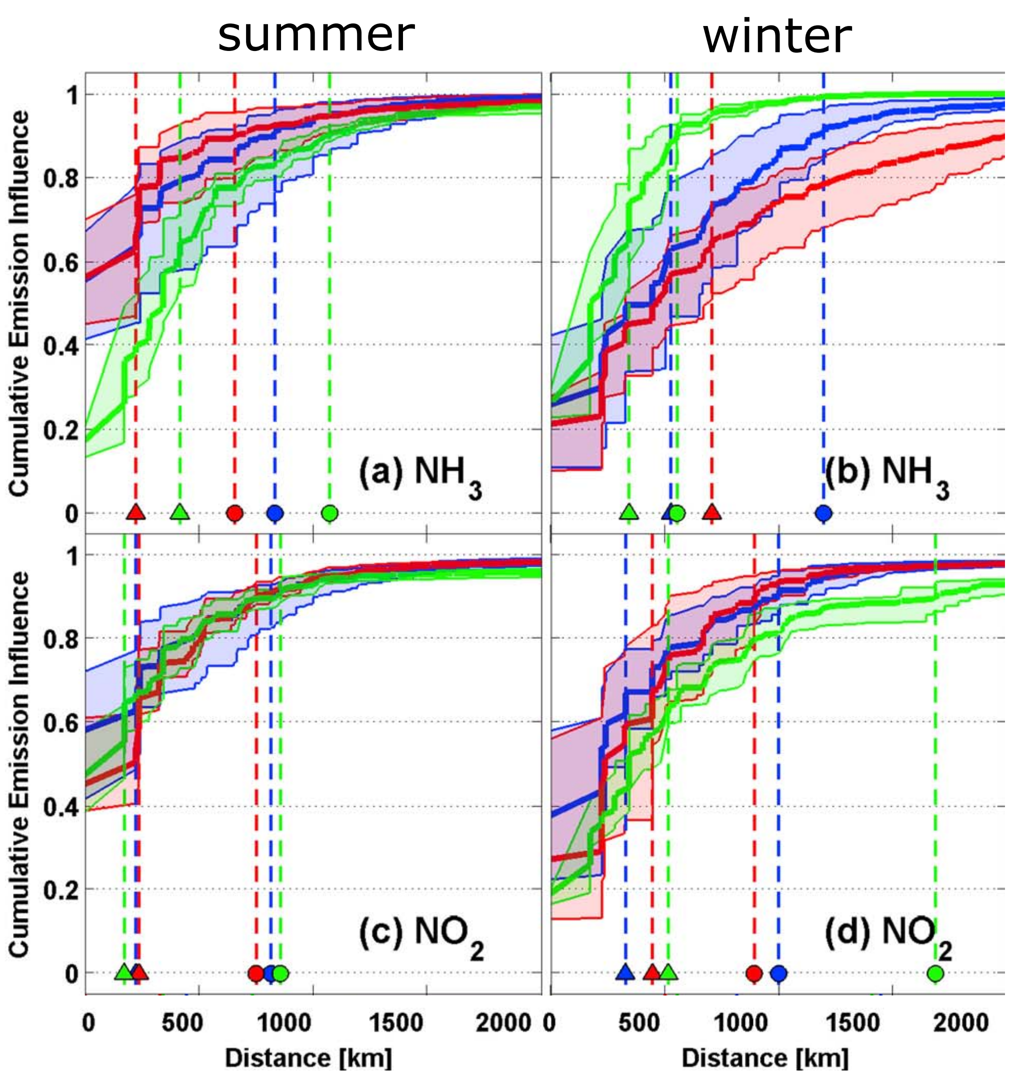
example responses: model error, statewide mortality,...





Spatial extent of short-lived species on column concentrations

Fraction of column governed by emissions

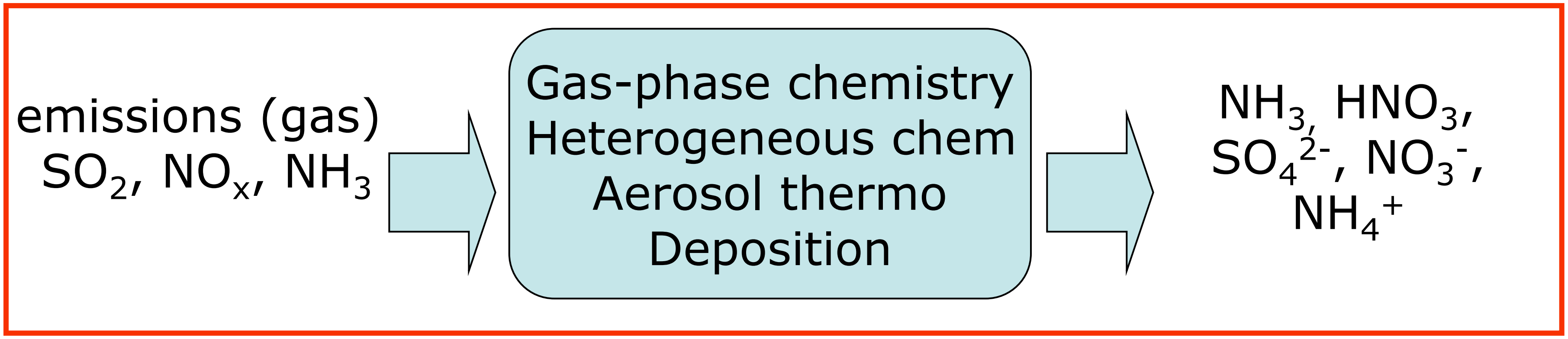


- "mass balance" approach assumed to work with 2x2.5 degree models and coarse satellite footprints may have significant errors
- Emissions of short-lived species can have long-range impacts
- Sophisticated data assimilation tools needed
- Need is exacerbated by higher resolution models and higher density of data e.g., geostationary (TEMPO)

Turner et al., *GRL*, 2012

Constraining $PM_{2.5}$ sources with 4D-Var approach

Air quality models (**M**) GEOS-Chem / CMAQ predict speciated $PM_{2.5}$



4D-Var approach: constrain emissions through inverse modeling

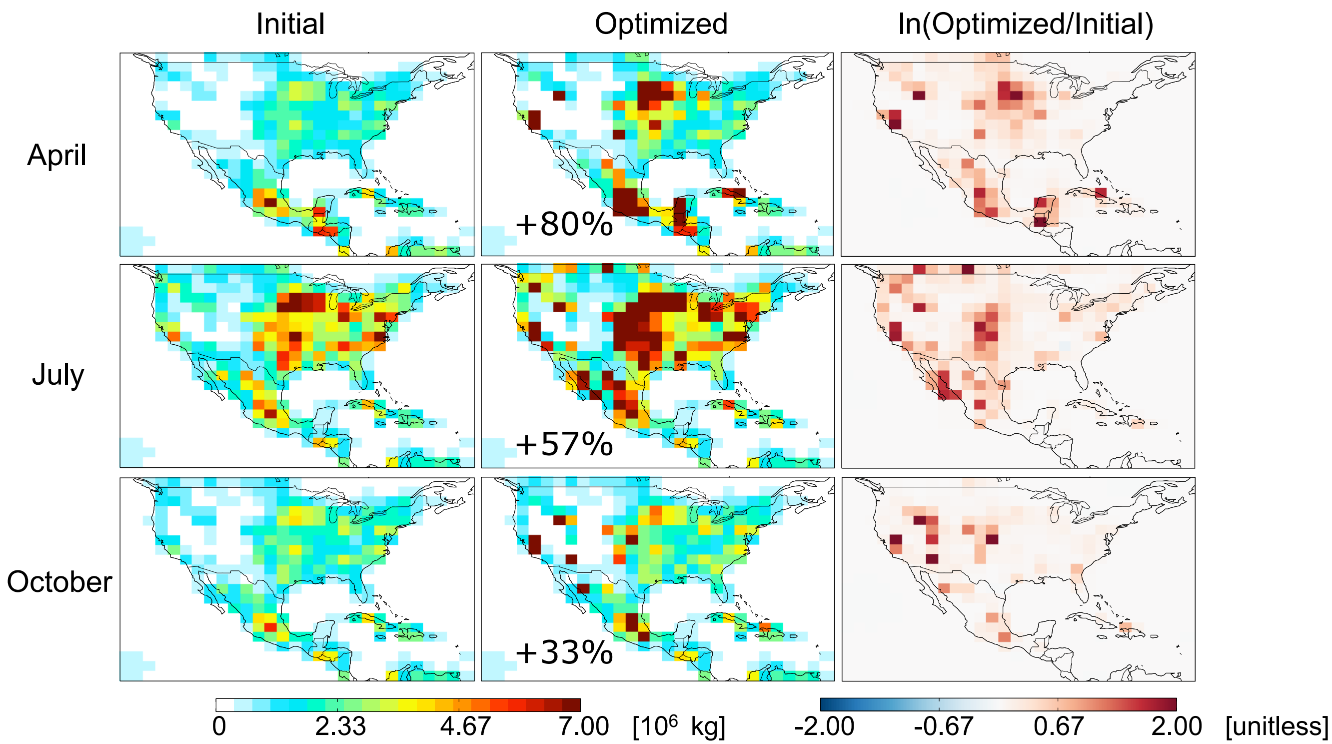
- uses adjoint model (e.g., Henze et al., 2007)
- assimilates observations (**y**)
- adjusts emissions (**x**) at the grid-scale to minimize J:

$$\min_{\mathbf{x}} \mathcal{J}(\mathbf{x}) = \sum_i (M(\mathbf{x}) - \mathbf{y}_i)^T \mathbf{S}_y^{-1} (M(\mathbf{x}) - \mathbf{y}_i) + (\mathbf{x} - \mathbf{x}^a)^T \mathbf{S}_x^{-1} (\mathbf{x} - \mathbf{x}^a)$$

model error *a priori constraint*

Constraining emissions of NH_3 in GEOS-Chem using 4D-Var technique

NH_3 emissions in GEOS-Chem

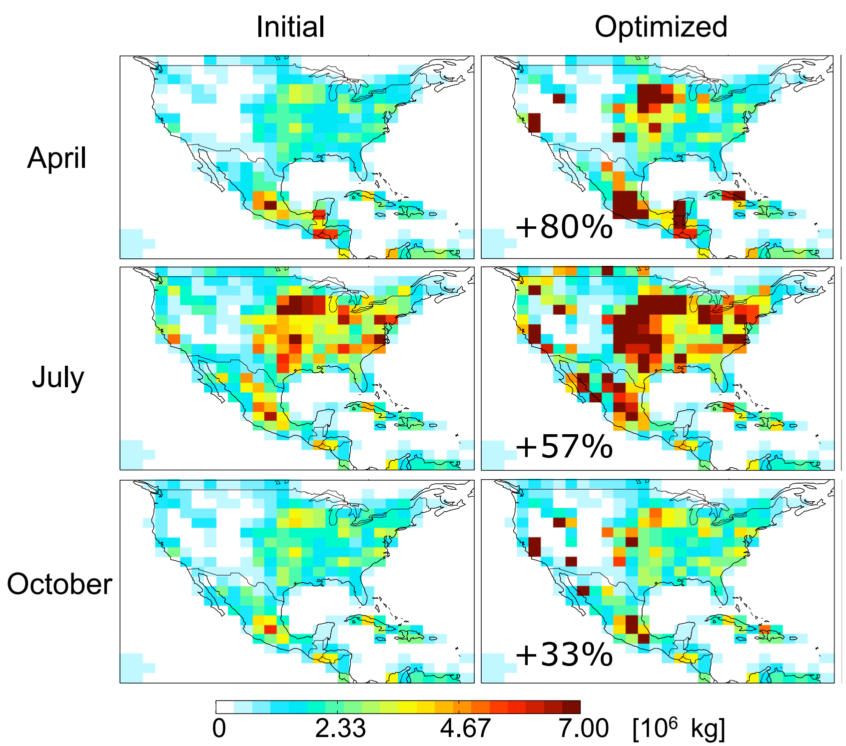


- Corrections to NH_3 sources highly spatially variable
- Recent field campaigns (CalNex) confirm some sources underestimated by x3 (e.g, Nowak et al., 2012)

Zhu et al., 2013

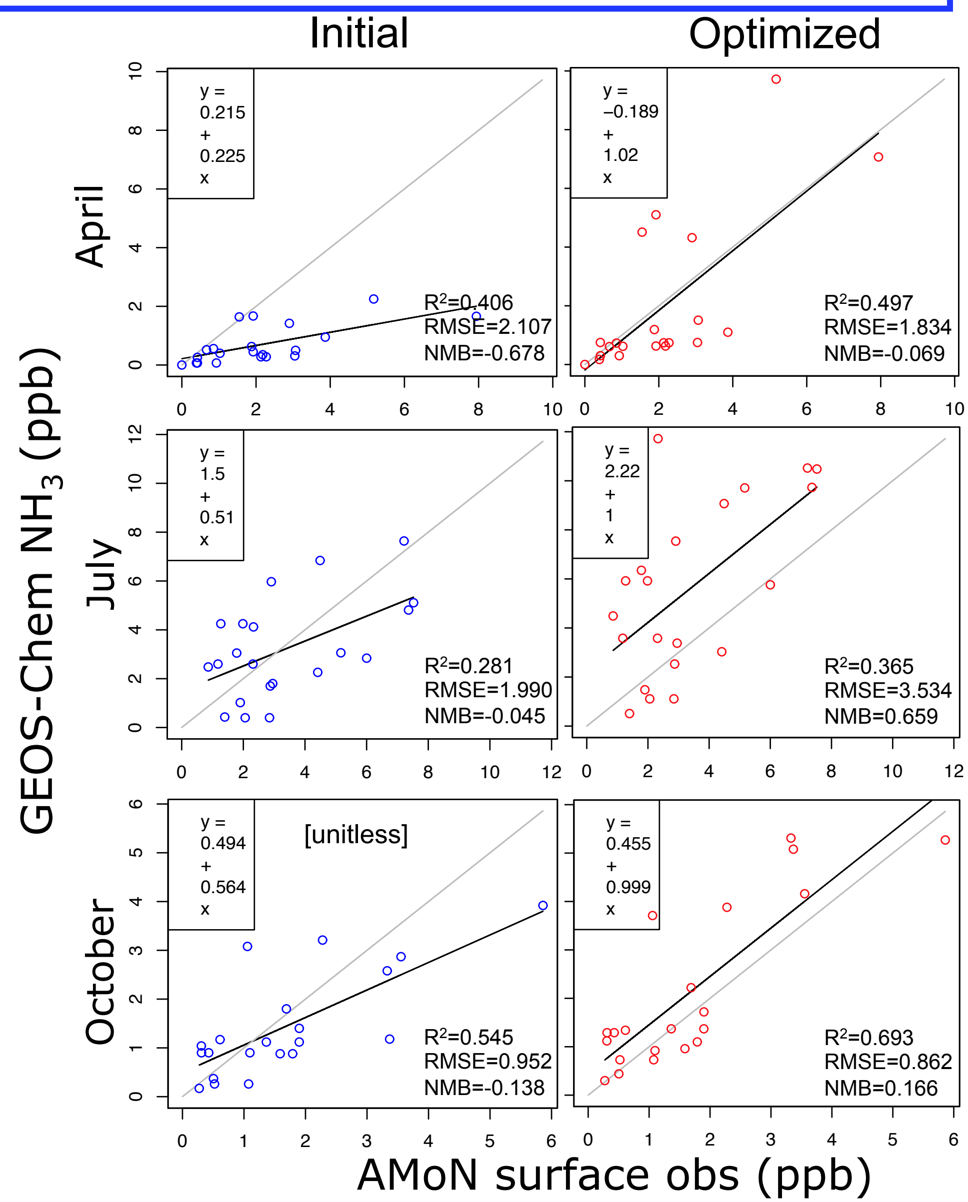
Constraining emissions of NH_3 in GEOS-Chem using 4D-Var technique

NH_3 emissions in GEOS-Chem



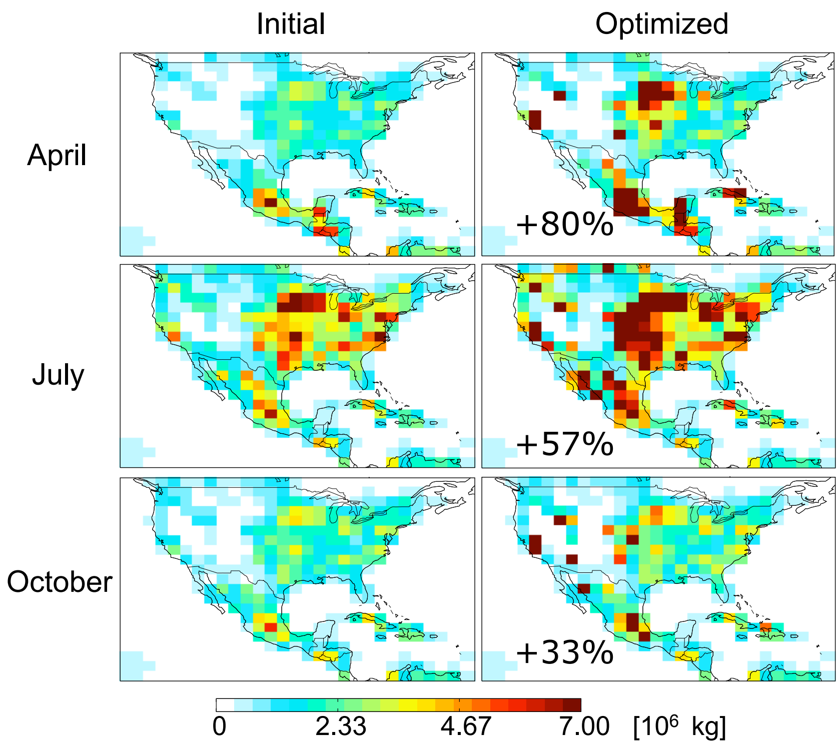
Constraints from TES improve estimates of NH_3 at AMoN sites in April and October. Contradicting in July.

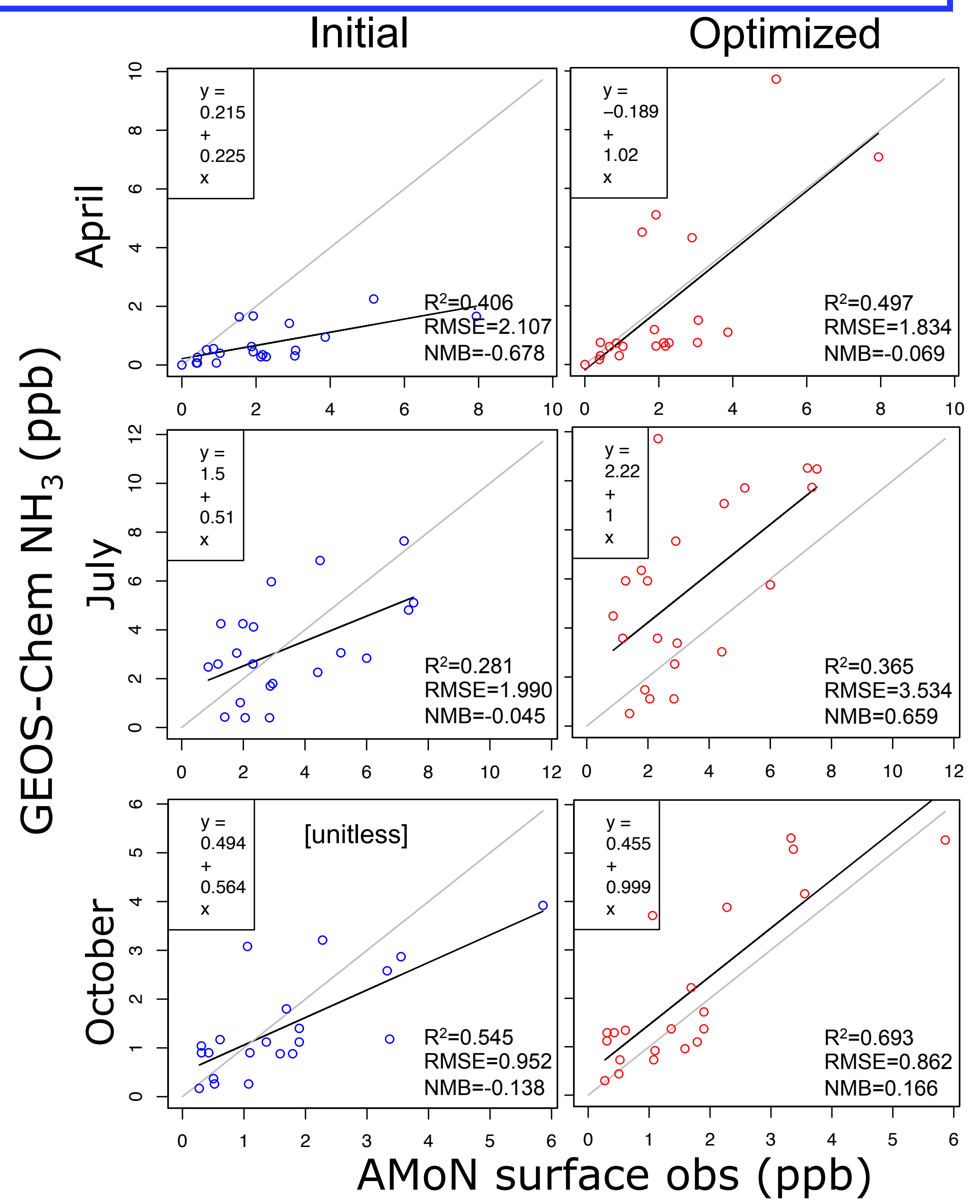
Zhu et al., 2013



Constraining emissions of NH_3 in GEOS-Chem using 4D-Var technique

NH_3 emissions in GEOS-Chem

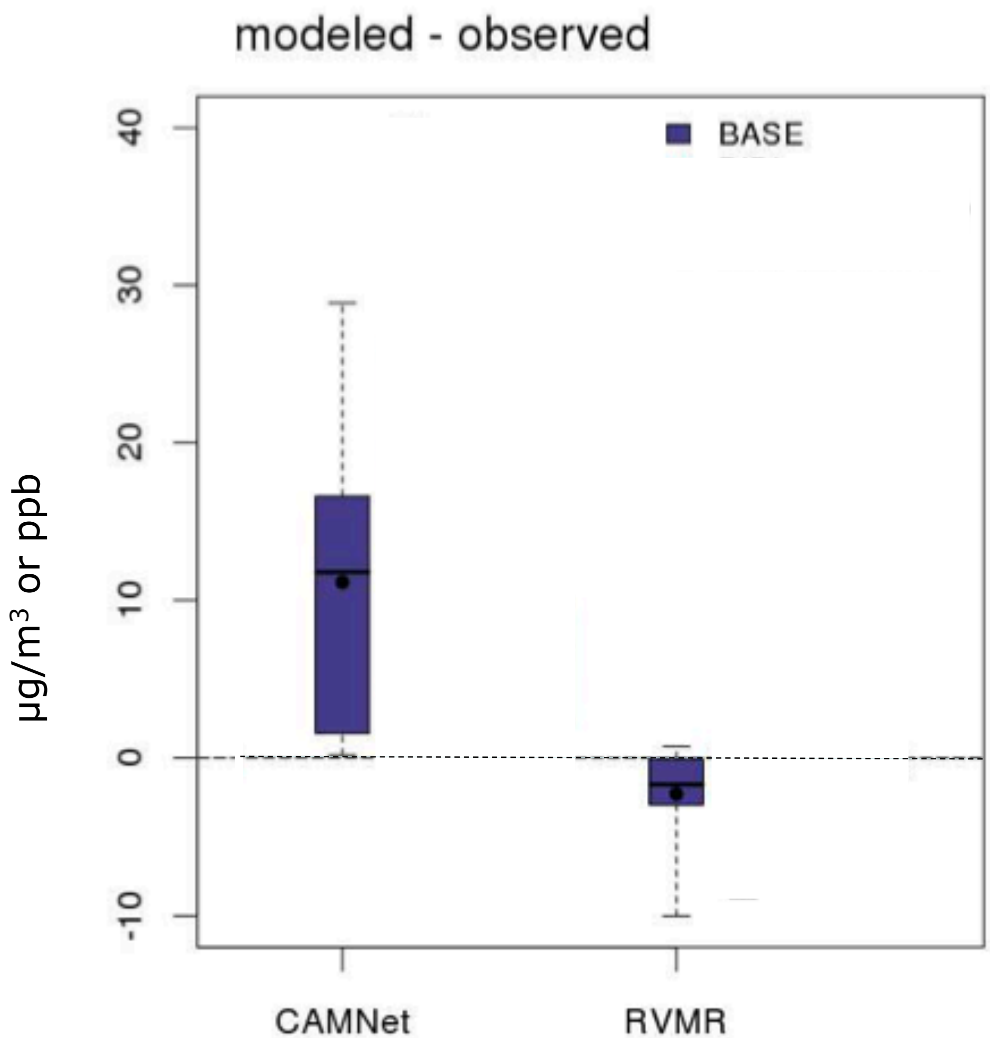




Agrees with constraints using NH_x deposition from Paulot et al. (submitted) in April (+/- 20%) but not in July

Zhu et al., 2013

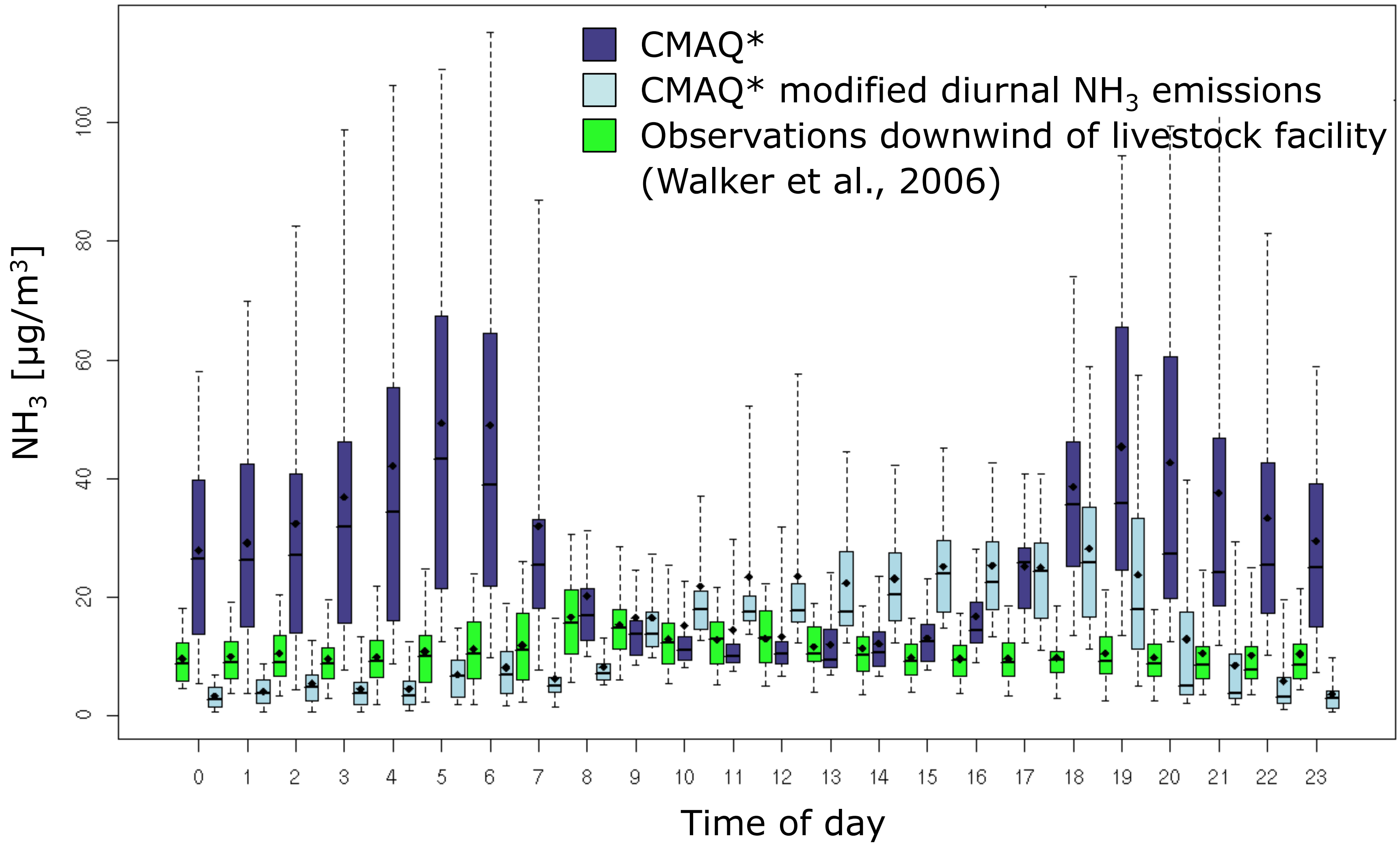
NH_3 too high at the surface, too low at ~2km also in CMAQ over NC



(surface, 2 week average) (TES, ~2km, 1:30 pm)

Jeong et al., submitted

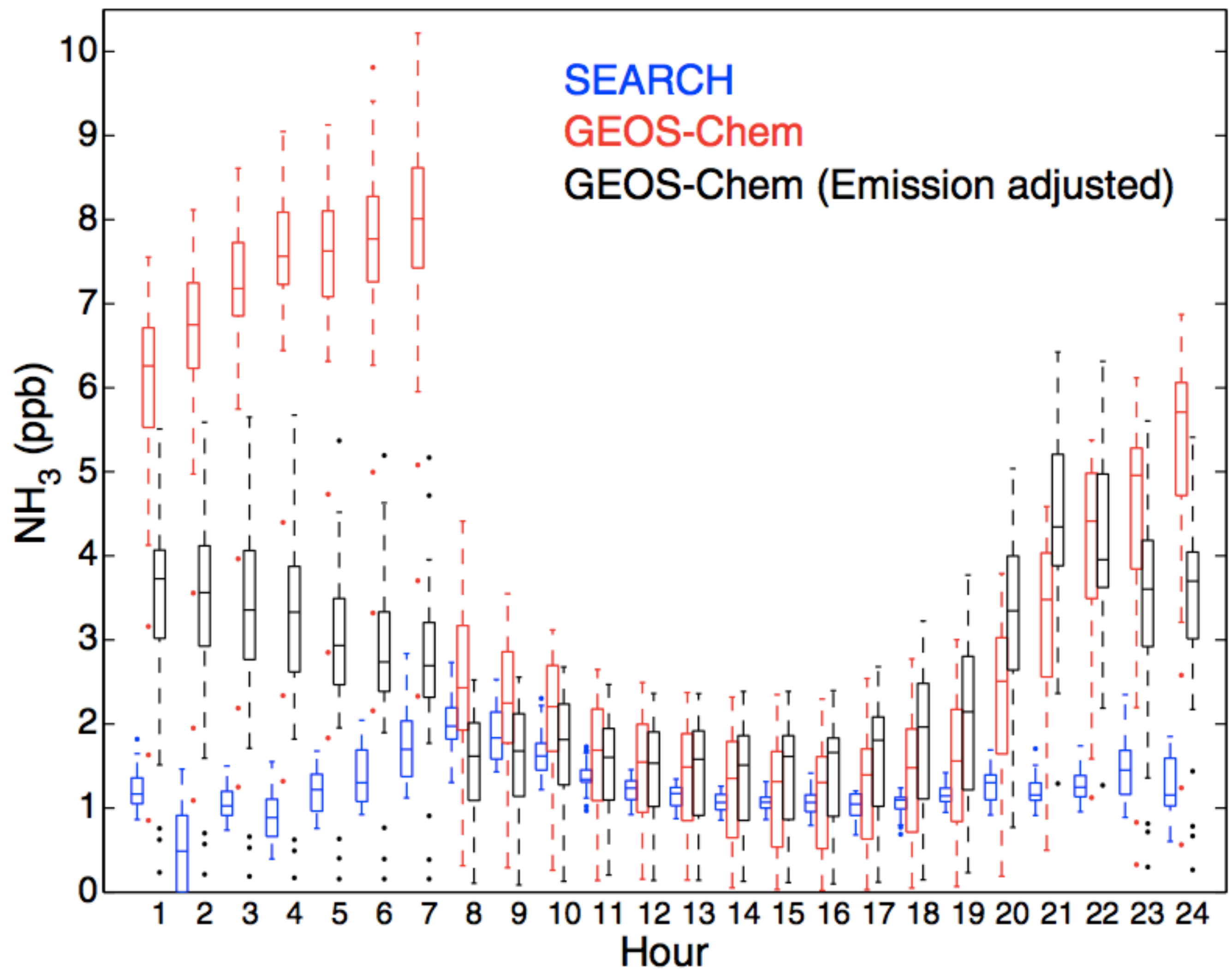
Diurnal variability of NH_3: case study in Warsaw, NC, with CMAQ regional model



* Using NEI05 emissions, simulated year not same as observations

Jeong et al., submitted

Diurnal variability of NH_3 in GEOS-Chem global model in Yorkville, GA (forest/agriculture)



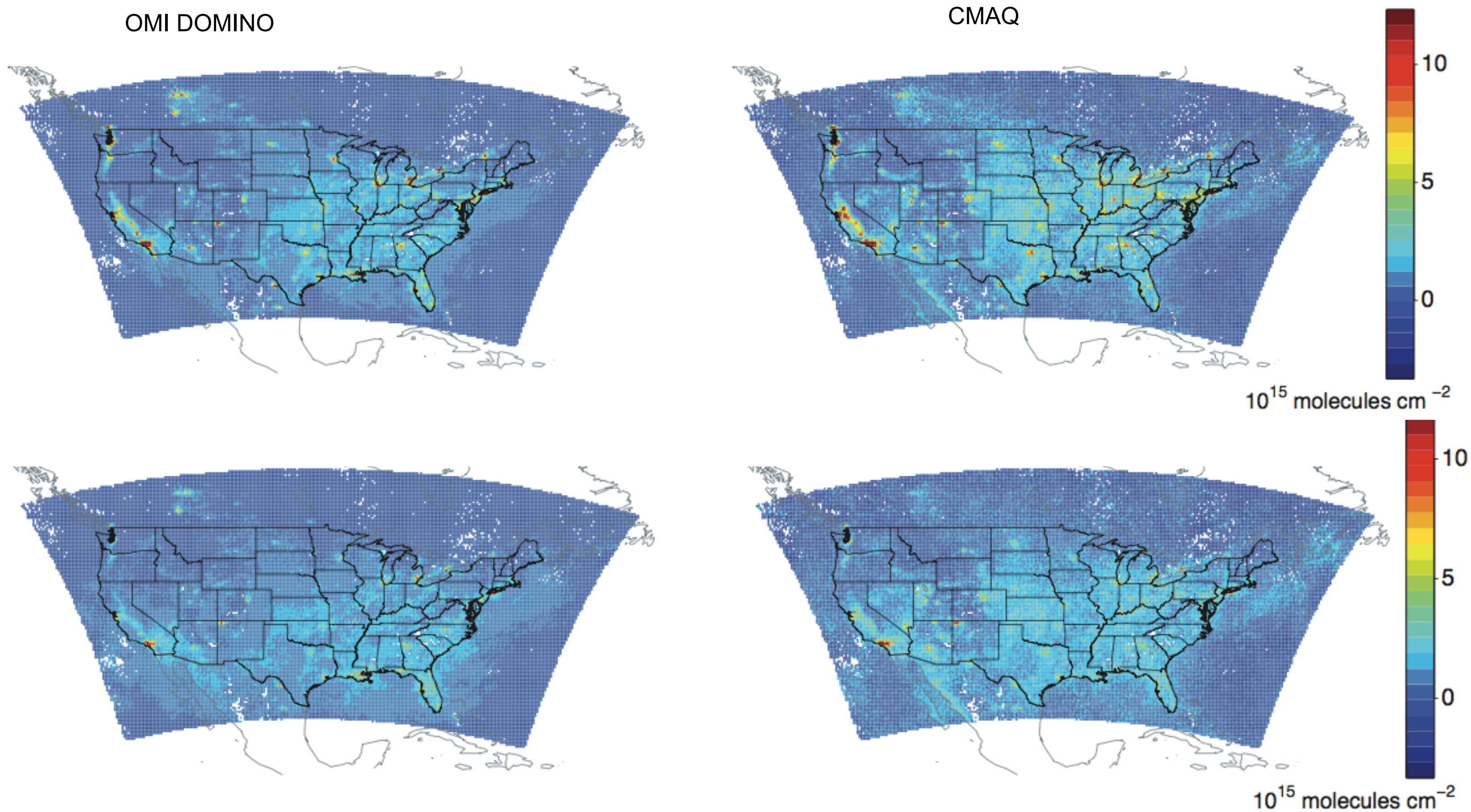
Zhu et al., 2013

New scheme tested in National Emission Inventory at EPA

Online observation operator for assimilating NO_2 into CMAQ using 4D-Var

1. compare vertical columns

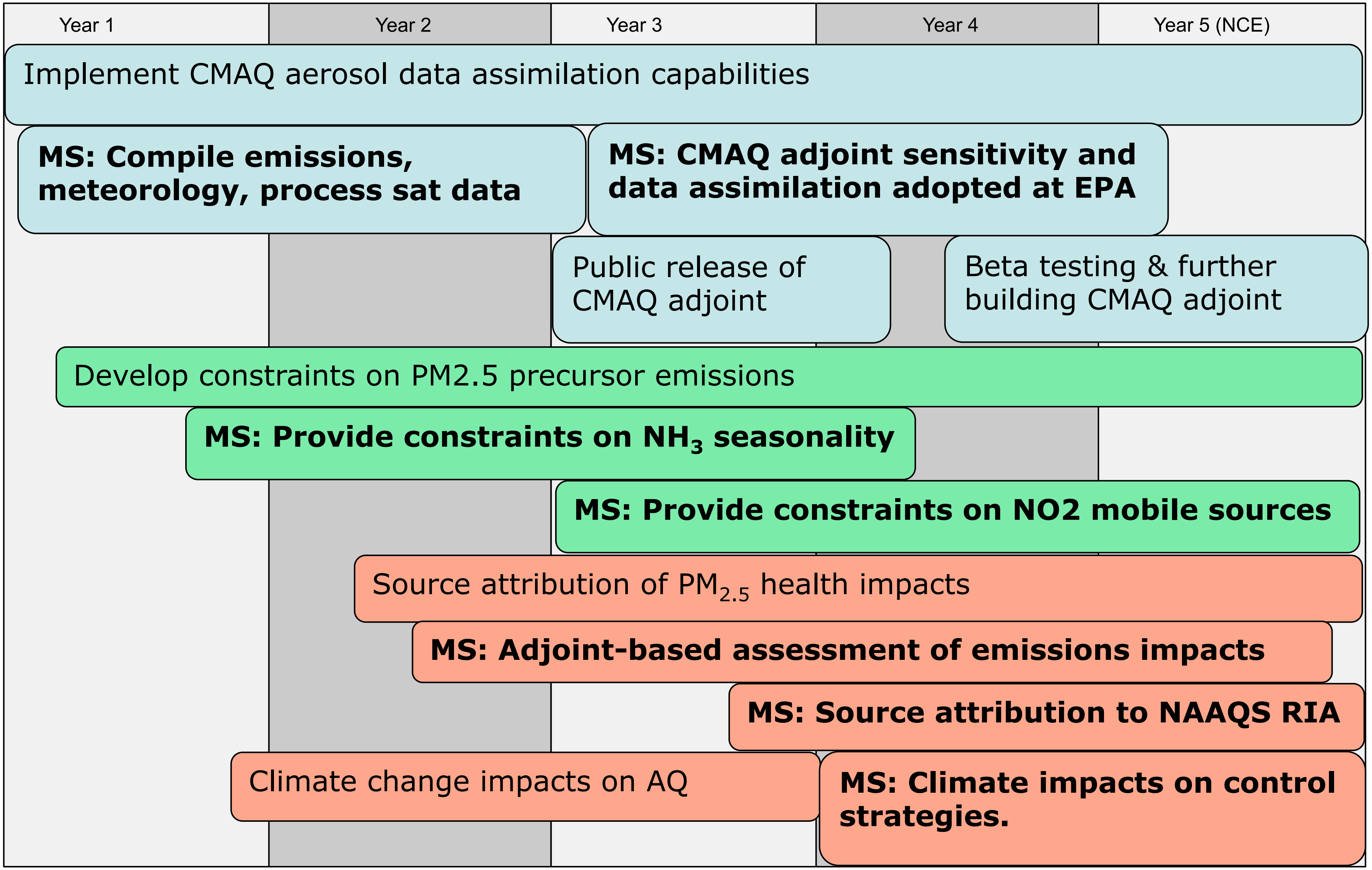
2. compare slant columns



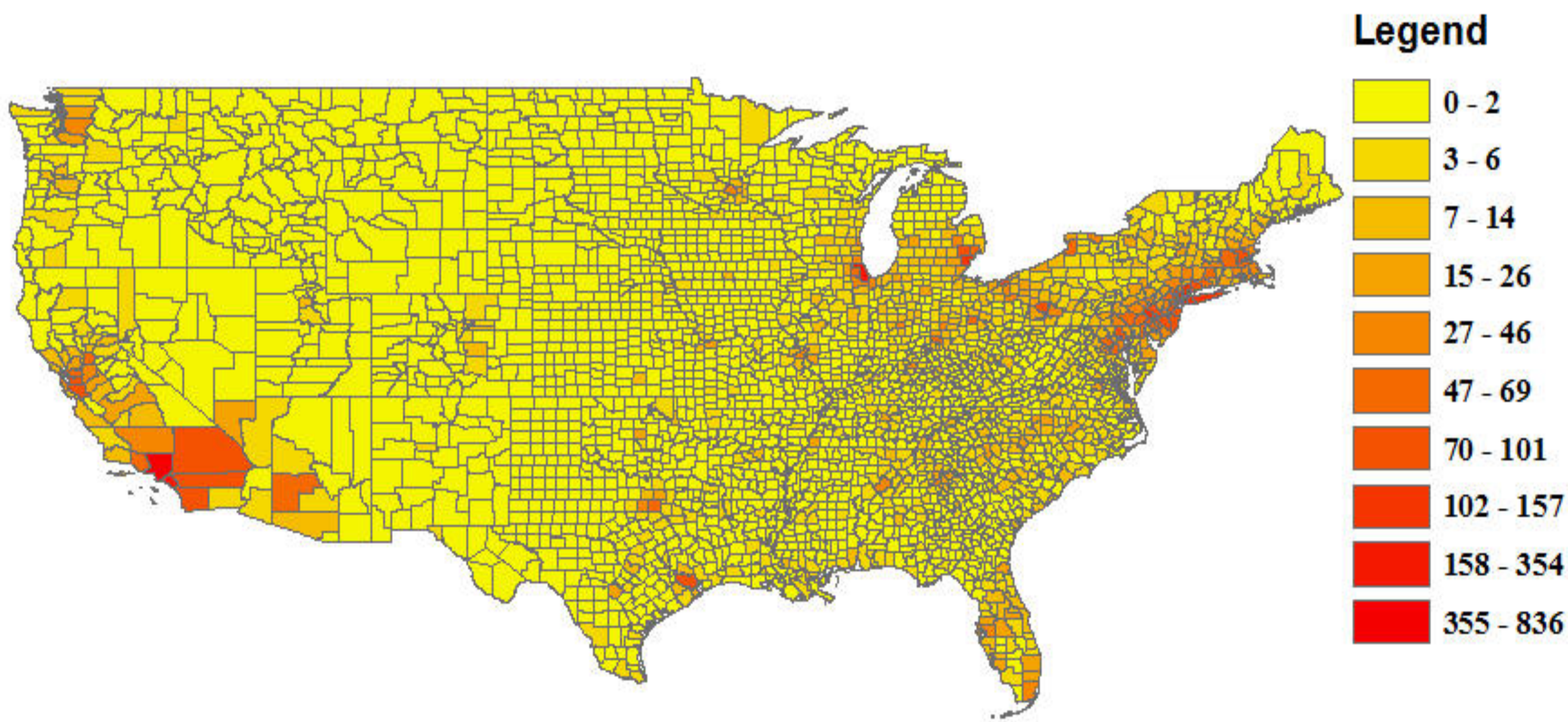
1. Leads to more rigorous slant vertical column comparison
2. Emphasizes altitudes where OMI is most sensitive
3. Facilitates eventual remote sensing assimilation in CMAQ

Pye et al., 2011

Project timeline and milestones



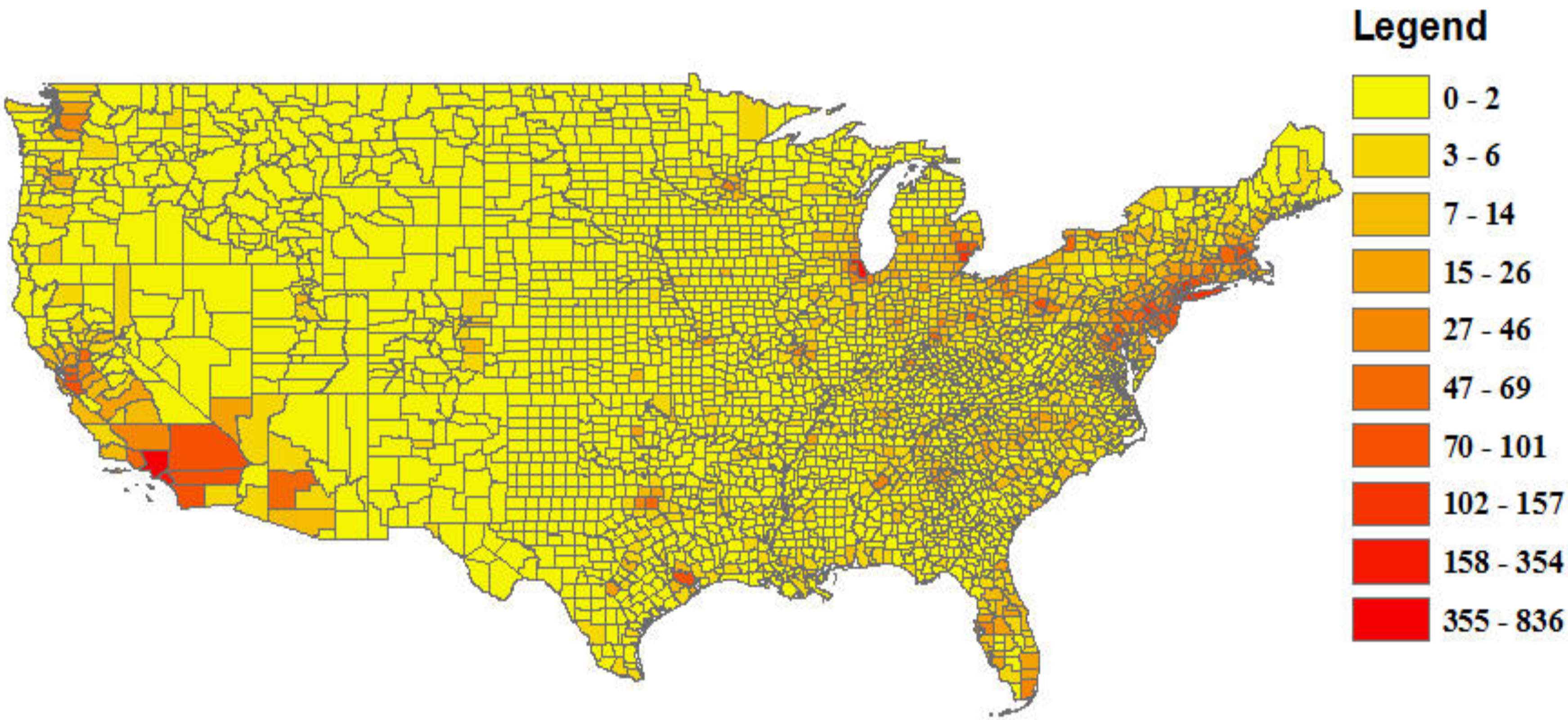
Health impacts of PM2.5: mortality associated with black carbon (BC) in the US



National mortality related to BC exposure: Using the all-cause mortality risk estimate for $PM_{2.5}$ among adults aged over 30 drawn from the Krewski et al (2009), we estimate (BenMAP) approximately 14,000 BC-related premature deaths to result from 2010 air quality levels.

Ying Li, Columbia U.

Health impacts of PM2.5: mortality associated with black carbon (BC) in the US

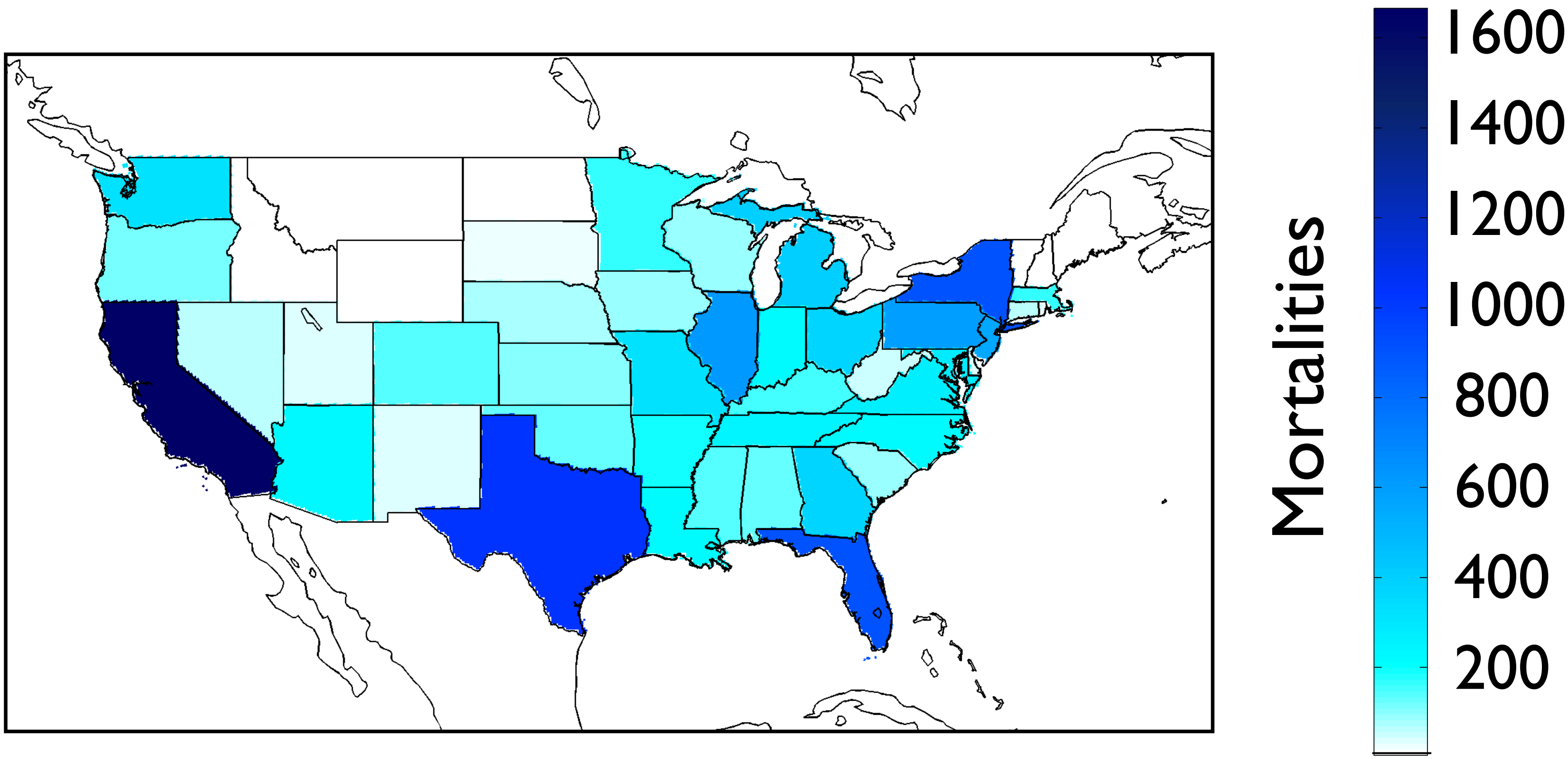


National mortality related to BC exposure: Which emissions contribute the most to this health burden?

Ying Li, Columbia U.

Health impacts of PM2.5: mortality associated with black carbon (BC) in the US

Adjoint estimate of BC source contributions (CMAQ 12 km)



Preliminary results based on June, Matt Turner CU Boulder

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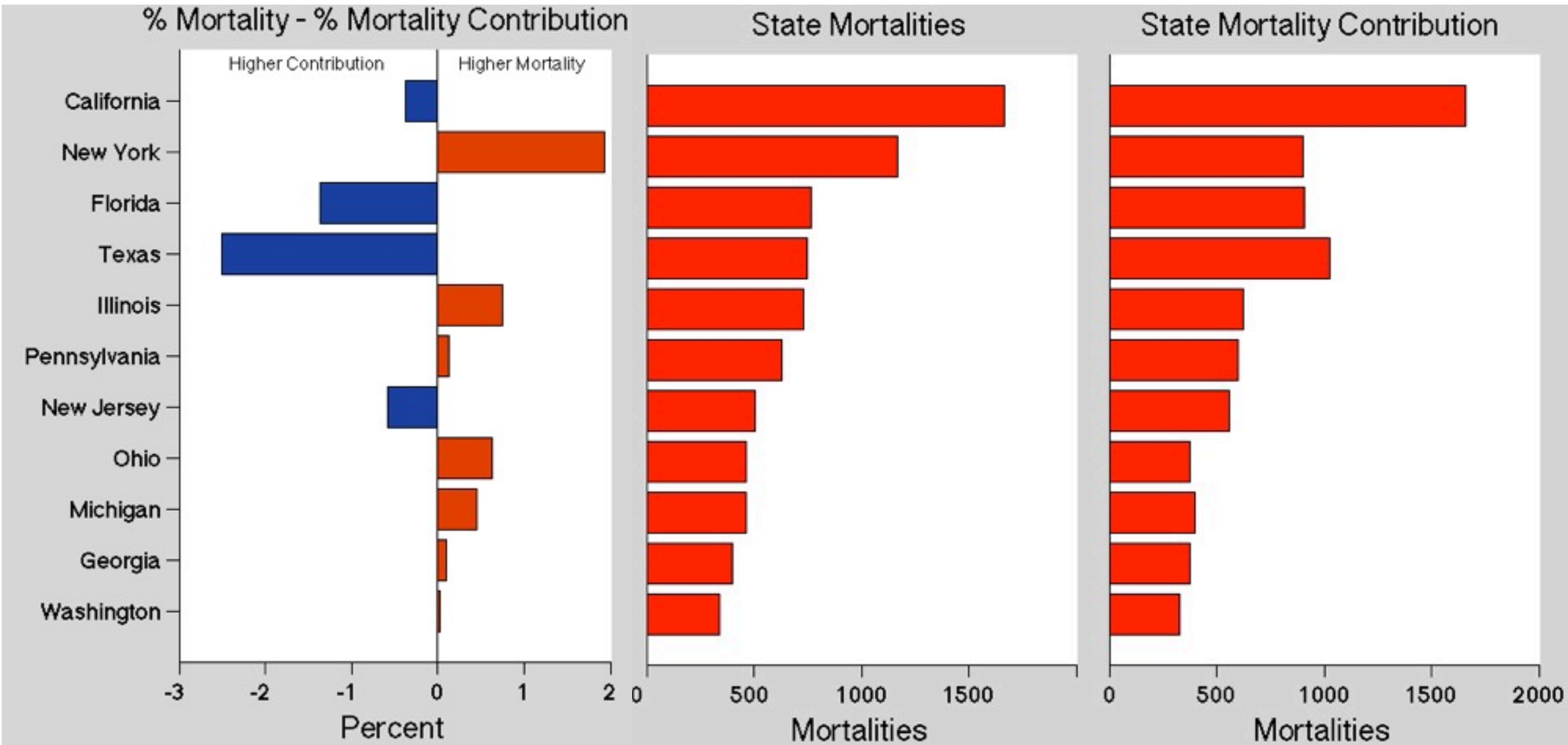

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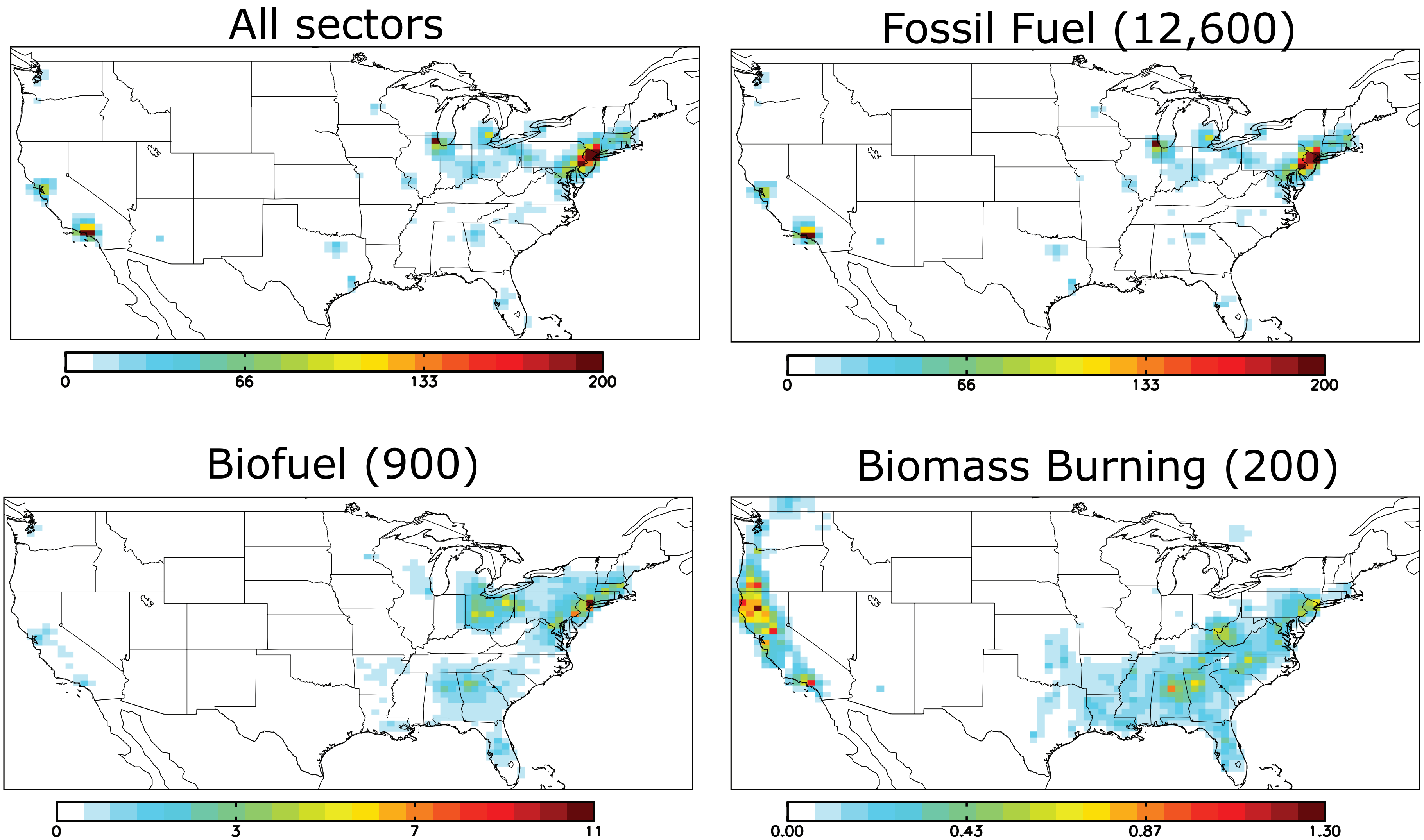
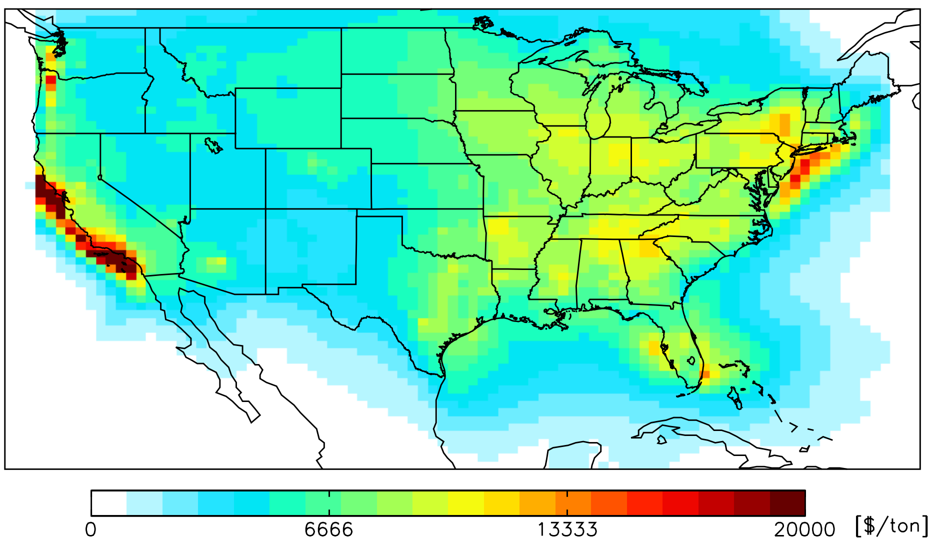


Annual mortalities from BC emissions in each grid cell

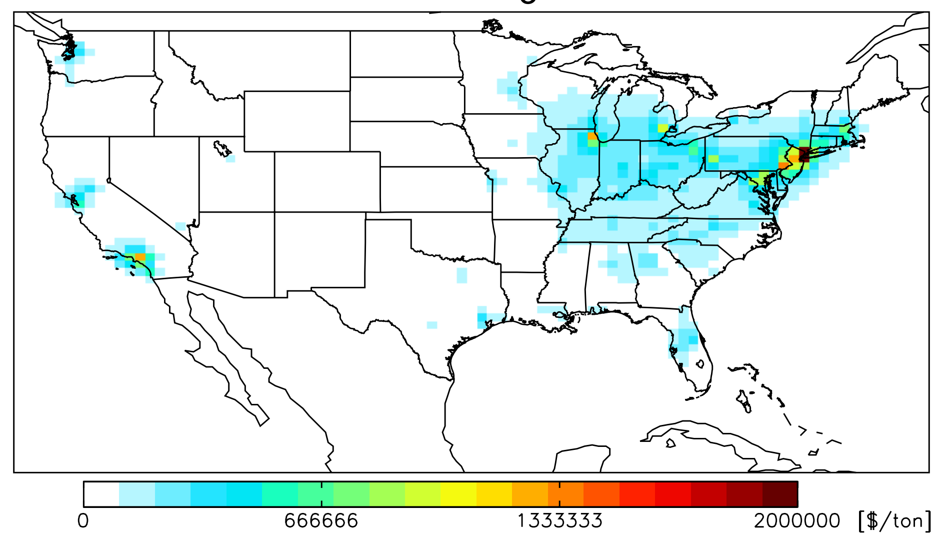
Health impacts of PM2.5: mortality costs of SO2 and NH3 sources in the US

Adjoint estimate of BC source contributions (GEOS-Chem ½° x 2/3°)

Surface SO_2



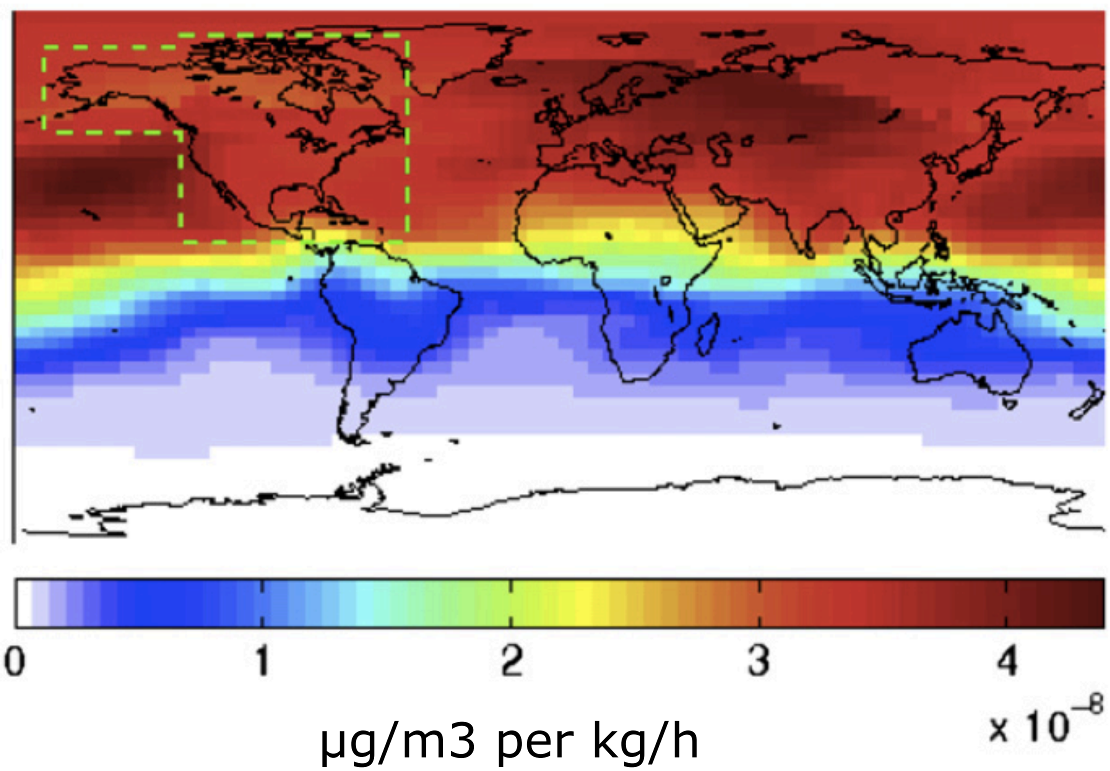
NH_3



- overall risk compare well to 130,000 / yr from Fann et al. (2012)
- mean $/ton benefits similar to Fann et al. (2009) for SO2
- much larger for NH3

Source attribution of population-weighted PM concentrations

- Impacts of cruise-altitude NOx emissions on NA surface PM



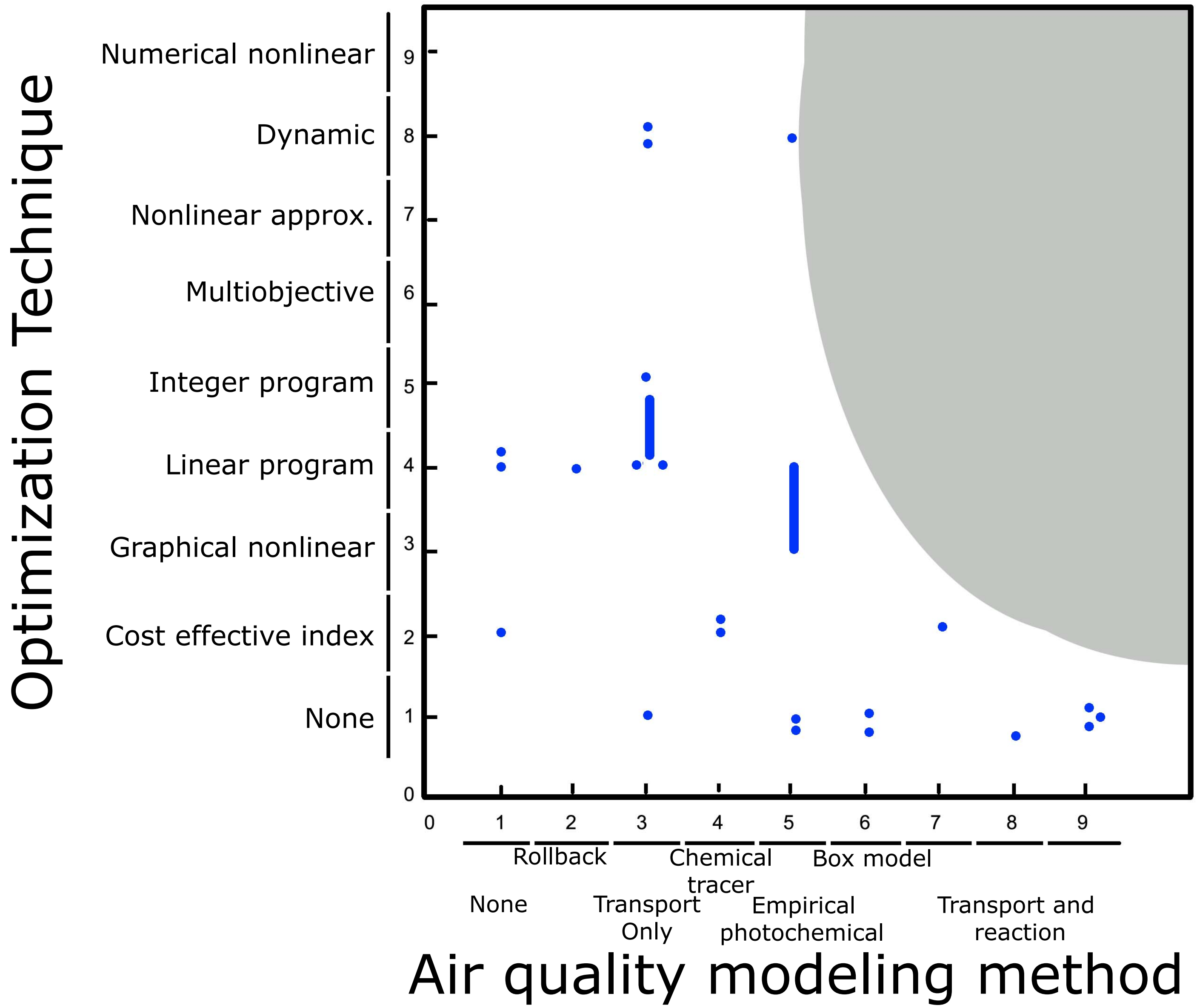
Impact by location:
- 1 kg NOx @ 9 – 12km

Total global impact on NA mortality:
- 320 (120, 580) deaths

Koo et al., *AE*, 2013

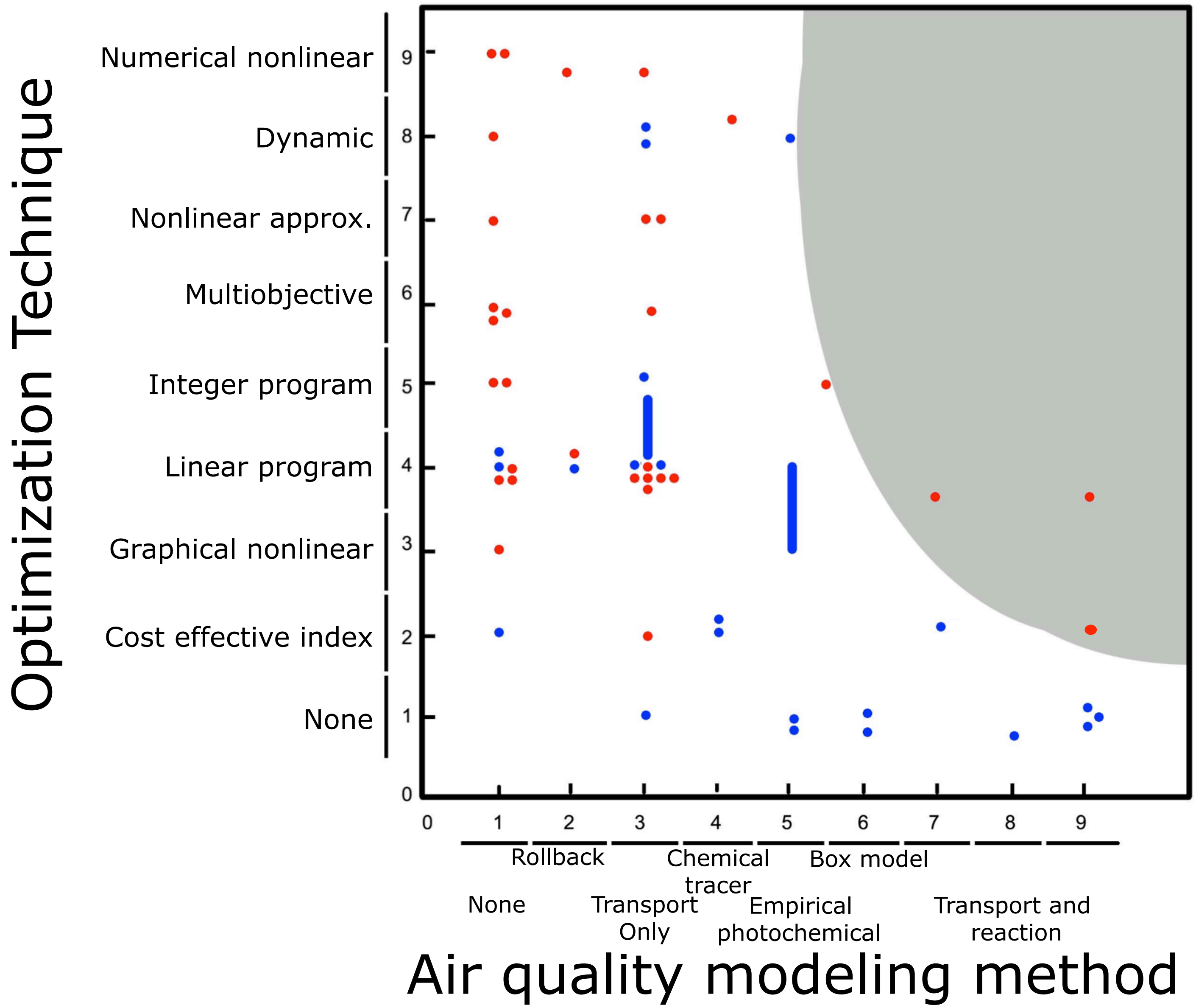
- Impacts of NOx vs SO2 vs NH3 emissions on PM2.5 concentrations in China and India (Kharol et al., *GRL*, 2013)
  - NOx plays a persistent role throughout the year

Optimal AQ control strategy design: past



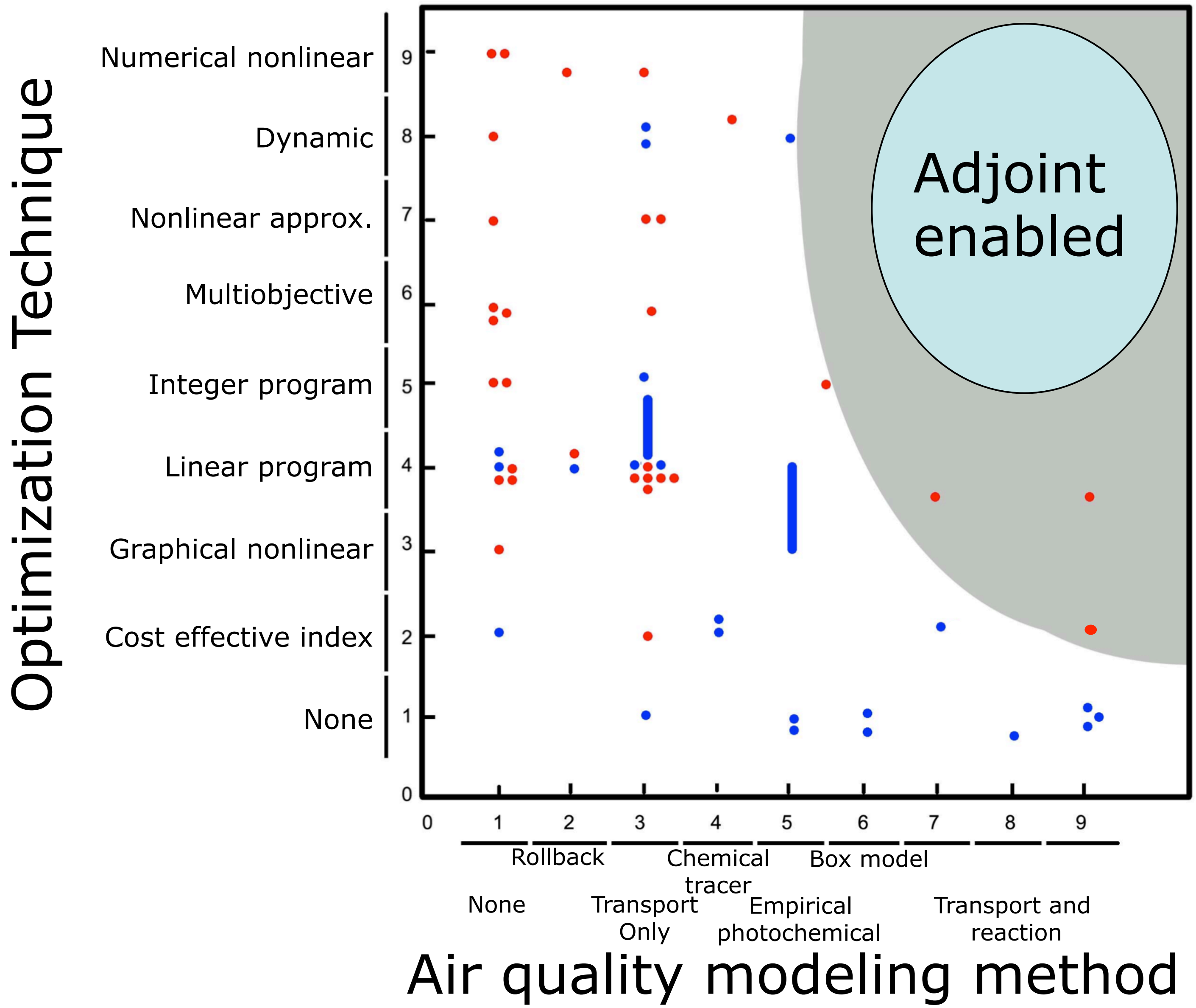
McRae and Cass (1981)

Optimal AQ control strategy design: present



Mesbah and Hakami (2011)

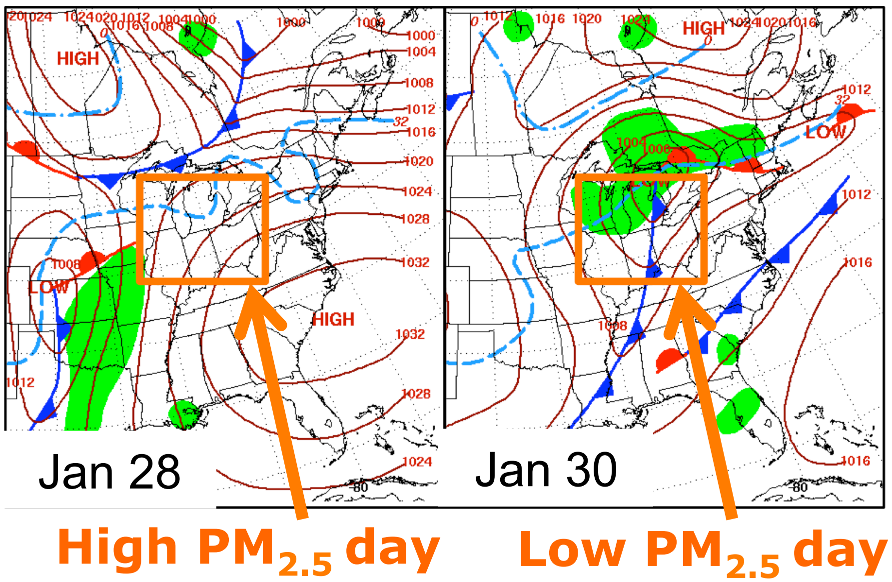
Optimal AQ control strategy design: future



Mesbah and Hakami (2011)

Rapid calculation of $PM_{2.5}$ response to 2000-2050 climate change in a suite of climate models

Step 1: Use principal component analysis (PCA) of to identify the dominant meteorological modes driving day-to-day $PM_{2.5}$ variability by region



Example of the meteorological conditions driving $PM_{2.5}$ in Midwest on two winter days in 2006.

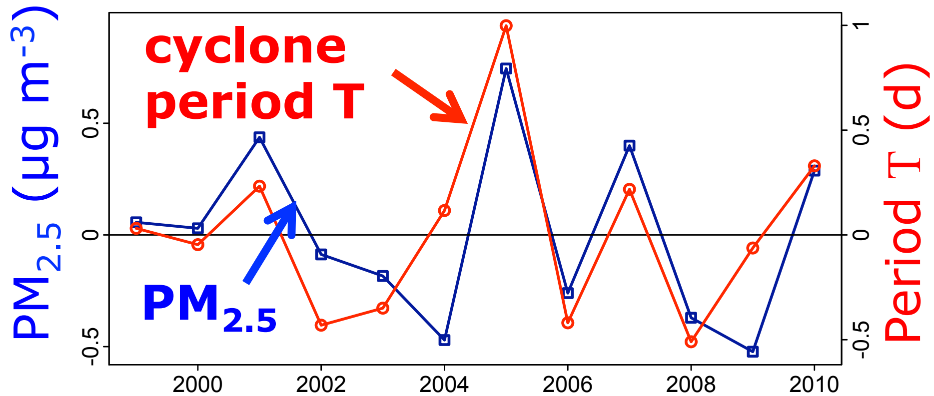
- High PM2.5 on warm, dry day at tail end of anticyclone.
- Low PM2.5 when cold front brings cool, moist air.

Main transport modes controlling $PM_{2.5}$ in the United States:

- Eastern US: mid-latitude cyclone and cold front passage
- Pacific coast: synoptic-scale maritime inflow

Tai et al., 2012

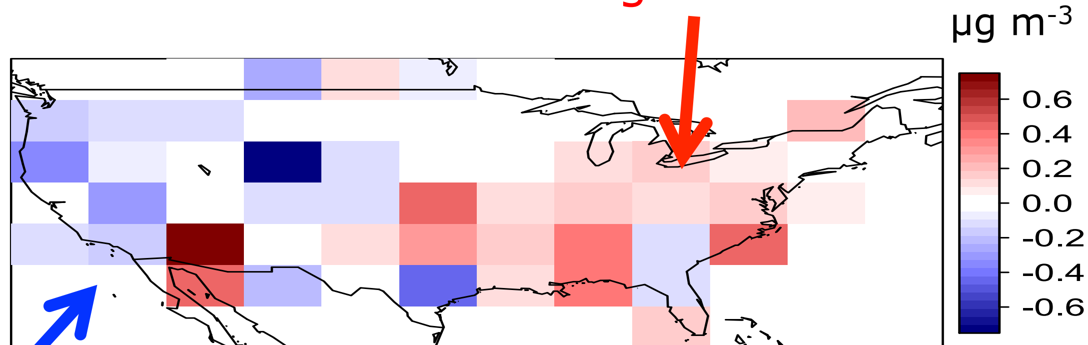
Step 2. Identify the dominant meteorological mode in each region whose mean period T is most strongly correlated with annual mean $PM_{2.5}$.



Example: Anomalies of the annual mean $PM_{2.5}$ and period of cyclone passage for US Midwest

Climate penalty:
Increased stagnation

Step 3. Diagnose trends in these meteorological modes in an ensemble of climate output for 2000 to 2050. Apply observed $PM_{2.5}$ sensitivities ($dPM_{2.5}/dT$) to these trends.

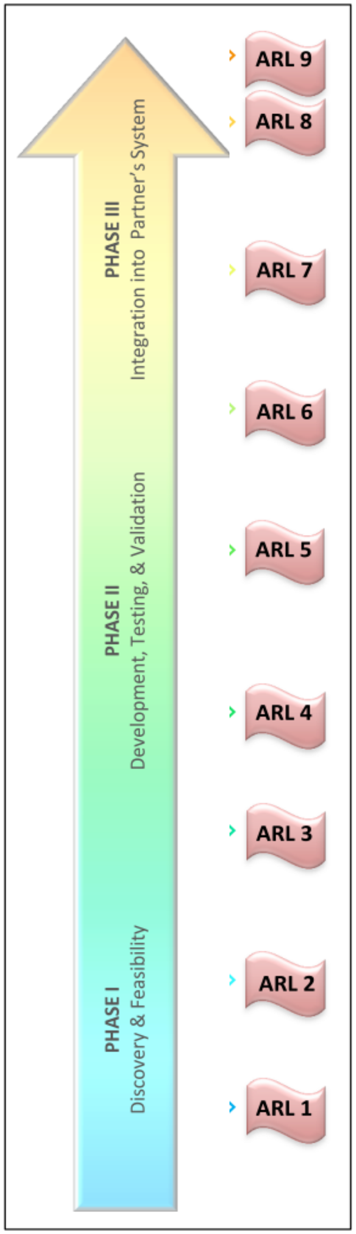


2000-2050 change in annual mean $PM_{2.5}$ diagnosed from a suite of climate models

Climate benefit:
Increased maritime flow

Tai et al., 2012

Current and projected ARL

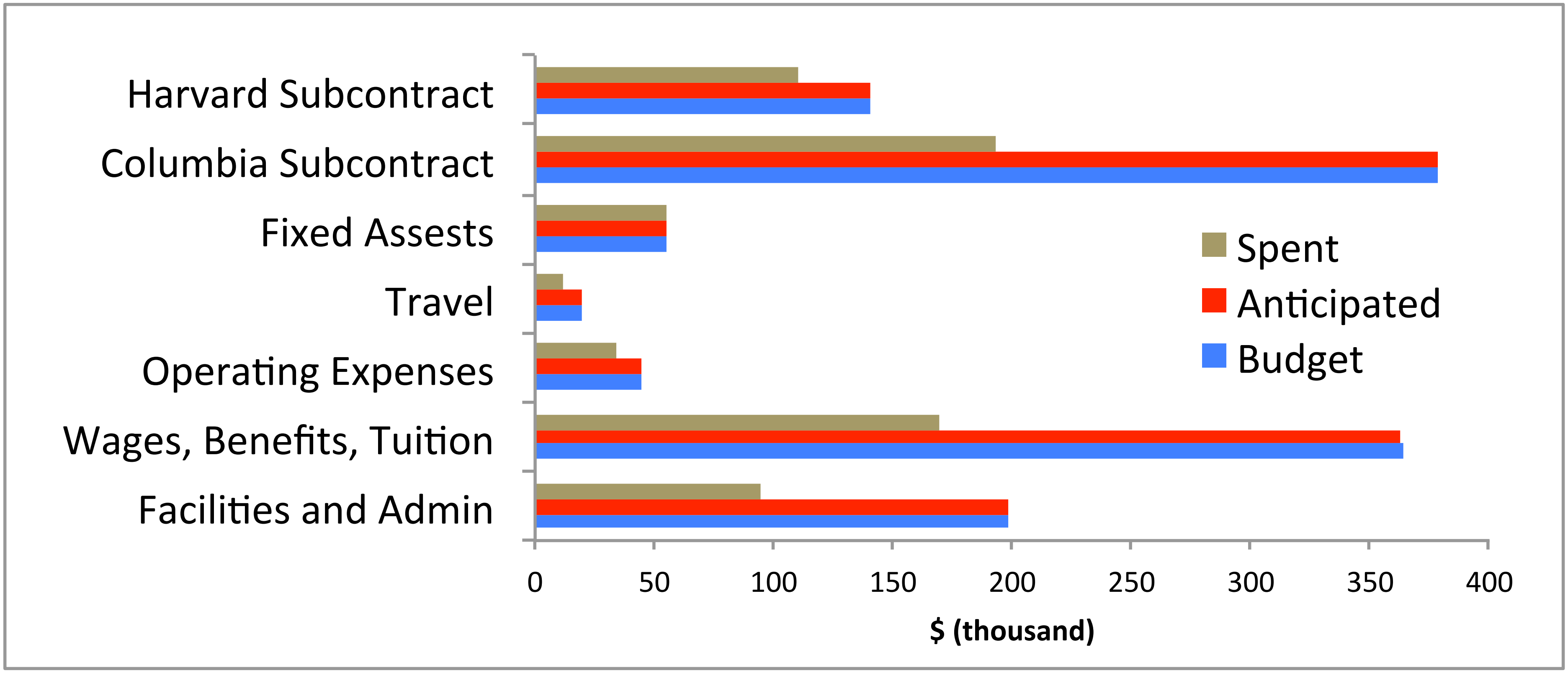


- Start of project ARL = 1 *(08/01/2009)*
- Current ARL = 6
  - Nested GEOS-Chem adjoint applied to estimate PM25 health related damages per ton of emission for each species, sector and grid-cell. Species-specific health impacts are being assessed at the county level for the year 2008.
  - Health impacts of BC emissions using CMAQ adjoint in progress.
  - GEOS-Chem NH3 assimilation and CMAQ ozone adjoint being used by EPA AMD (Shannon Capps).
- Expected Ending ARL = 8 *(Date)*
  - Emissions constraints implemented in CMAQ and attainment analysis considered in EPA NAAQS review
- Final ARL with no cost extension = 8
  - Will prepare seasonal constraints on NH3 emissions for incorporation into future National Emissions Inventory.

Recent and future milestones

Milestones	Deadline	Team	Status
NO_2 assimilation operator for CMAQ	01/2011	US EPA	Complete
Model input data prepared for CMAQ 12 km analysis	08/2012	CU Boulder / US EPA	Complete
Remote sensing constraints on NH_3	01/2013	CU Boulder	Complete
Adjoint-enabled satellite data assimilation and attainment analysis adopted at EPA	01/2013	US EPA	Complete
Forward model evaluation of aerosol health impacts	08/2013	Columbia U. / CU Boulder	Complete
Aerosol health impacts / attainment applied to PM2.5 NAAQS	08/2014	Columbia U. / CU Boulder	NCE
NO_2 inverse modeling constraints in CMAQ	07/2014	CU Boulder	NCE
Climate impacts on PM2.5 attainment	08/2014	Harvard / CU Boulder	NCE

Obligations and cost status



- Harvard subcontract nearly completed
- Columbia subcontract will be exhausted by Feb, 2014; funding to support postdoc Ying Li , Co-PI Pak Kinney and Co-I Darby Jack
- CU Boulder funding proceeding for support of graduate students and PI. Facilities, Admin, and Op expenses track personnel expenditures.

Risks and Issues

- Management challenges
  - Challenge to hire / maintain personnel at EPA
    - Postdoc Shannon Capps there for 9 months using CMAQ adjoint & TES NH3
  - Took time to hire postdocs / students in Henze group
    - After slow start, group now tackling all project areas.
- Technical risk
  - CMAQ adjoint still not stable
    - Complete inversions / attainment with GEOS-Chem; use CMAQ forward model for downscaling results.
  - TES NH3 data challenges owing to sparse sampling
    - Consider IASI or AIRS products, which have poorer signal/noise but wider coverage
  - TES/OMI lifetime is limited
    - Consider CrIS, TROPOMI, GEO-CAPE for future remote sensing constraints on emissions
  - Driving GEOS-Chem with high resolution GISS climatology difficult
- Schedule risk: Low to medium
  - NAAQS review process at EPA a moving target

Publications and Presentations

Turner, M., D. K. Henze, A. Hakami, S. Zhao, J. Resler, G. Carmichael, C. Stanier, J. Baek, P. Saide, A. Sandu, A. Russell, G. Jeong, A. Nenes, S. Capps, P. Percell, R. W. Pinder, S. Napelenok, H. Pye, J. O. Bash, T. Chai, D. Byun, Aerosols Processes in the CMAQ Adjoint, International Aerosol Modeling Algorithms Conference, Davis, CA, Nov 30 - Dec 2, 2011a.

Turner, M., D. K. Henze, A. Hakami, S. Zhao, J. Resler, G. Carmichael, C. Stanier, J. Baek, P. Saide, A. Sandu, A. Russell, G. Jeong, A. Nenes, S. Capps, P. Percell, R. W. Pinder, S. Napelenok, H. Pye, J. O. Bash, T. Chai, D. Byun, High Resolution Source Attribution of PM Health Impacts with the CMAQ Adjoint Model, 10th Annual CMAS Conference, Chapel Hill, NC, Oct 24-26, 2011b.

Capps, S. L., D. K. Henze, A. G. Russell, and A. Nenes, Quantifying relative contributions of global emissions to PM2.5 air quality attainment in the U.S, A54C-07, or presentation, AGU Fall Meeting, San Francisco, CA, 5-9 Dec, 2011. *Best Student Poster Award.*

Pye, H. O. T., S. L. Napelenok, R. W. Pinder, D. K. Henze, R. V. Martin, and K. W. Appel, Evaluation of CMAQ NO2 predictions over the U.S. using ground-based and satellite observations, 5th International GEOS-Chem Conference, Cambridge, MA, May 2-5, 2011.

Li, Y., Henze, D., Jack, D., & Kinney, P. Assessing the national public health burden associ- ated with exposure to ambient black carbon in the United States. Poster. 2013 Conference Environment and Health Bridging South, North, East and West, Basel, Switzerland August 19-23, 2013.

Jiang, Z., D. B. A. Jones, H. M. Worden, M. N. Deeter, D. K. Henze, J. Worden, and K. W. Bowman, C. A. M. Brenninkmeijer, and T. J. Schuck, Impact of model errors in convective transport on CO source estimates inferred from MOPITT CO retrievals, J. Geophys. Res., 118, doi:10.1029/jgrd.50216, 2013.

Koo, J., Q. Wang, D. K. Henze, I. A. Waitz, S.R.H. Barrett, Spatial sensitivities of human health risk to intercontinental and high-altitude pollution, *Atmos. Environ.*, 71, 140-147, 2013.

Capps, S. L., D. K. Henze, A. Hakami, A. G. Russell, and A. Nenes, ANISORROPIA: the adjoint of the aerosol thermodynamic model ISORROPIA, *Atmos. Chem. Phys.,* 12, 527-543, 2012.

Turner, A., D. K. Henze, R. V. Martin, and A. Hakami, Modeled source influences on column concentrations of short-lived species, *Geophys. Res. Lett., 39,* L12806, doi:10.1029/2012GL051832, 2012.

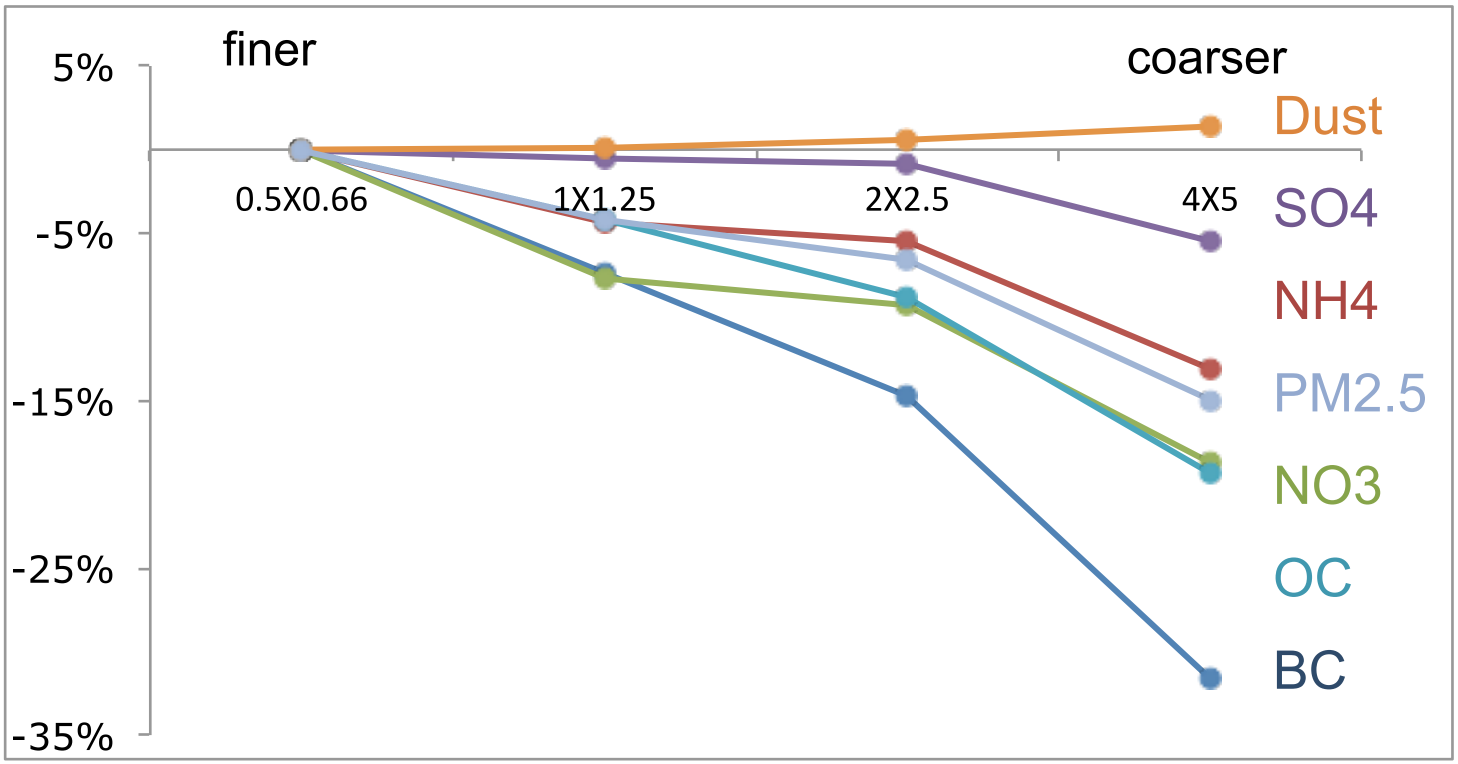
Kharol, S., R. V. Martin, S. Philip, S. Vogel, D. K. Henze, D. Chen, Y. Wang, Q. Zhang, C. L. Heald, Persistent sensitivity of Asian aerosol to emissions of nitrogen oxides, Geophys. Res. Lett., 40, 1021-1026, doi:10.1002/grl.50234, 2013

L. Zhu, D. K. Henze, K. E. Cady-Pereira, M. W. Shephard, M. Luo, R. W. Pinder, J. O. Bash, G. Jeong, Constraining U.S. ammonia emissions using TES remote sensing observations and the GEOS-Chem adjoint model, *submitted to J. Geophys. Res.,* 118, doi:10.1002/jgrd.50166, 2013.

bonus slides!

Health impacts of PM2.5: impacts of black carbon (BC) in the US

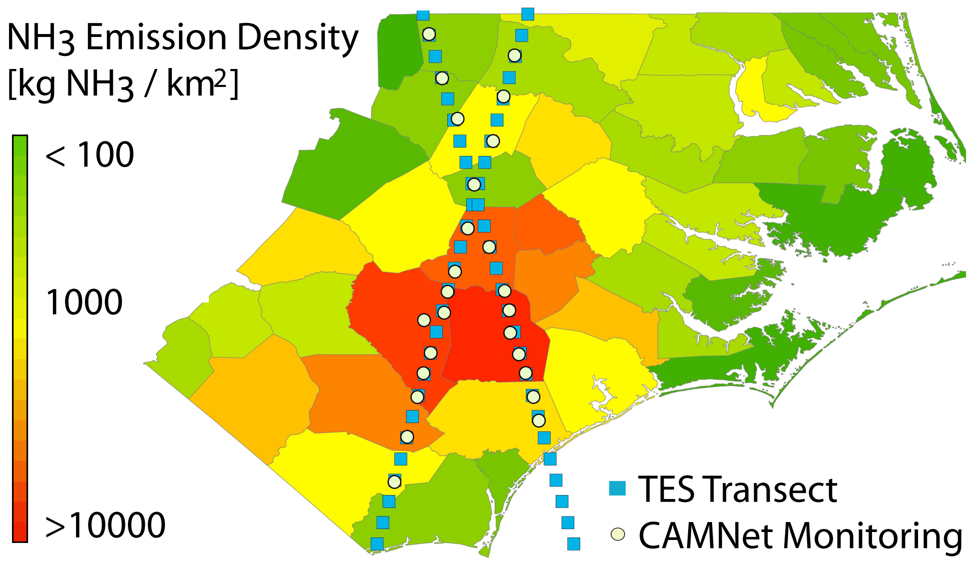
Error in estimating US $PM_{2.5}$ associated mortality relative to finest simulation (Ying Li, Columbia U.):

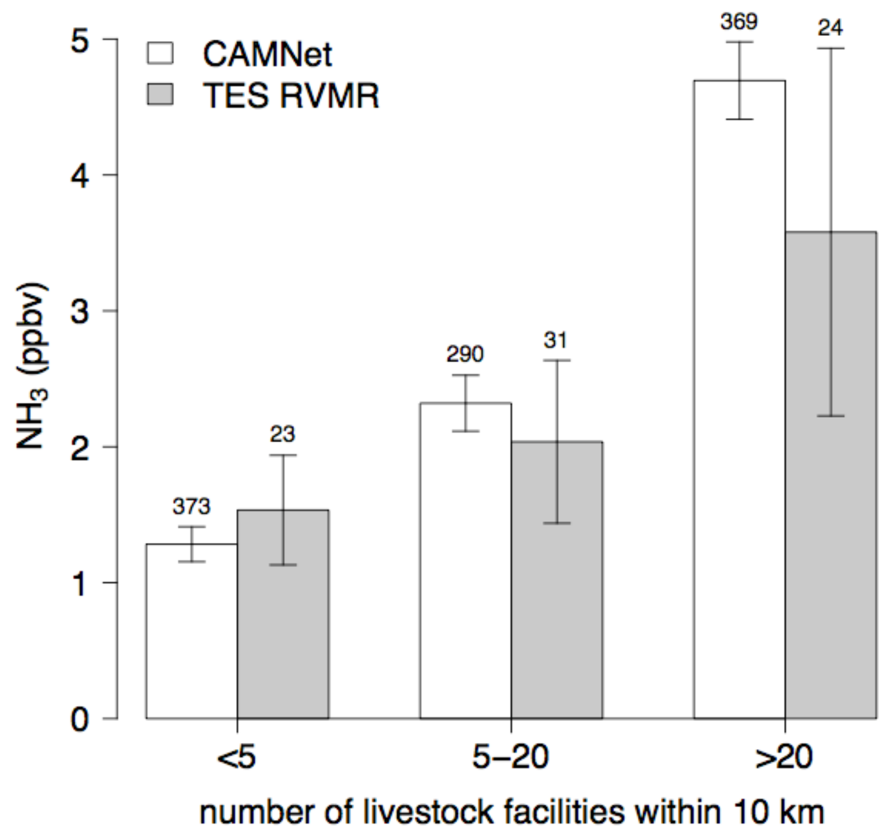


- Greater resolution error for PM2.5 when using threshold (x2 w/5.8 ug/m3)
- Aggregated model values ≠ coarse simulation, but close (<0.3 ug/m3)
- Results similar, but less dramatic, than Punger and West (2013)

Validating TES NH_3 with surface observations

Overlap surface obs with TES Transects for 2009:



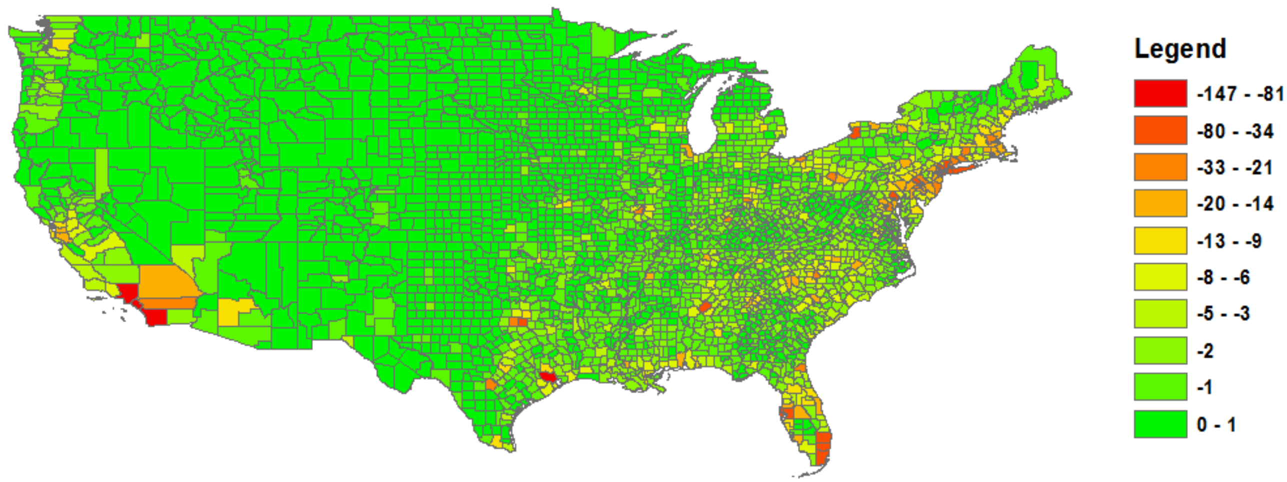


TES reflects real-world spatial gradients and seasonal trends

Pinder et al., 2011

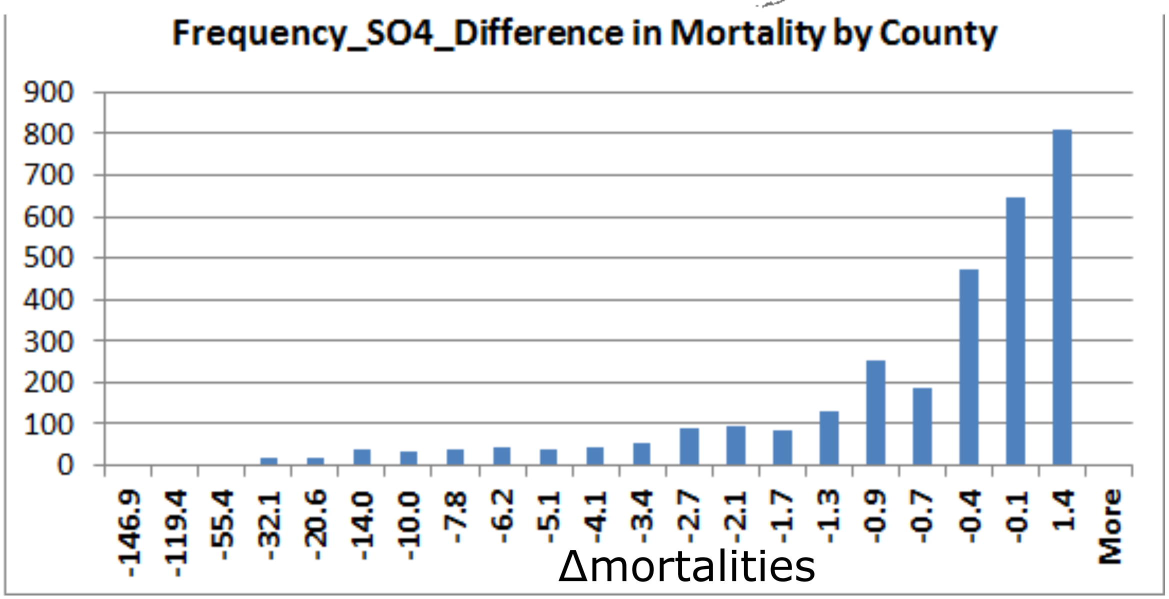
Resolution effects on model estimates of health impacts: US

GEOS-Chem 2x2.5 – GEOS-Chem 0.5 x 0.67: deaths from sulfate



mortalities

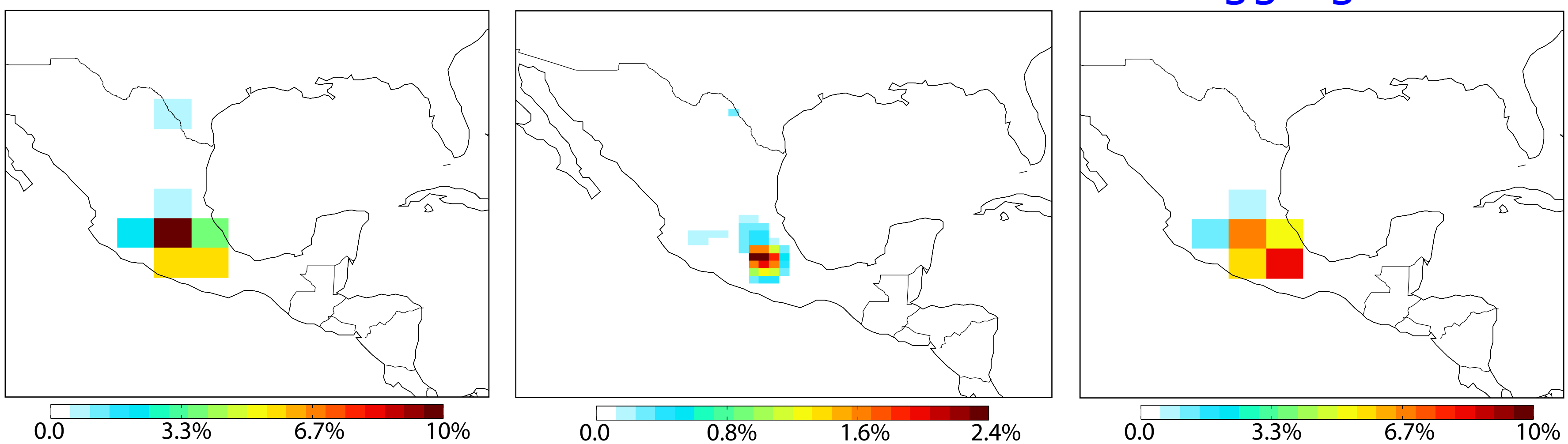
counties



Resolution effects on ***response coefficients***: Mexico

Contribution of SO_2 emissions per grid-box to Mexico's pop-$PM_{2.5}$:

coarse | fine | fine aggregated

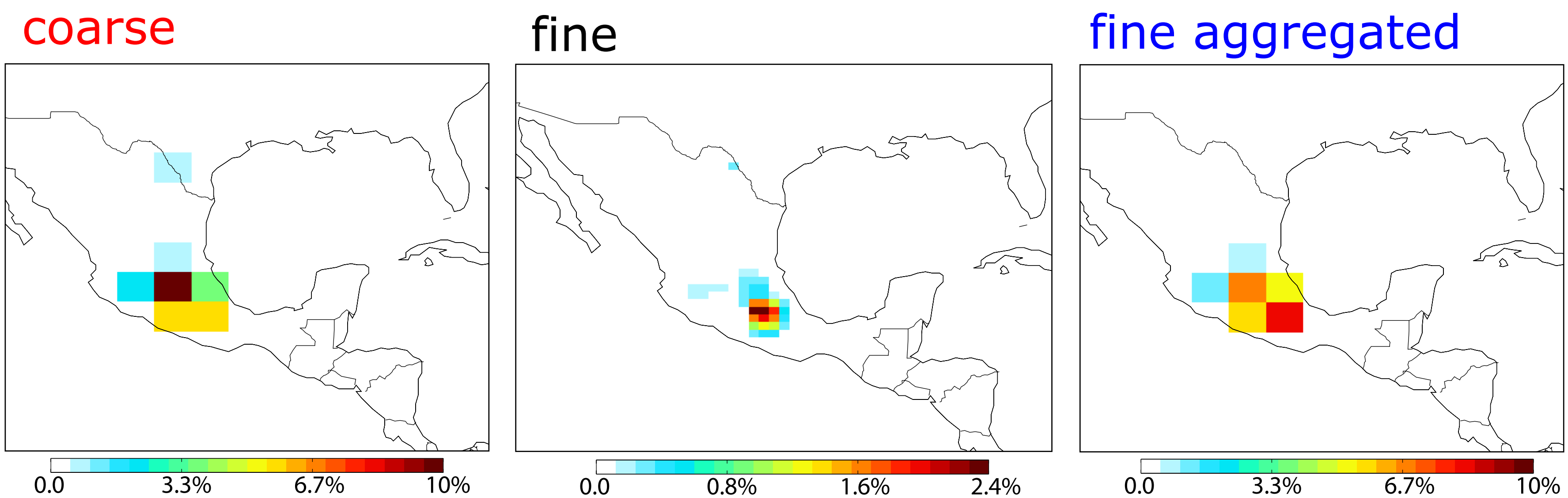


Coarse simulation overestimates impacts of emissions near urban center and underestimates in surrounding areas

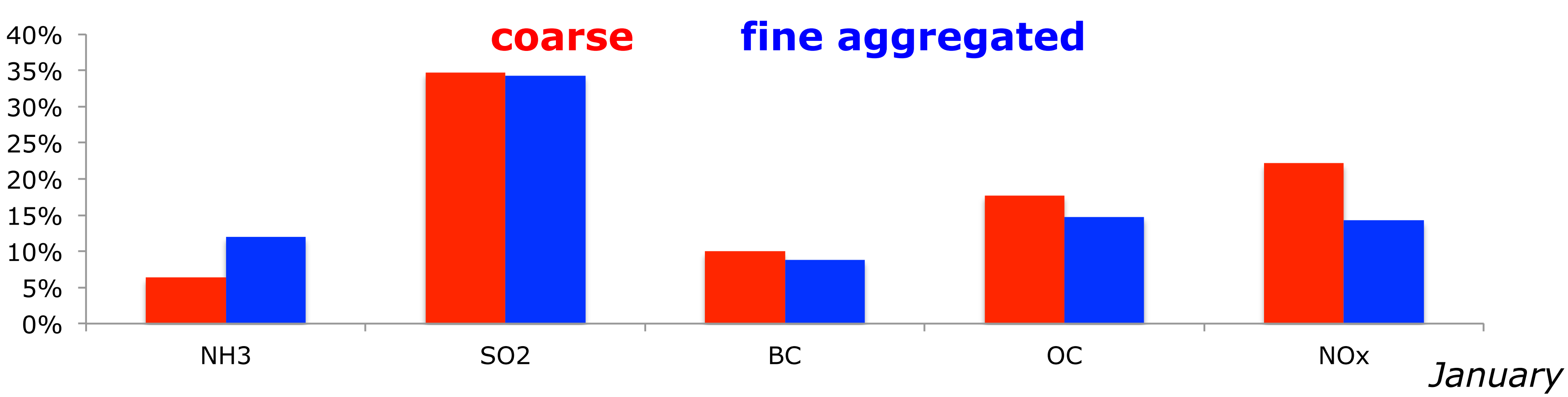
January

Resolution effects on ***response coefficients***: Mexico

Contribution of BC emissions per grid-box to Mexico's pop-$PM_{2.5}$:

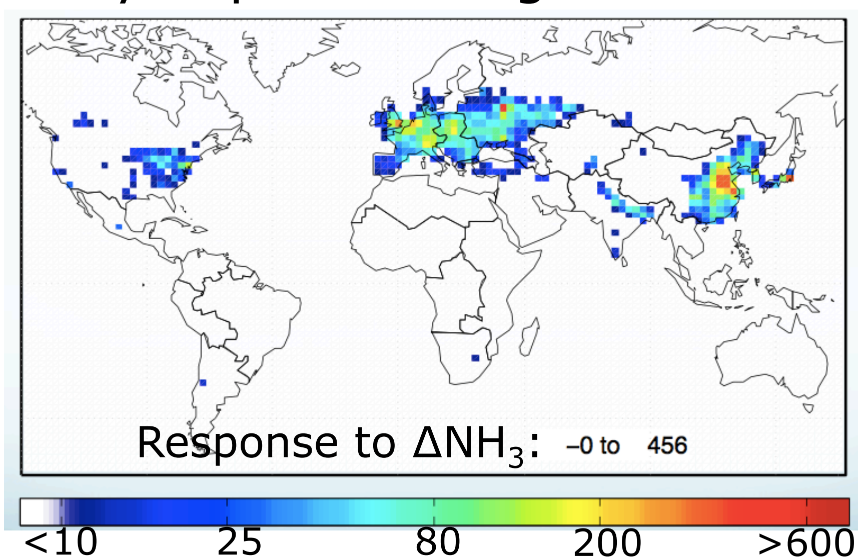


Total contribution of species to % of Mexico's pop-$PM_{2.5}$:



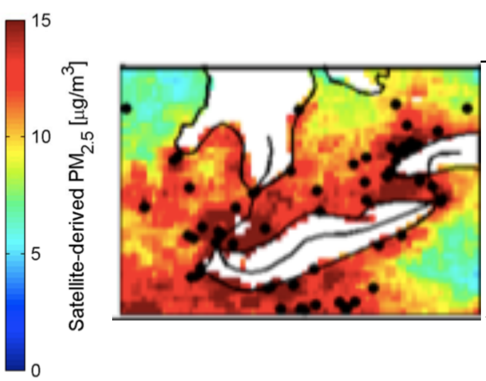
Source attribution of $PM_{2.5}$ related global mortality

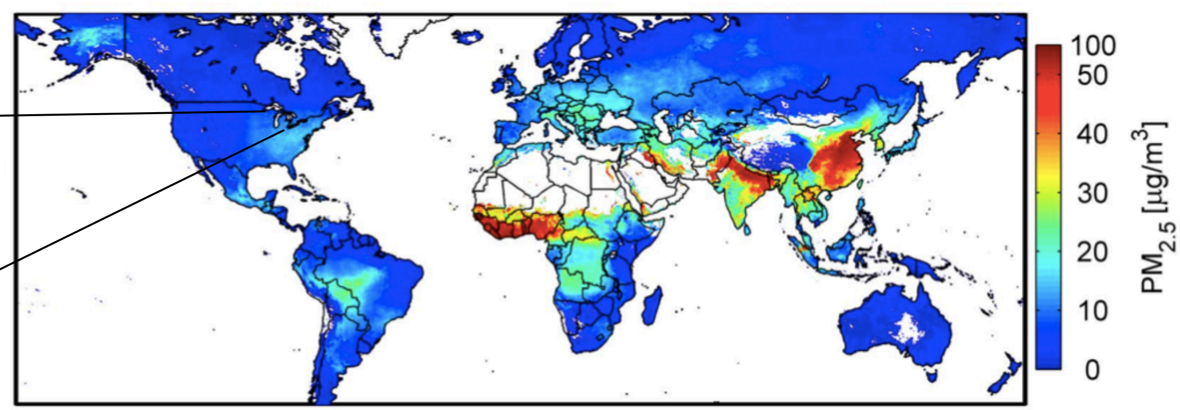
Mortality impacts owing to 10% $PM_{2.5}$ precursor emissions reductions:



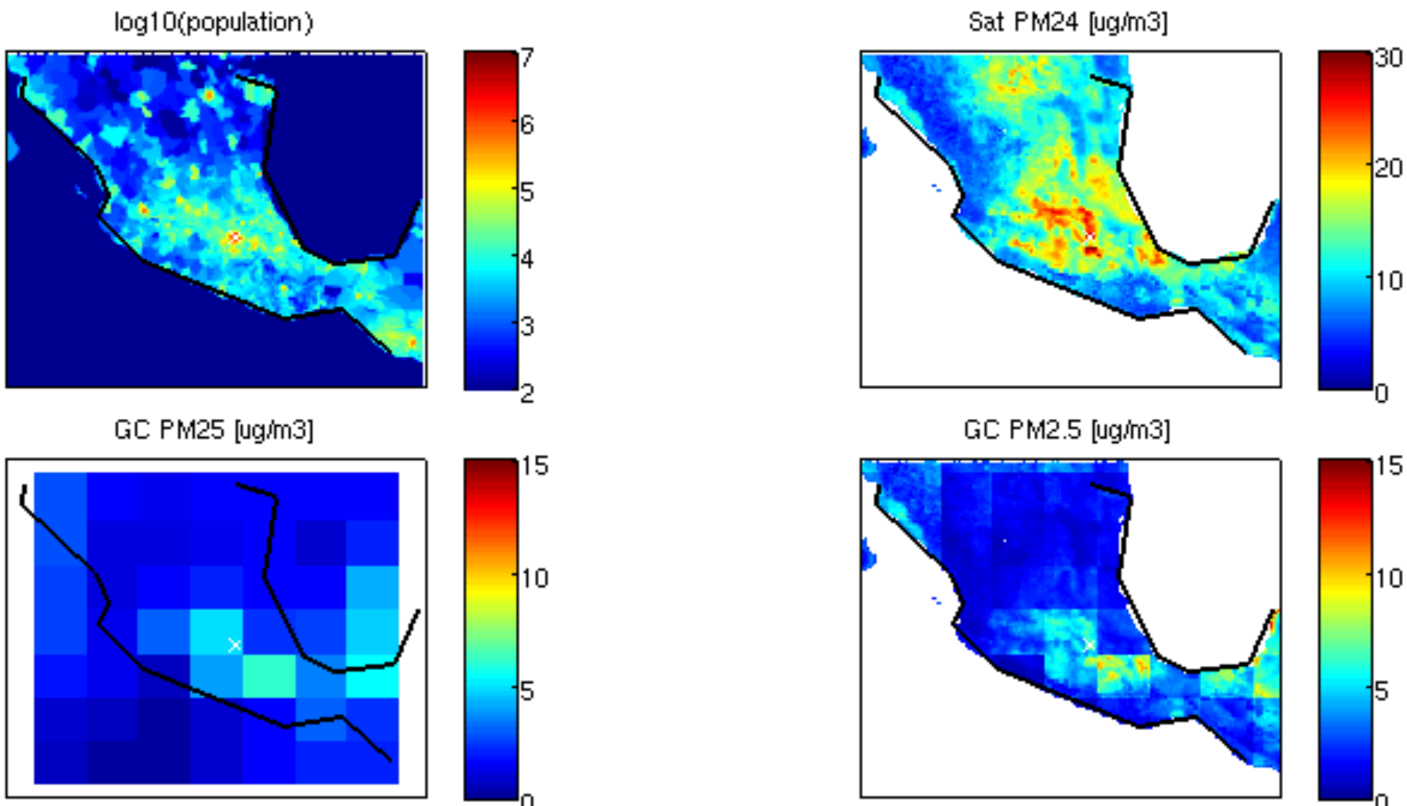
Mortality rates and dose-receptor relationships from Global Burden of Disease, GEOS-Chem adjoint at 2° x 2.5° (Colin Lee, Dalhousie).

$PM_{2.5}$ subgrid variability (0.1° x 0.1°) resolved using MODIS AOD (van Donkelaar et al., 2013)





Impacts of sub-grid variability (MODIS) on SO_2 emissions global mortality response coefficients

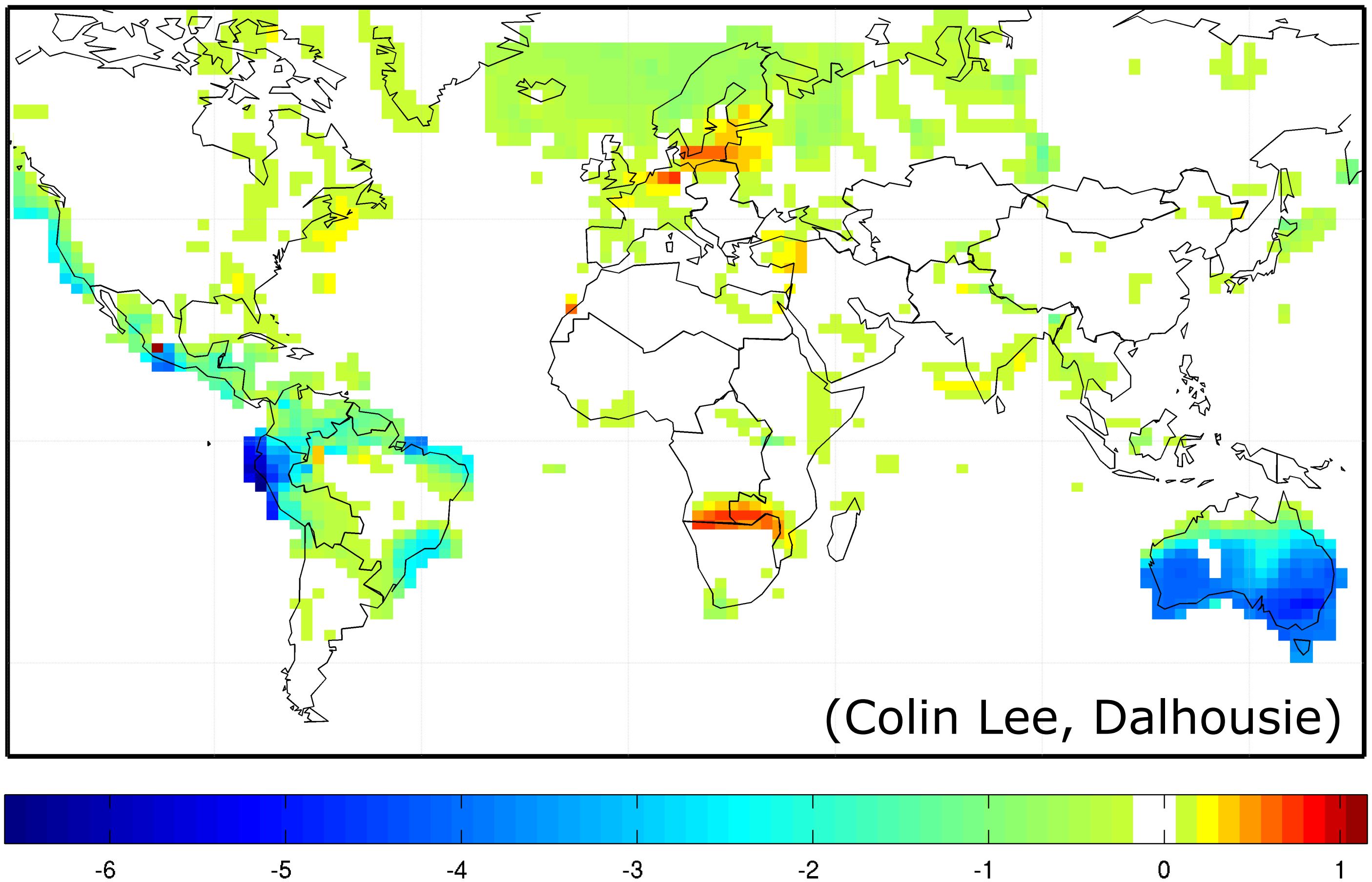


(Colin Lee, Dalhousie)

- GEOS-Chem subgrid variability determined by satellite
- GEOS-Chem values retained
- GEOS-Chem values clearly very low

Impacts of sub-grid variability (MODIS) on SO_2 emissions global mortality response coefficients

$\log_{10}$(with downscaling / without)

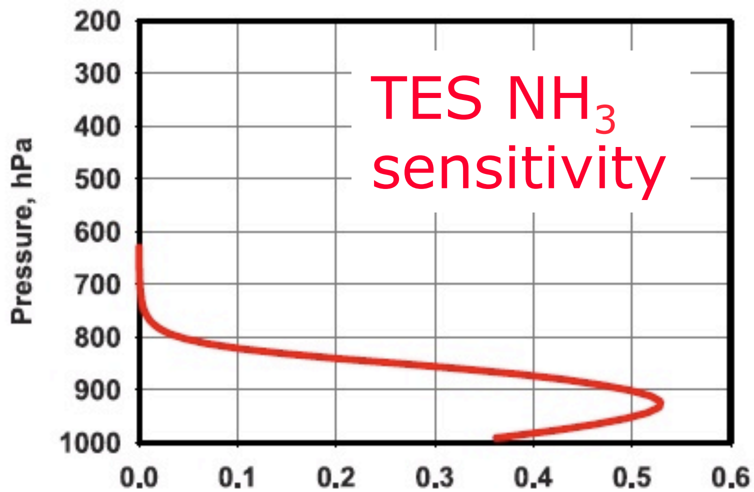


Impacts of resolution can be dramatic, increasing or decreasing, and depend upon environmental condition.

Remote sensing constraints on NH_3

NH_3 is important for understanding $PM_{2.5}$ and Nr dep but sources are not well constrained.

Remote sensing with TES and IASI:



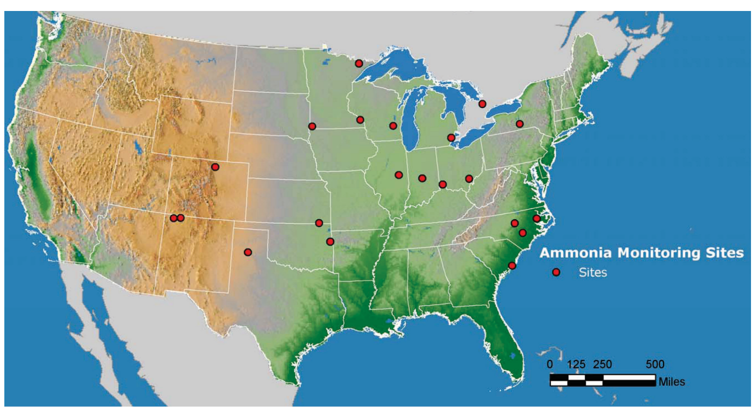
TES:

- sensitive to BL
- detection limit of ~ 1 ppb
- bias of +0.5 ppb

more precise & sparse than IASI

(Beer et al., 2008; Clarisse et al., 2009; Clarisse et al., 2010; Shephard et al., 2011)

Passive surface measurements:

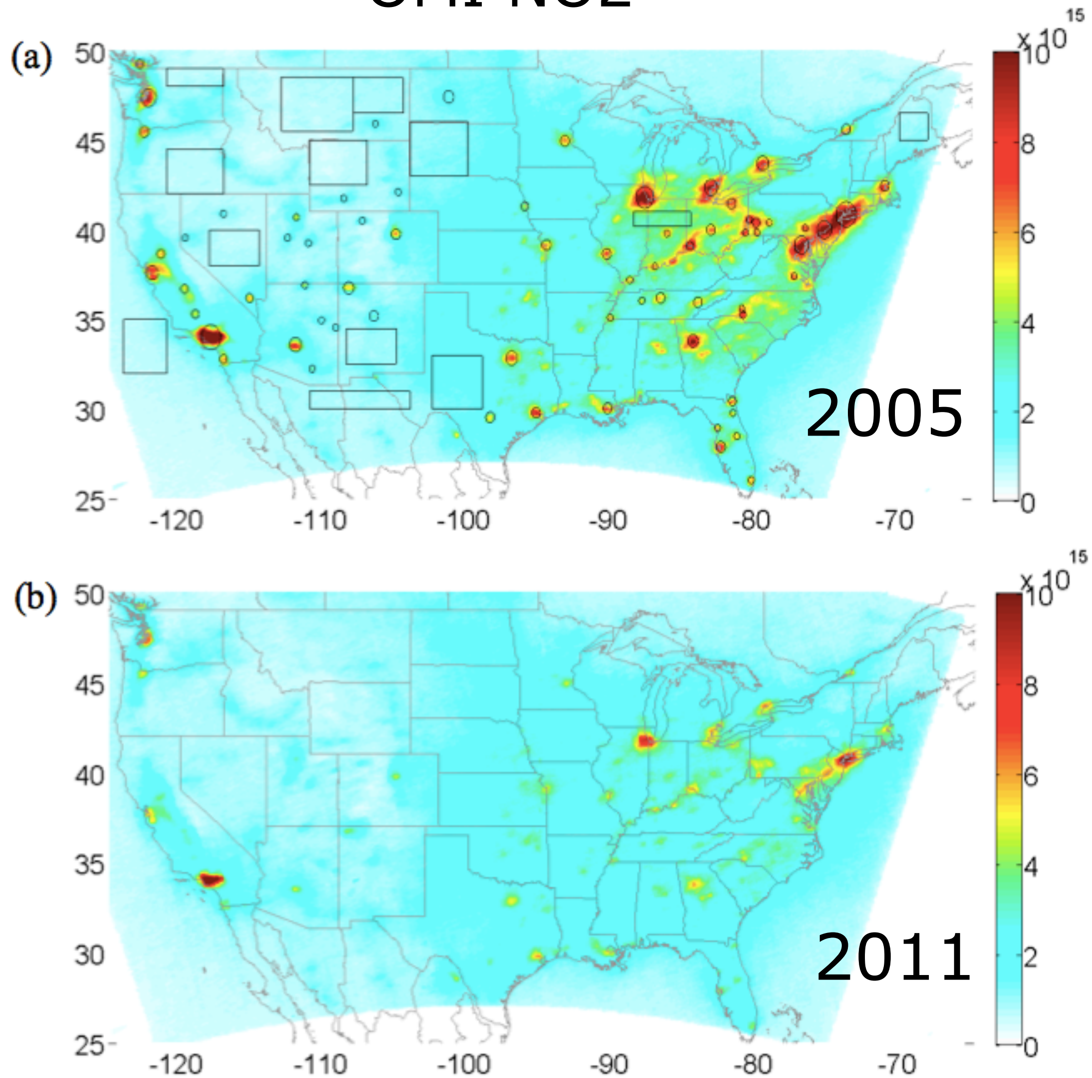


EPA's AMoN sites (>2007)
(Puchalski et al., 2011)

Also LADCO, SEARCH, CSU, ANARChE

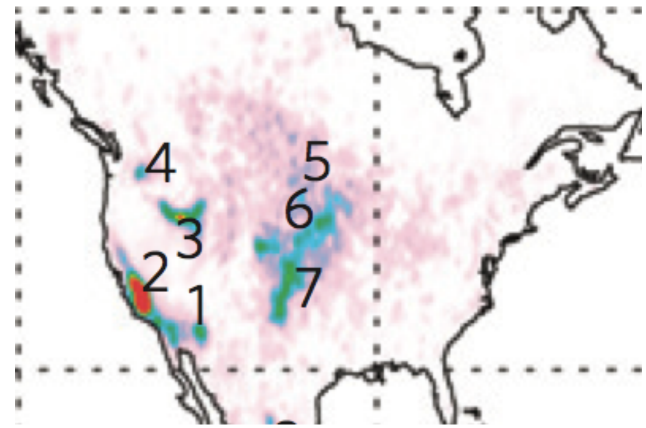
Remote sensing of $PM_{2.5}$ precursors

OMI NO2



Russell et al., 2012

NH_3



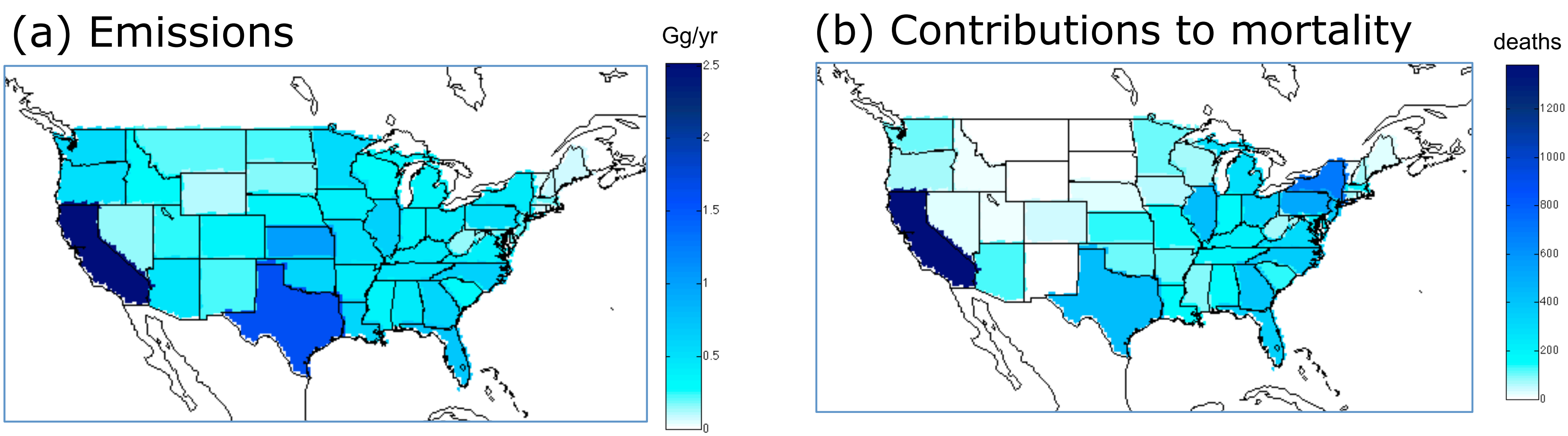
Clarisse et al., 2009

$PM_{2.5}$ AQ model simulations undermined by emissions uncertainties.

How to use remote sensing to constrain emissions?

Milestone #3: Source attribution of aerosol related health impacts

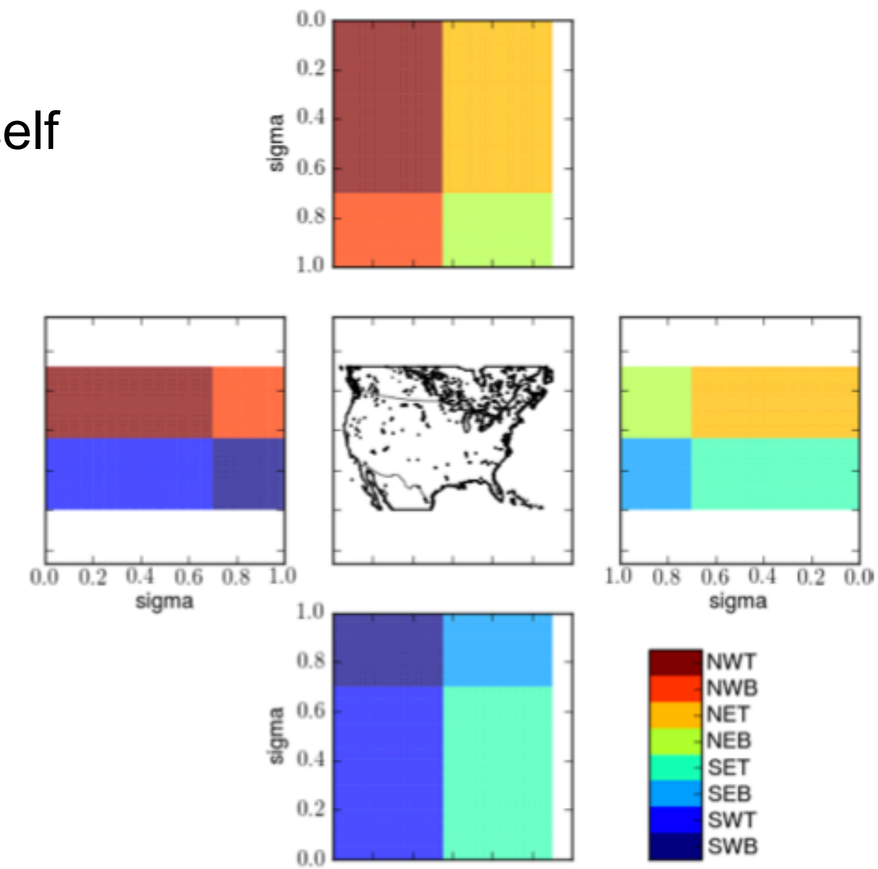
Sensitivity of U.S. BC related mortalities to emissions estimated in CMAQ for April, 2008



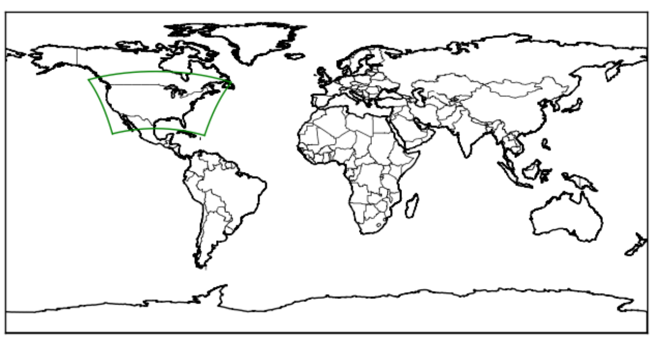
Adjoint sensitivities reveal differences between locations with the highest emissions (Fig a) and locations where emissions have the largest impact on health (Fig b)

Instrumented AQ modeling: boundary sensitivity case study (in collaboration with EPA AMD)

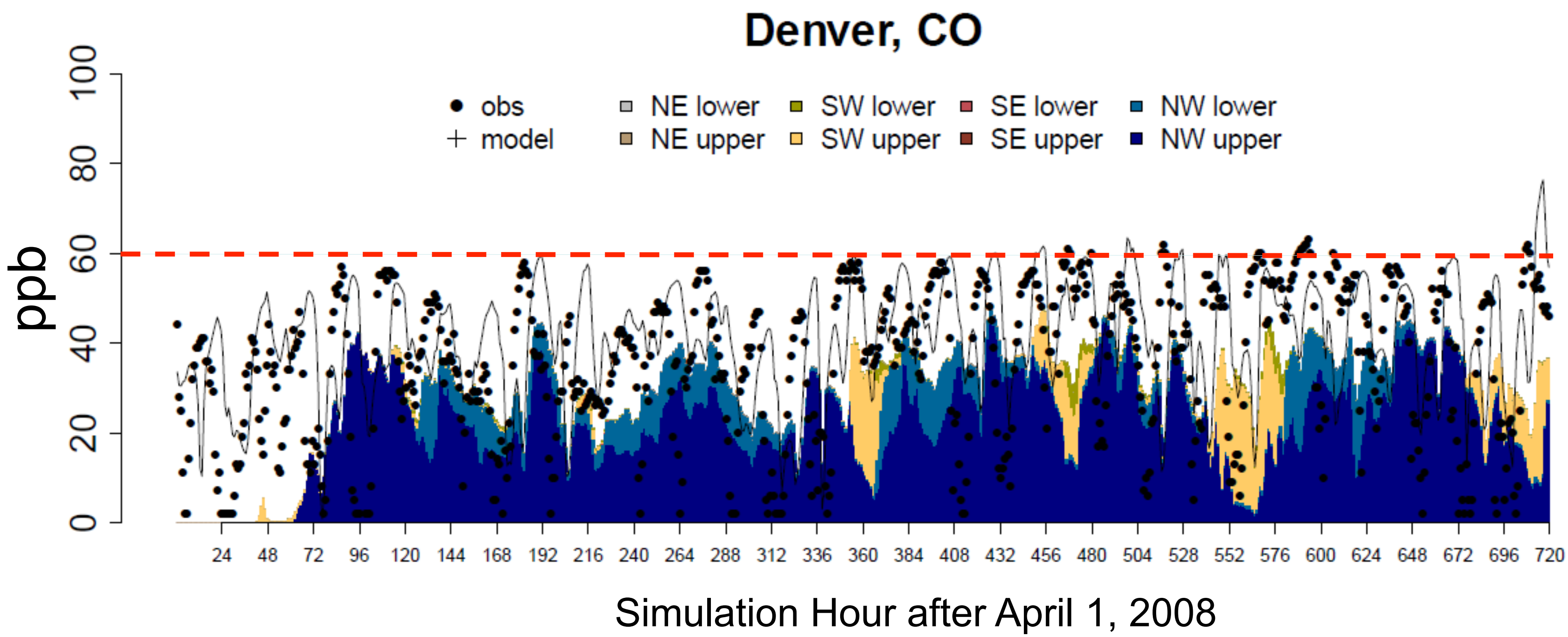
- Designation of "background O_3" important for potential revisions to O_3 standards (e.g., Zhang et al., 2011; Lin et al., 2012)
- Oltmans et al. (2010): episode of high influence of international transport on ozone over western North America during April, 2008.
- CMAQ uses GEOS-Chem boundary conditions – where does that ozone itself come from?



GC adjoint calculations to define boundary subregions:



Denver O_3 sensitivity to boundary O_3 concentrations

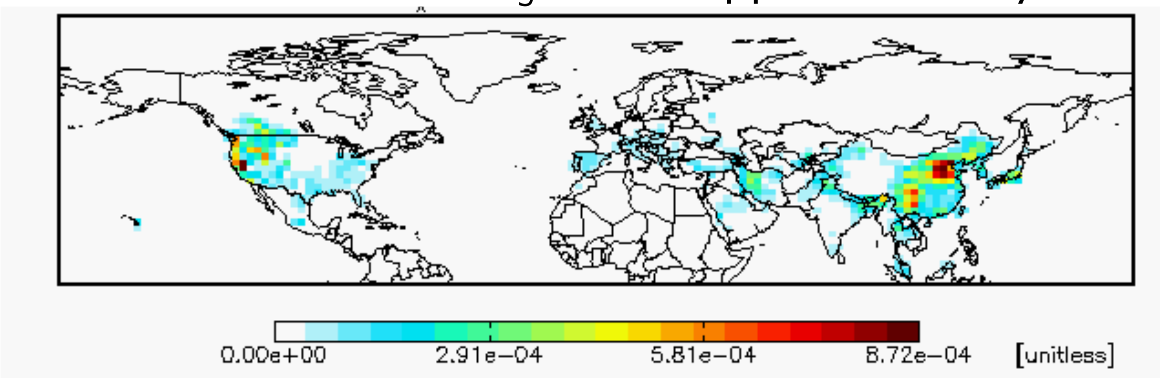


Ozone concentrations are most sensitive to O_3 concentrations at the Northwestern boundaries, primarily the upper boundary.

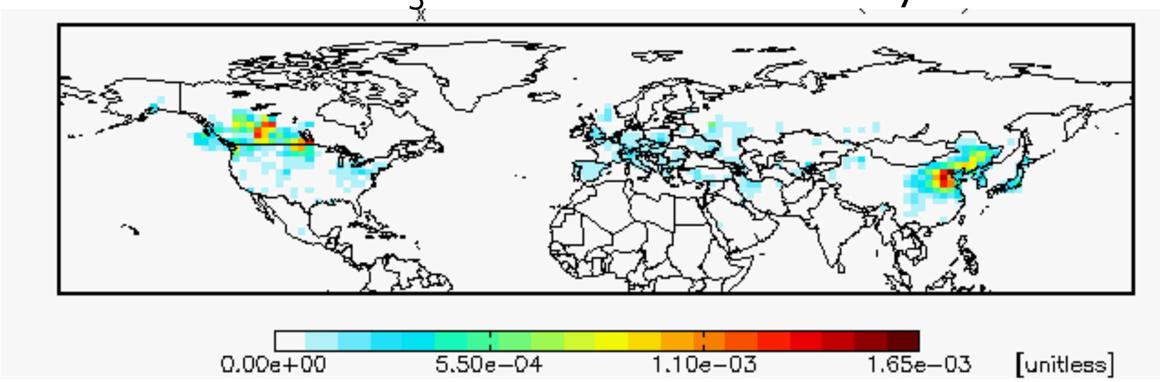
*-*NOTE: Results are specific to period of high ozone transport and should not be extrapolated to other times or seasons*

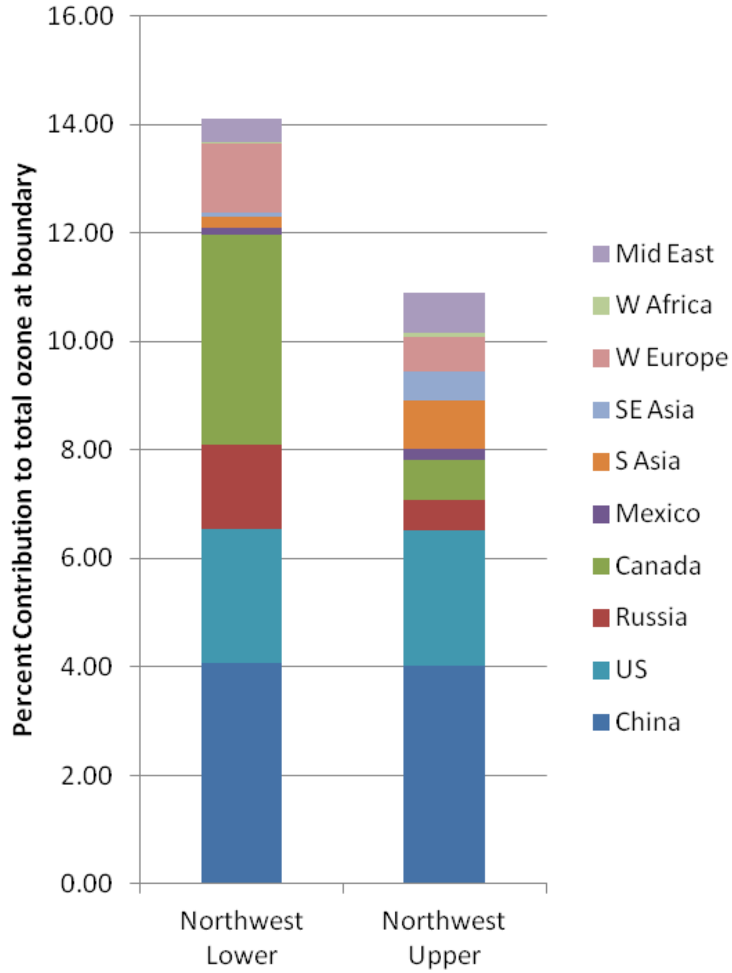
Northwest O_3 sensitivity to global NO_x emissions

Influences on total O_3 at NW upper boundary



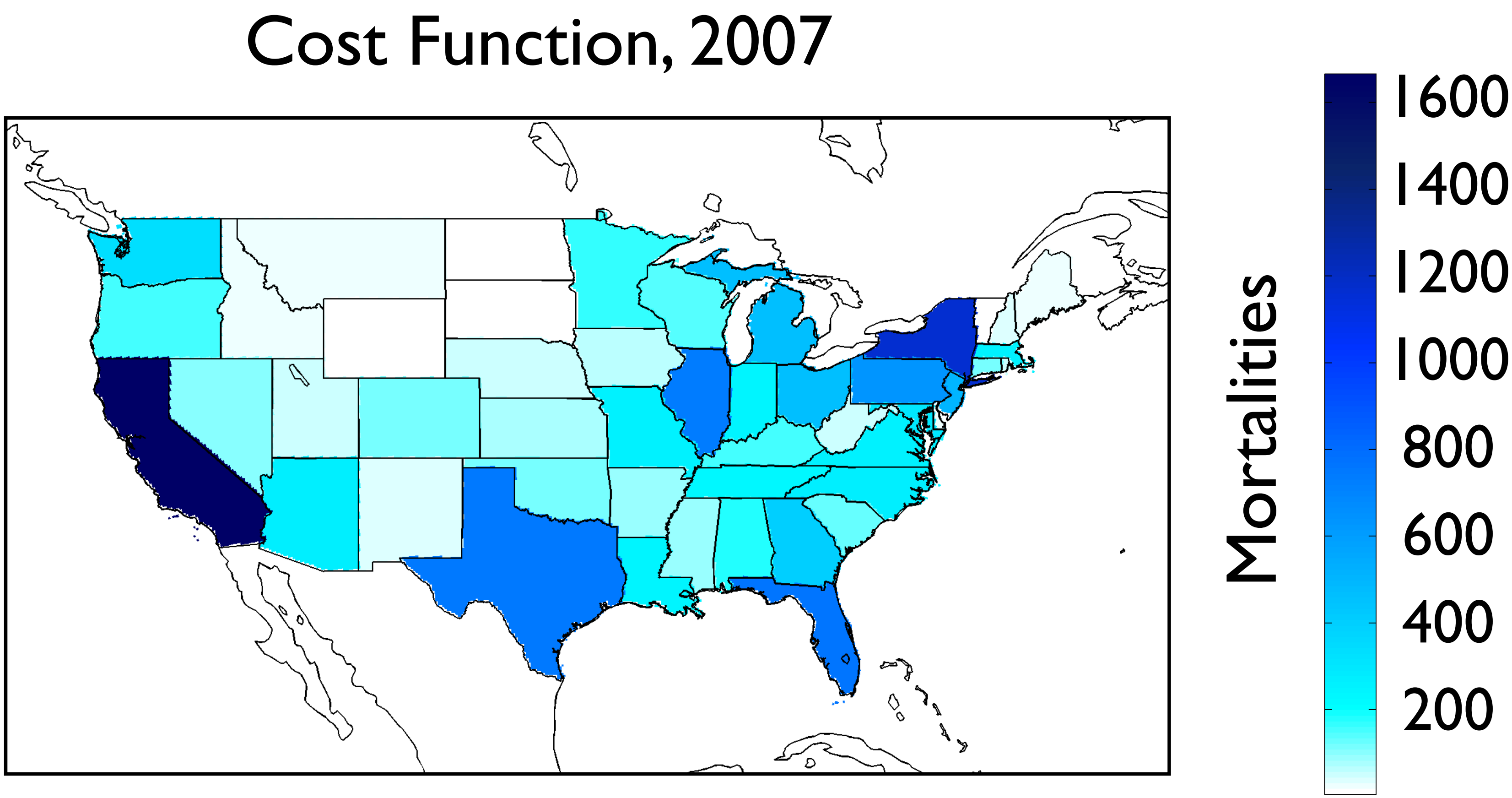
Influences on O_3 at NW lower boundary





Northwest boundaries are impacted by China, United States, Russia, and Canada.

Health impacts of PM2.5: mortality associated with black carbon (BC) in the US



~12,356 mortalities in the US attributed to exposure to BC in 2007