Assessing the ability of OMI NO_2 to detect interannual variability and trends in anthropogenic versus natural sources of NO_x

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1. Motivation

- The Ozone Monitoring Instrument (OMI) on the Aura satellite has provided daily, near-global coverage of total and tropospheric columns of NO_2 since Oct 2004 (Bucsela et al., 2006). Its data has been extensively used to constrain anthropogenic and natural sources of NO_x (e.g., Martin et al., 2006, Boersma et al., 2008, Hudman et al., 2012).
- Columns generally are dominated by anthropogenic (surface) sources. Anthropogenic sources tend to vary on longer time scales than "background" sources – e.g., lightning, soils, fires.
- Here, we exploit the near-decadal length of the OMI NO_2 record to determine if monthly and daily departures from climatological mean values can provide useful constraints on trends and daily, seasonal, and yearly variability in these smaller background sources.

2. Multi-year, global, sensitivity simulations: 2004-2012

Complete: An ensemble of 9-year base and sensitivity simulations in support of this and any other projects investigating recent trends and IAV in tropospheric composition.

- GEOS-Chem v9.02 global 3D chemical transport model (http://www.geos-chem.org)
- MERRA meteorology, at 2° × 2.5° resolution (also 4° × 5°)
- N. American Emissions: NEI2005 (US), CAC (Canada), BRAVO (Mexico)
- Base simulation, standard O_3-NO_x-CO-VOC-aerosol simulation, but
  - Lightning flash rates prescribed from National Lightning Detection Network (NLDN)
  - Monthly surface methane prescribed from kriged NOAA GMD flasks
- Sensitivity tests include
  - No US anthropogenic emissions ('US Background')
  - No North American anthropogenic emissions ('North American Background')
  - No global anthropogenic emissions + preind. CH_4 levels ('Natural Background')
  - No lightning emissions (NO_x)
  - No soil emissions (NO_x)
  - No fire emissions (NO_x, CO, VOCs, aerosols and precursors)
  - Zero emissions of individual states for individual episodes (e.g., WI, June 2007)
- Diagnostics archived for each scenario include
  - Monthly means of all available diagnostics
  - Daily 3-D NO_y budget terms saved at 10:00h LT (SCIAMACHY/GOME-2) and 13:30h LT (Aura)
  - Daily 24-h mean 3-D NO_y budget
  - Hourly 2-D surface concentrations of NAAQS species
  - Instantaneous sampling along field campaigns, passenger aircraft programs, and sonde profiles
  - North American boundary conditions for regional model simulations

References

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2. Bucsela, E J, E A Celarier, M O Wenig, et al., Algorithm for NO_2 vertical column retrieval from the ozone monitoring instrument, *IEEE Geosci Remote Sens Lett*, 44(5), 2006.
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4. Hudman, R C, N E Moore, A K Mebust, et al., Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints, *Atmos Chem Phys*, 12, 7779-7795, 2012.
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3. Reproduces dominant mode of interannual variability

- Model well-reproduces spatial, vertical and seasonal variability (not shown).
- EOF analysis demonstrates the ability of the model to reproduce the dominant observed mode of IAV: anthropogenic-driven decreases in the Northeast following emission reductions (e.g., Duncan et al., 2013).

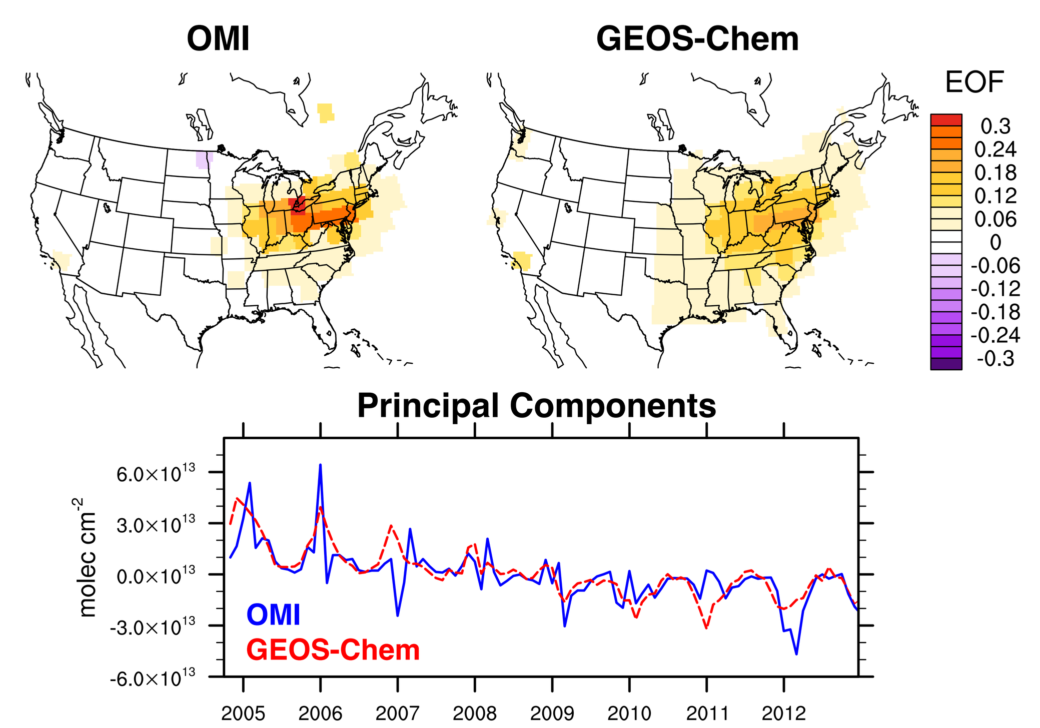

Figure 1: Maps show spatial patterns of dominant mode of variability in de-seasonalized monthly anomalies of tropospheric NO_2 columns from OMI (top left panel) and GEOS-Chem sampled during satellite overpasses (top right panel). The bottom panel shows the time series component of the change. The model and observations are highly consistent in time ($R^2 > 0.6$) and space.

4. Lightning dominates N American Background variability

- While anthropogenic emissions decline, background NO_2 increases in the model (especially since 2010), in part reflecting increases in lightning and soil emissions.
- Background increases are several times less than contemporaneous anthropogenic decreases, but occur in regions where columns are more sensitive to the background.

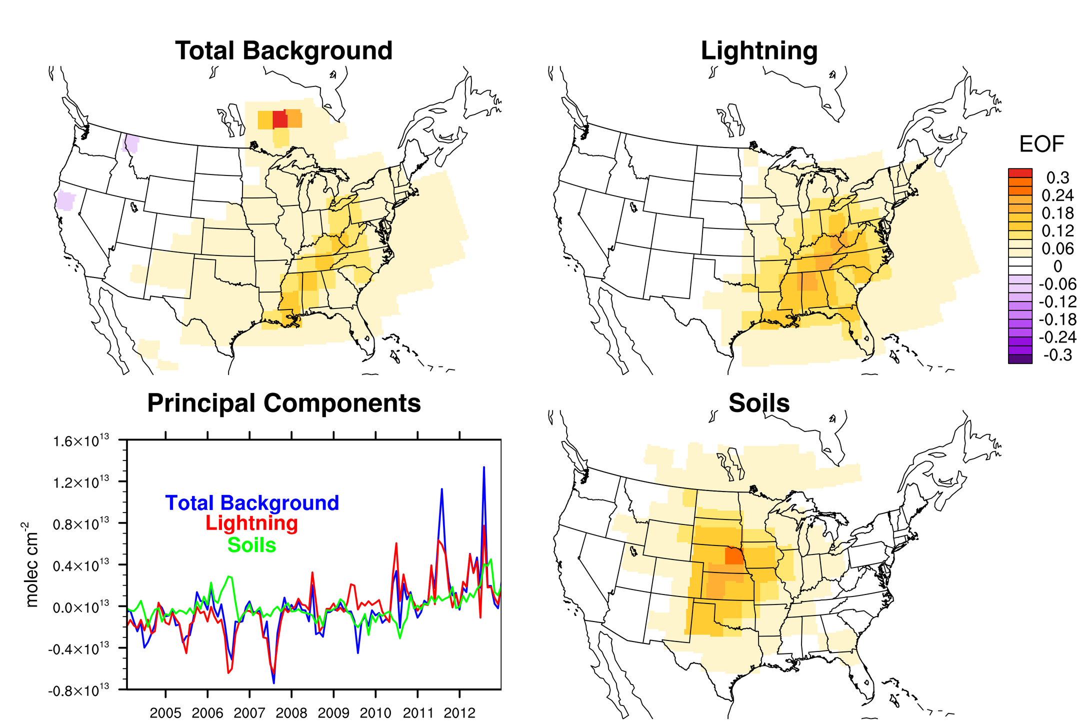

Figure 2: Dominant modes of variability in simulated contributions from lightning, soil, and the total North American background to tropospheric NO_2 columns. Lightning and soil contributions are estimated by subtracting a simulation with each source removed individually from a full simulation with all sources turned on.

- Spatial patterns of column sensitivity to lightning and soil emissions are relatively distinct from each other, and from the fossil fuel patterns (previous section).
- Modes with these natural spatiotemporal signatures not immediately apparent in lower EOFs of either OMI or GEOS-Chem full simulations.

5. Can NO_2 column anomalies constrain natural variability?

- Although anthropogenic sources dominate the mean tropospheric NO_2 columns, there are seasons (summer) and regions (southern and western) with half or more of their column NO_2 contributed by background sources.

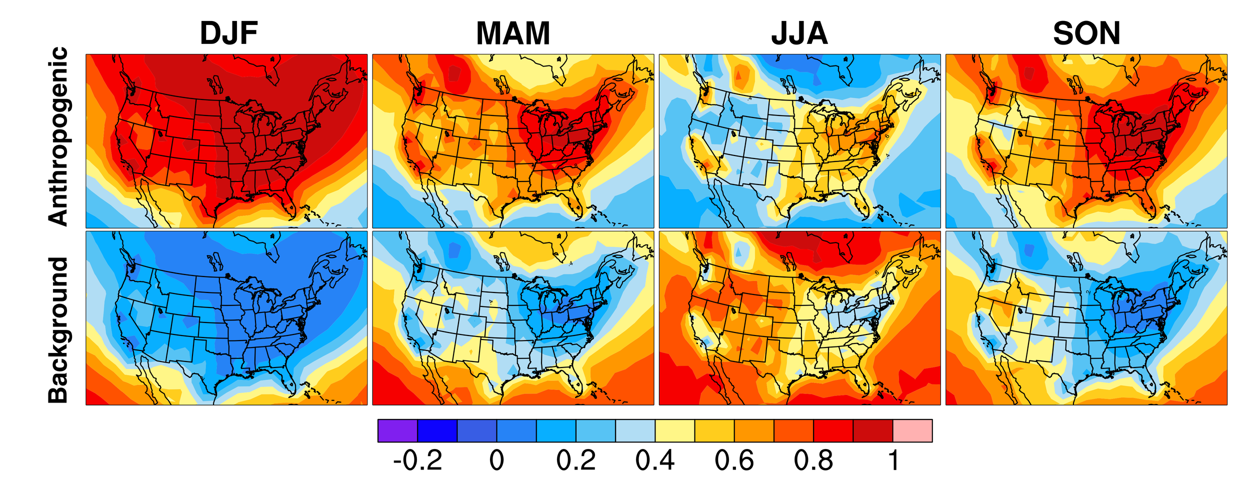

Figure 3: Simulated fraction of the mean tropospheric NO_2 column by source and season.

- Futhermore, there are regions and seasons where the variability in the anomalies from natural processes exceeds that from anthropogenic sources; can we exploit this variability to constrain natural processes?

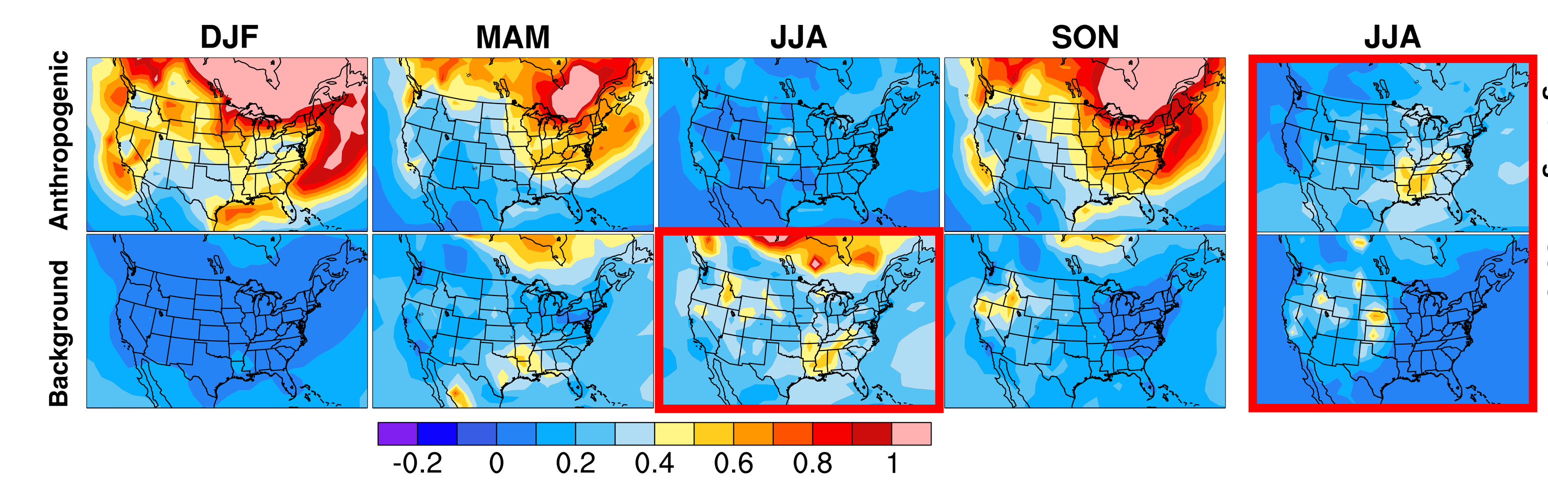

Figure 4: Ratio of standard deviation in daily columns to total ($\sigma_{\text{source}}/\sigma_{\text{total}}$), aggregated by season

- Best candidates will exhibit strong sensitivity of column to background processes (Fig. 5, left panel), while regions whose monthly anomalies are already well correlated with OMI will offer most immediate information (middle panel).

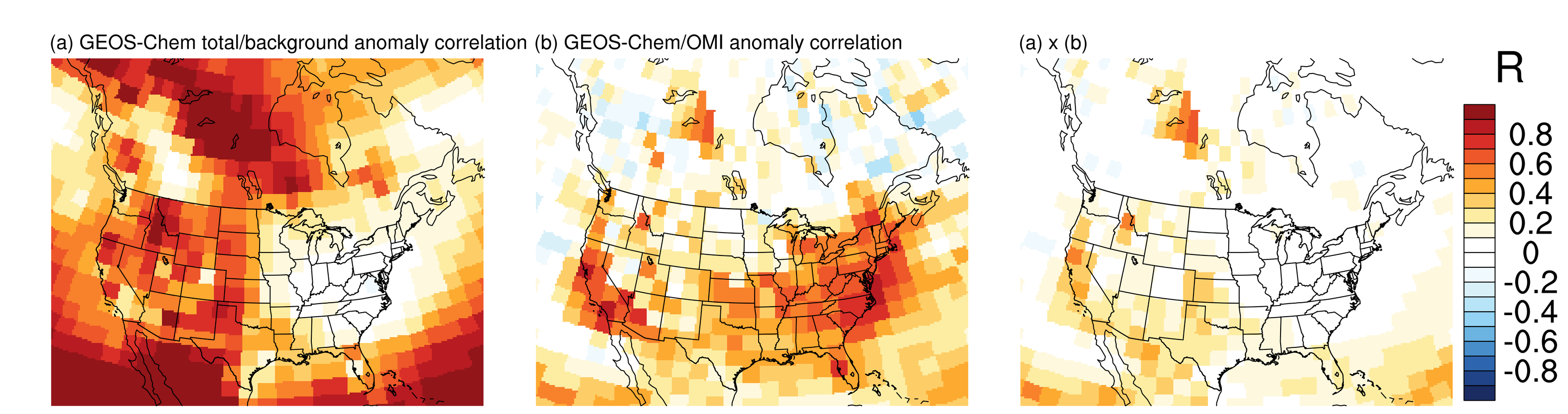

Figure 5: Candidate regions of North America for constraining background processes.

- Areas that exhibit both properties (Fig. 5, right; N Florida, Mexico, N Calif., Idaho, W Central Plains, Canadian taiga) will be targeted for local time series analyses.
- Lastly, note that North America is relatively unique globally in model skill; GEOS-Chem poorly reproduces IAV in most non-urbanized areas (Fig. 6), affirming need for better constraints on variability in natural sources of NO_x.

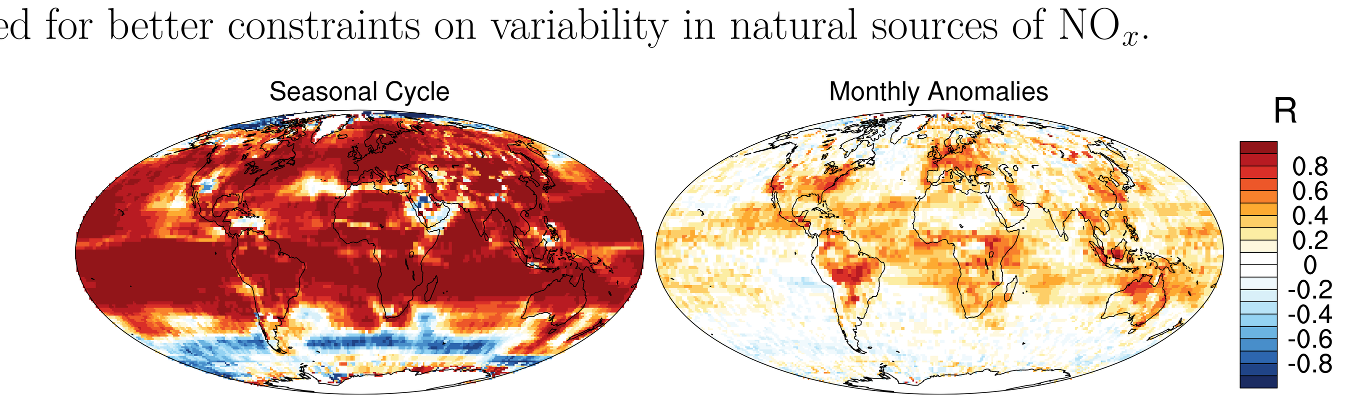

Figure 6: Local GEOS-Chem/OMI correlations in NO_2 climatological seasonality (left) and in IAV (right)