Modeling decadal trends in continental US air pollution, with a focus on $\text{NO}_x$

Progress on Project 1, Objective 1

Rachel Silvern, Loretta Mickley, Daniel Jacob
Atmospheric Chemistry Modeling Group, Harvard University
Large uncertainties in EPA National Emission Inventory (NEI) NO\textsubscript{x} emissions used in models

Fuel-based inventory shows NEI 2011 2.2x too high

- Diesel EF within ~20%
- Gasoline EF high by factor of ~2

Medians vertical profiles from SEAC\textsuperscript{4}RS aircraft campaign show impact of reducing NEI NO\textsubscript{x} emissions by 60%

Can we use satellite observations of NO\textsubscript{2} columns to better understand US NO\textsubscript{x} emissions and their trends?
US NO\textsubscript{x} emissions show linear decrease while satellite NO\textsubscript{2} observations show slowdown

Jiang et al. (2018) argue discrepancy in trends must be due to emission errors

Do we expect the response of NO\textsubscript{2} columns to changes in emissions to be constant over time?

EPA NO\textsubscript{x} emissions = -6.4\% a\textsuperscript{-1}  
OMI NO\textsubscript{2} columns = -8.8\pm1.0\% a\textsuperscript{-1}  
EPA NO\textsubscript{x} emissions = -5.3\% a\textsuperscript{-1}  
OMI NO\textsubscript{2} columns = -1.7\pm1.4\% a\textsuperscript{-1}
High resolution North American GEOS-Chem simulations

- 2004-2015 simulations with consistent meteorology
- Updated chemistry for isoprene and monoterpenes (Travis et al., 2016; Fisher et al., 2016) and SOA formation (Marais et al., 2016)
- 2011 National Emission Inventory scaled by species and year; 60% NO$_x$ emission scaling

Daily surface output passed off to Joel Schwartz (Project 1)

Model results can be used as validation for Project 5
Anthropogenic NO\textsubscript{x} emissions decreasing linearly, natural sources show large interannual variability.
Observed nitrate wet deposition suggest GEOS-Chem captures decadal US NO$_x$ surface emissions

**US nitrate wet deposition, summertime (JJA) 2004-2015**

60% anthropogenic NO$_x$ emission scaling required to reconcile modeled and observed nitrate wet deposition
Previous work on satellite NO$_2$ trends did not account for importance of the upper troposphere.

Scattering by surface and atmosphere

Satellite observed slant column of backscattered solar radiation

Tropospheric NO$_2$ vertical column density

Need information about vertical distribution of NO$_2$

Scattering Weight

GEOS–Chem NO$_2$ vertical profile (10$^{14}$ molecules cm$^{-2}$)  

Normalized NO$_2$ profile

Silvern et al. (2018); For more see our Project 1 poster
GEOS-Chem can generally reproduce flattening trend due to decreasing contribution from the boundary layer as emissions decline.

Flattening trend in satellite NO$_2$ columns in part due to increasing importance of the upper troposphere, not recognized previously.