Contrasting Spatial Responses in Air Pollution to Hourly Anthropogenic Emissions in the Contiguous United States

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Research Questions

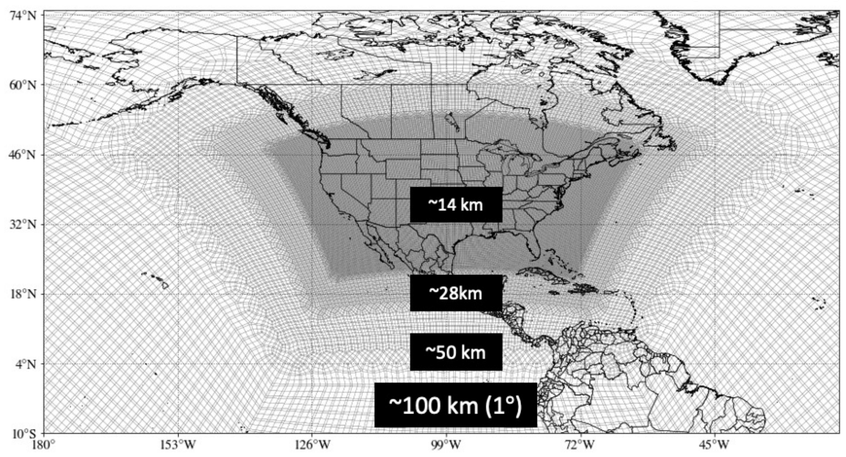
Motivation: Accurate simulations of air pollutant concentrations and variations are essential for model applications in attributing pollution sources and assessing exposure risks, with anthropogenic emissions being an essential component.

For July 2018 over the Contiguous U.S. (CONUS):

How do the temporal characteristics of anthropogenic emissions, including weekday versus weekend differences and diurnal patterns, affect the concentrations of ozone (O_3) and its precursors, as well as their diurnal variations across different regions?

Methods

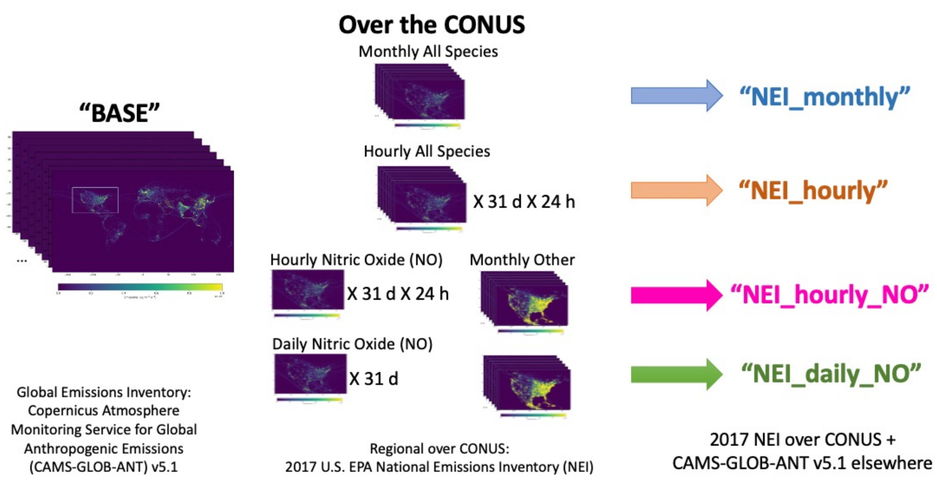
Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICAv0)



= CESM/CAM-chem-SE with ne0CONUSne30x8

(Pfister et al., *Bull Am Meteorol Soc*, 2020, Schwantes et al., *J Adv Model Earth Syst*, 2022)

Sensitivity Simulations



Impact of Incorporating Daily and Hourly Nitric Oxide (NO) Emissions

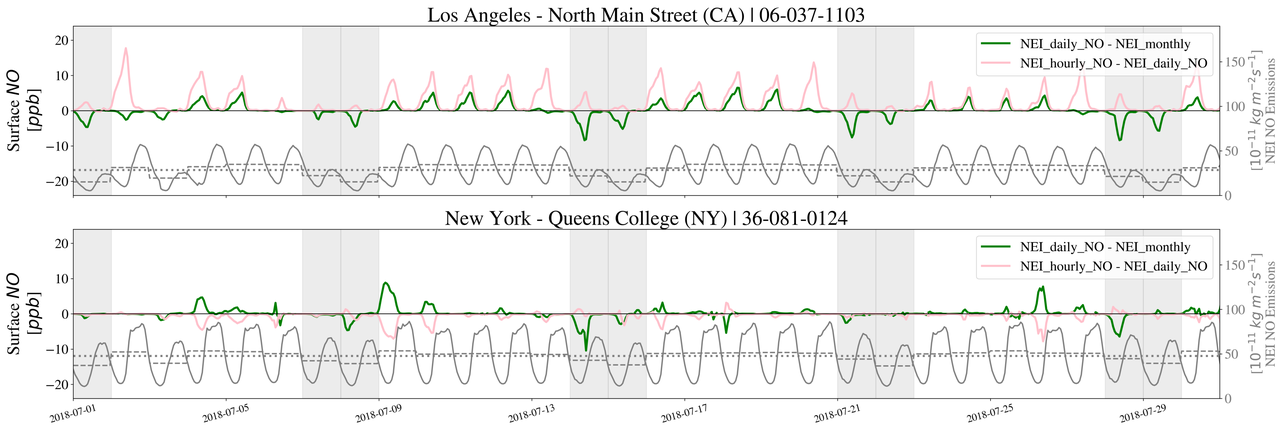
***Surface NO, NO_2, and O_3 concentrations respond differently despite similar changes in emissions diurnal cycles.**

- In West Coast urban regions such as Los Angeles, during daytime hours, monthly mean surface concentrations of NO and NO_2 increase by up to 8 ppb and 6 ppb, respectively, while O_3 decreases by up to 7 ppb.
- Conversely, in Northeast urban regions such as New York, monthly mean surface NO_2 during daytime hours decreases by up to 1 ppb, and O_3 decreases by up to 2 ppb. The changes in time mean are considerably smaller compared to those observed in Los Angeles.

***Spatial variations in the impact of including diel cycles in anthropogenic emissions on model evaluation with observations.**

- MUSICA captures some of the spatial variation (r_s > 0.5), but performance varies by region. Surface O_3 and column HCHO are biased high, while column NO_2 is typically biased low.
- MUSICA overestimates the daily range in both surface O_3 and NO_2 concentrations compared to SLAMS observations. The modeled weekday-weekend differences are more aligned with observed conditions after incorporating daily and hourly variations in emissions.

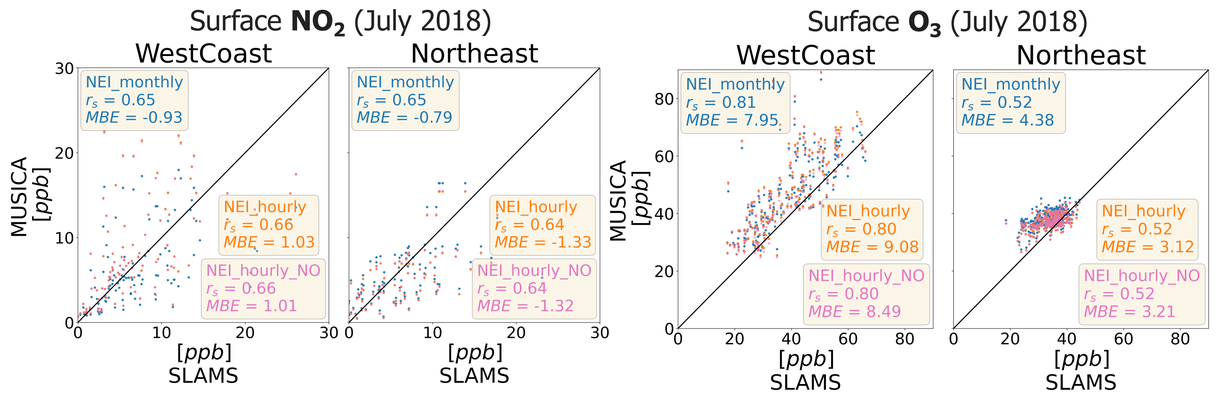
Changes in Surface NO Concentrations & NO Emissions in Different Temporal Resolutions



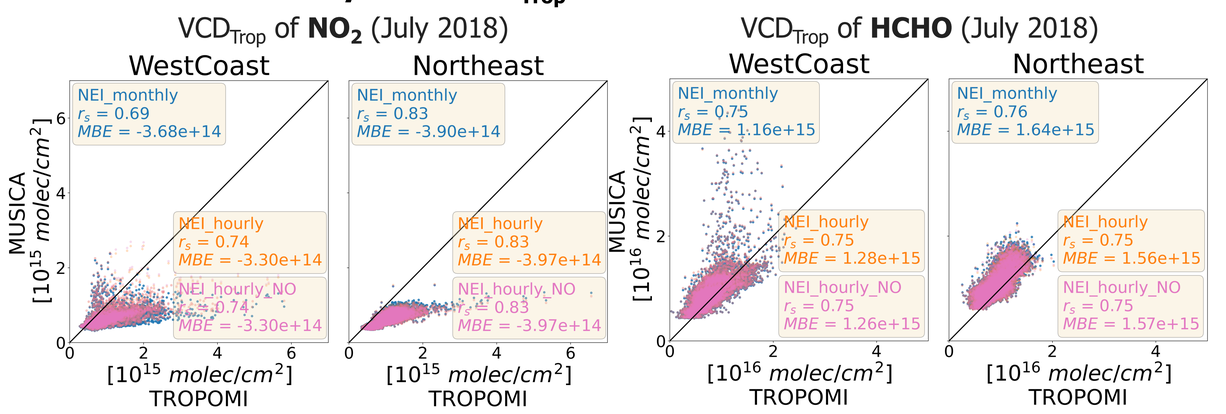
Changes in surface NO concentrations due to switching to daily (green: NEI_daily_NO – NEI_monthly) and then hourly (pink: NEI_hourly_NO – NEI_daily_NO) NO emissions, alongside hourly NEI NO emissions (gray) approximated at two urban monitoring stations. Weekends are shaded gray. **The shift from daily mean to hourly varying emissions typically increases daytime NO and decreases nighttime NO, with more pronounced differences on weekdays.**

Comparison of July mean surface concentrations (NO_2 and O_3; top) against State and Local Air Monitoring Stations (SLAMS) observations or tropospheric vertical column densities (NO_2 and HCHO VCD_{Trop}; bottom) against TROPOspheric Monitoring Instrument (TROPOMI) retrievals across West Coast and Northeast. Colors indicate different model simulations with Spearman's rank correlation coefficient (r_s) and mean bias error (MBE).

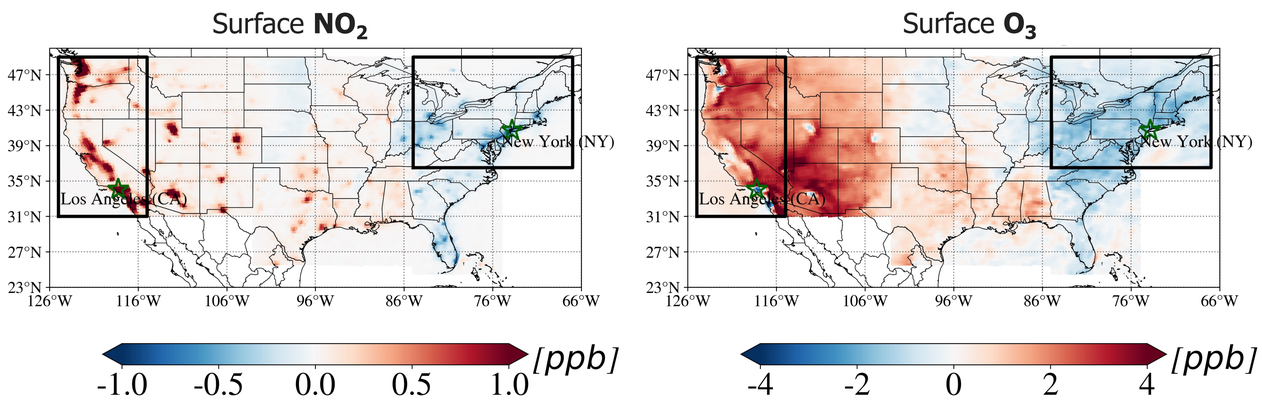
July Mean Surface Concentrations in MUSICA vs. SLAMS



July Mean VCD_{Trop} in MUSICA vs. TROPOMI

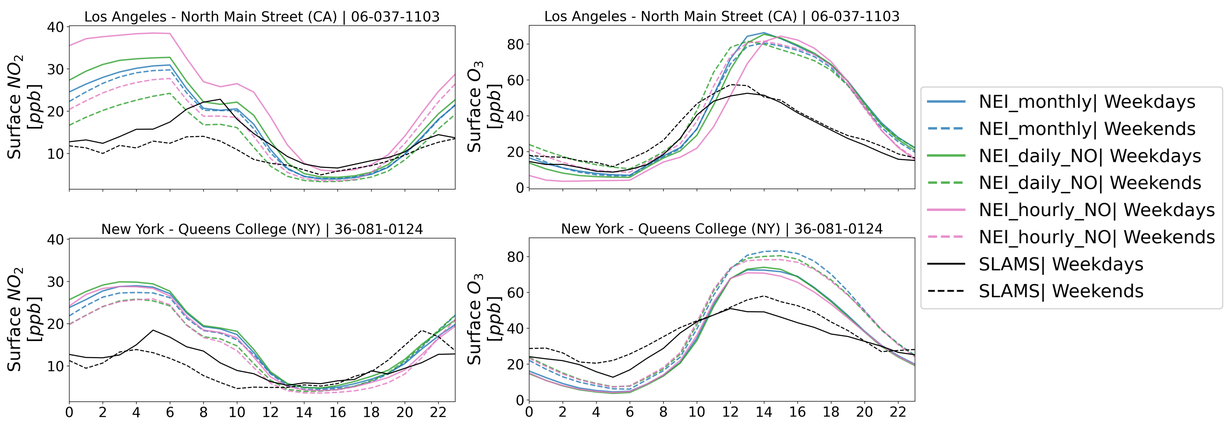


Daytime (9AM-5PM) July Mean NEI_hourly_NO – NEI_daily_NO



The spatial changes in NO (not shown) are similar to those of surface NO_2.

Hourly variations in surface NO_2 and O_3 concentrations from observations (SLAMS in black) **and model simulations** (colored for different simulations) averaged for July 2018, shown separately for weekdays (solid lines) and weekends (dashed lines).



Implications and Future Directions

- The sensitivity of monthly mean NO_x concentrations to the hourly variations in NO_x emissions underscores the importance of careful accounting for diurnal changes when inferring emissions from concentrations.
- Additional work is necessary to identify the specific factors driving these different regional photochemical environments.

in prep for submission to ACP

This work was supported by the NASA Health & Air Quality Applied Sciences Team (80NSSC21K0509).

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