Decadal trends in US OMI NO$_2$ observations and the role of the upper troposphere

Rachel Silvern
Atmospheric Chemistry Modeling Group, Harvard University

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Previous work showed 60% reduction of EPA National Emission Inventory (NEI) NO\textsubscript{x} required to reconcile GEOS-Chem with observations.

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

Emission reduction consistent with OMI NO\textsubscript{2} observations.

Remaining model bias compared to satellite observations due to underestimate of NO\textsubscript{2} in upper troposphere.
Observations show elevated NO$_2$ concentrations in the upper troposphere not captured by models

- GEOS-Chem overestimates the NO/NO$_2$ ratio by over a factor of 2 in the upper troposphere.
- Boundary layer (<2 km) accounts for only 20-35% of the total NO$_2$ column, the upper troposphere (>8 km) accounts for 35-50%.
Conversion of NO to NO\(_2\) balances only half of NO\(_2\) photolysis and cannot be explained by errors in GEOS-Chem radical concentrations.

NO-NO\(_2\) cycling in the upper troposphere during SEAC\(^4\)RS

Underestimate in peroxy and BrO radicals would have to be factor of 5 and 21 to close NO-NO\(_2\) budget

Silvern et al. (2018)
Errors in model NO$_2$ concentrations in the upper troposphere results in biases in the interpretation of satellite observations

- GEOS-Chem may underestimate NO$_2$ due to kinetic errors or would not if a missing organic NO$_x$ reservoir were present.
- GEOS-Chem simulates upper troposphere lightning NO$_x$ consistent with observations.
- If observed NO$_2$ is correct, the NASA retrieval may be biased high by 30%.

Silvern et al. (2018)
US NO\textsubscript{x} emissions show linear decrease while OMI NO\textsubscript{2} observations show slowdown.

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

Assume 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}
Contribution of natural sources to total NO$_x$ budget varies by region and season.

Seasonal mean NO$_x$ emissions in GEOS-Chem, 2004-2015

- Anthropogenic
- Fires
- Lightning
- Soil
- Fertilizer

Midwest (37-47°N, 96-80.5°W)

Northeast (37-46°N, 80.5-70°W)

West (33-49°N, 124-109°W)

Southeast (30-37°N, 94.5-76°W)
Nitrate wet deposition shows 60% anthropogenic NO\textsubscript{x} emission scaling applies to 2004-2015 summertime and consistency with linear EPA trend

- Agreement consistent across US for summertime
- GEOS-Chem underestimates nitrate wet deposition by 20-30% in seasons other than summer indicating emissions or NO\textsubscript{x} lifetime errors
We simulate daily NO$_2$ profiles to improve representation of the upper troposphere in interpretation of OMI observations.

Standard retrievals underestimate NO$_2$ in the upper troposphere.

Sample GEOS-Chem at local OMI overpass time.

Calculate new air mass factor with GEOS-Chem NO$_2$ profile.
GEOS-Chem can generally reproduce observed flattening trend due to decreasing contribution of 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.
Implications for TEMPO

- \( \text{NO}_2 \) in the free and upper troposphere makes a large contribution to the total \( \text{NO}_2 \) vertical column observed from satellites
  - TEMPO retrieval will need to properly account for upper tropospheric \( \text{NO}_2 \)
  - Large discrepancy between observed and modeled \( \text{NO}_2 \) in the upper troposphere, which may bias satellite retrievals

- Cloud-slicing technique developed for OMI \( \text{NO}_2 \) (Choi et al., 2014; Belmonte Rivas et al., 2015) could be leveraged by TEMPO to observe upper tropospheric \( \text{NO}_2 \) with higher temporal and spatial resolution
Current cloud-sliced NO$_2$ products provide constraint on upper tropospheric NO$_x$ but only at coarse spatial resolutions.

Correlations between upper troposphere NO$_2$ and aircraft observations:

TEMPO could offer improved constraints on upper tropospheric NO$_2$ at higher spatial and temporal resolution than current cloud-slicing products.

OMI-derived seasonal mean lightning NO$_x$ production rate:

Marais et al., submitted