Nitrate Wet Deposition as a Constraint on NO_x Emissions and Impacts on Modeled Secondary Pollutants

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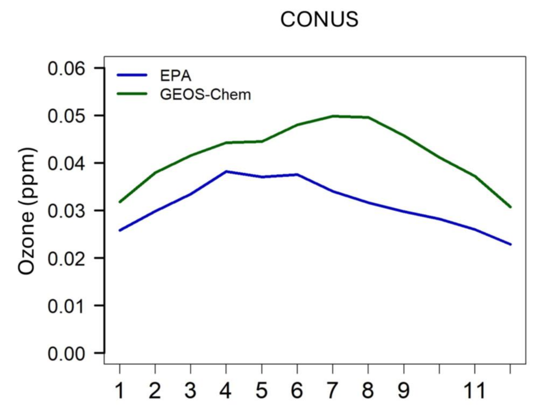
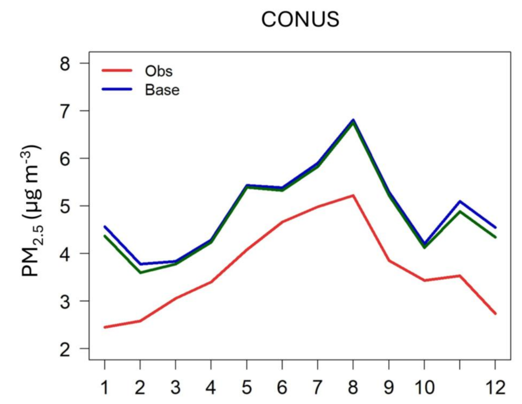
Introduction

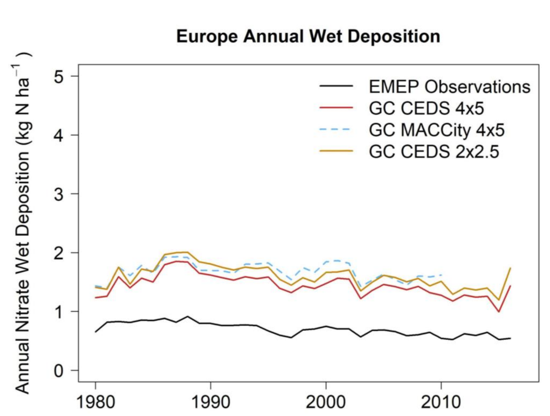
Current chemical transport models struggle to reproduce observations of secondary pollutants such as ozone and fine particulate matter ($PM_{2.5}$).[1-3]



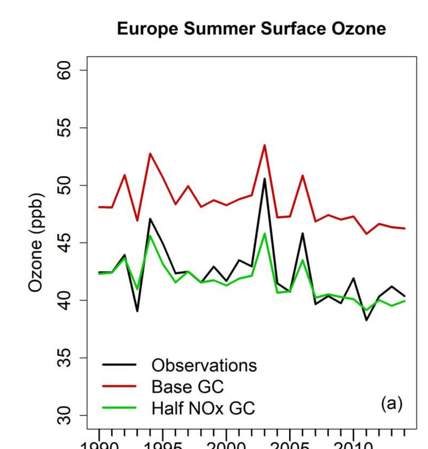


Emissions are a major uncertainty in models, impacting secondary pollutant reproduction.[4]

Prior work has identified nitrate wet deposition (NWD) as a useful constraint on emissions of nitrogen oxides (NO_x = NO + NO_2) that has improved model ozone reproduction over Europe.[4]



Are NO_x emissions over Europe too high by ~2x? Let's try reducing anthropogenic NO_x by half…



Goals

The goals of this work are:

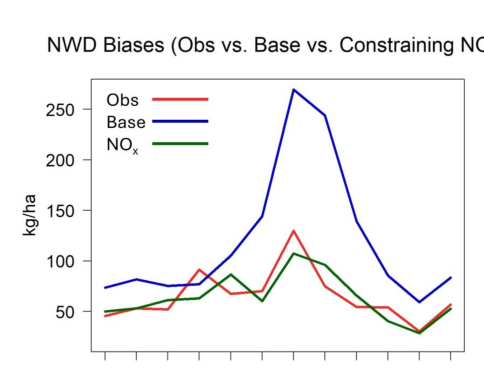
1. Expand the use of NWD as a NO_x constraint to the contiguous United States (CONUS) on a seasonal basis to improve ozone representation.
2. Expand this methodology to include other deposited species such as ammonium and sulfate to improve particulate matter representation.

Methodology

After NO_x is emitted, it is oxidized to nitric acid and partitions to aerosol nitrate. Both are efficiently scavenged by precipitation.

NWD can be related to NO_x emissions on a regional scale since the lifetime of NO_x is short.

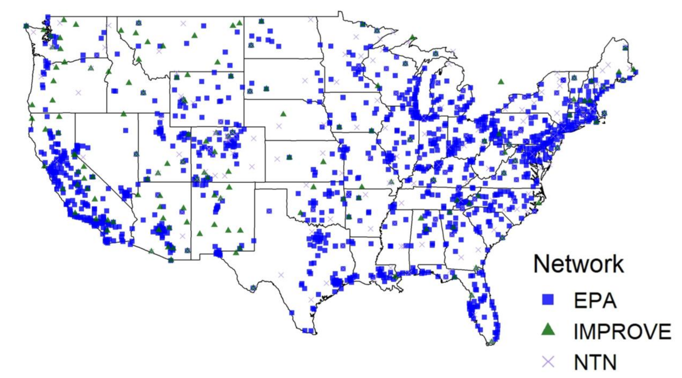
By combining NWD observations with GEOS-Chem simulations, we can assess if NO_x emissions or trends are biased in emissions inventories.



Season	NO_x Constraint from Model-Obs NWD Diffs
Winter (DJF)	0.66
Spring (MAM)	0.84
Summer (JJA)	0.43
Fall (SON)	0.52

Methodology

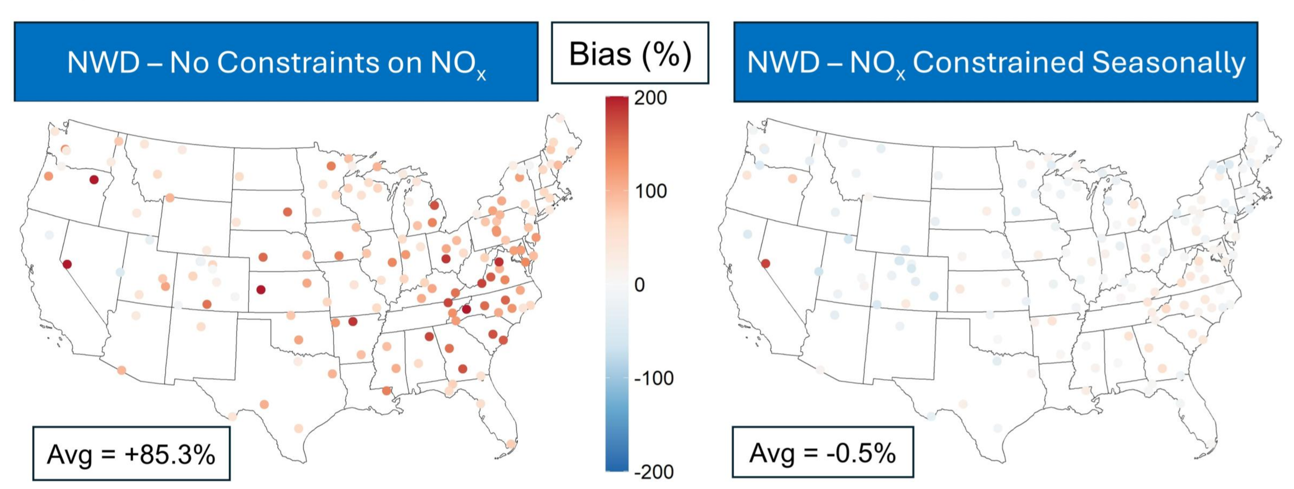
We use data from National Trends Network (NTN) for wet deposition;[5] Environmental Protection Agency (EPA) for ozone and particulate matter concentrations;[6] Interagency Monitoring of Protected Visual Environments (IMPROVE) for particulate matter and its chemical composition;[7] and GEOS-Chem version 14.4.2 for model-observation comparisons.[8]



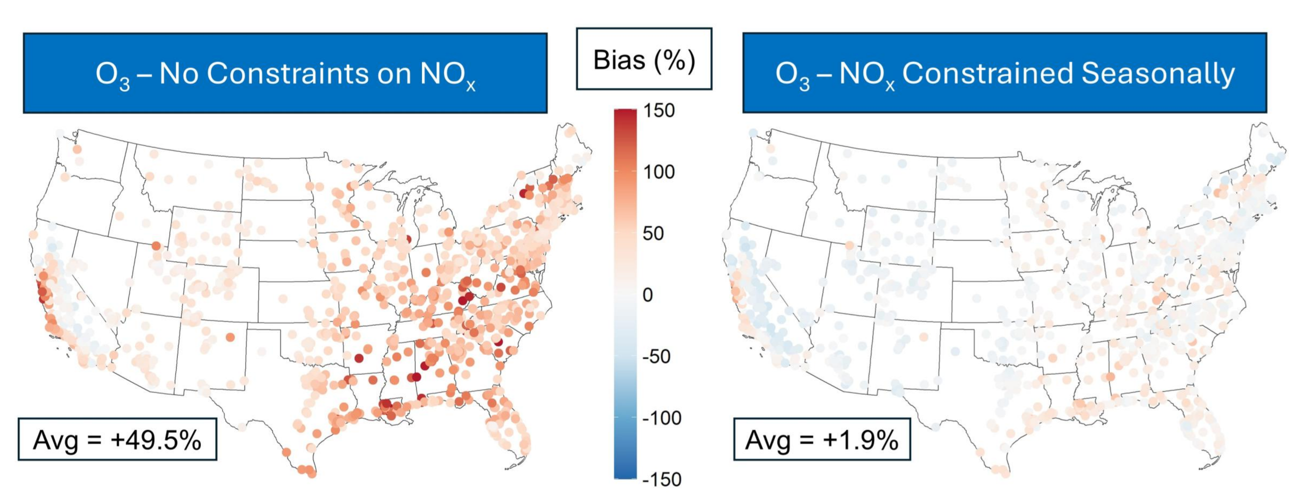
GEOS-Chem version 14.4.2	
Horizontal Resolution (latitude x longitude)	4°x5° for preliminary evaluations
Meteorology	MERRA-2
Anthropogenic Emissions	CEDS
Biomass Burning Emissions	QFED2

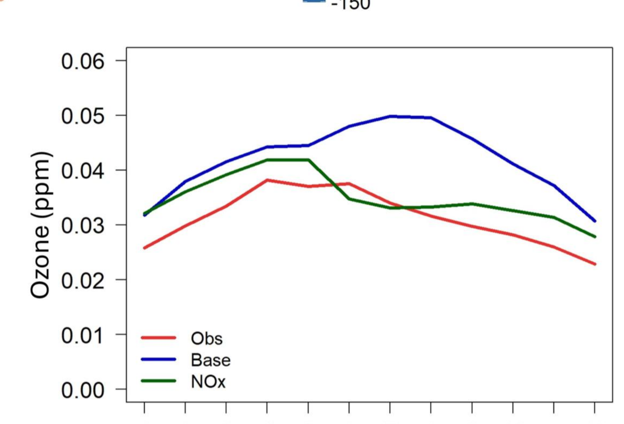
Results: 2016 as Test Year

Seasonal NO_x constraints reduce overestimation of annual NWD at NTN sites.



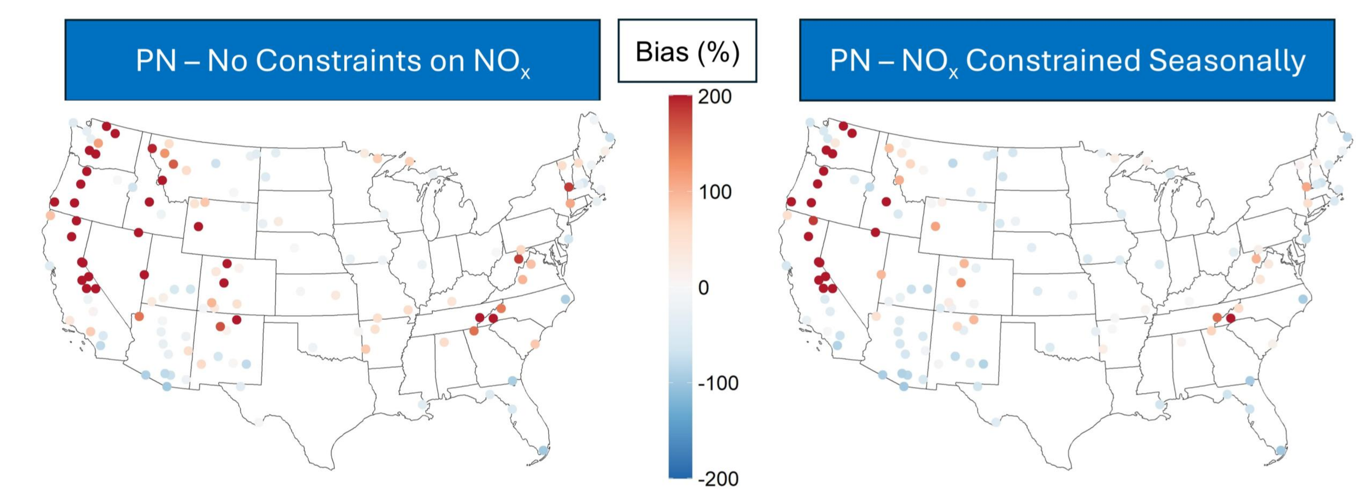
NO_x constraints are effective in reducing the overestimation of summertime ozone at EPA sites.



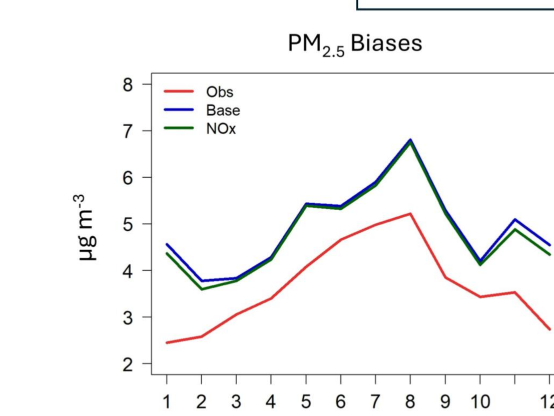


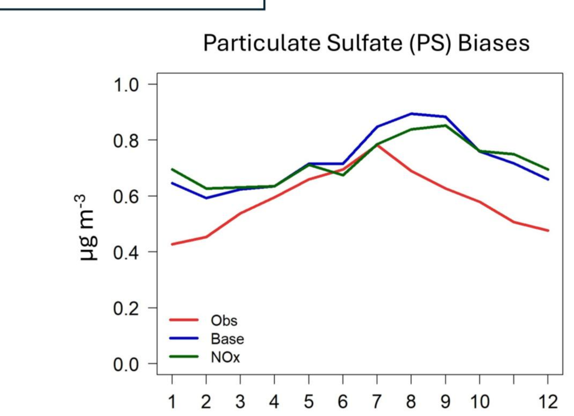
Results: 2016 as Test Year

NO_x constraints help marginally with annual particulate nitrate (PN) overestimates at IMPROVE sites, but $PM_{2.5}$ overestimates will require more or different constraints.



Avg bias reduction = 0.2 μg m^{-3} (95%)





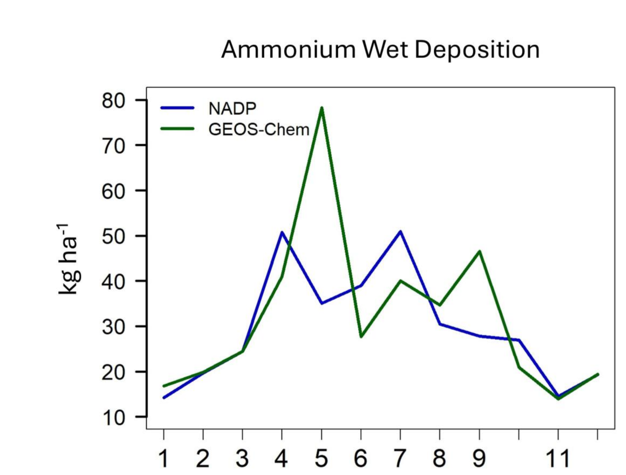
Conclusions and Future Work

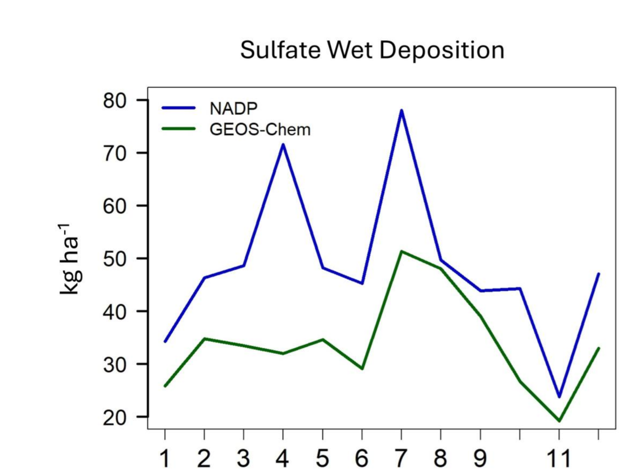
Using NWD as a constraint on NO_x emissions is effective for improving ozone representation across the CONUS.

Ozone biases are reduced by 48% at the surface during summer.

More work is needed to improve representation of $PM_{2.5}$ and its chemical components.

Other emissions constraints will be similarly implemented based on wet deposition of other inorganic species (ammonium, sulfate).





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