Modeling Analysis of **Multi-Decadal Trends** in Ozone and Precursor Species Across the **Northern Hemisphere** and the **United States**

Rohit Mathur, Jia Xing, Daiwen Kang, Sergey Napelenok, Christian Hogrefe, Golam Sarwar, Jonathan Pleim

Computational Exposure Division
National Exposure Research Laboratory
U.S. Environmental Protection Agency

ENV-VISION Conference
Washington, DC
May 10-11, 2016

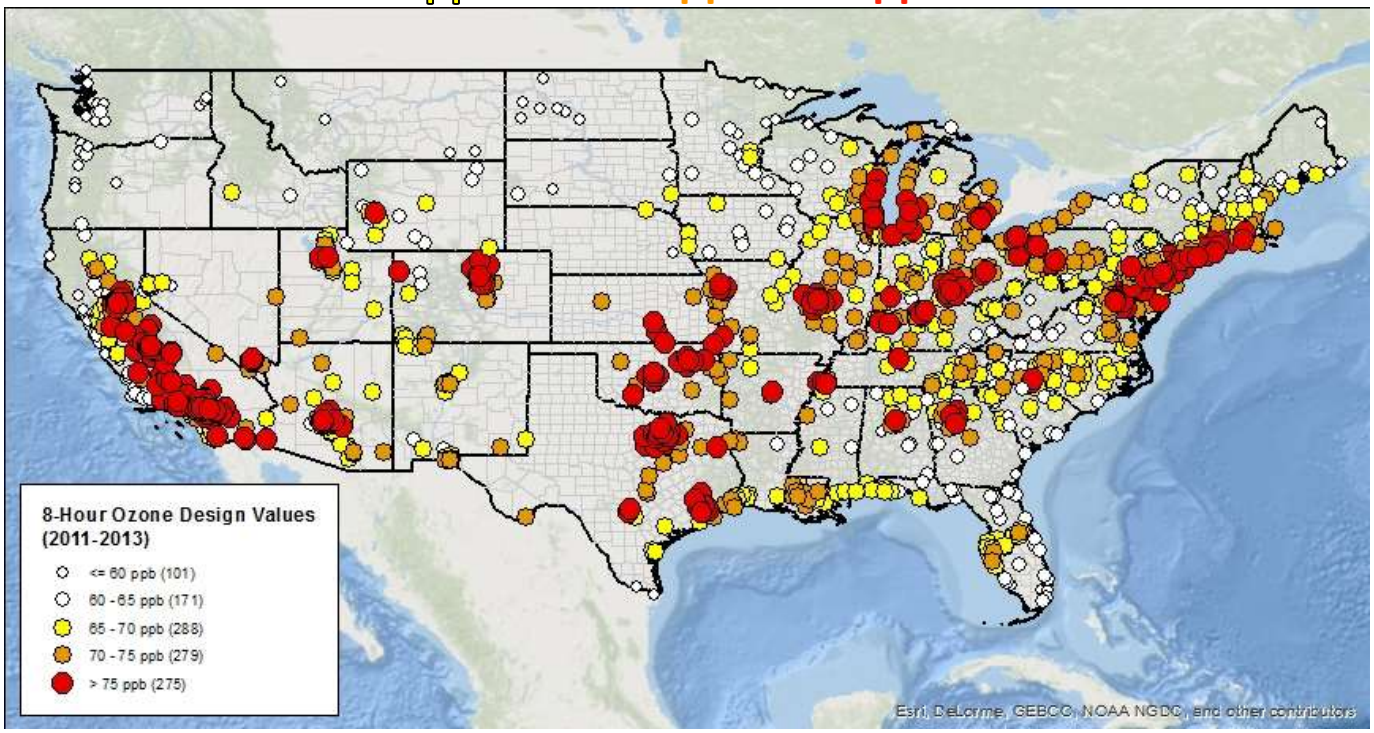
Outline

- Importance of accounting for long-range transport (LRT) in regional air pollution analysis
- Multi-decadal (1990-2010) trends in model & observed surface-level O_3 across the northern hemisphere
- Initial look at how changing emission patterns across the northern hemisphere may be impacting LRT and background pollution
- A call for better observational characterization of LRT of pollution – an "aloft network"

Motivation

2013 8-Hour Ozone Design Values across the U.S.

65-70ppb **70-75ppb** **>75ppb**



Courtesy: P. Dolwick; (http://www.epa.gov/airtrends/values.html)

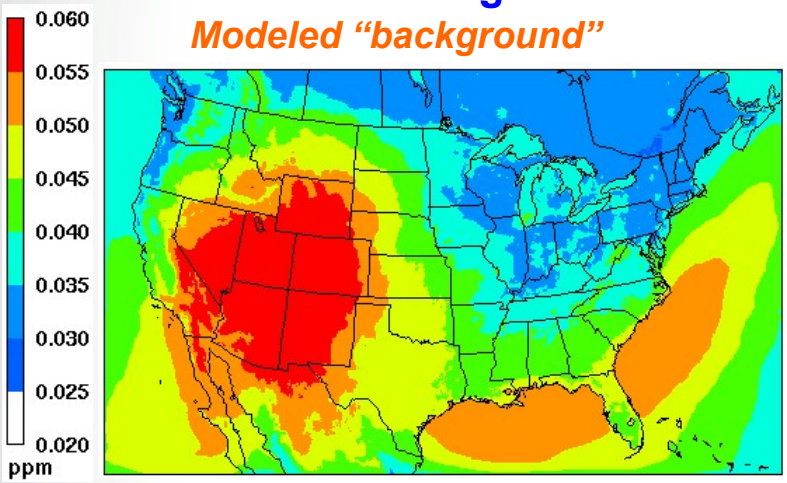
- Tightening of air quality standards places greater emphasis on the ability of air quality models to simulate the entire spectrum of concentrations
- The relative importance of "Background pollution" to the NAAQS becomes more important

Model's view of "Background" Pollution: *Spatial Variability*

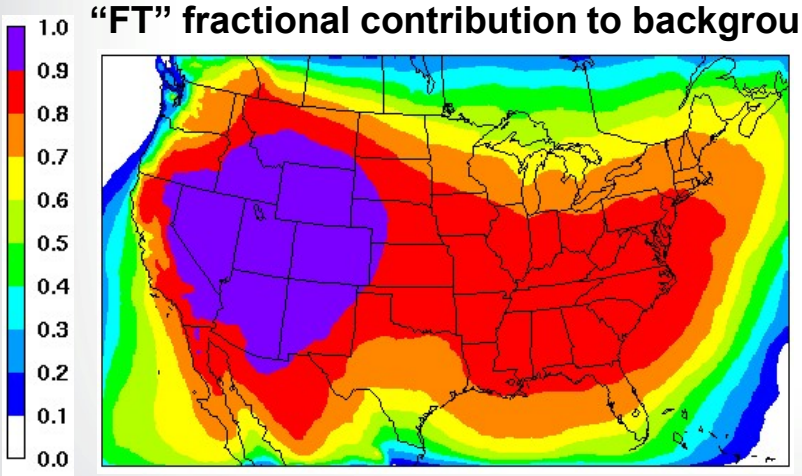
- Added diagnostic tracers to track impact of lateral boundary conditions: surface-3km (***BL***), 3km-model top (***FT***), top-model layer (***ST***) – mixing, transport, deposition mimics O_3

Summer average: 2006

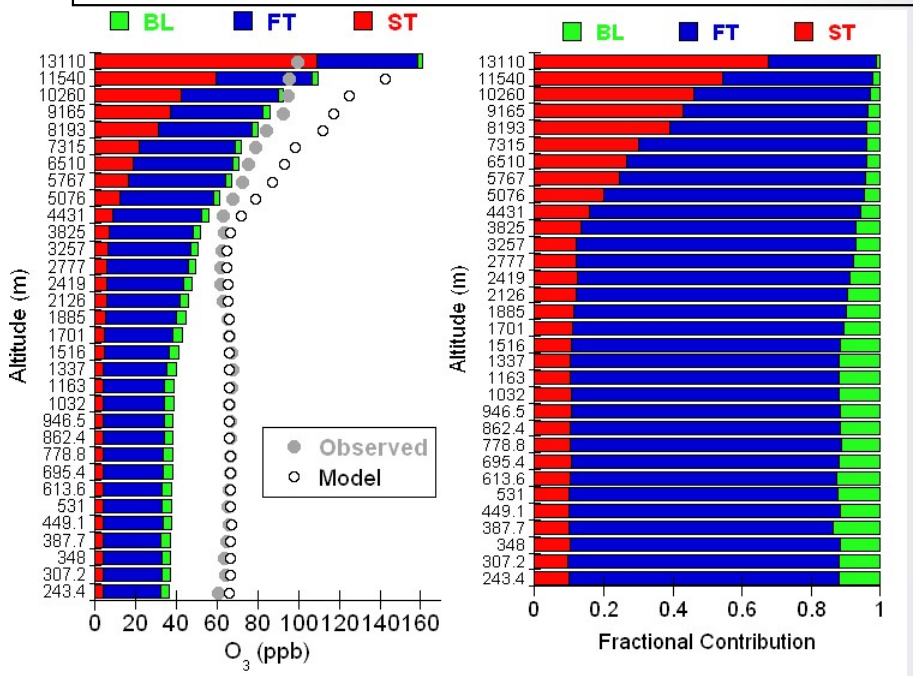
Modeled "background"



"FT" fractional contribution to background



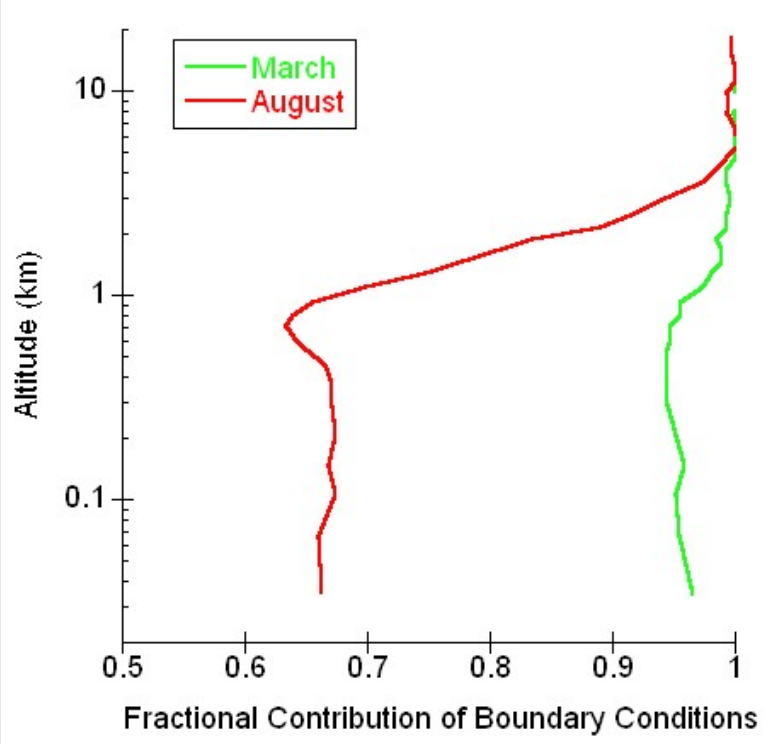
O_3 vertical profile at Huntsville, Alabama



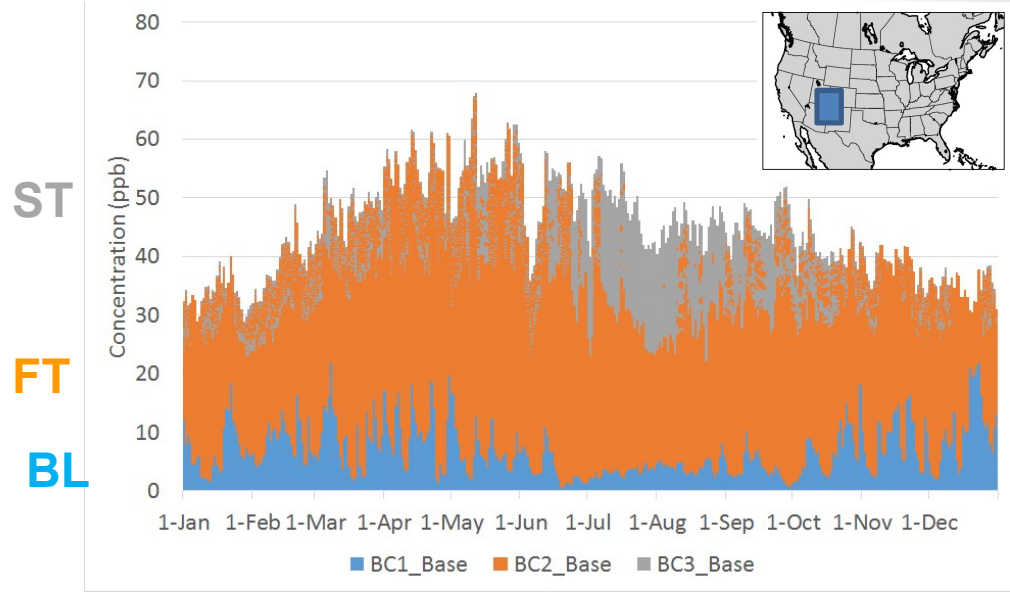
- Spatial variability in "background" value
- ~80% of the surface/BL background is dictated by LRT occurring above the nominal daytime BL

Model's view of "Background" Pollution: *Temporal Variability*

Seasonal variation in LBC Fractional Contribution to simulated O_3 at Trinidad Head

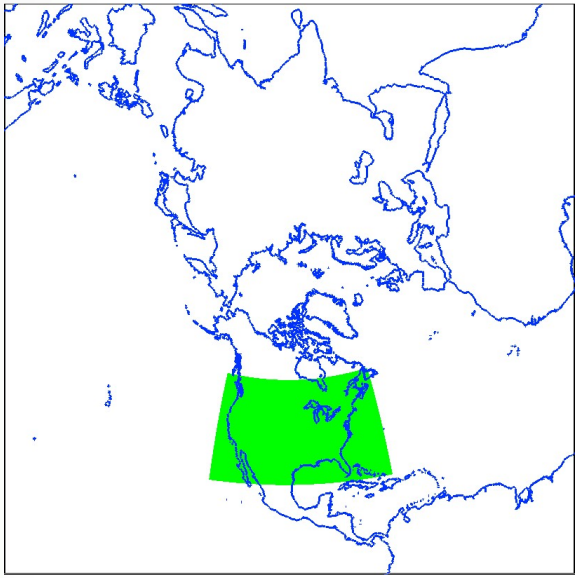


Hourly Tracer Concentrations over the Four Corners Region



- "Background" values vary on daily to seasonal time-scales
- Contributions of LRT and stratosphere vary seasonally

WRF-CMAQ Coupled Model Setup



Horizontal

- **Northern Hemisphere**
  - Polar Stereographic projection
  - 108 km resolution (187x187)
- **Nested CONUS: 36km resolution**

Vertical

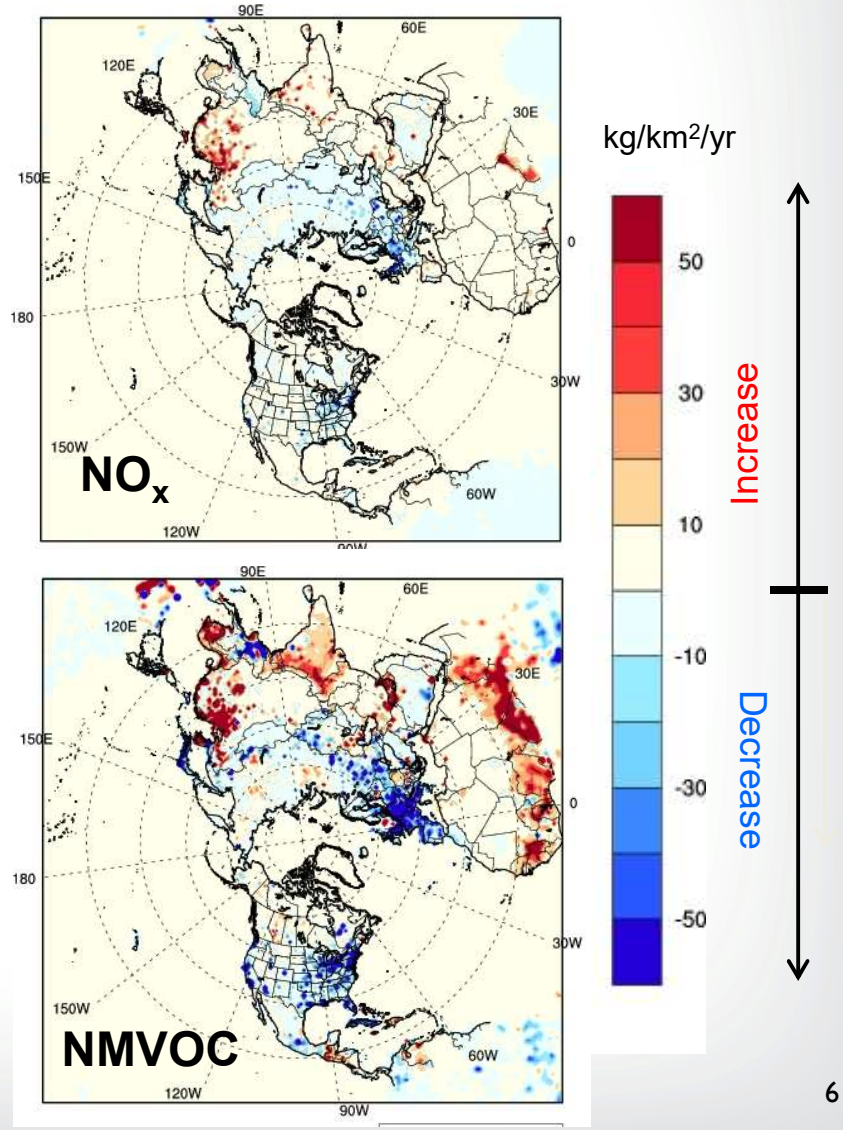
- 44 layers between surface & 50mb

Emissions

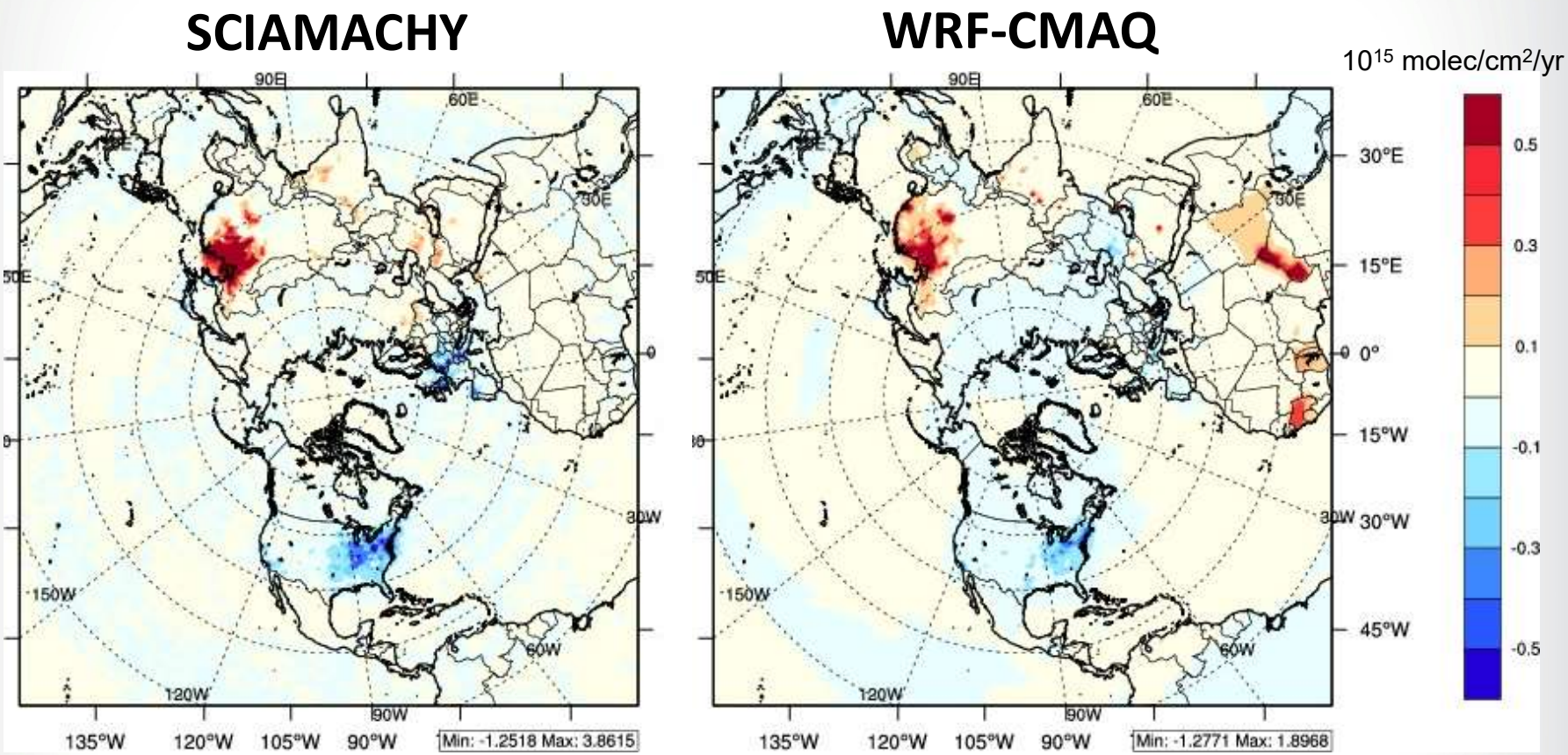
- NH: EDGARv4.2; *Xing et al, 2015 (ACP)*
- CONUS: *Xing et al., 2013 (ACP)*

Time Period: 1990-2010

1990-2010 Emission Trends

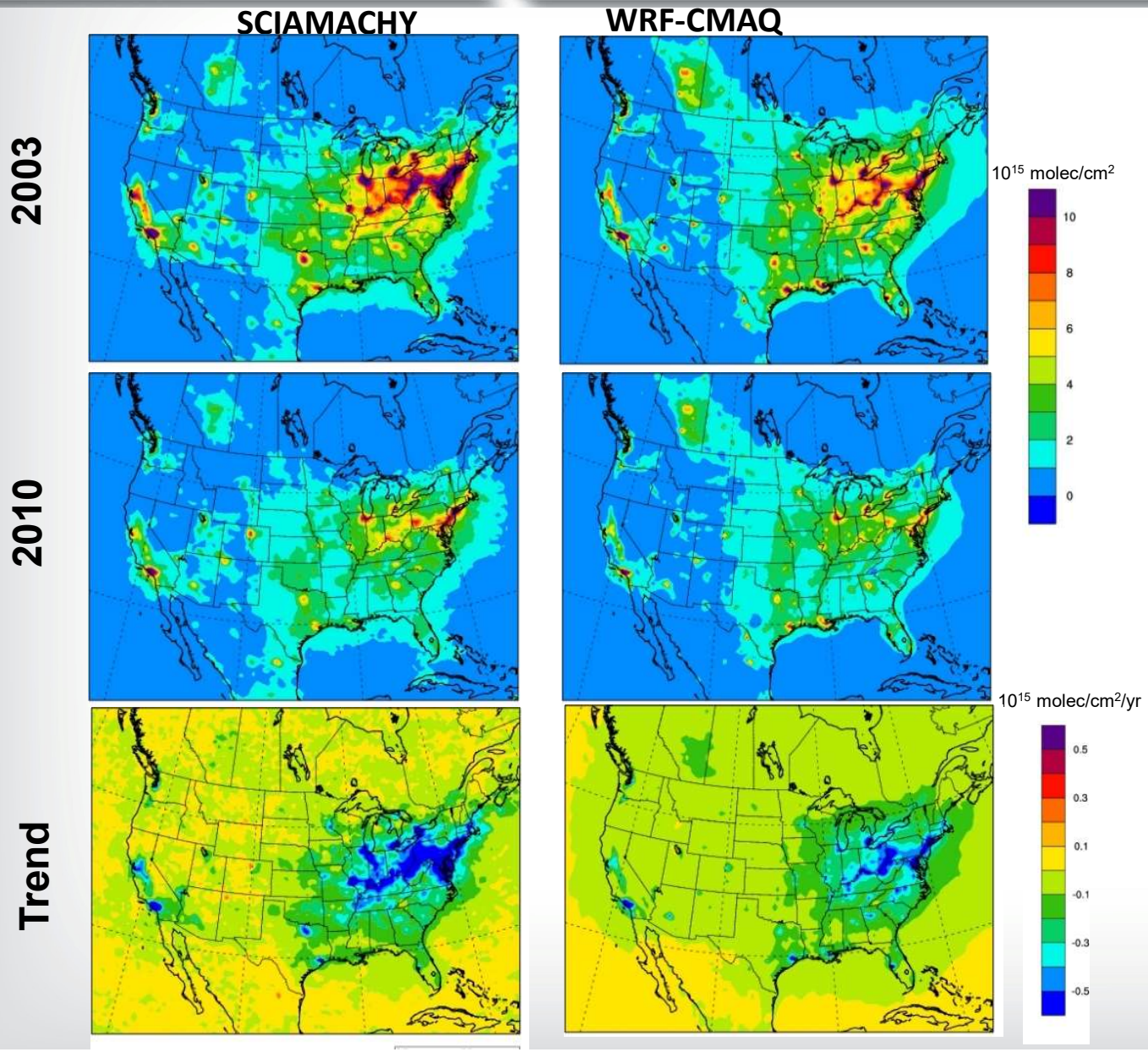


Simulated and Observed Trends in NO_2 Column across the Northern Hemisphere:2003-2010



- Model captures decrease in the U.S & increase in E. China and N. India
- Discrepancies in Africa

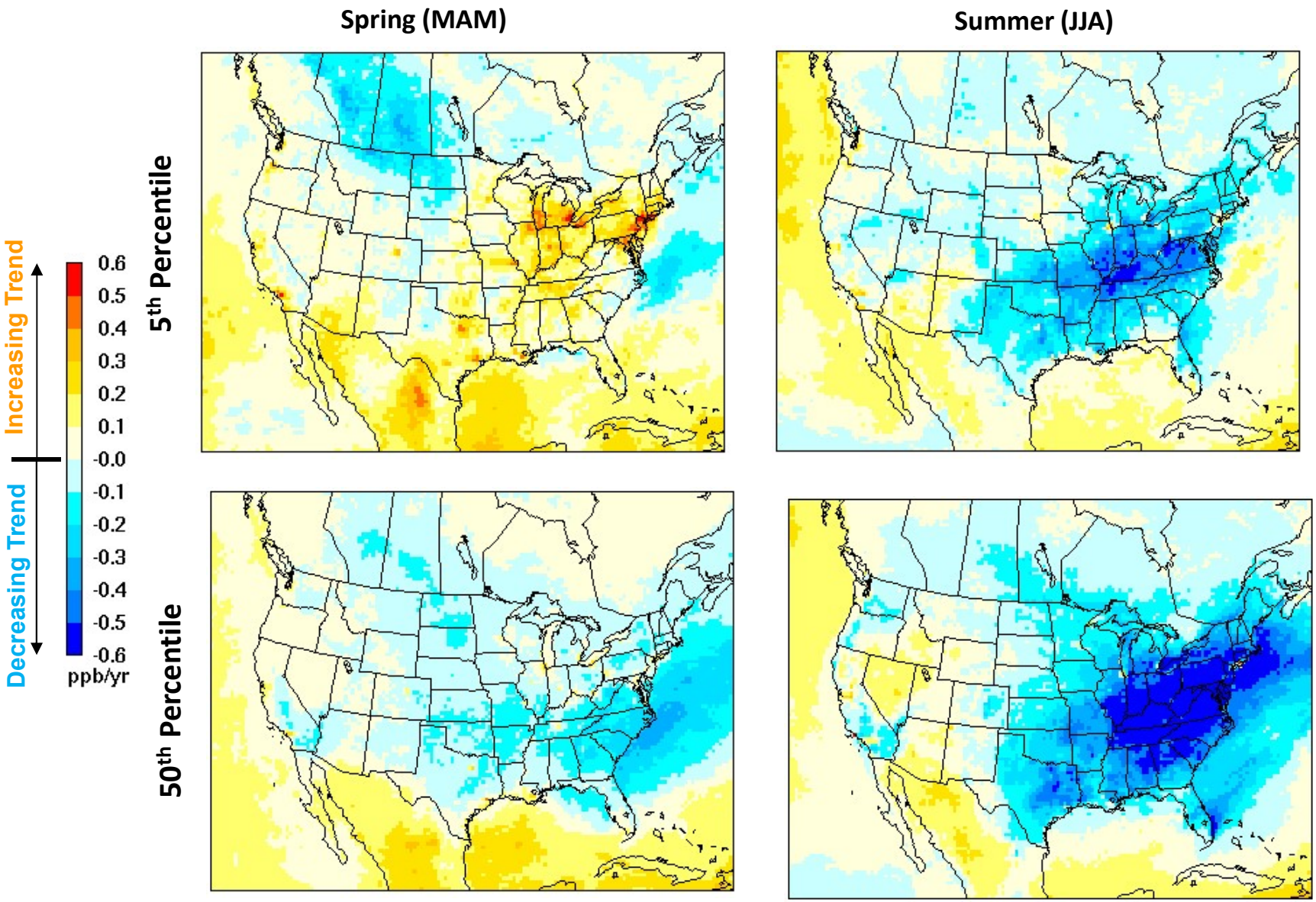
Simulated and Observed Trends in NO_2 Column across the U.S: 2003-2010



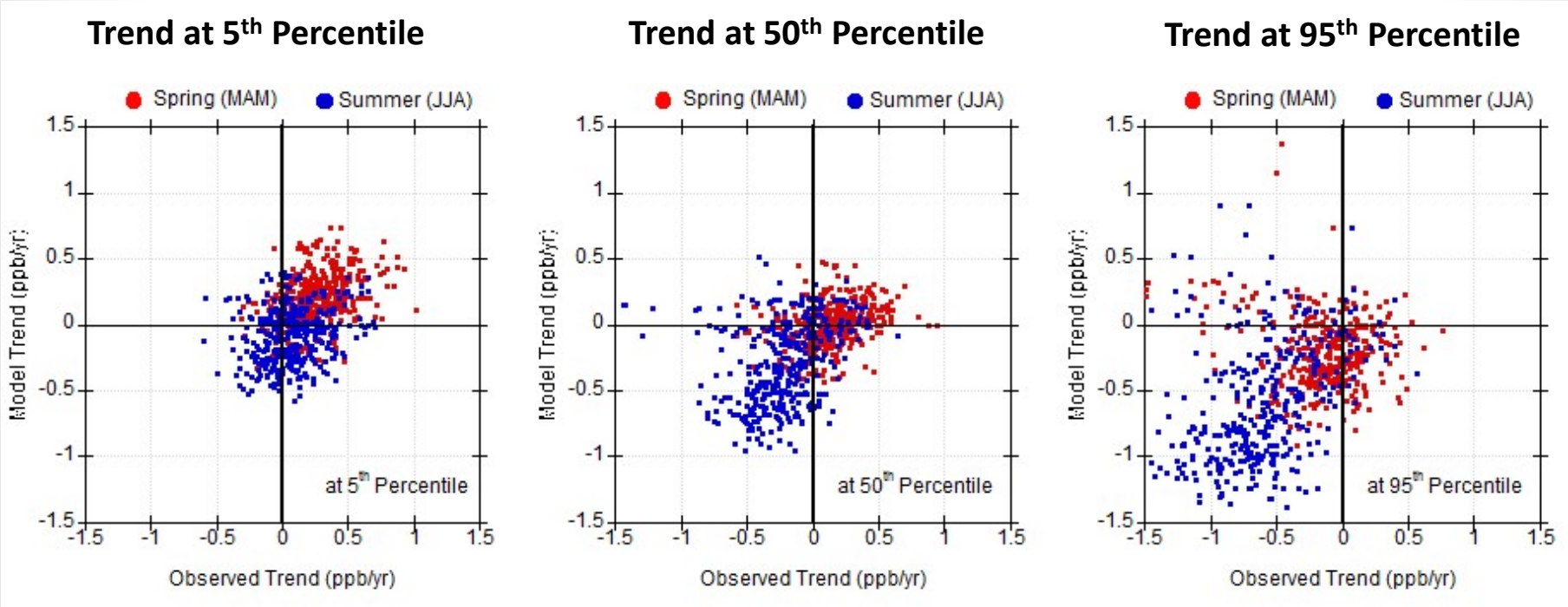
Both observations & model show reductions in NO_2 in urban areas and regionally

Model estimated NO_2 column as well as trend are lower than retrievals

1990-2010 Trends in Surface O_3 at Different Percentiles; 36km resolution Domain



Comparison of Model and Observed DM8hr O_3 Trend at Different Percentiles: Seasonal Contrast at AQS sites in the U.S

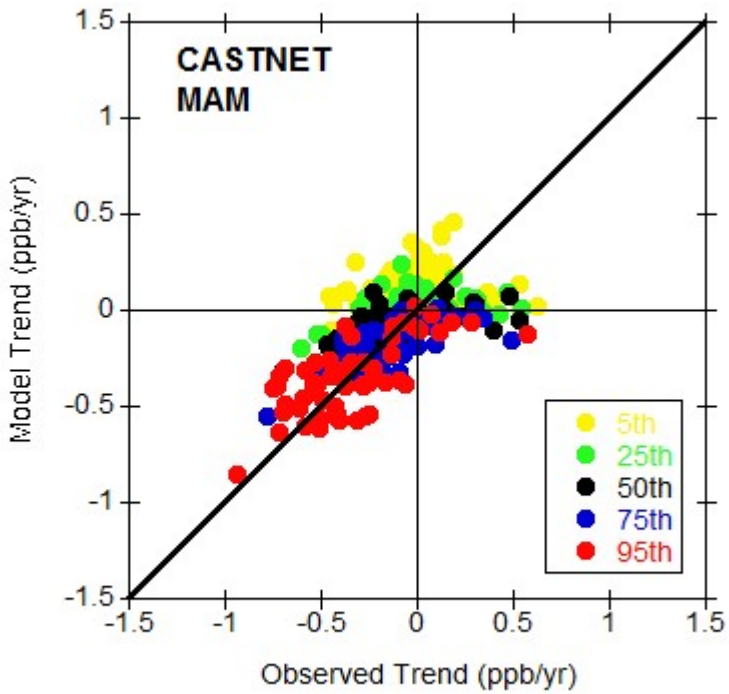


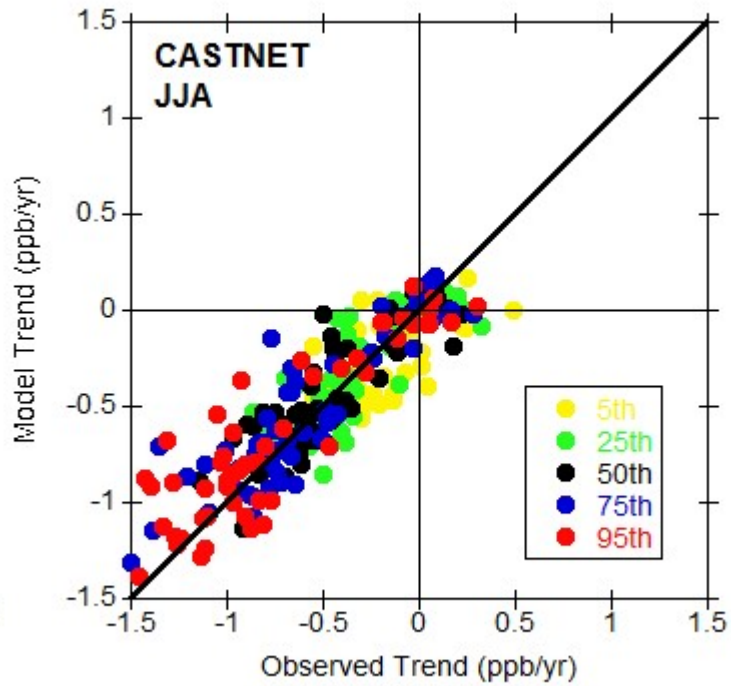
- During Spring, both model & observations show increases at lower end of the concentration distribution at majority of the sites

- Summer trends at the median concentration and high ranges show decreases across majority of sites
- Increasing trend at many locations during Spring

- At the upper end of the concentration distribution, decreasing trends at most locations during the warmer months

Comparison of Model and Observed DM8hr O_3 Trend at Different Percentiles: Seasonal Contrast at CASTNET sites in the U.S

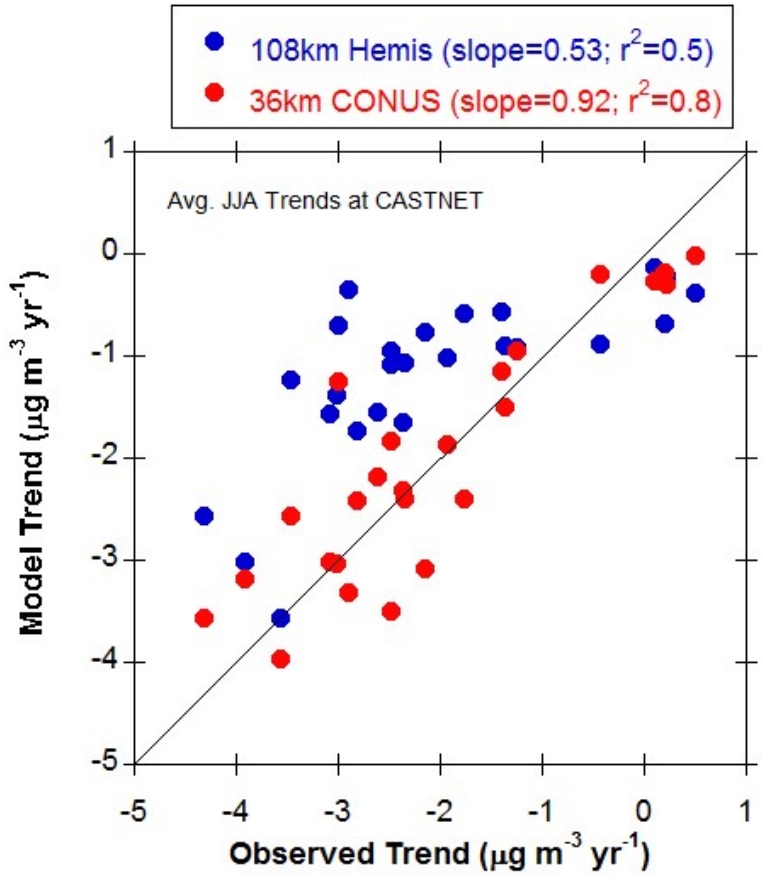




- During Spring, both model & observations show increases at lower end of the concentration distribution at many sites
- At the upper end of the concentration distribution, decreasing trends at most locations during the warmer months

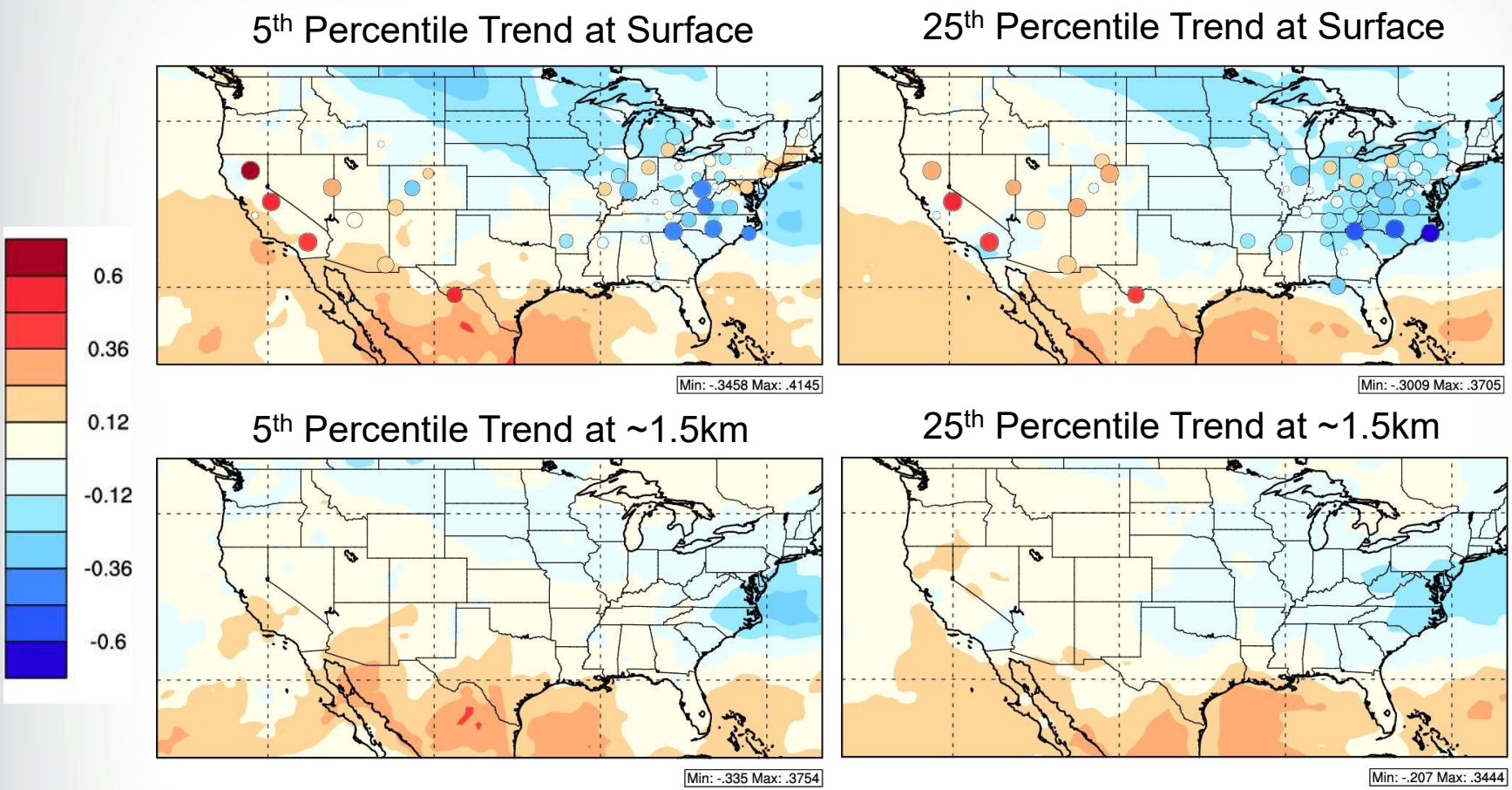
Impact of Resolution and Emission Improvements on Simulated Trends (JJA-avg at CASTNET)

Daily Max. 8-Hr O_3



- Finer resolution and improved emission trends results in *better capturing spatial variability in observed trends*

Trends in Springtime DM8hr O_3 at the Surface & Aloft



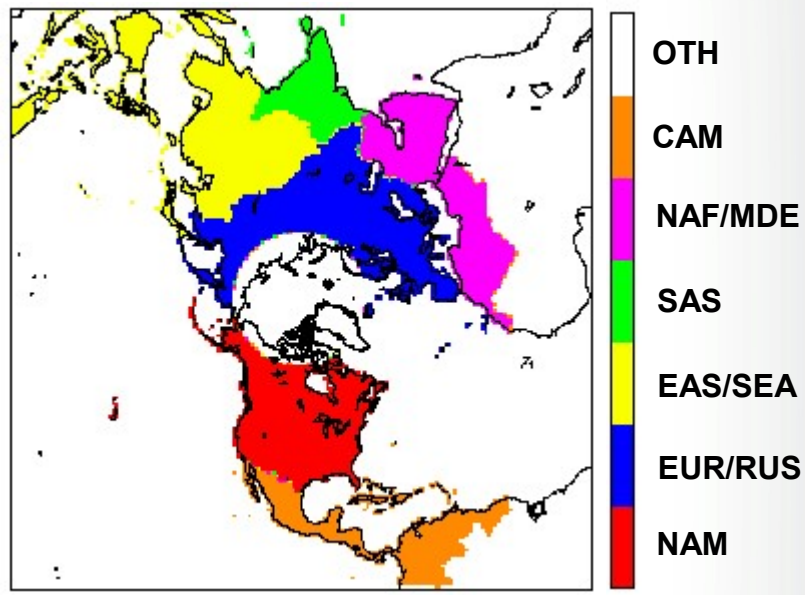
- Increasing trend in surface O_3 at several CASTNET monitor locations in the western U.S.
  - Model simulations also show increasing trends at many of these locations
- Increasing trends aloft (~1.5km) suggestive of influence of LRT effects

How do different source regions influence background pollution?

Approach

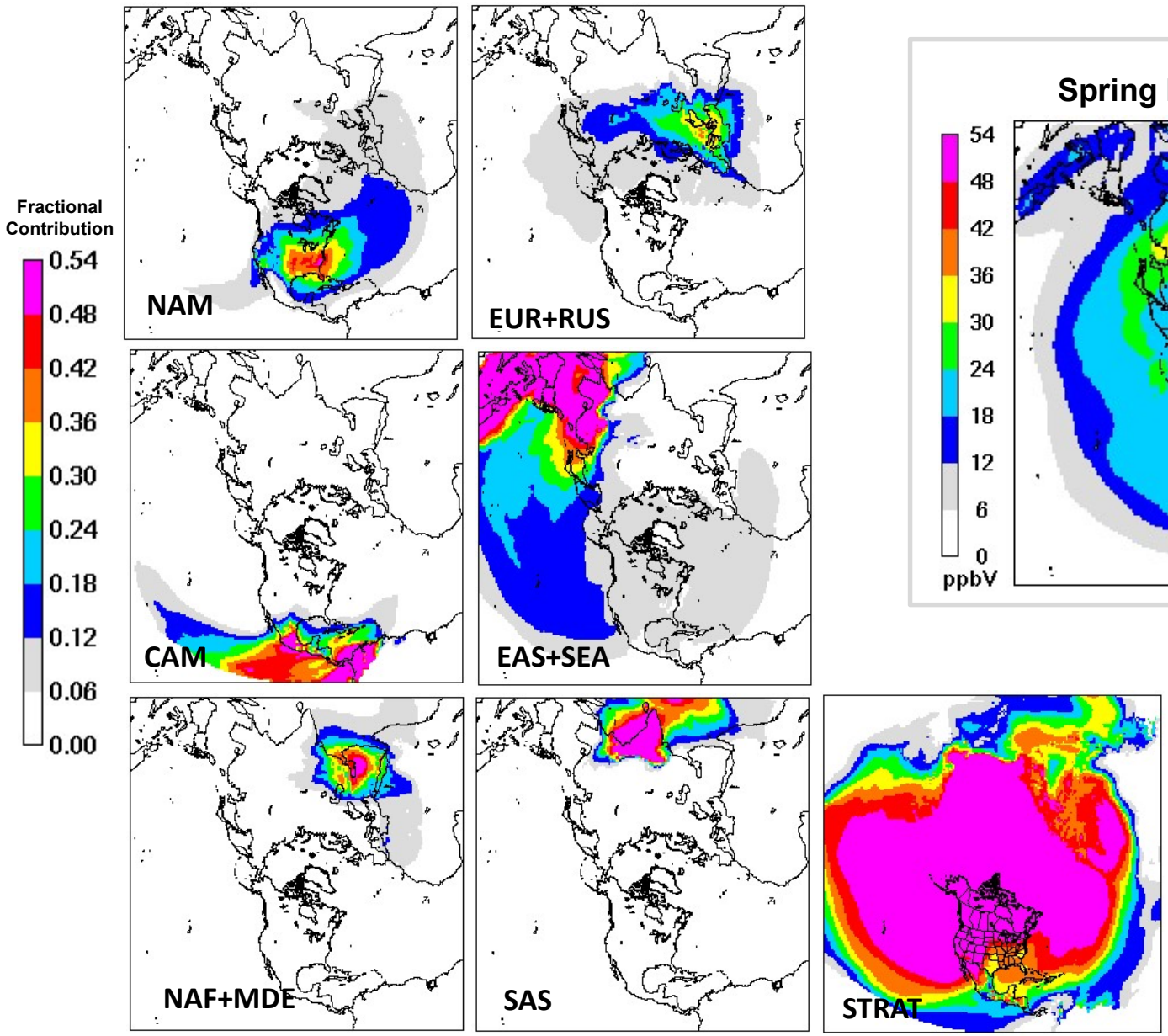
- Use the direct decoupled method (DDM) to track sensitivity (S_j) of O_3 to emissions from source region *j*
- Estimate long-range transport O_3 to a receptor region n as $\sum_{j \neq n} S_j$
- Examine the relative contributions of the different source regions to the background for the receptor region

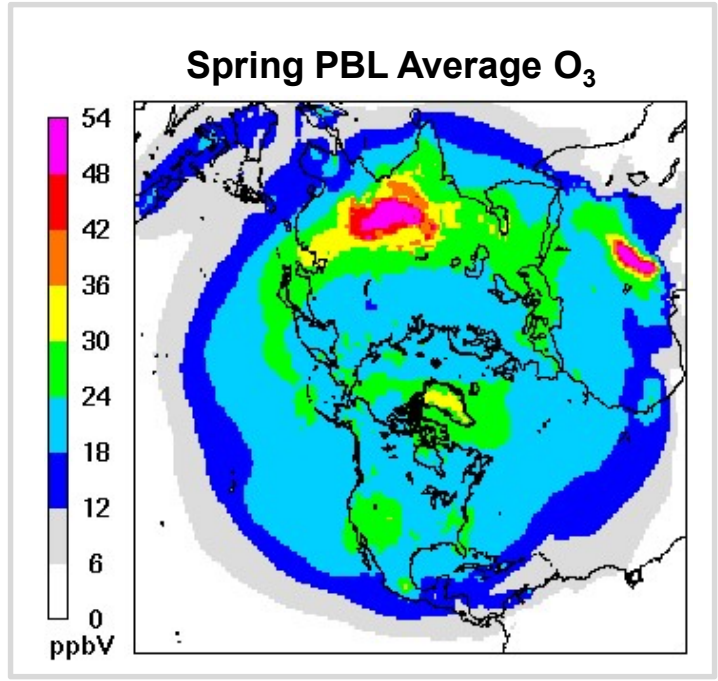
Seven source regions



(following the HTAP source region definitions)

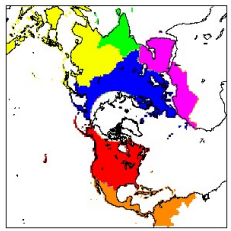
PBL & Seasonal-average O_3 & Source-region Contributions : Spring 2006





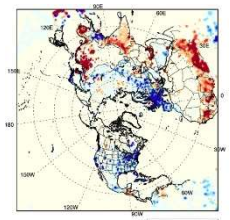
Are changing emission patterns impacting LRT and background pollution-levels?

Combine:



Single-year source sensitivities

+



Multi-decadal Emission and Concentration Changes

Concentration due to LRT at cells in receptor *m* for year *N* ***approximated*** as*:*

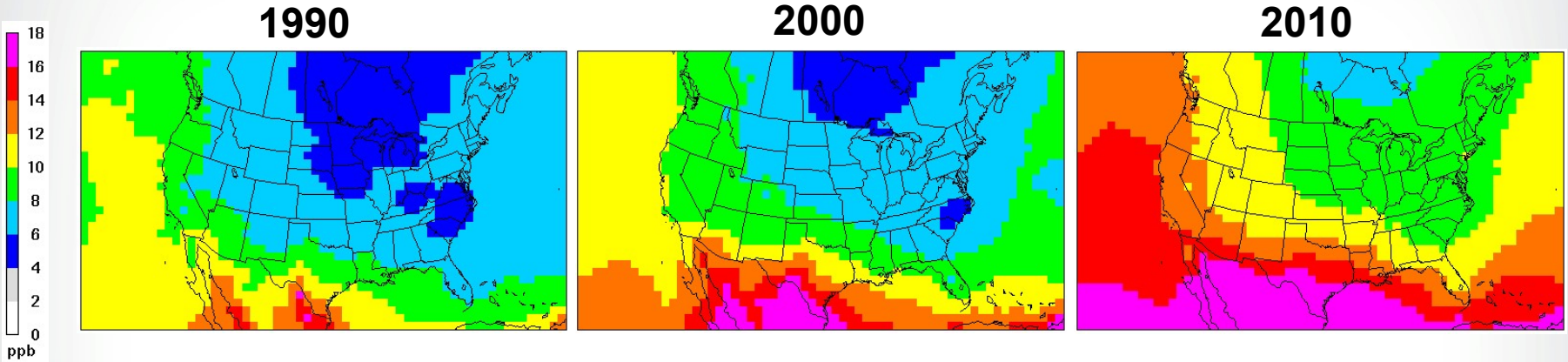
$$C^{N}_{LRT,m} = \underbrace{\left(\sum_{j \neq m} S_j \ \Delta E_j^N\right)}_{\text{first-order Taylor series}} \underbrace{\left(C^N \Big/ \sum S_j \ \Delta E_j^N\right)}_{\text{re-normalizing factor}}$$

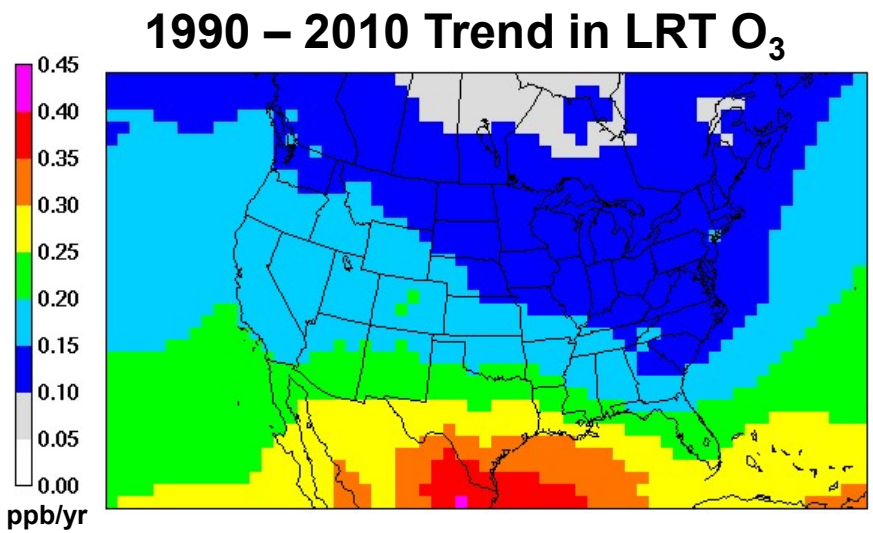
S_j – normalized sensitivity
ΔE_j – Emission change
C^N – simulated conc. for year N

Approximate O_3 from Long-Range Transport: Spring

Seasonal and PBL mean

Preliminary Results



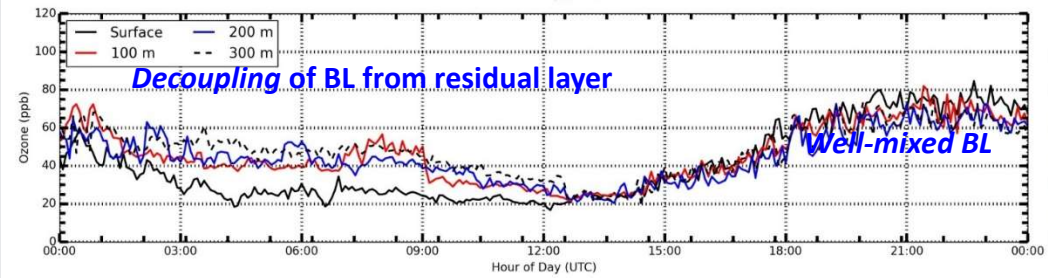


- Increasing contributions of LRT to surface O_3 over the past two decades

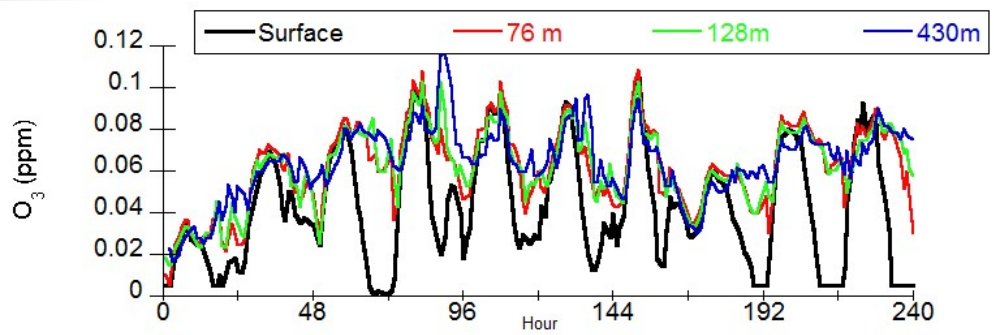
Need for Observational Information on LRT

Characterizing Nocturnal Aloft O_3 is Important

NOAA BAO Tower, CO: 2014 DISCOVER-AQ



WRAL Tower Observations, Raleigh, NC: 1996



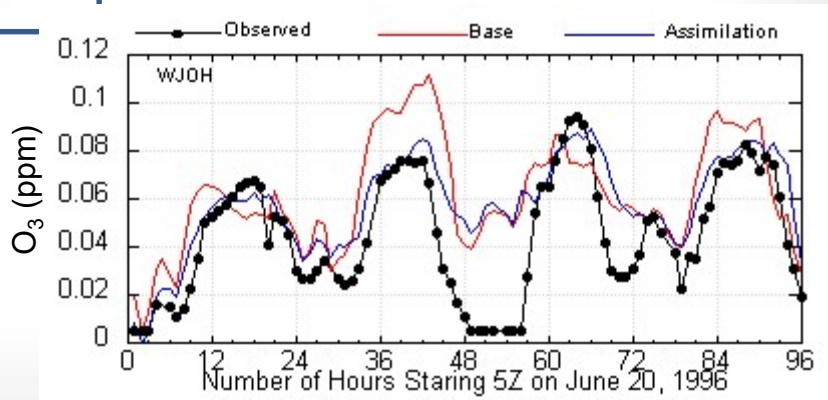
- Surface O_3 = Background [*transport*] + In-situ production
- Transport constitutes a large fraction of night time O_3 in residual layer
- Local background influenced by downward mixing of O_3 from the residual layer

Impact of Assimilation of Aloft O_3

Downwind of WRAL site

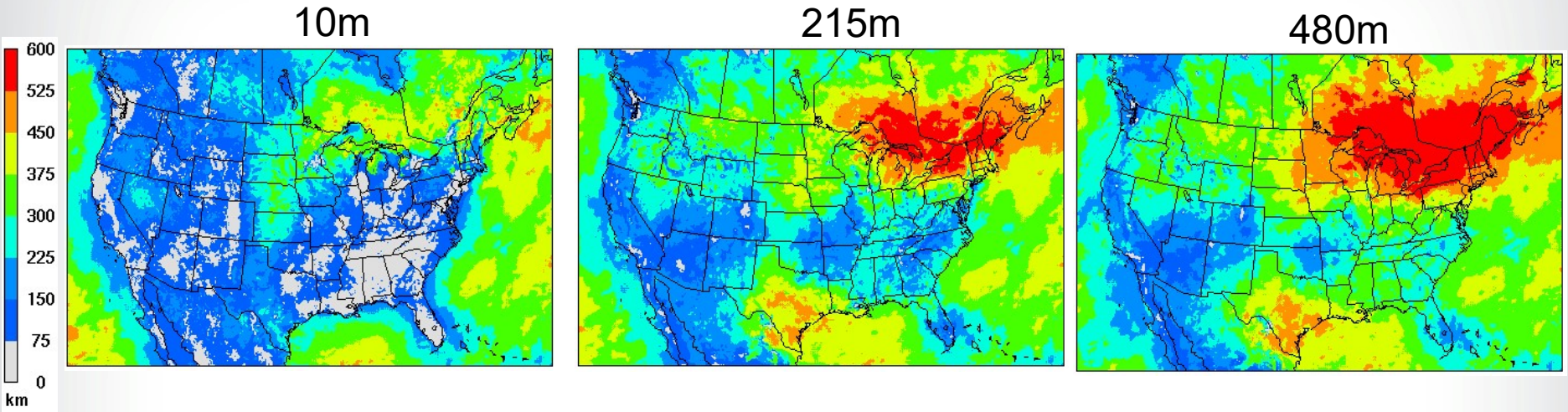
Reduces over prediction (6/21)

Reduces under prediction (6/22)

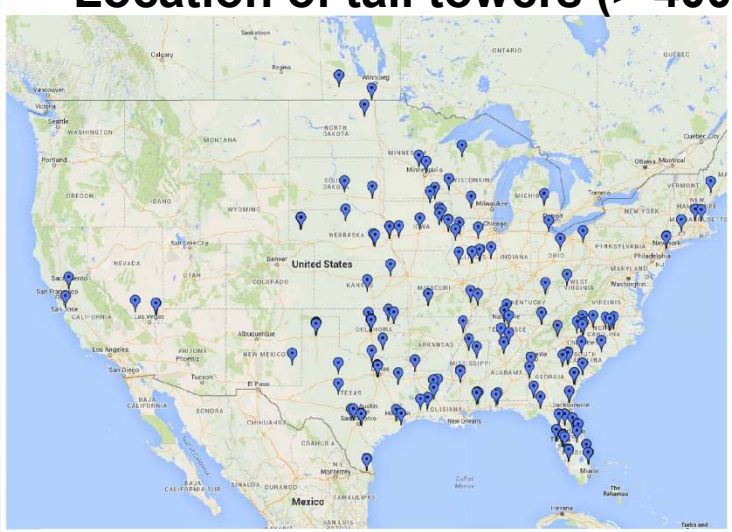


Can Lower Cost Technology Fill a Critical Observational Gap?

CMAQ estimated e-folding distance for nocturnal O_3 (7 GMT, May-September 2010)



Location of tall towers (> 400m)



[Wikipedia: List of tallest structures in the world - 400-500 meters]

- e-folding distances aloft nocturnally are several hundred of kilometers
- Can existing tall structures be instrumented with “lower cost” sensors to provide a critical continuous aloft observation?
- ~20x20 array of aloft measurements

Summary

- **Multi-decadal (1990-2010) model calculations suggest large (& contrasting) changes in tropospheric composition over the northern hemisphere**
  - Significant reductions in air pollution over N. America and western Europe and increases in large portions of Asia
  - Quantitative trends inferred from the model agree well with those from measurements
- **The model is able to capture the changing seasonal distributions in surface O_3 in the U.S. during 1990-2010 arising from changing emissions and LRT**
  - Increasing trends during Spring and Winter and at the lower end of concentration distributions
- **Changing emission patterns across the northern hemisphere will impact background pollution in a region. Sensitivity analysis indicates that**
  - the background is increasing in N. America and varies seasonally
  - the source region contributions and their relative importance also vary seasonally
- **Measurements to characterize aloft transport (and subsequent downward mixing) are needed**