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Linking CO_2 Emissions Verification to Air-Quality Monitoring: *Spatiotemporal Variability of CO_2 Simulated with CMAQ*

Sandia Climate Research Program

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Highlights

- ❑ We for the first time applied the EPA CMAQ chemical transport model (CTM) to simulate CO_2 [1]
- ❑ We examined the spatial correlations between CO_2 and NO_2, CO, and SO_2, and found that the correlations vary with season and sampling strategy (e.g., surface concentrations or vertical column densities)
- ❑ The CMAQ-CO_2 model will be used for developing a multi-species emissions verification system

Background

❑ **Two key challenges in fossil-fuel CO_2 emissions verification**

(1) A signal-to-noise issue due to the large and yet uncertain interference by the biosphere
(2) Limited understanding of fine-scale spatial variability of CO_2 and its fossil-fuel component

❑ **Motivation and rationale of studying CO_2 in conjunction with air-quality species**

(1) Sources almost always emit an array of pollutants together
(2) All trace chemical constituents are transported in the atmosphere simultaneously
(3) Abundant data are available from the air-quality monitoring networks and satellites
(4) Inverse modeling theory provides the foundation for multi-species flux inversion [2,3]

CMAQ Model Configuration

(refer to [1] for details)

CMAQ model setup

❑**CMAQ**	:	v5.0, CONUS/36km/22L
❑**Meteorology**	:	WRF v3.1.1
❑**Simulation year**	:	2007

CO_2 simulation

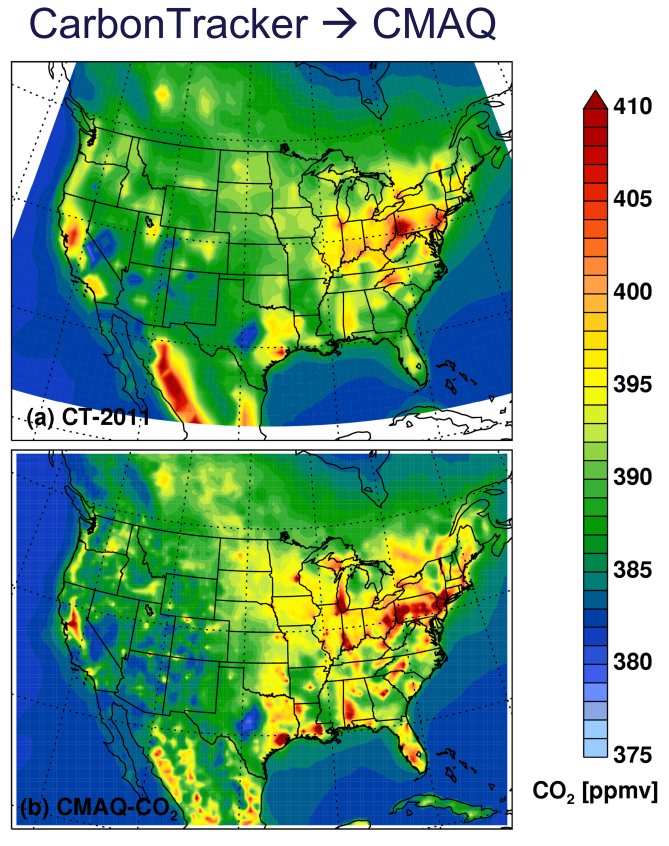
❑**BC and IC**	:	CT-2011 (1°× 1°; 3-hourly)
❑**Biosphere fluxes**	:	CT-2011 (1°× 1°; 3-hourly)
❑**Fossil fuel (FF) emissions**	:	Vulcan (2002; 10km; hourly) in the US; CDIAC outside
❑**Fire emissions**	:	GFEDv3.1 (0.5°× 0.5°; 3-hourly)
❑**Oceans fluxes**	:	CT-2011 (1°× 1°; 3-hourly)

Atmospheric chemistry simulation

❑**BC and IC**	:	GEOS-Chem v8.2.1
❑**Biogenic emissions**	:	MEGAN v2.1
❑**Anthropogenic emissions**	:	NEI 2005
❑**Fire emissions**	:	Blue Sky + FEPS
❑**Chemistry**	:	CB-05

CMAQ CO_2 Simulation Evaluation

CarbonTracker → CMAQ



CMAQ simulations vs Tower observations

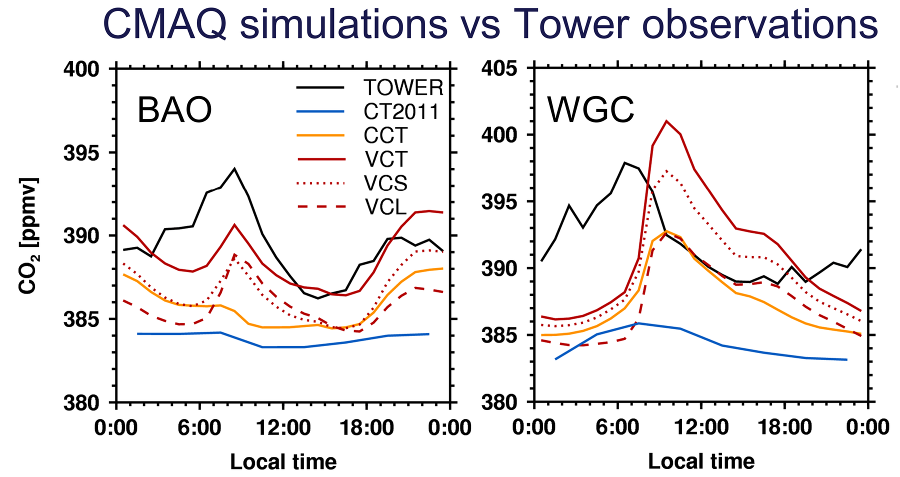


Figure 1. Monthly mean diurnal profiles of CO_2 in October 2007 observed at Boulder Atmospheric Observatory (BAO) and Walnut Grove (WGC) (TOWER) and simulated by CT2011 and CMAQ with different configurations. CCT uses CDIAC FF emissions and CT2011 NEE; VCT uses Vulcan and CT2011 NEE; VCS uses Vulcan FF emissions and CASA NEE; and VLM uses Vulcan FF emissions and CLM4VIC NEE. For model grids outside the US, Vulcan has no values and CDIAC is used instead.

Figure 2. Monthly mean CO_2 concentrations near the surface simulated by (a) CT2011 and (b) CMAQ using CT2011 NEE, and fossil-fuel emissions from CDIAC and Vulcan for model grids outside and inside the US, respectively.

Spatial Distributions of CO_2 and NO_2 – *Surface Concentrations*

Summer

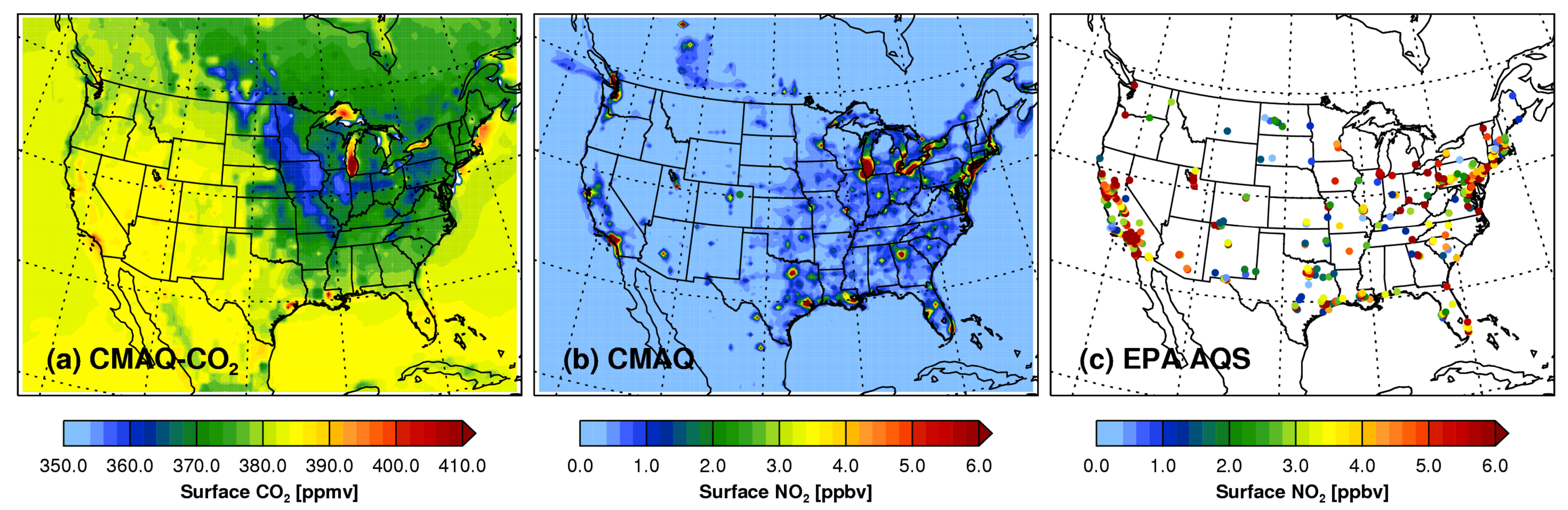


Figure 3. Spatial distributions of (a) CMAQ simulated CO_2, (b) CMAQ simulated NO_2, and (c) EPA AQS observed NO_2 near the surface at 14:00 local time in July 2007.

Winter

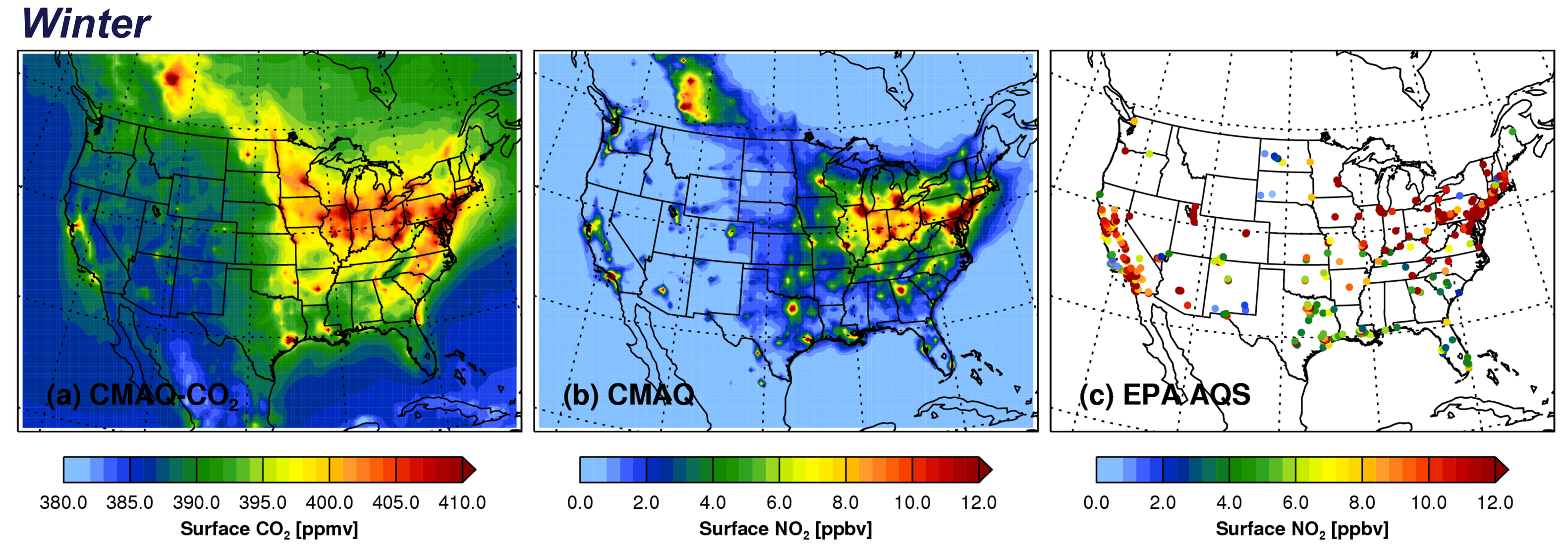


Figure 4. Same as Figure 3, but for December 2007.

Spatial Distributions of CO_2 and NO_2 – *Vertical Column Densities*

Summer

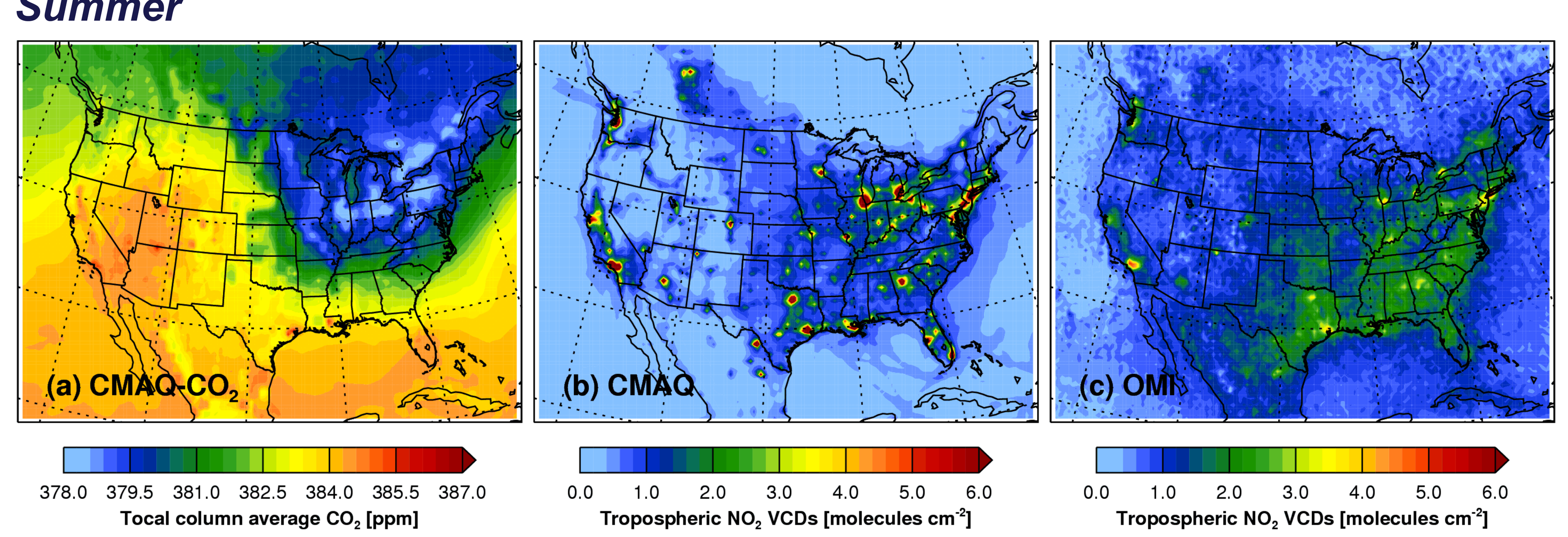


Figure 5. Spatial distributions of (a) CMAQ (trop) and CT-2011 (strat) simulated total column average CO_2, (b) CMAQ simulated tropospheric vertical column densities (VCDs) of NO_2, and (c) Aura OMI observed NO_2 VCDs at 14:00 local time in July 2007.

Winter

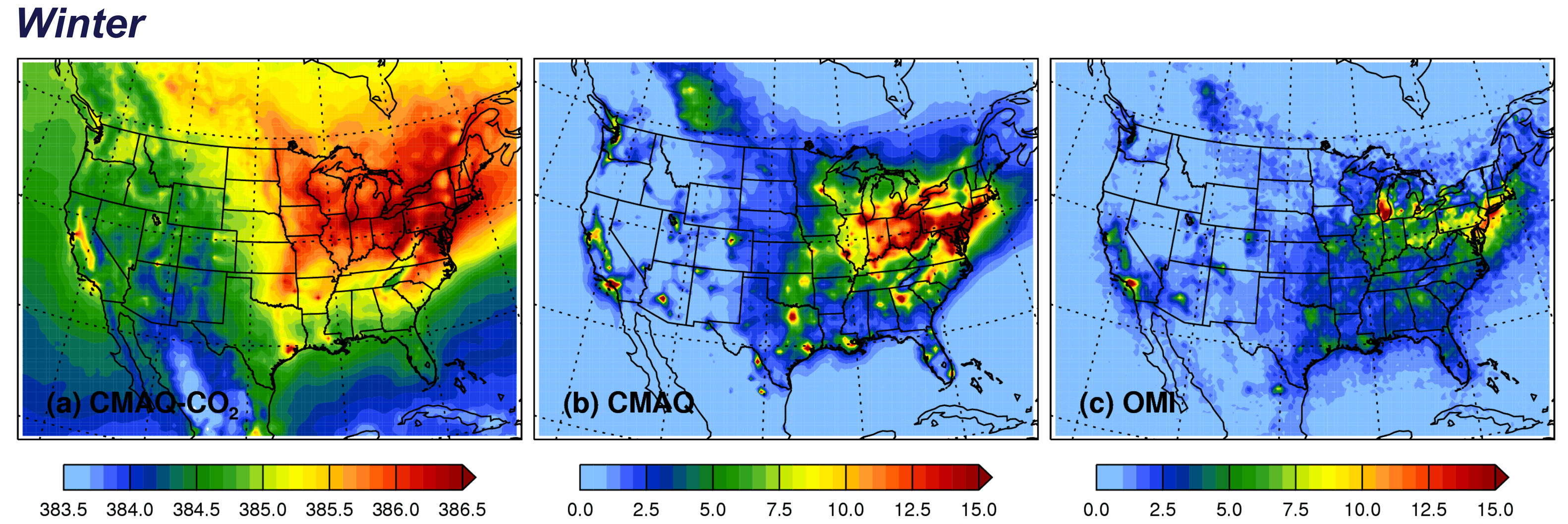


Figure 6. Same as Figure 5, but for December 2007.

Simulated Seasonal Variations of CO_2:NO_2,CO,SO_2 Spatial Correlations

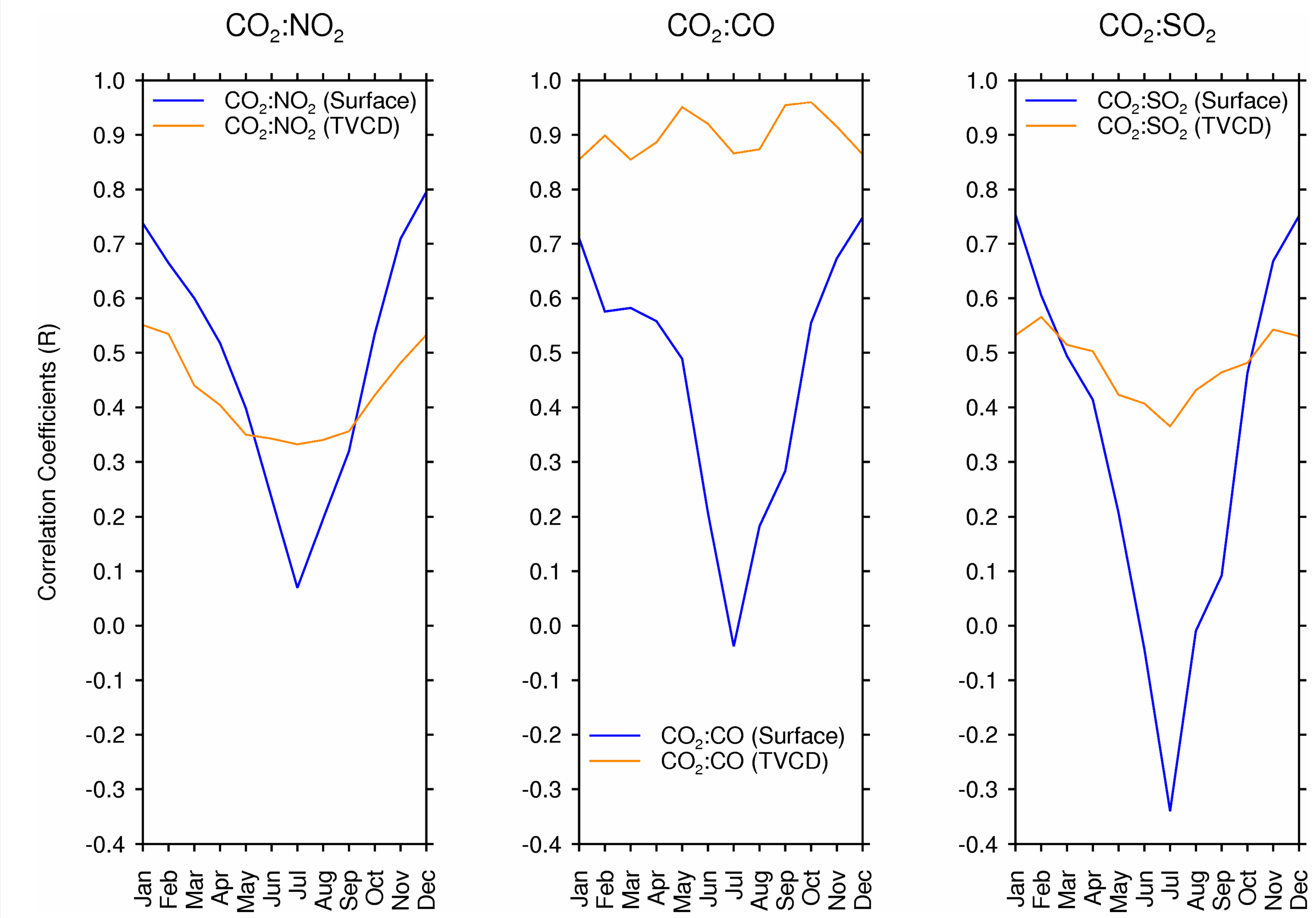


Figure 7. CMAQ simulated spatial correlation coefficients between CO_2 and NO_2, CO, SO_2, respectively. Correlations are examined for monthly mean surface concentrations (Surface) and tropospheric integrated vertical column densities (TVCDs) sampled from CMAQ native grids at 14:00 local time.

- ❑ Among all combinations, the best correlation is found for CO_2:CO TVCDs
- ❑ In all cases but one (CO_2:CO TVCD), the best correlation is in winter and the worst in summer
- ❑ R between vertical column densities show much weaker seasonality than surface concentrations

Ongoing and Future Work

- ❑ Further understanding the spatiotemporal variability of CO_2 (the role of fluxes and transport)
- ❑ Modeling CO_2 emissions together with air quality species using SMOKE
- ❑ Developing a multi-species flux inversion framework
- ❑ Performing OSSEs to assess the information gained from multi-species flux inversion

References

[1] Liu, Z. et al. (2013) *J. Air & Waste Manag. Assoc. CMAS-2012 special issue.*, in press.
[2] Palmer, P. I. et al. (2006) *J. Geophys. Res.*, 111, D12318.
[3] Wang, H. et al. (2009) *Atmos. Chem. Phys.*, 9, 7313-7323.

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