Simulating Diurnal Variation of Fine Particulate Matter Composition over Southeastern US using GEOS-Chem

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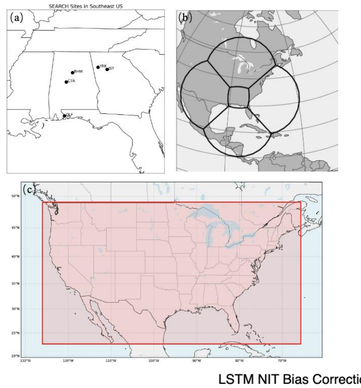
Introduction

Fine particulate matter, also known as $PM_{2.5}$, affects public health, visibility and Earth's climate system. Accurately representing the $PM_{2.5}$ variation over the course of a day, namely, diurnal variation, is essential for exposure assessment and air quality modeling. Although diurnal variation of total mass concentration of $PM_{2.5}$ has been widely measured and simulated at different times and scales, studies investigating the diurnal variation of its chemical composition are relatively rare.

Methodology

We simulates the diurnal variation of $PM_{2.5}$ composition over the Southeastern US for January and July 2015 using GEOS-Chem driven by the offline GEOS-FP meteorological datasets (GCHP) and coupled with the online WRF model (WRF-GC). The simulation are evaluated using hourly measurements of $PM_{2.5}$ composition from the Southeastern Aerosol Research and Characterization Study (SEARCH).



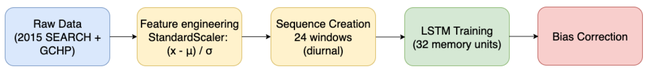
Urban Sites: JST, BHM

Rural Sites: YRK, CTR

Coastal Site: OLF

(a) Five SEARCH sites with hourly in-situ observations for mass concentrations of sulfate, nitrate, ammonium, organic matter and black carbon. (b) Stretch grid GCHP simulation focusing Southeaster US. (c) WRF-GC domain over CONUS.

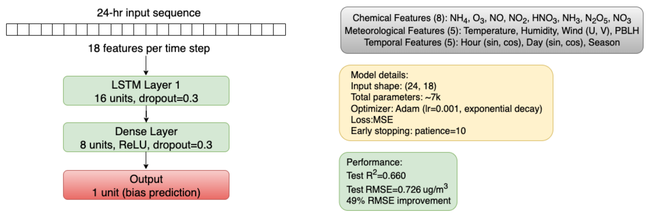
LSTM NIT Bias Correction Work Flow



Balanced Seasonal Split: Days 1-21 (train 70%) | Days 22-25 (Val 15%) | Days 26-31 (Test 15%)
Universal model (no site-specific features)

Target: Nitrate Bias = GCHP_NIT_2m - SEARCH_NIT (observed)

LSTM Model Architecture



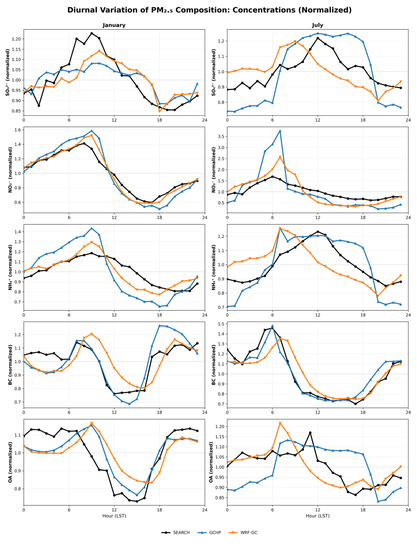
Chemical Features (8): NH_4, O_3, NO, NO_2, HNO_3, NH_3, N_2O_5, NO_3
Meteorological Features (5): Temperature, Humidity, Wind (U, V), PBLH
Temporal Features (5): Hour (sin, cos), Day (sin, cos), Season

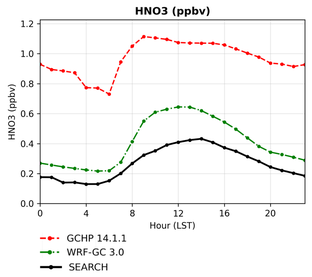
Model details:
Input shape: (24, 18)
Total parameters: ~7k
Optimizer: Adam (lr=0.001, exponential decay)
Loss:MSE
Early stopping: patience=10

Performance:
Test R^2=0.660
Test RMSE=0.726 ug/m^3
49% RMSE improvement

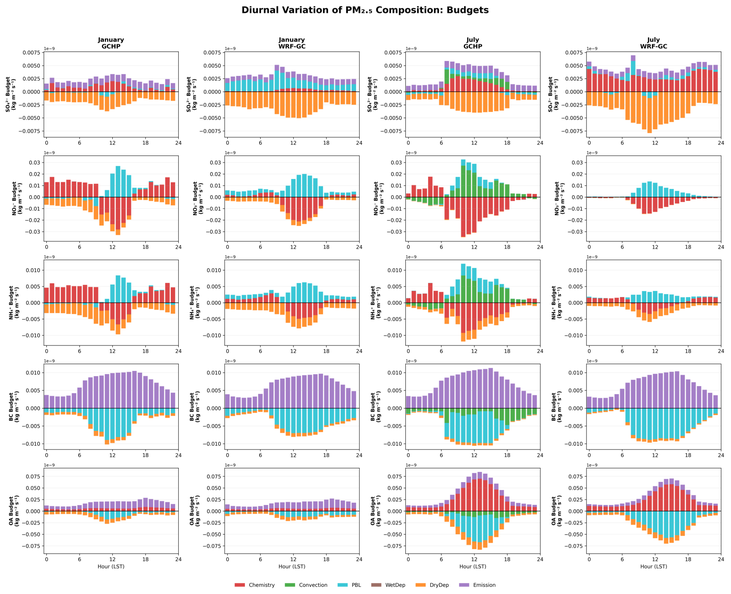
A LSTM deep learning frame work for reducing nitrate simulation bias in GCHP.

Diurnal $PM_{2.5}$ Composition & Budgets



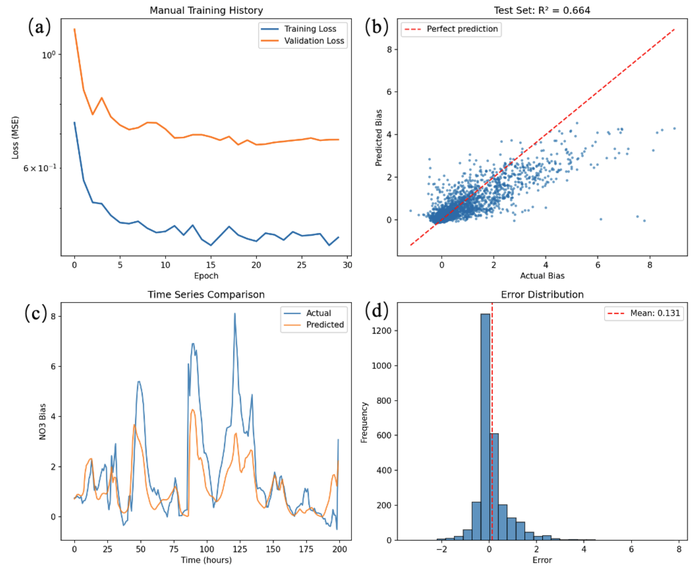


GCHP and WRF-GC exhibit mixed skill reproducing observed diurnal $PM_{2.5}$ composition. GCHP has higher positive biases on the overnight nitrate accumulation in July, likely contributed by overestimated HNO_3 concentration.

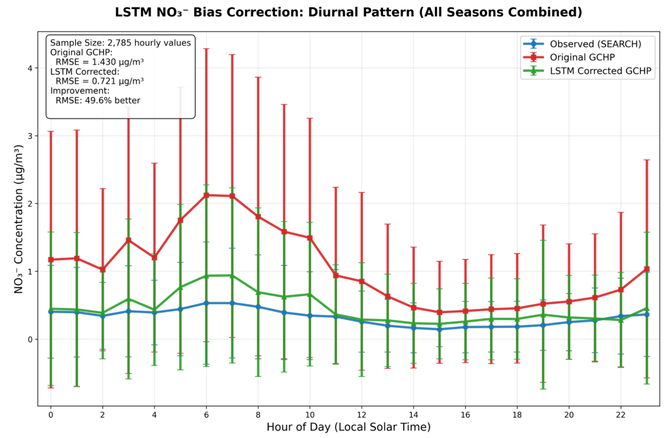


Both configurations reproduce the observed diurnal variation of black carbon and organic matter better than other secondary components, especially for January. Budget analysis indicates that the diurnal variation of black carbon and organic matter is mainly driven by the competition between emission, boundary layer mixing and chemical production. The diurnal variation of nitrate is mainly driven by bidirectional gas-aerosol thermodynamic partitioning and chemical production. The diurnal variation of sulfate is affected by fumigation, dry deposition, boundary layer mixing and chemical production.

Reducing Nitrate Bias in GCHP using LSTM



Both training loss and validation loss decreases along with epochs.



Improvements on diurnal variation of nitrate in GCHP

Summary

We conducted GEOS-Chem simulations on diurnal variation of fine particulate matter composition at ~27km resolution driven by the offline GEOSFP meteorological datasets (GCHP) and the online WRF model (WRF-GC) over Southeastern US. The simulations are evaluated using hourly in-situ measurements from SEARCH. Budget analysis is conducted to reveal the underlying driving forces. A LSTM deep learning approach is proposed to further reduce the modeling bias of diurnal nitrate in GCHP.