Joint assimilation of NO_2 and HCHO observations from TEMPO and TROPOMI for constraining NO_x and VOC emissions

- **Chia-Hua Hsu (CUB, NIES),** Daven K. Henze (CUB)
- Arthur P. Mizzi (NASA)
- Owen Cooper, Brian McDonald (NOAA CSL)
- Kelley Wells, Dylan Millet (UM)
- Jennifer Kaiser (GIT)

Improving US air pollution assessment and forecasting with TEMPO

Motives

- O_3 nonattainment (increasing in Denver)
- US air pollution health burden ~10^4 premature deaths/yr
- uncertainties in NOx, VOC sources
- limited air pollution forecast skill

Objectives

- Improve O_3 forecast and inventories / models of NO_x and VOC emissions
- Quantify benefits of joint assimilation and GEO vs LEO satellite data

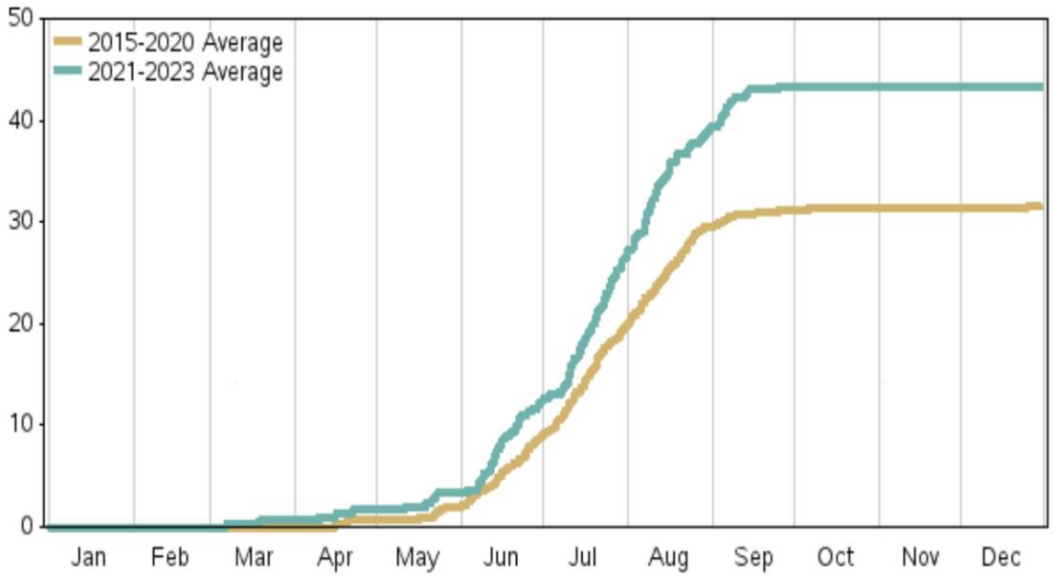

***Fig 3**: Cumulative number of day whin 8-r Ozone daily max > 70 ppb in the Denver-Aurora-Lakewood region of CO, based on all sites from U.S. EPA AirData (www.epa.gov/air-data, accessed July, 2024).*

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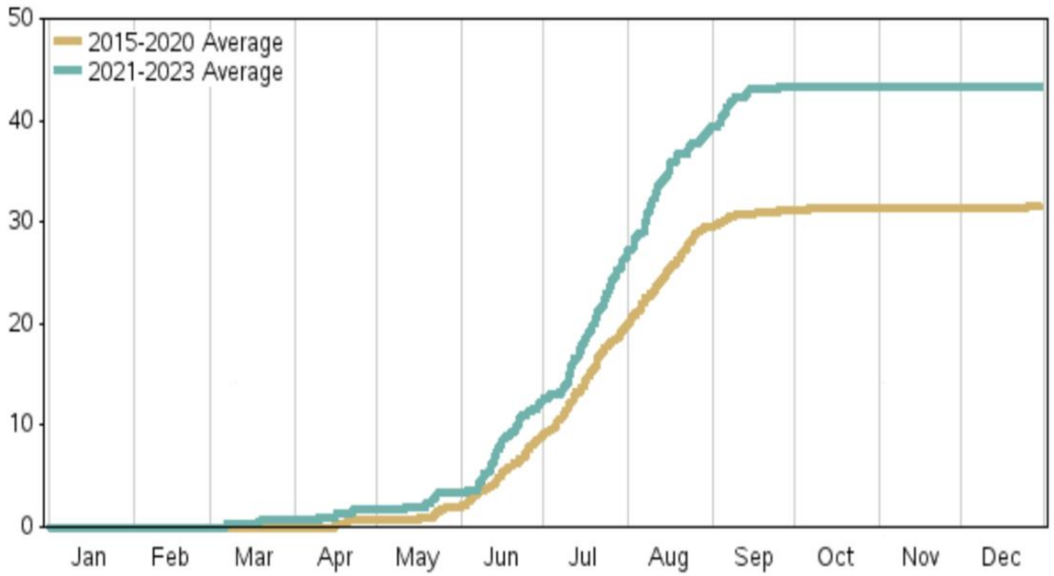

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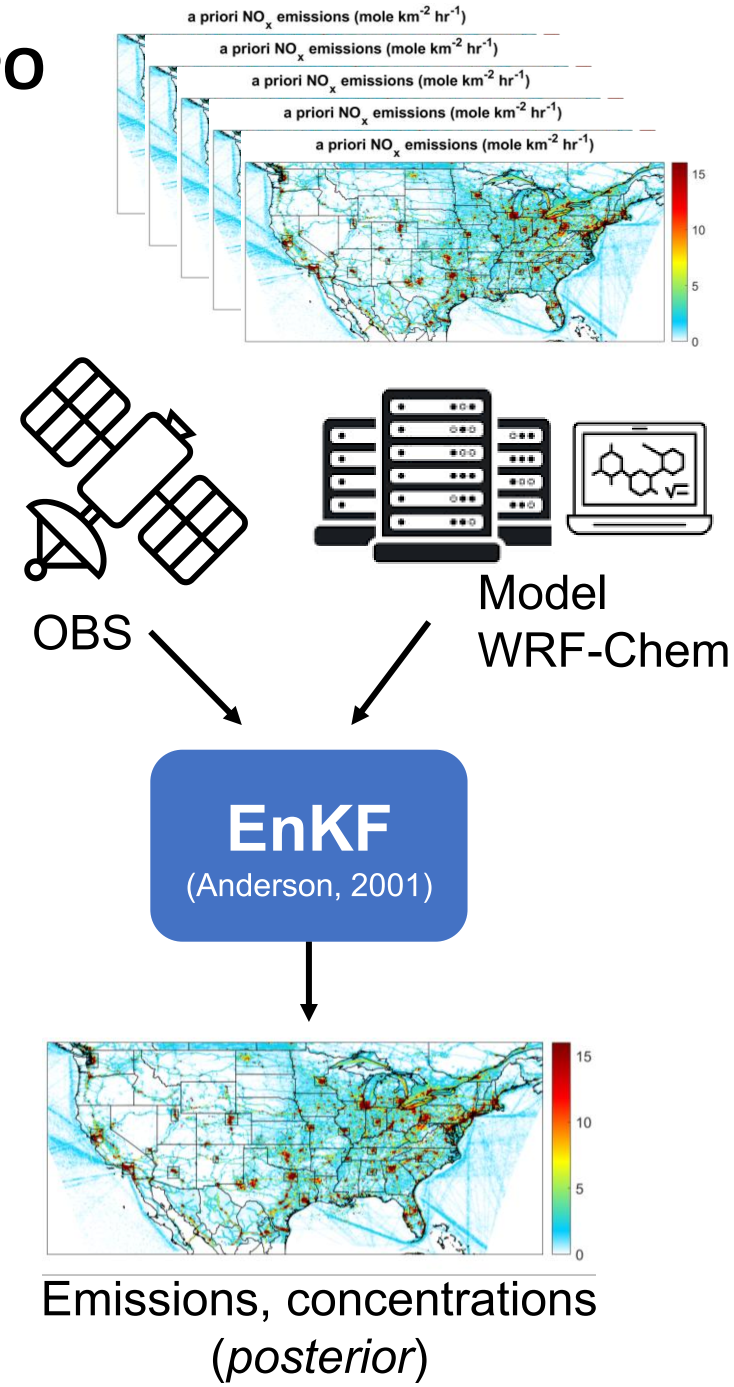

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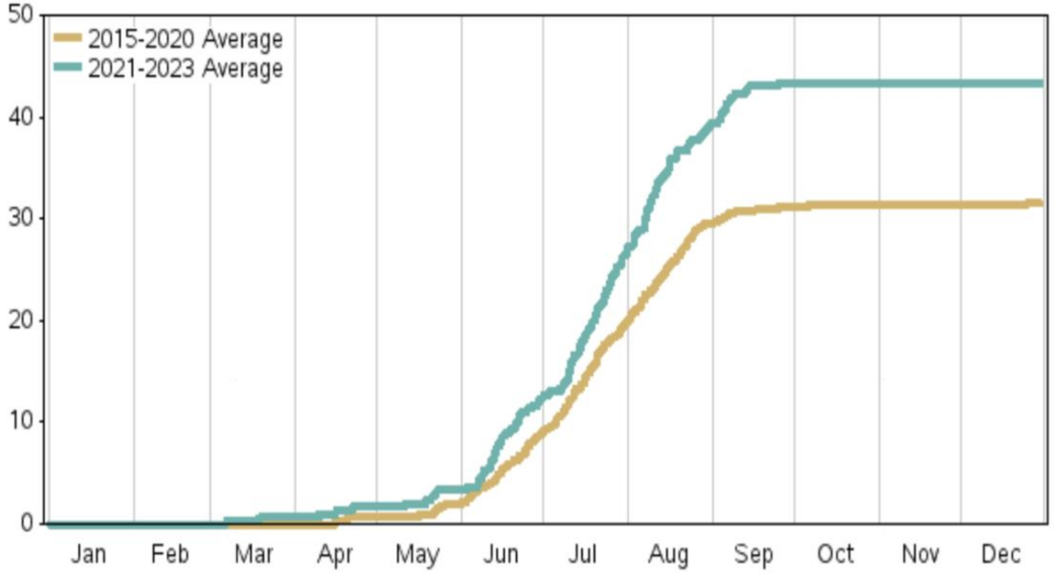

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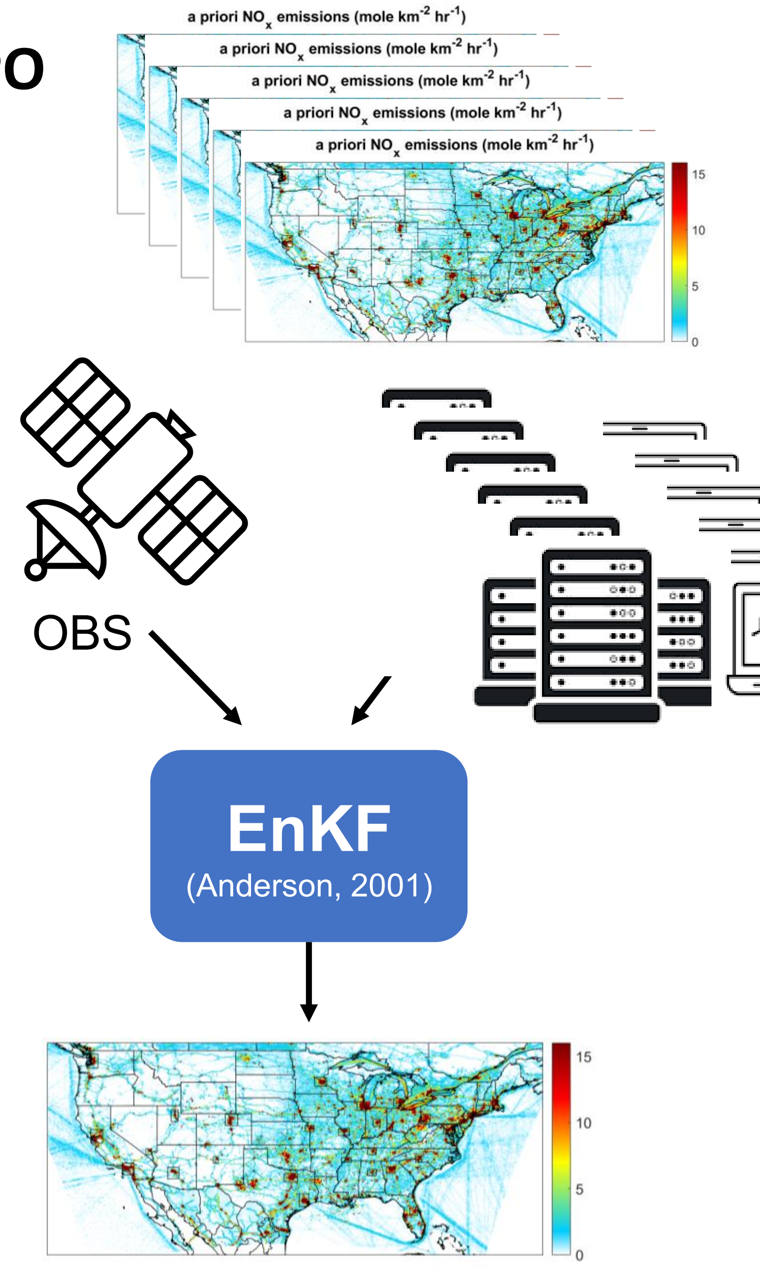


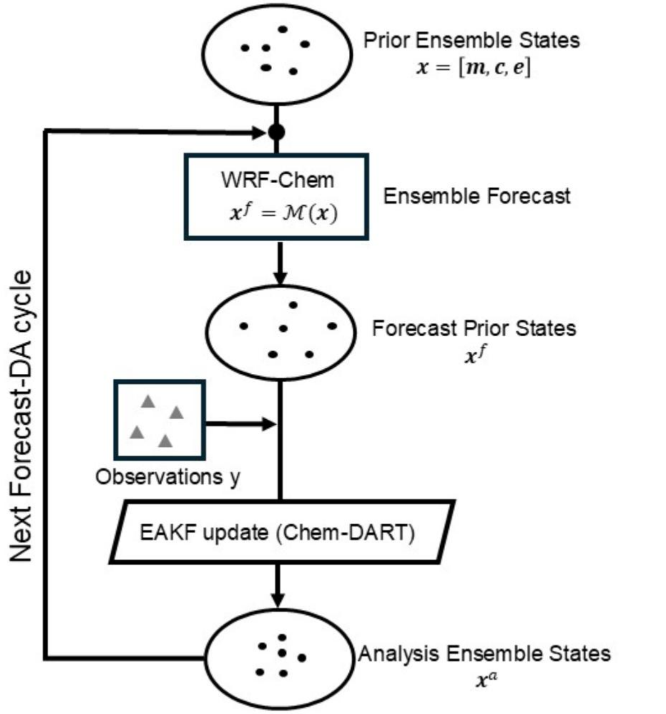
Emissions, concentrations (*posterior*)

Part I – An Observing System Simulation Experiment analysis of how well geostationary satellite trace-gas observations constrain NO_x emissions in the US (Hsu et al., *JGR*, 2024)

✓ Best practices for assimilating TEMPO NO_2 data (WRF-Chem/Chem-DART EnKF):

- short DA window (e.g., 30 min).
- high frequency assimilation cycling (e.g., 1 or 2 hours).
- **simultaneous chem and meteorological DA, while jointly constraining concentrations and emissions, mitigates model resolution and transport errors**

✓ TEMPO-exp retrieves true NO_x emissions at least twice as fast as TROPOMI-exp and provides more accurate NO_x emissions estimates.



Anderson (2001), Mizzi et al. (2016)

Part II – Top-down estimates of U.S. NO_x emissions using TEMPO and TROPOMI NO_2 remote sensing observations with WRF-Chem/Chem-DART (Hsu et al., *JGR*, 2026)

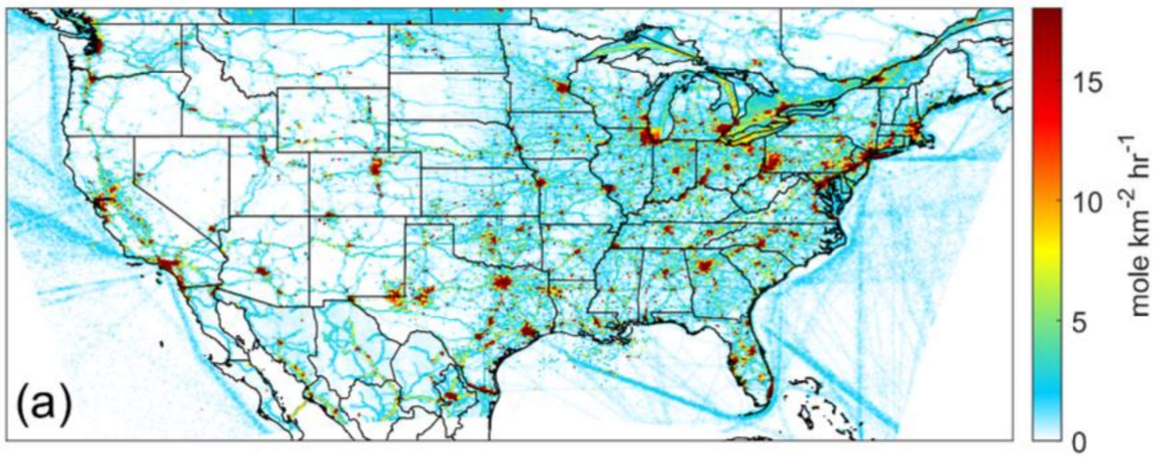
Launched — April 2023

First Light — August 2023

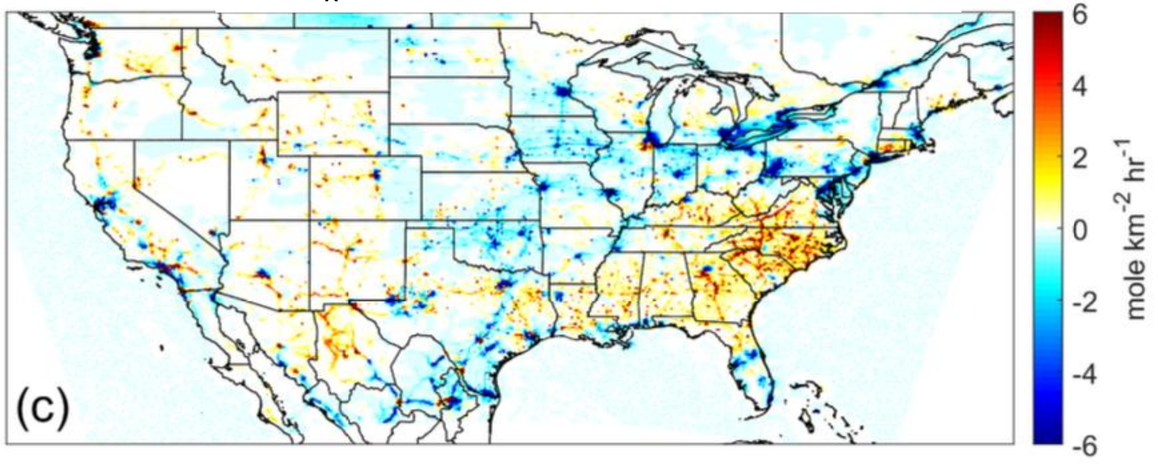
V03 Data Release — May 2024

Assimilation of TEMPO NO_2 (v03) for Aug 7 - 26, 2023

Initial NO_x emissions (a priori)



ΔNO_x emissions (posterior - a priori)



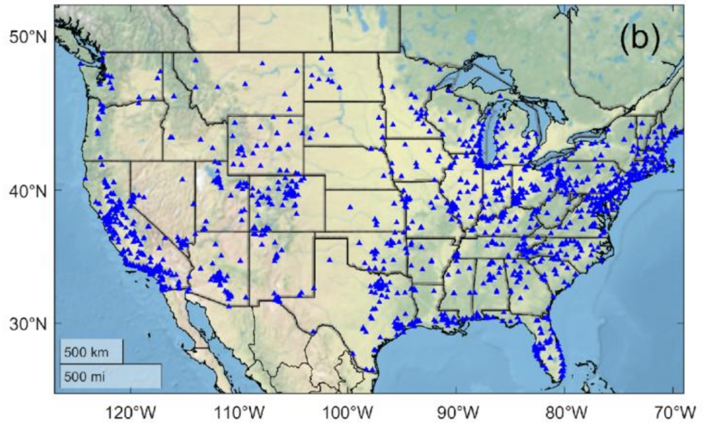
Model (Top-down Emissions) Evaluations

- 1. Aircraft measurement: DC-8

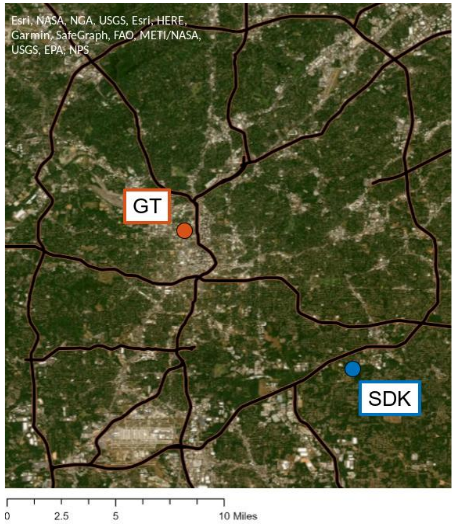
https://csl.noaa.gov/projects/aeromma/data.html

- 2. Ground-based measurements:

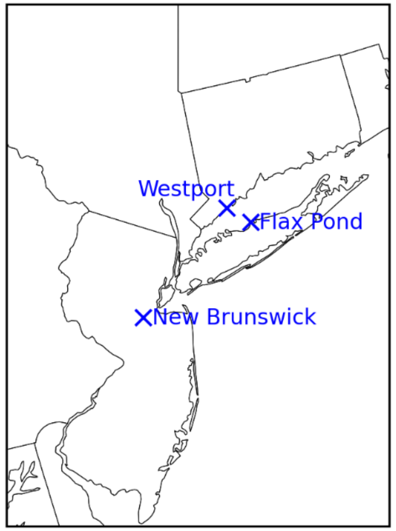
EPA AQS O_3, NO_2, isoprene



HCHO: Prof. Jennifer Kaiser (South Dekalb) & NASA STAQS 2023



Adopted from Masut et al. (2024)



3. Satellite isoprene retrievals (Cross-track Infrared Sounder)

https://satnews.com/2023/02/16/first-global-image-from-noaa-21s-cris-instrument/

4. Bottom-up inventories

GRA^{2}PES
Greenhouse Gas And Air Pollutants Emissions System

Anthropogenic emissions from NOAA CSL
Prior = GRA^{2}PES 2021.
Compare posterior to GRA^{2}PES 2023.

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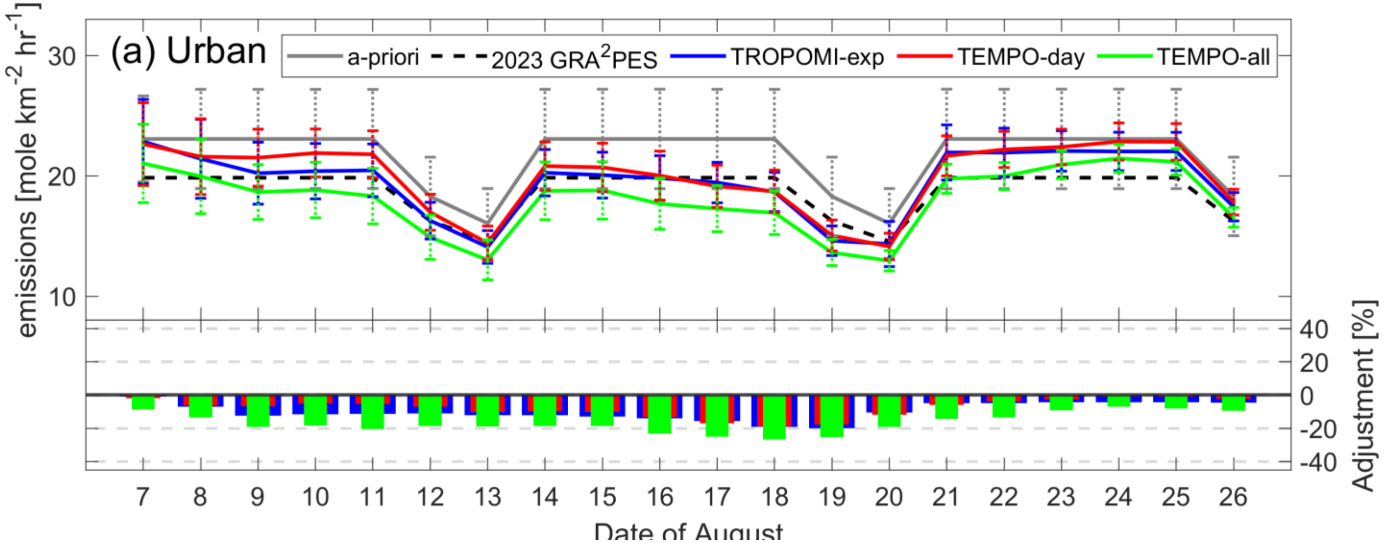
TEMPO v3 midday obs provide better constraints on NO_x emissions than TROPOMI, while morning/late afternoon TEMPO data subject to uncertainties.

Top-down NO_x emissions generally agree well with new bottom-up (i.e., 2023 GRA^2PES), improve O_3 and NO_x simulations (particularly in LA), but may over-correct in urban cores



Part III – Joint NO_x and NMVOC Emissions Inversion over the US by Assimilating Multi-Constituent TEMPO and TROPOMI Satellite Measurements (Hsu et al., in prep)

Launched — April 2023

First Light — August 2023

V03 Data Release — May 2024

V04 Data

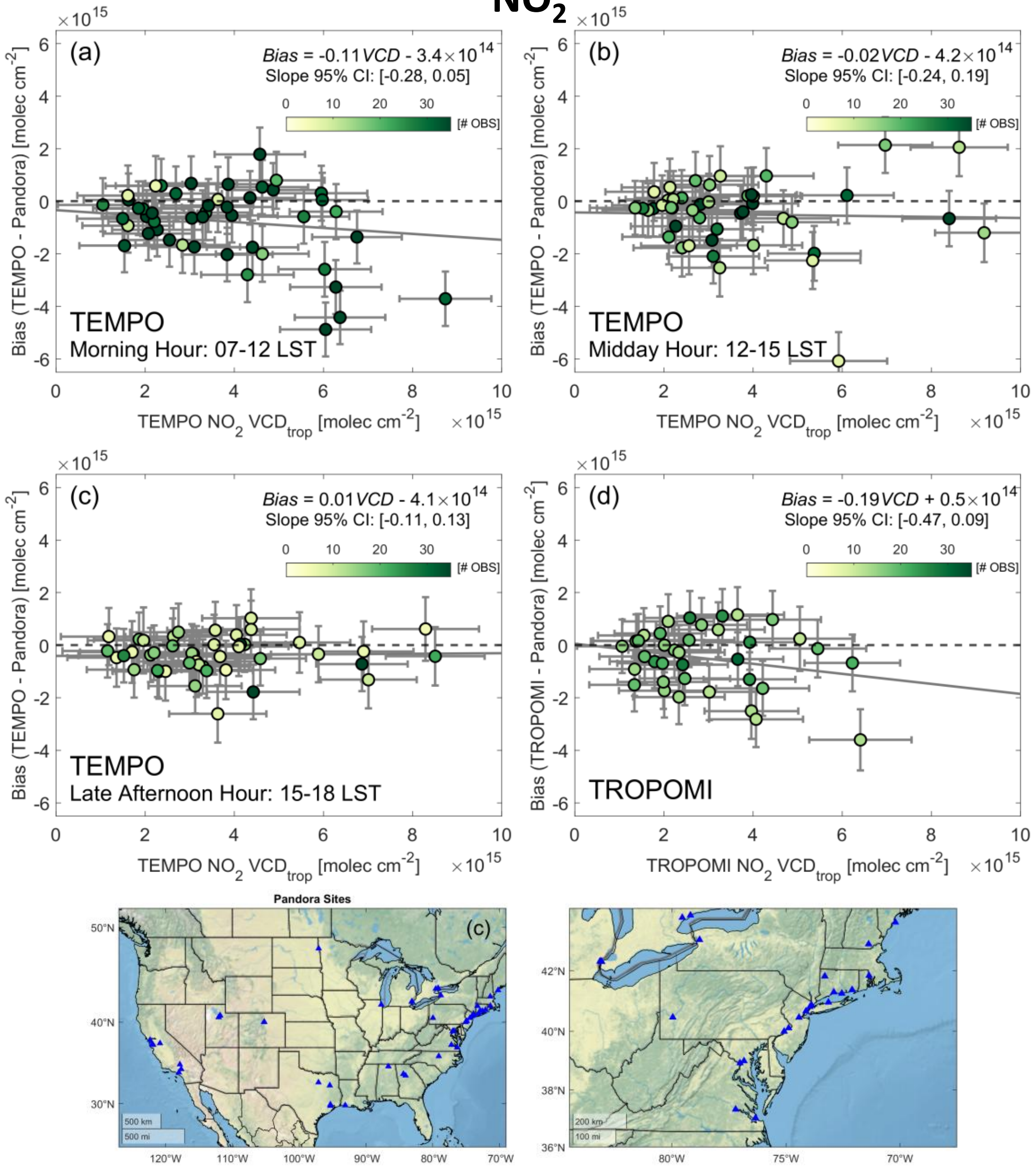
Objectives:

1. *Evaluate observations against Pandora, compare v03 and v04 retrievals*
2. *Jointly optimize NO_x and NMVOC emissions and investigate the benefits and challenges of multispecies emissions inversion*

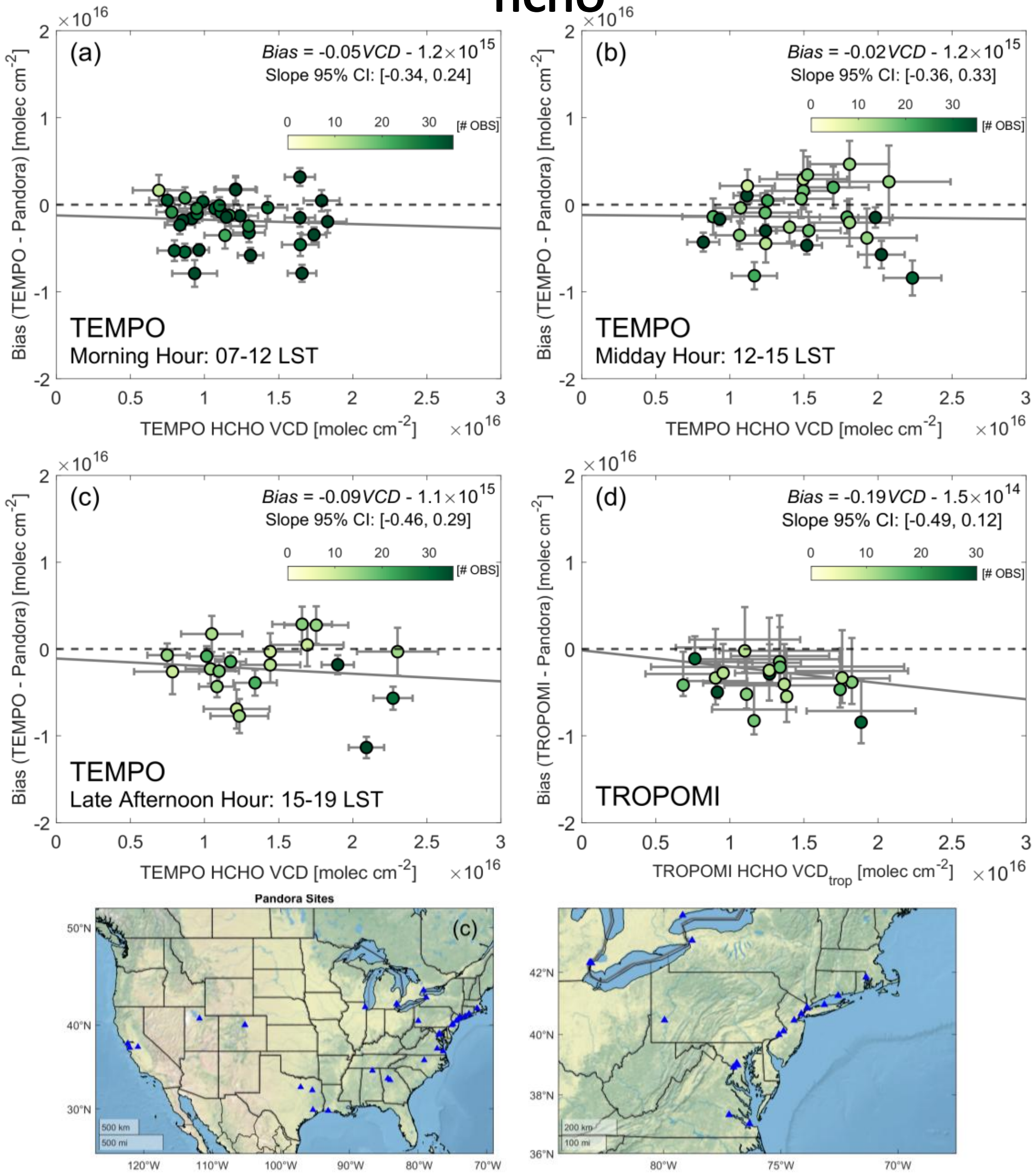
Method: Joint assimilation of NO_2 and formaldehyde (HCHO). The latter produced largely in the atmosphere by oxidation of other VOCs

TEMPO v03 /TROPOMI vs Pandora (Aug, 2023)

NO_2



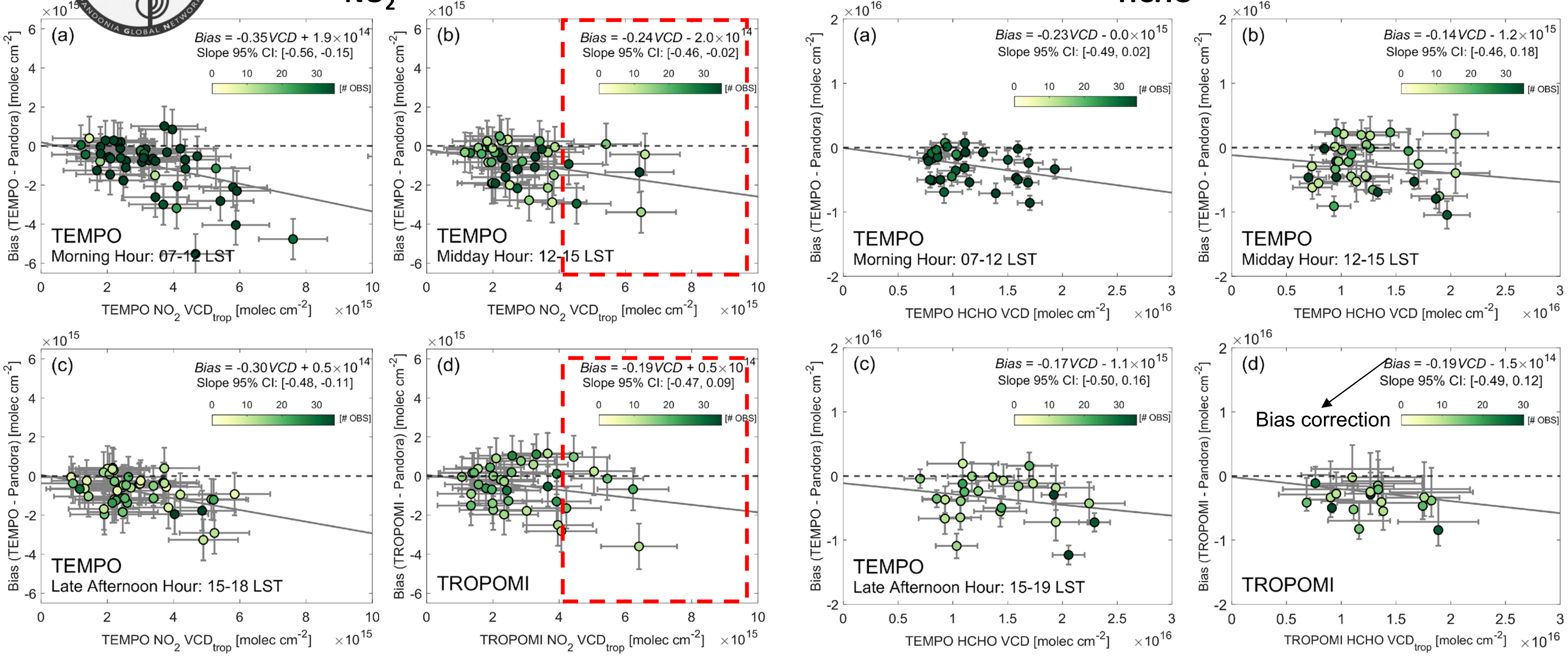
HCHO



HCHO site selection, data screening: Rawat et al. (2024; 2025)

Evaluate satellite NO_2 & HCHO column (TEMPO v04)

NO_2 **HCHO**

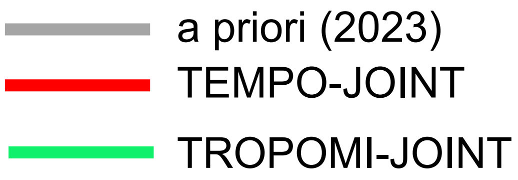


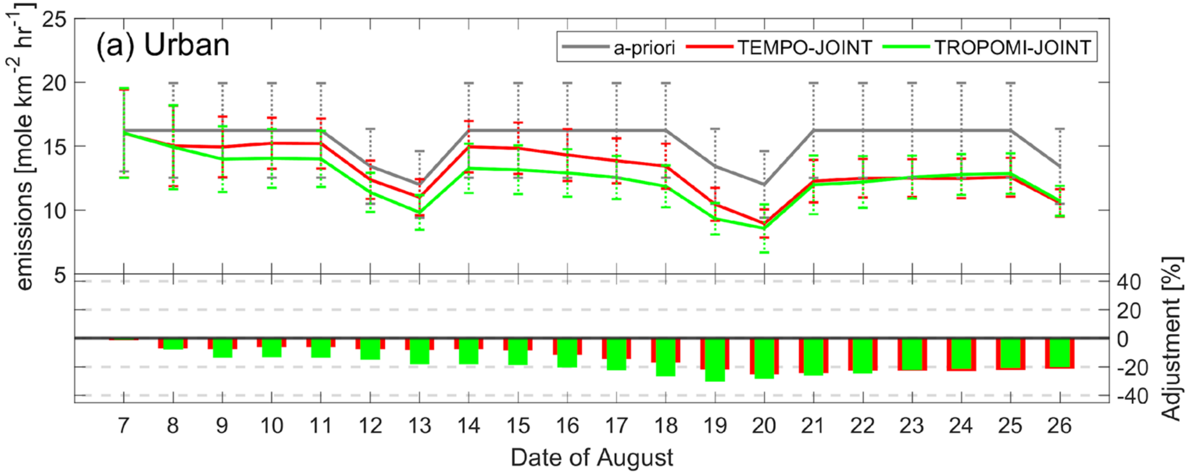
→ TROPOMI and TEMPO data are biased low over polluted regions.

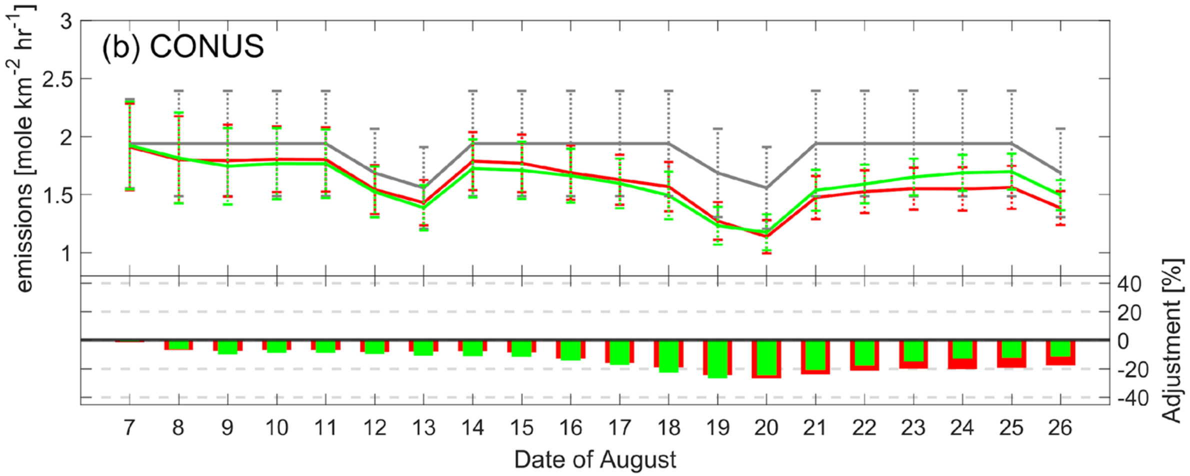
→ TEMPO HCHO show better data quality.

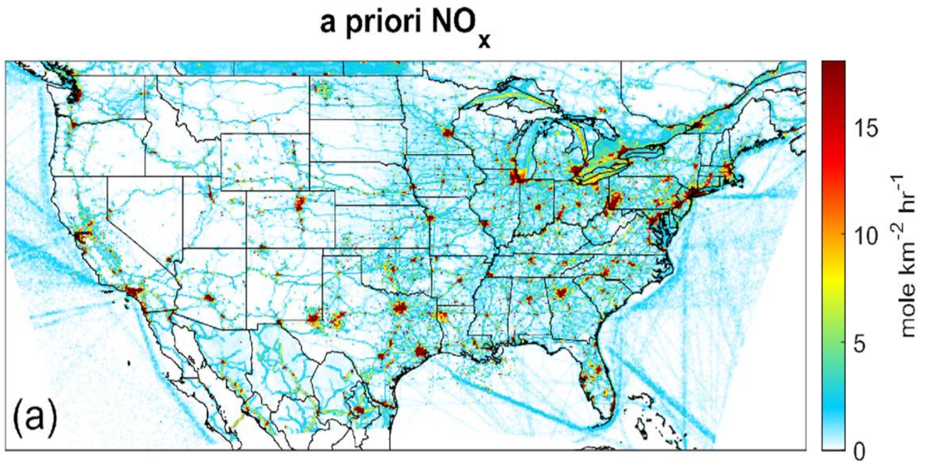
Challenges: Satellite data bias/accuracy

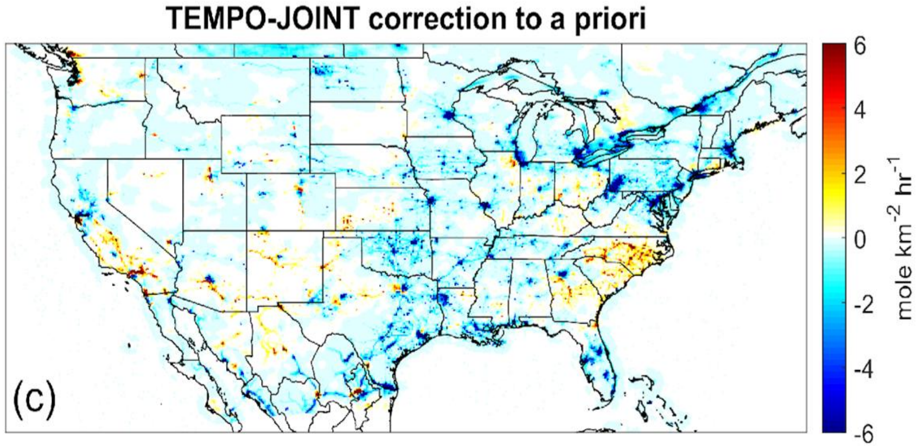
Top-down NO_x emissions





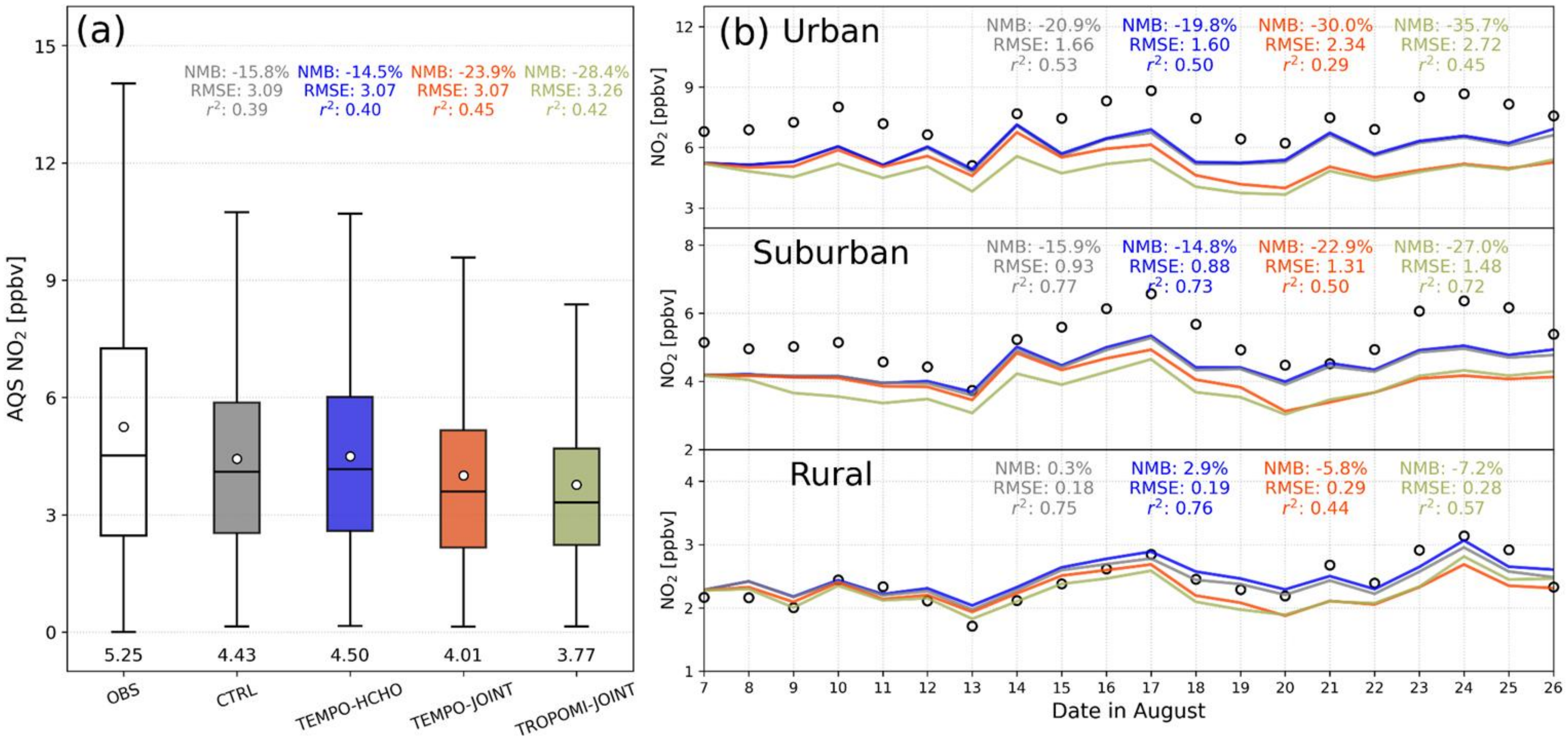




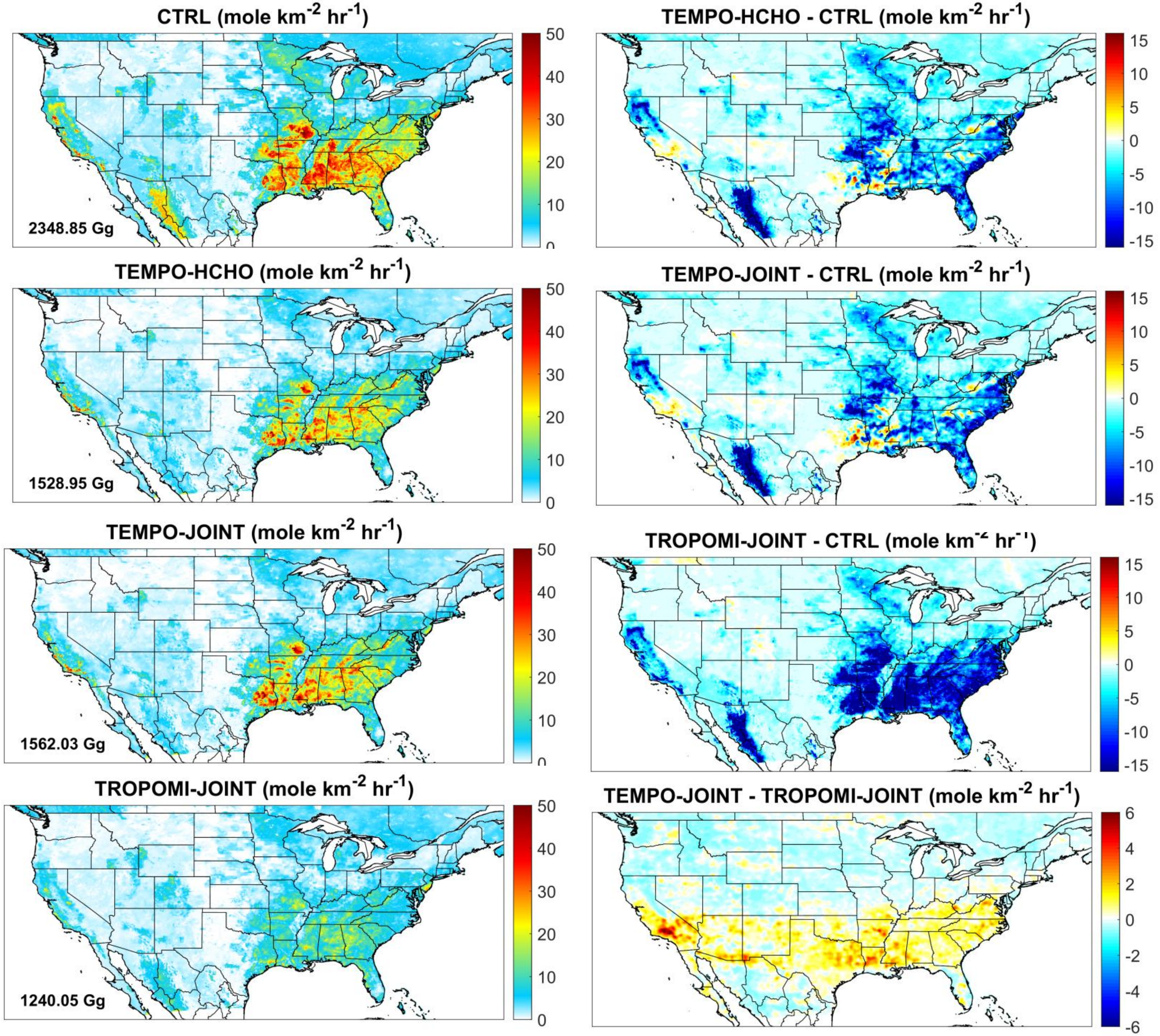


- Top-down NO_x emissions show consistent adjustments over urban (-10-20%).
- Regional difference existed

Comparison to surface (AQS) NO_2

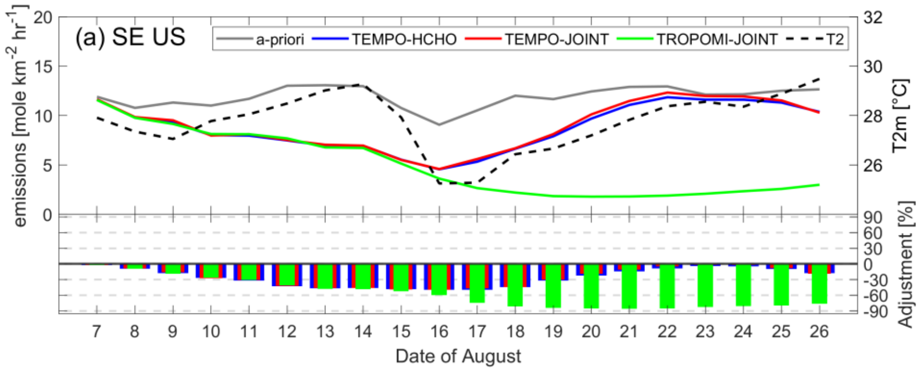


- Bias correction needed
- Evaluation in other months ongoing

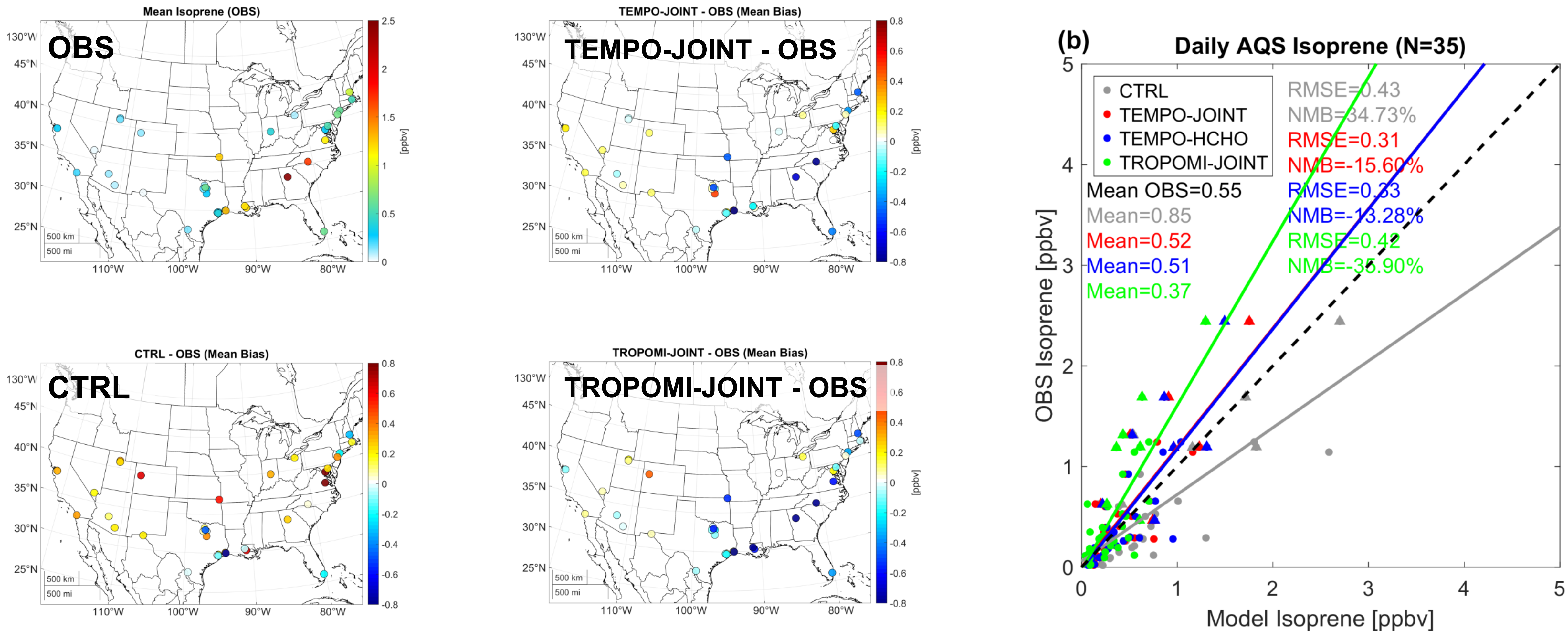


Top-down Biogenic Isoprene Emissions

- Top-down isoprene emissions show a ~30-50% reduction compared to the CTRL.
- Substantial daily variation
- TROPOMI-derived (after bias-correction) isoprene emissions is still the lowest.
- TEMPO-derived isoprene emissions align better with temperature changes (e.g., T2m).



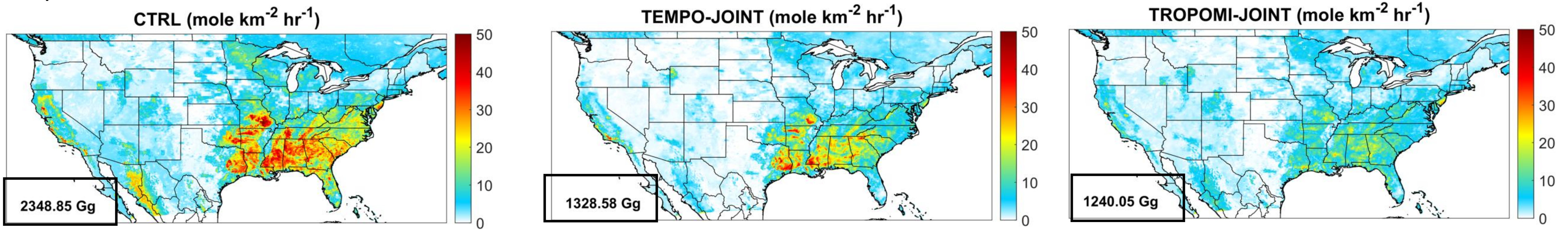
Evaluation with AQS isoprene (08/07 – 26, 2023)



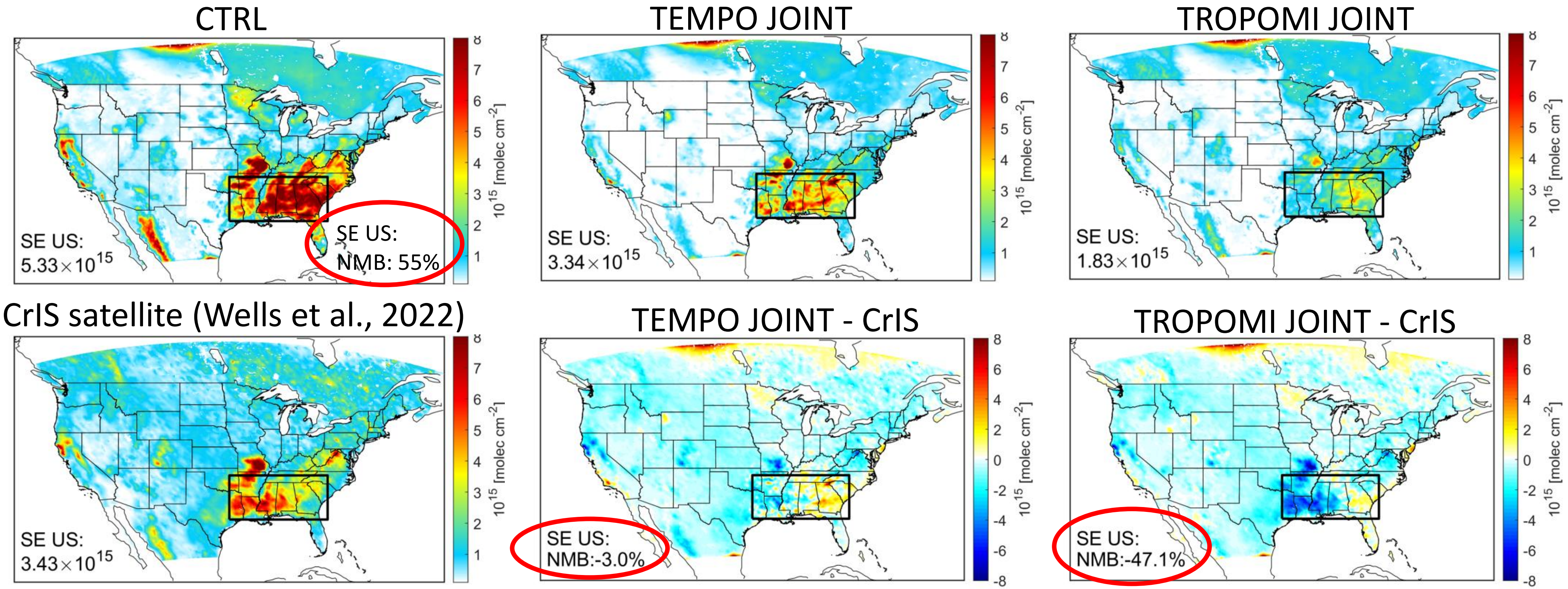
- TEMPO-JOINT and TEMPO-HCHO show consistent, and improved, agreement with AQS.
- TROPOMI-JOINT is too low.

Biogenic VOC emissions (isoprene): GEO (TEMPO) vs LEO (TROPOMI)

Isoprene emissions:

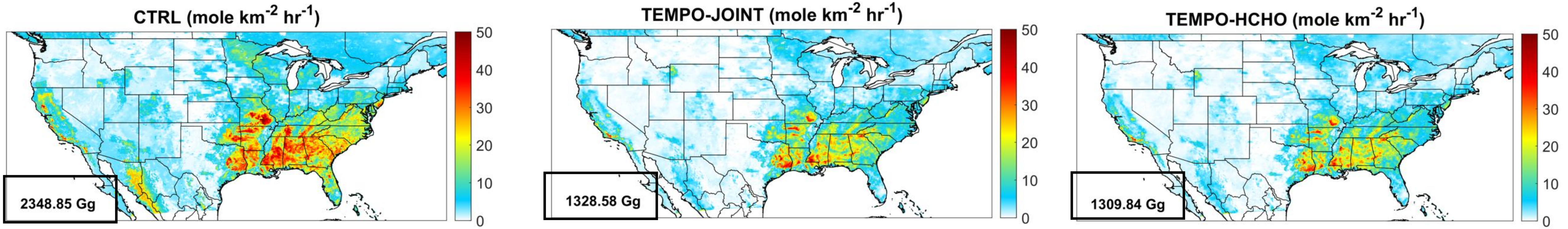


Isoprene concentrations:



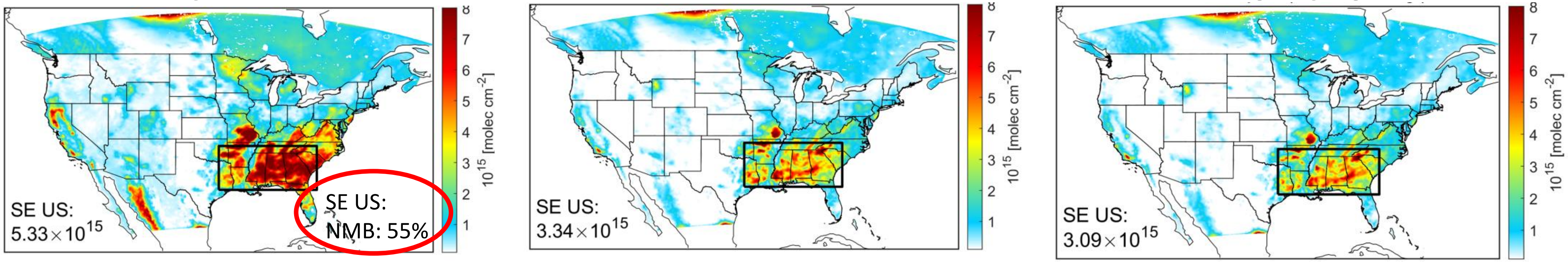
Biogenic VOC emissions (isoprene): joint (HCHO & NO2) vs HCHO

Isoprene emissions:

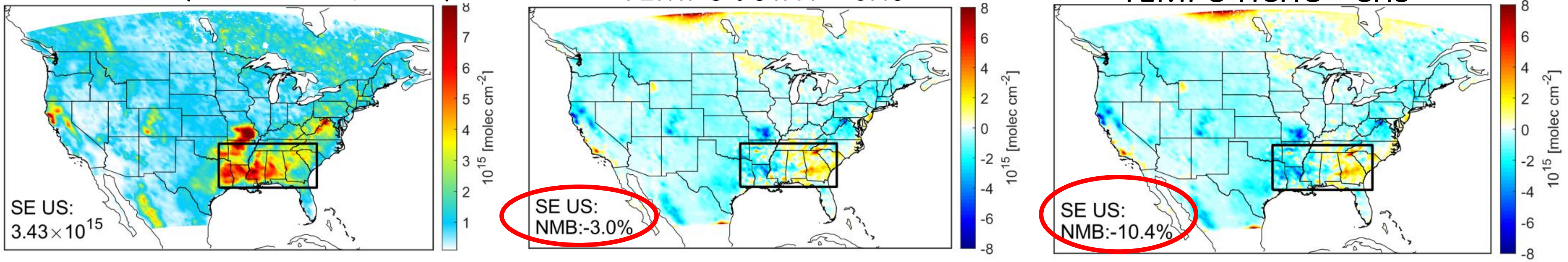


Isoprene concentrations:

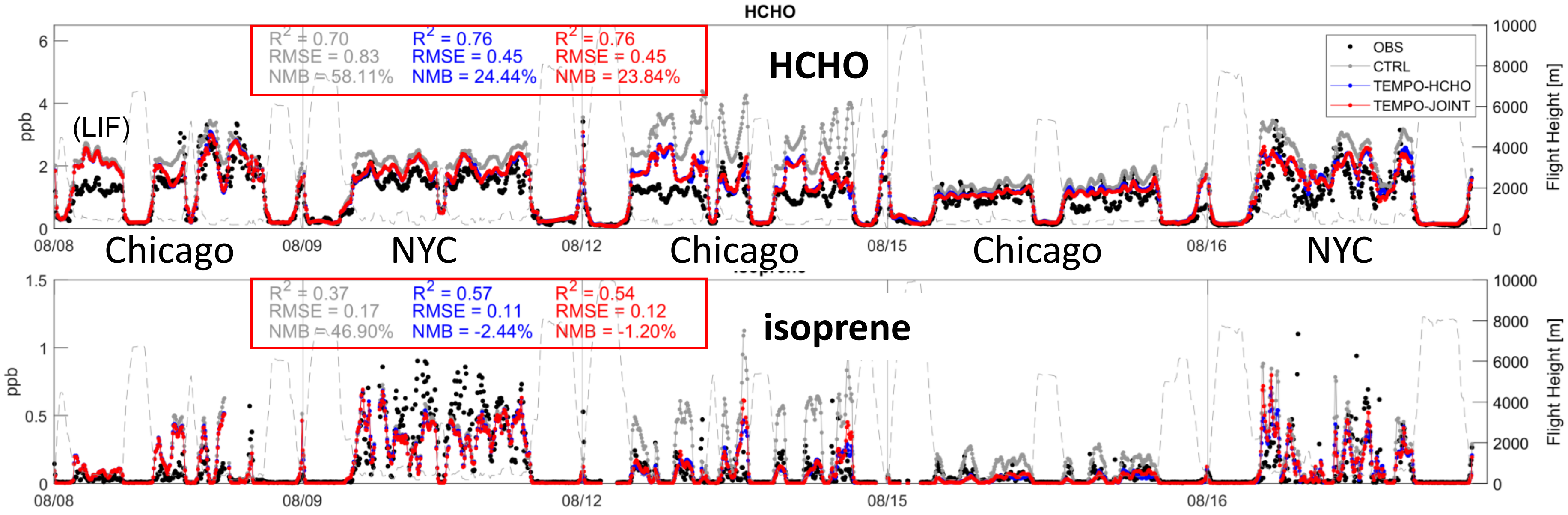
CTRL | TEMPO JOINT | TEMPO HCHO



CrIS satellite (Wells et al., 2022) | TEMPO JOINT - CrIS | TEMPO HCHO - CrIS

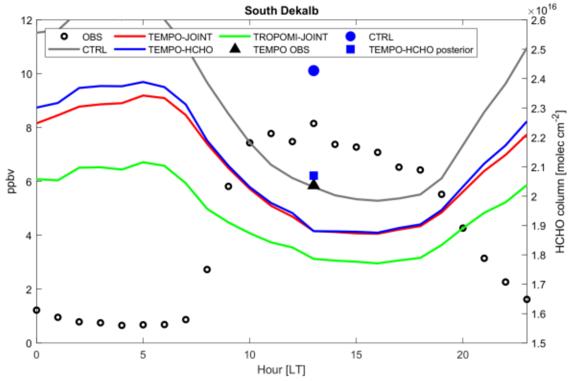


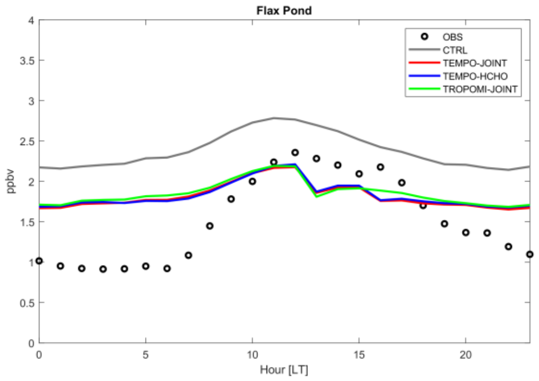
Evaluation against aircraft (AEROMMA DC-8)



- CTRL overpredicts VOC mixing ratio
- Top-down emissions improves HCHO, isoprene, benzene, methanol, ethene
- Mixed performance for other VOCs

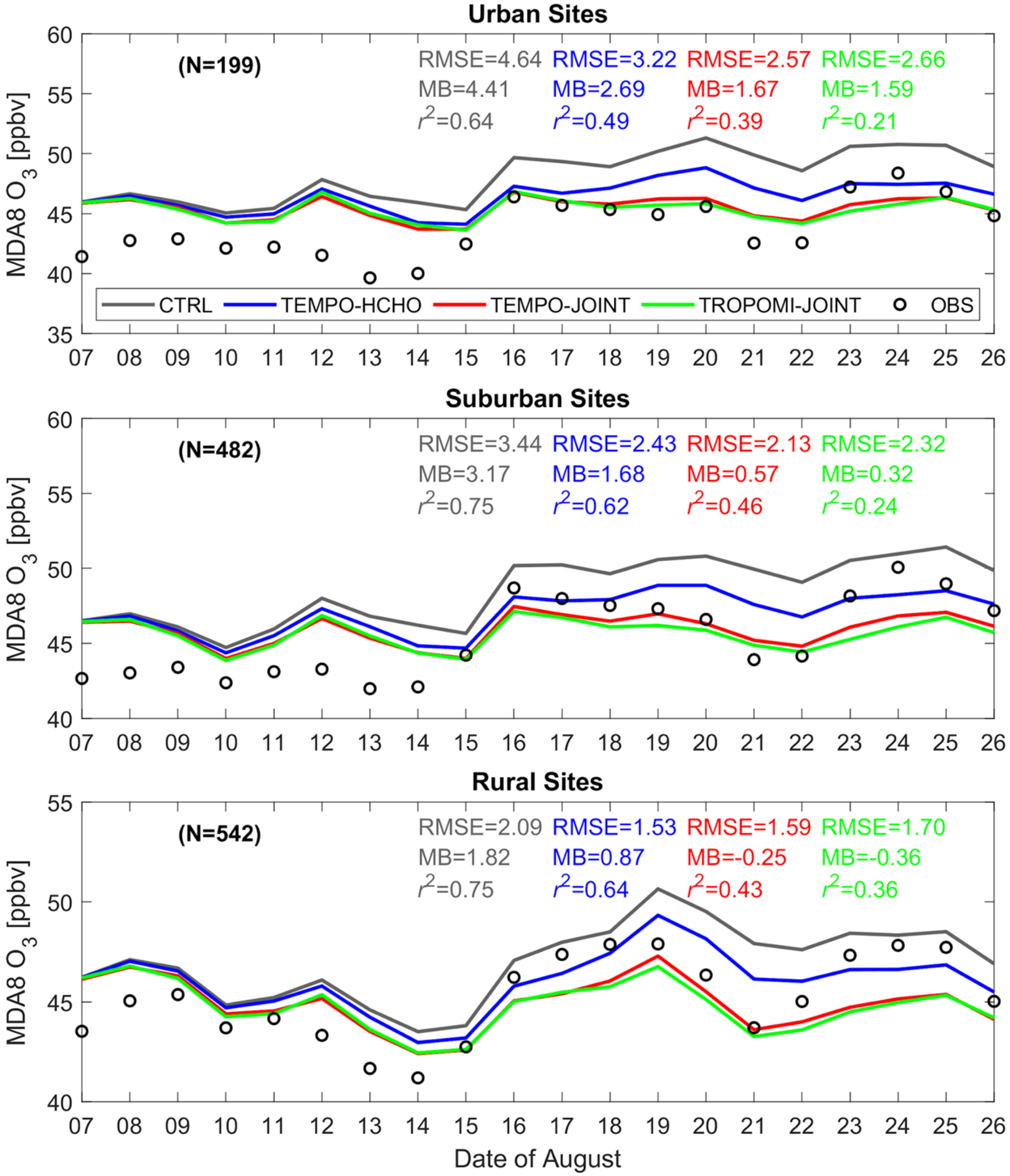
Hourly ground-based data: WRF-Chem can fail to capture diel variations of HCHO in GT and SDK, better for isoprene. HCHO bidi flux (Shutter et al. (2024))? PBL? analysis ongoing…



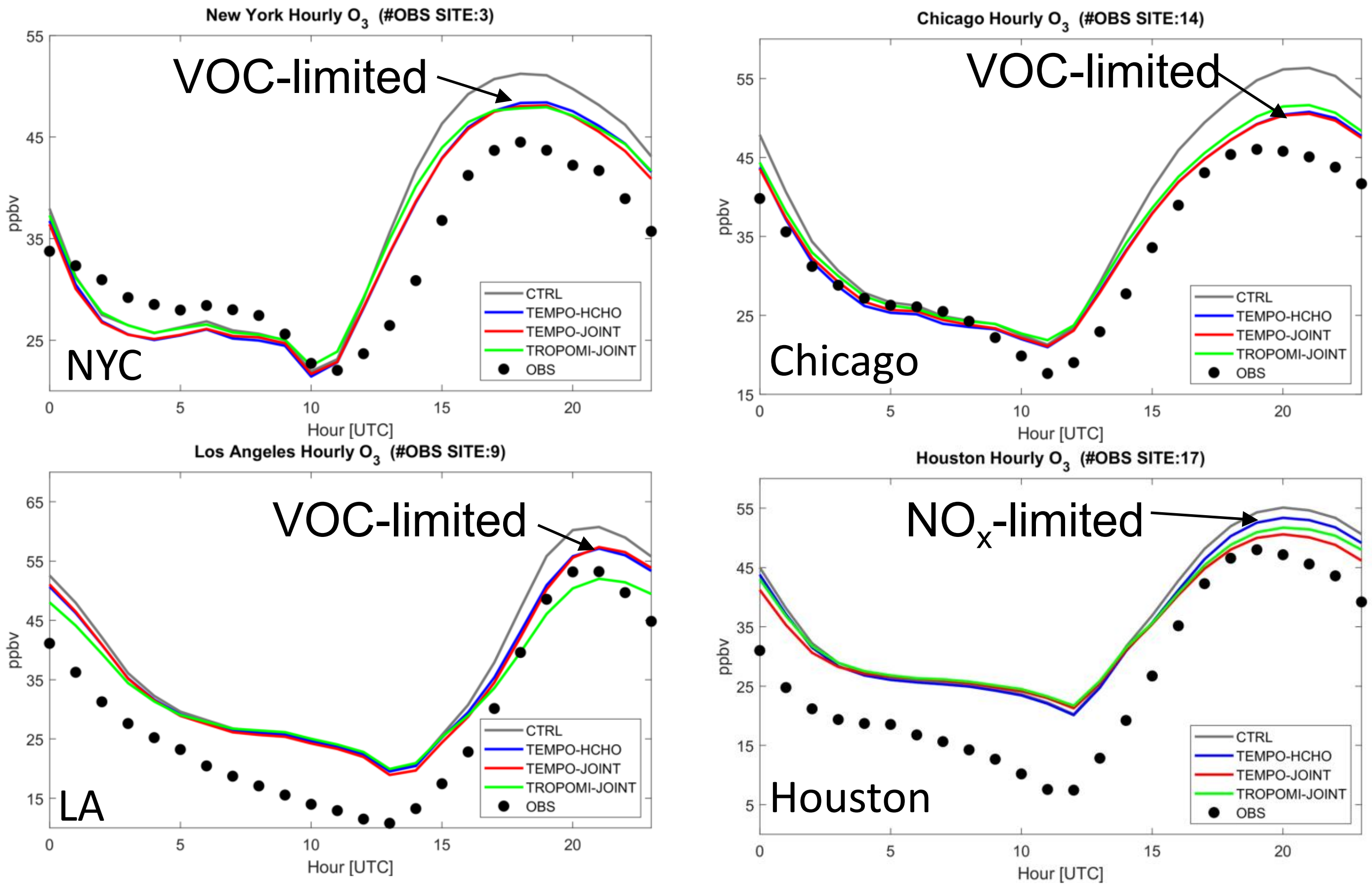


Evaluation of posterior simulations with AQS - MDA8 O_3

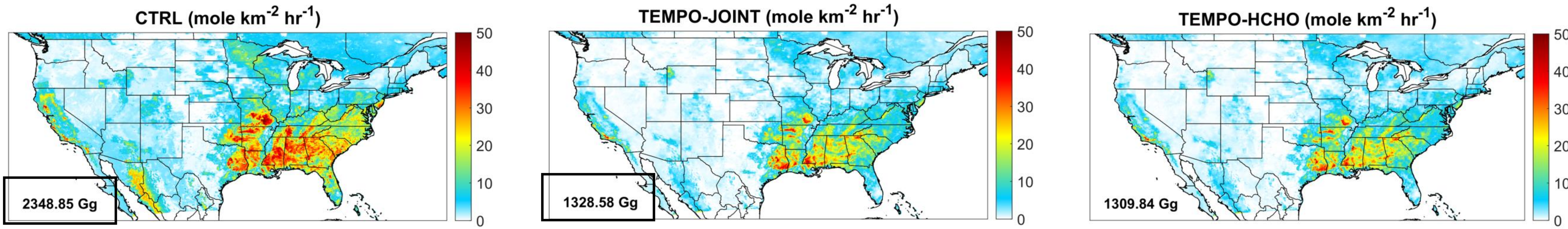
CNTRL TEMPO HCHO TEMPO HCHO+NO2 TROPOMI HCHO + NO2



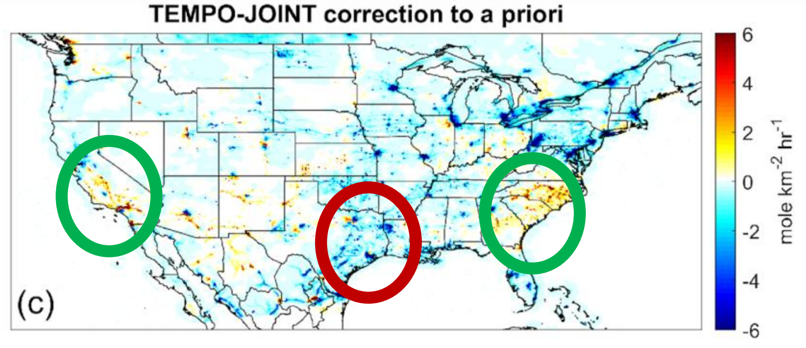
- Top-down emissions improves O_3 forecast
- Improvements in VOC and NOx-limited conditions
- TEMPO joint (HCO & NO_2) has lowest error and bias

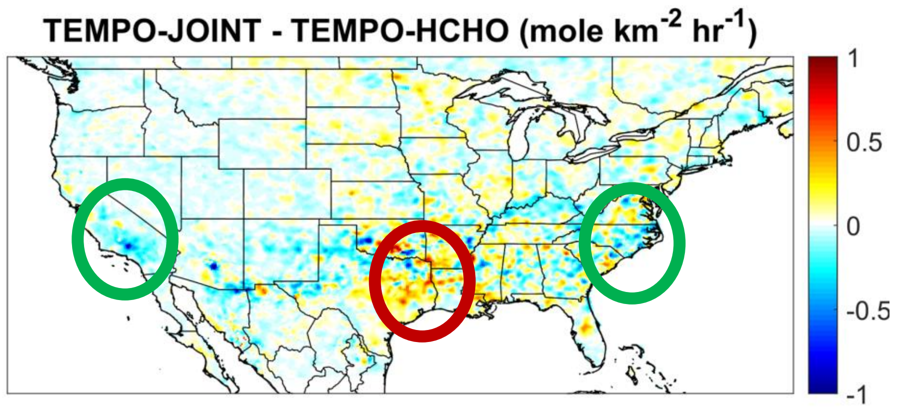


Biogenic VOC emissions (isoprene): joint benefit



NOx emissions





- Impacts of NOx on isoprene are anticorrelated

ΔNOx ~ ΔHCHO ~ -ΔISOP

NO_2 O - F < 0: NOx ↓ → isoprene oxidation ↓ → HCHO ↓ → smaller HCHO O - F → larger isoprene emissions (smaller reduction)

NO_2 O - F > 0: NOx ↑ → isoprene oxidation ↑ → HCHO ↑ → larger HCHO O - F → smaller isoprene emissions (larger reduction)

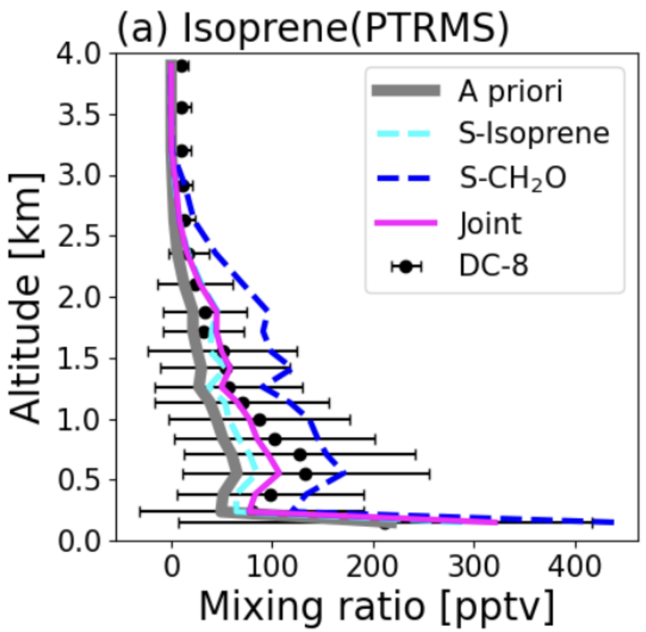
Summary

- TEMPO HCHO assimilation improves model estimates of HCHO, isoprene, and other VOCs
  - Reduced high-bias in prior (BEIS) biogenic isoprene and anthropogenic (GRA^2PES) VOC
  - Model still struggles with nighttime concentrations / diurnal variability
  - TROPOMI HCHO biased low, and assimilation over-corrects VOCs

- Joint assimilation of HCHO & NO_2 improves O_3 predictions over NO_2 or HCHO alone

- Despite other improvements, low bias in urban cores exacerbated in TEMPO v04 compared to v03
  - Top-down urban NOx emissions may be biased low
  - NO_2 bias correction needed prior to assimilation
  - Evaluation in additional months ongoing

- HCHO (UV/Vis) + ISOP (IR) proven useful for KORUS-AQ (Choi et al., 2025)



- GEO NO_2, HCHO, ISOP would be very useful!

extra

Modeling configurations & experimental design

- Modeling and DA system: WRF-Chem/Chem-DART
  - Ensemble Kalman Filter (Liu et al., 2017; Mizzi et al., 2016;2018, Ma et al., 2019)
- Four experiments:
- Time period: 08/07-26, 2023

Exp	Data
CTRL	MET
TEMPO-HCHO	MET + midday TEMPO HCHO VCD (18 & 21 UTC)
TEMPO-JOINT	MET + midday TEMPO NO_2 SCD & HCHO VCD
TROPOMI-JOINT	MET + TROPOMI NO2 SCD & HCHO VCD

- Appy bias-correction for TROPOMI HCHO VCD.
- TEMPO version V03, TROPOMI version V2.05
- Satellite data are aggregated to match WRF-Chem resolution (12 km)

- NMVOC emissions inversion setting:

→ Anthropogenic NMVOC emissions are aggregated based on the functional groups and optimized in bulk. (e.g., alkanes, alkenes, aldehydes, alcohols, aromatics,, ketones, other AVOC).

→ Biogenic isoprene emissions + other BVOC

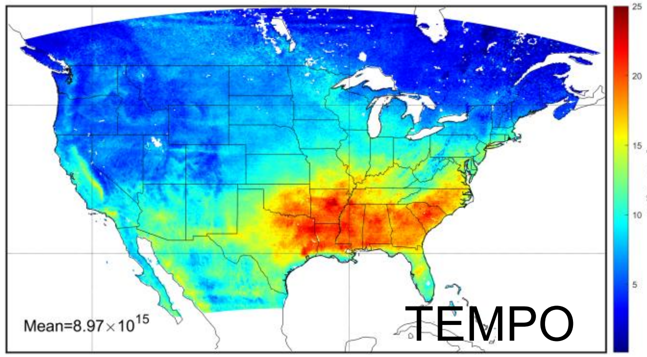
Model Configurations	
Model version	WRF-Chem v4.2.2
Anthropogenic emissions	GRA^{2}PES (based on 2023, developed by NOAA CSL)
Biogenic emissions	BEIS3.14
Fire emissions	RAVE
IC/BC	MET: NAM, CHEM: RAQMS
Gas phase chemistry	RACM2B_VCP
Aerosol module	MADE/SORGAM
Resolutions	12km with 51 vertical layers
microphysics	Morrison 2-moment scheme
longwave radiation	RRTMG scheme
shortwave radiation	RRTMG scheme
surface-layer	MYNN Scheme
Land Surface	Unified Noah Land Surface Model
PBL	MYNN
Cumulus Parameterization	Grell–Freitas Ensemble Scheme

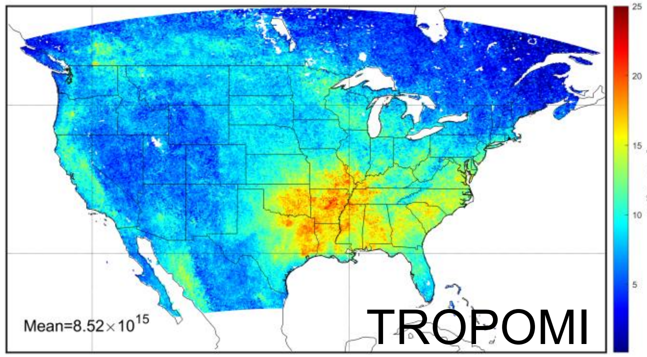
Hsu et al., 2024; 2026

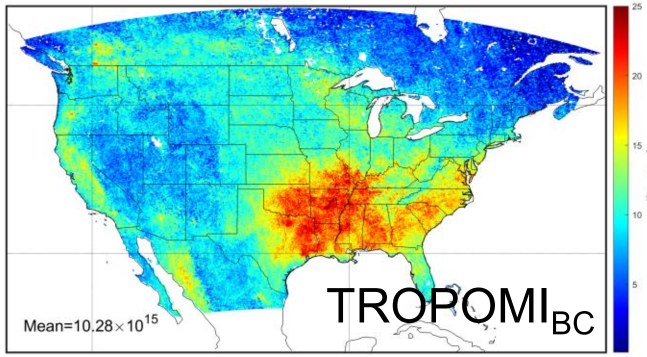
TROPOMI HCHO vs TEMPO HCHO

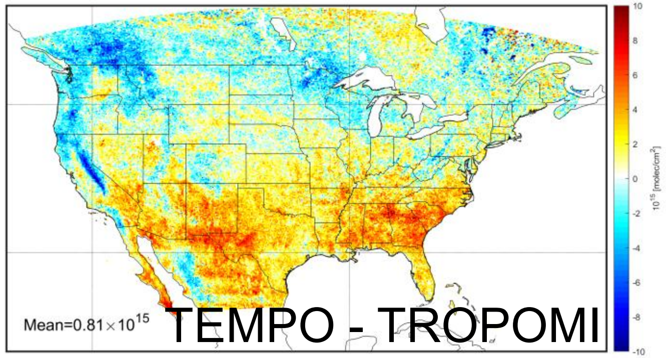
Bias correct TROPOMI based on Pandora data:

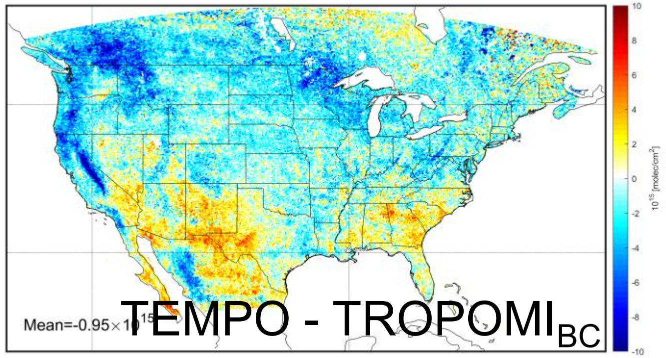
$$TROPOM_{BC} = TROPOMI \times 1.19 + 1.43 \times 10^{14}$$



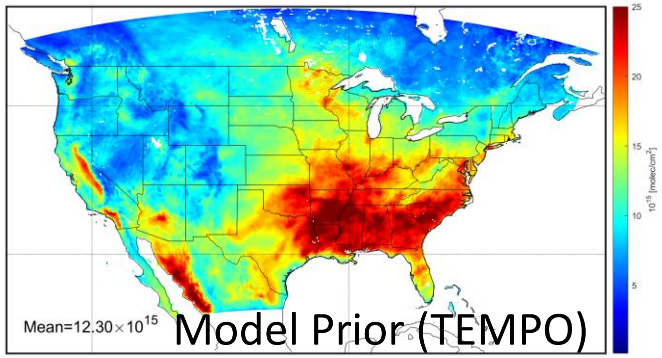


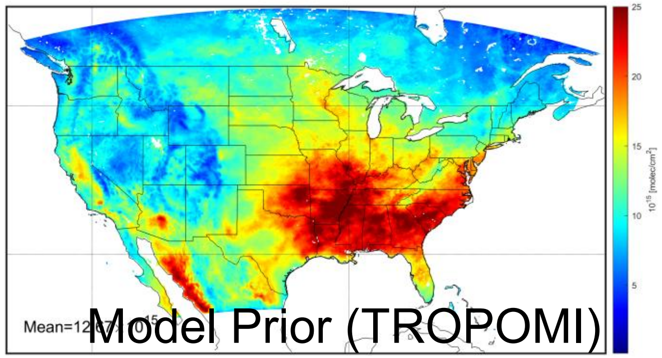






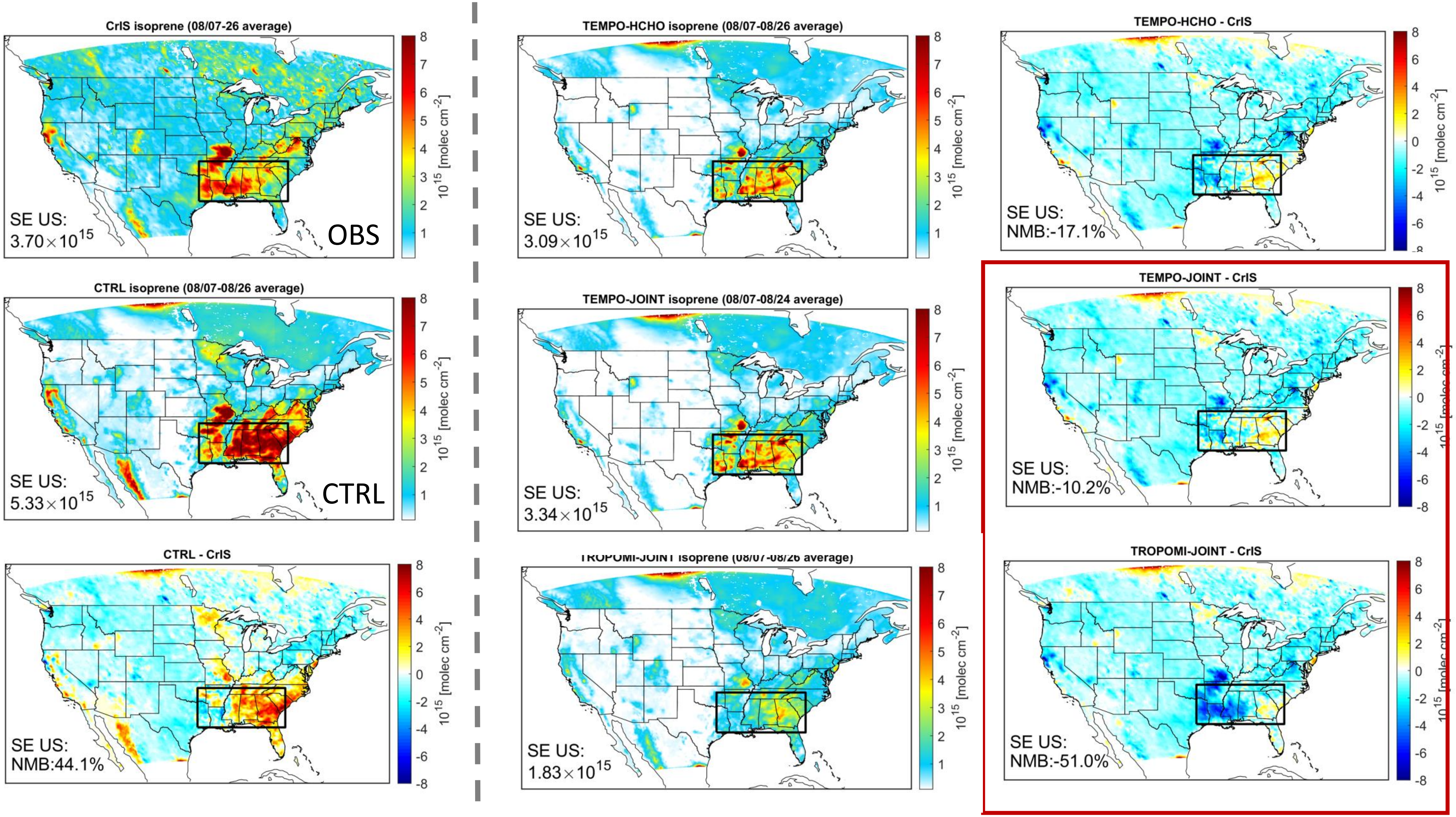
WRF-Chem HCHO





Model Evaluation: NOAA-20 CrIS isoprene

Data from Prof. Millet & Dr. Wells, UMN Preliminary/unpublish data



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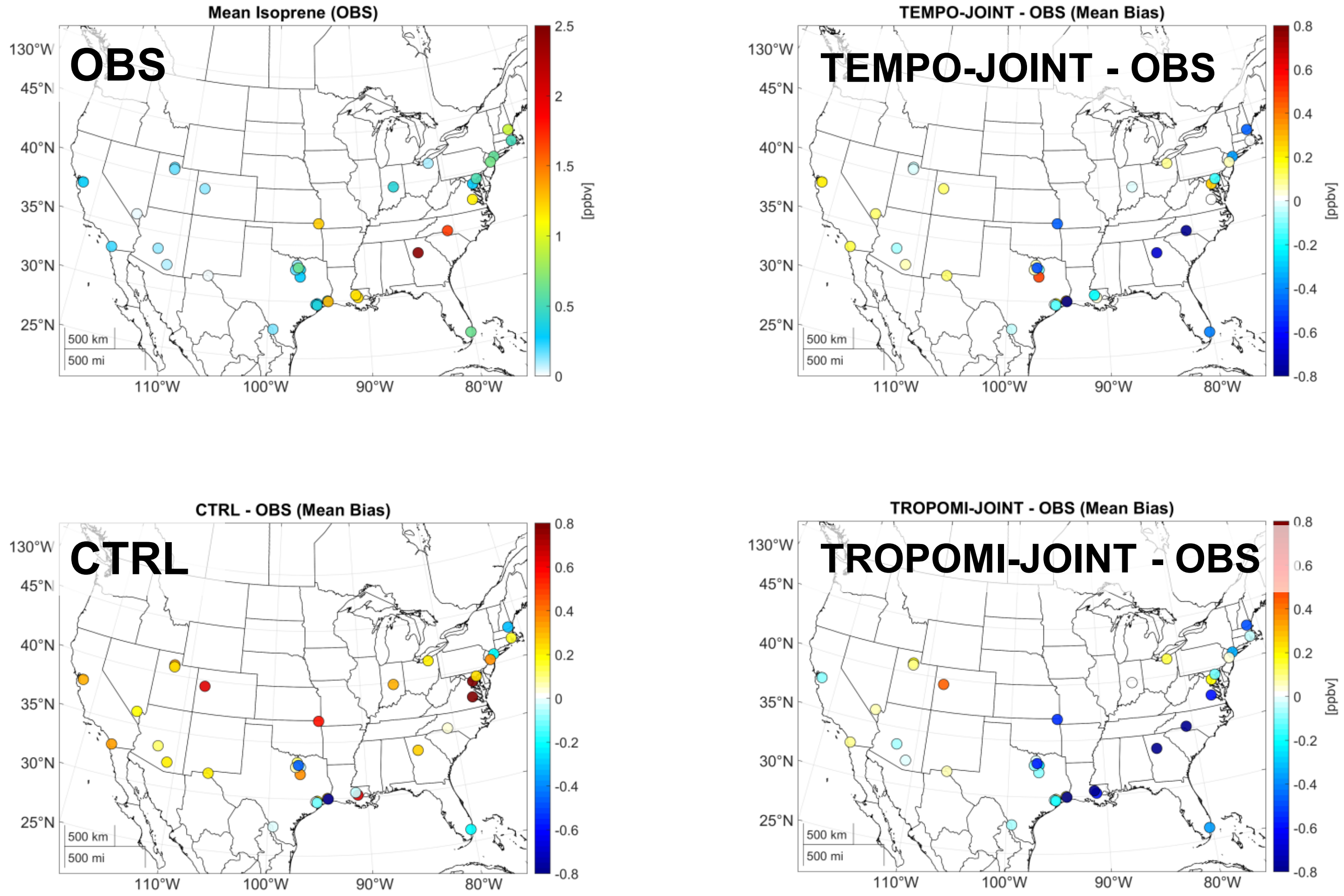
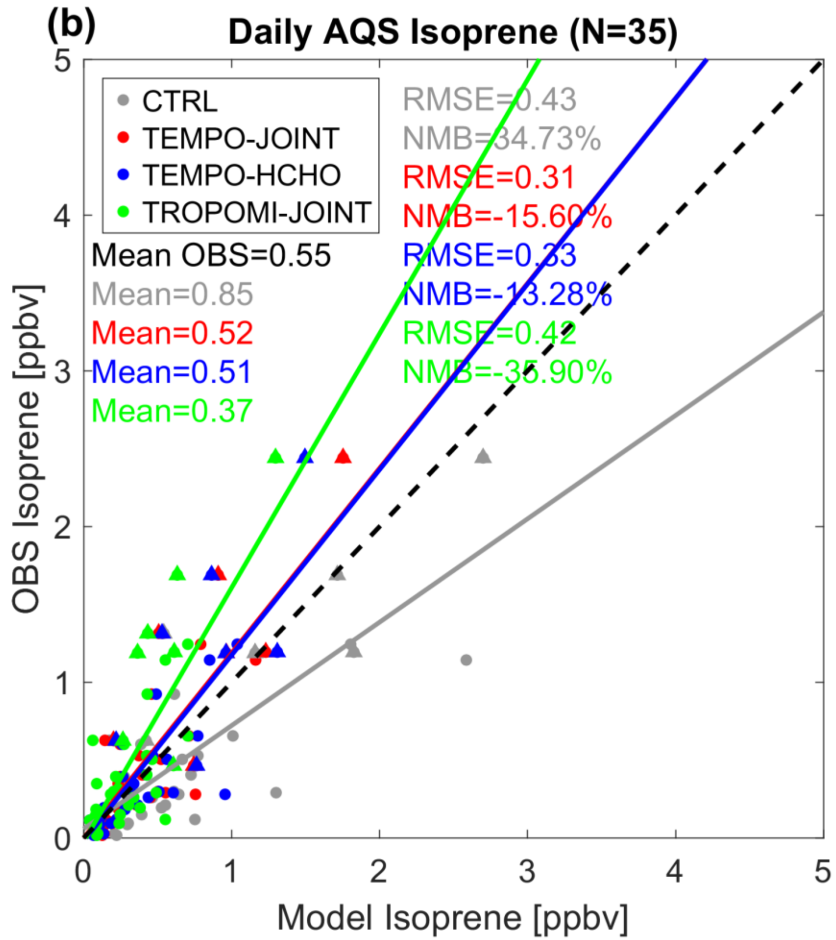
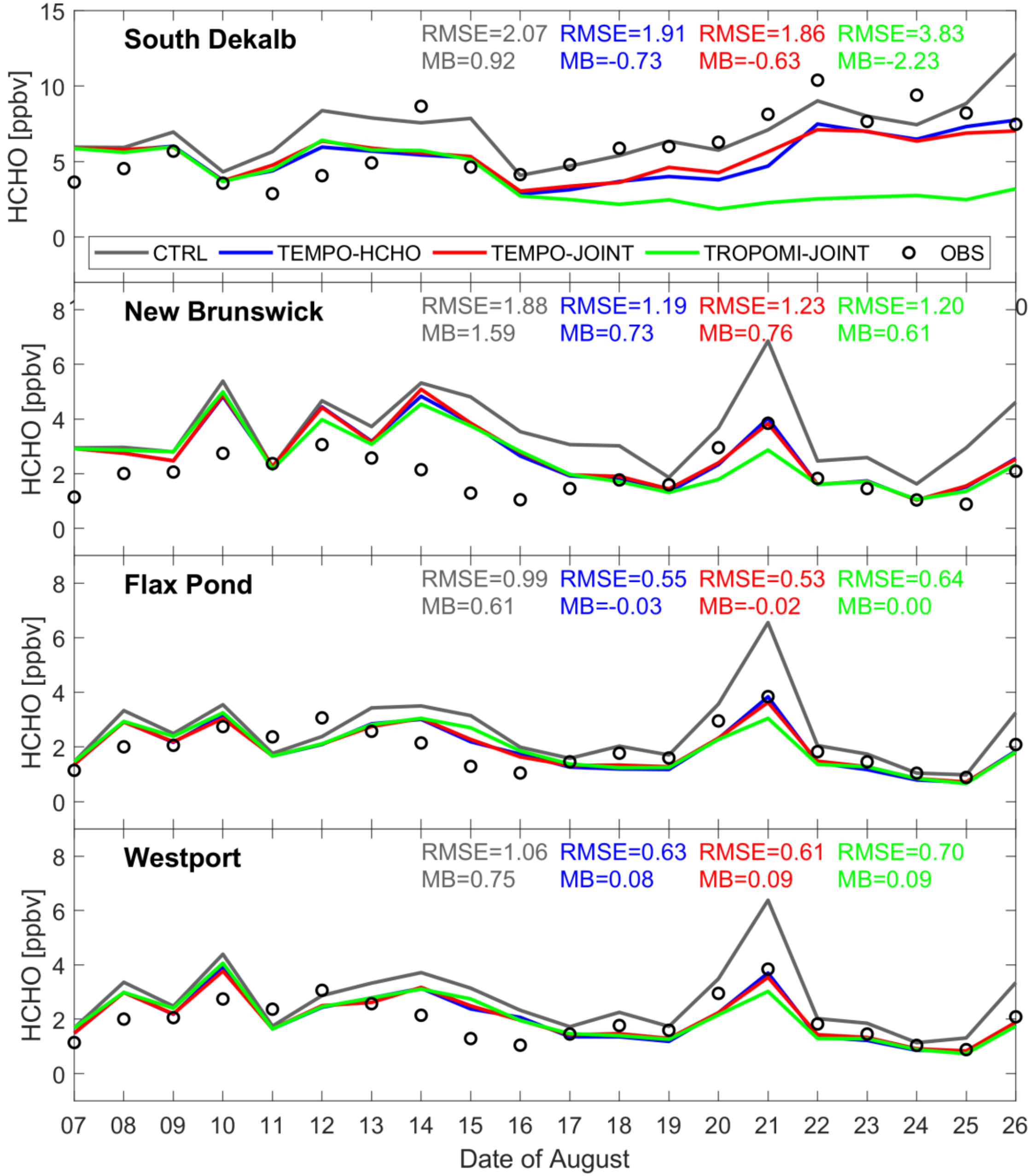
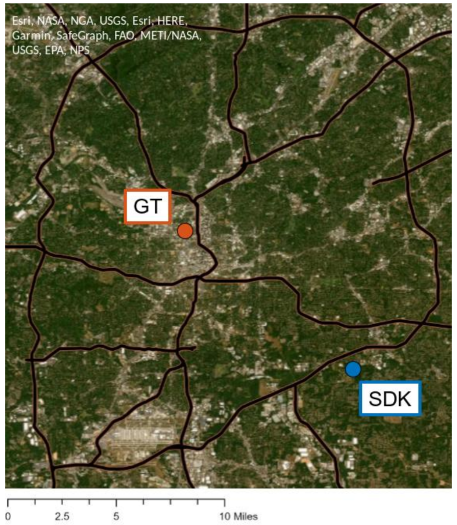




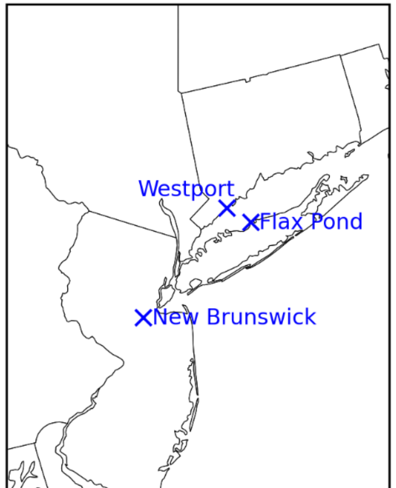
- TEMPO-JOINT and TEMPO-HCHO show consistent, and improved, agreement with AQS.
- TROPOMI-JOINT is too low.

HCHO evaluation (daytime average (07-17 LST))





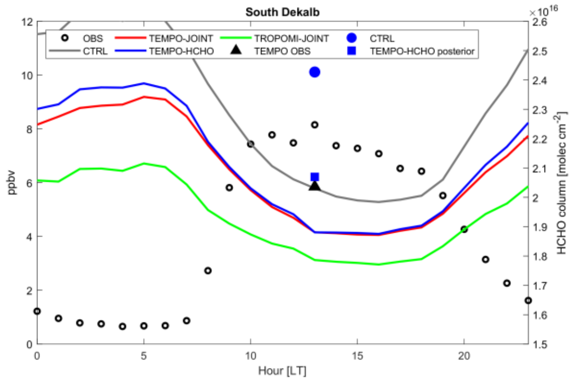
Adopted from Masut et al. (2024)

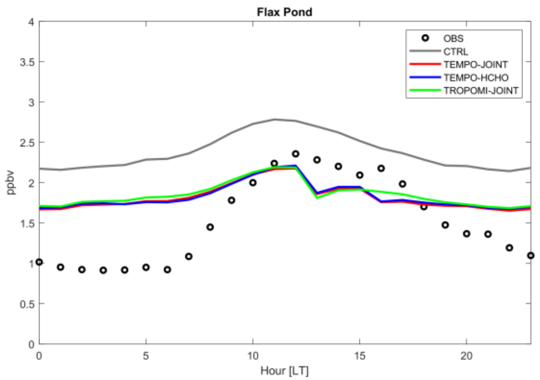


Adopted from Skipper et al. (2024)

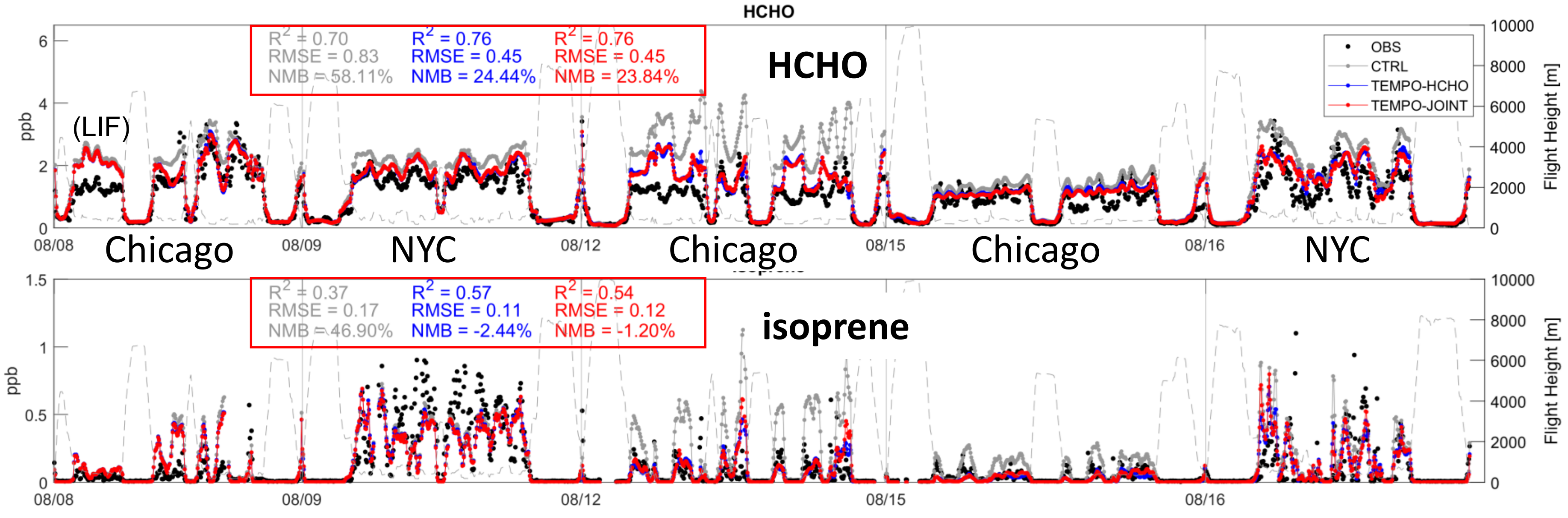
HCHO data courtesy: Prof. Jennifer Kaiser (South Dekalb) & NASA STAQS 2023

- Hourly data: WRF-Chem can fail to capture diel variations of HCHO in GT and SDK, better for isoprene, analysis (PBL, CO, NOx) ongoing…

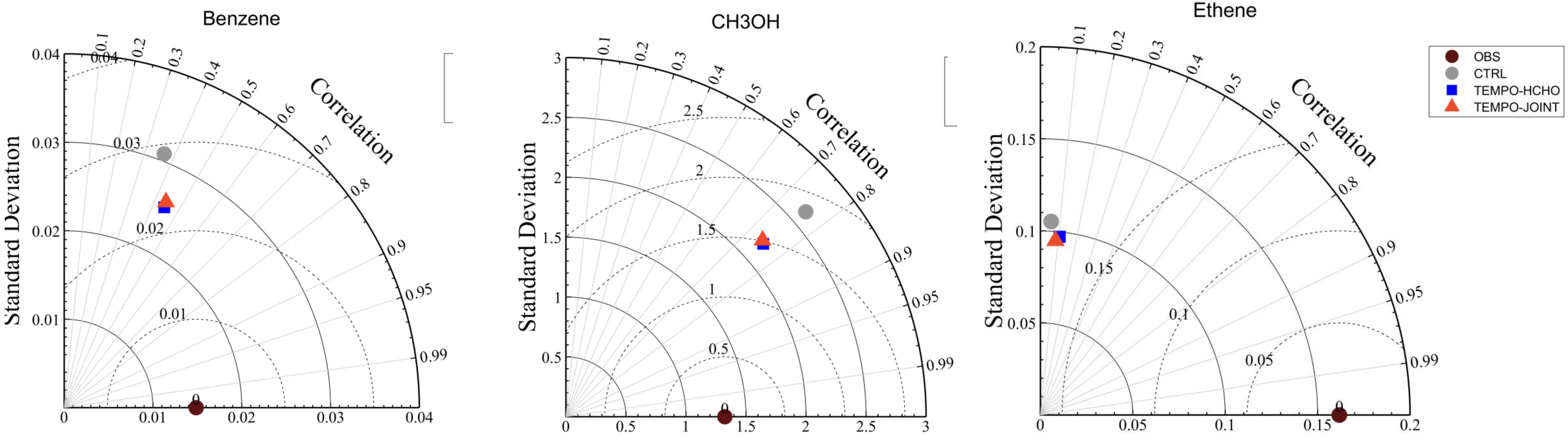




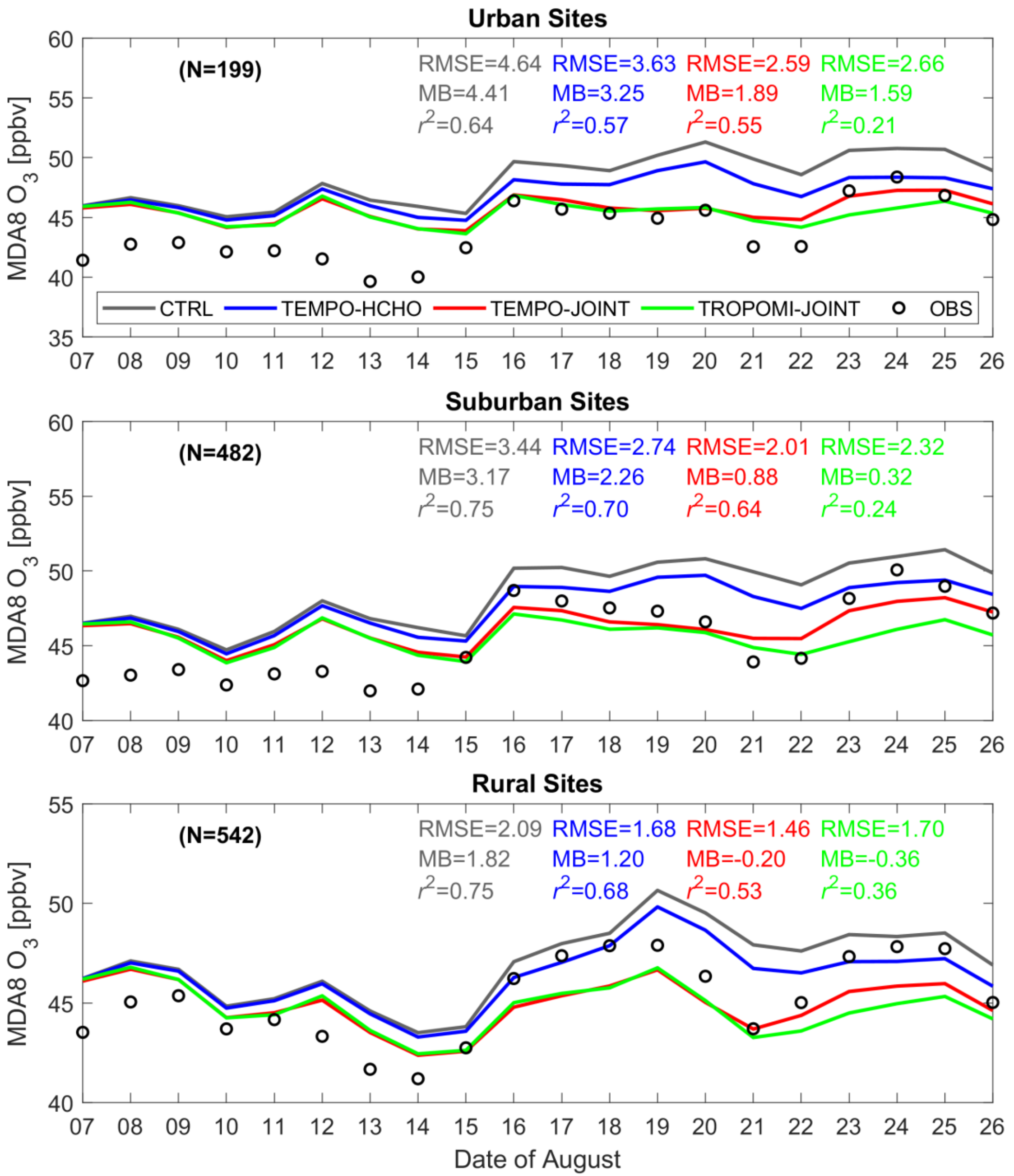
Evaluation against aircraft (AEROMMA DC-8)



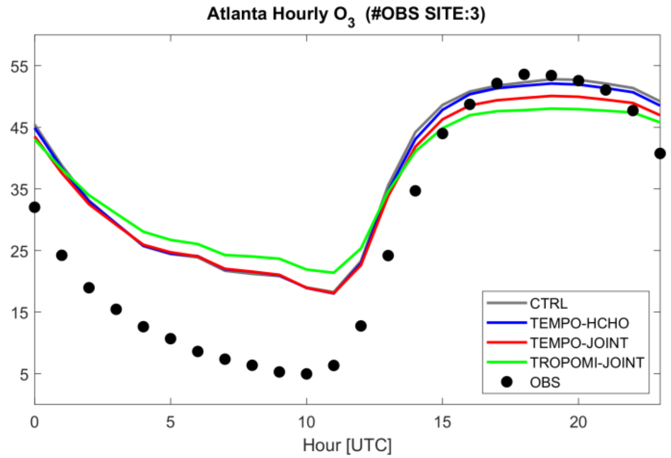
- CTRL overpredicts VOC mixing ratio
- Top-down emissions improves HCHO, isoprene
- Mixed performance for other VOCs



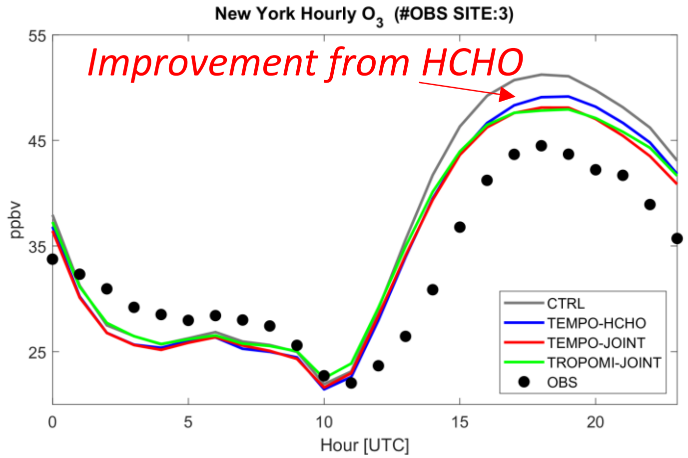
Evaluation with AQS O_3: MDA8 (left) & hourly (right)



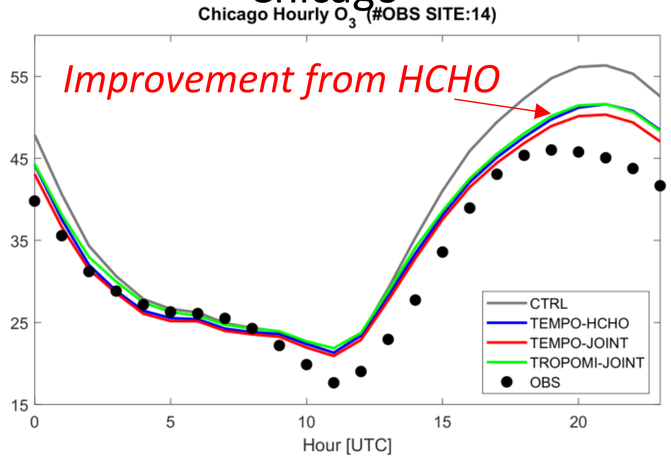
Atlanta



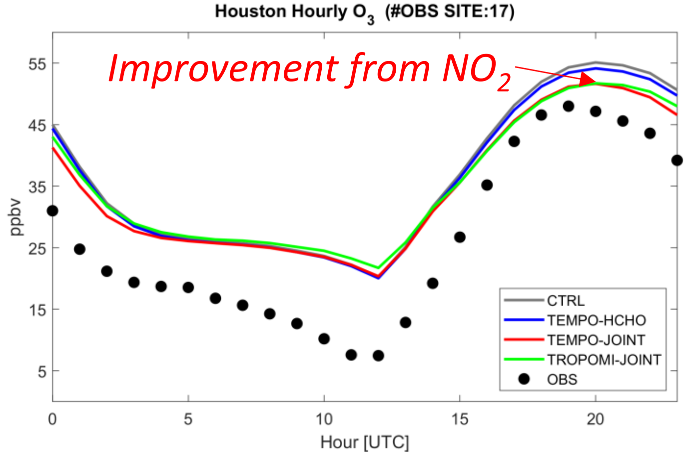
New York



Chicago



Houston



- JOINT assimilation has lowest RMSE and MB for MDA8 O3, with TEMPO often out-performing TROPOMI (particularly for r^2)
- Hourly O_3 shows daytime improvements, reflecting different regional sensitivities