Advancing sectoral emission estimates using TEMPO observations

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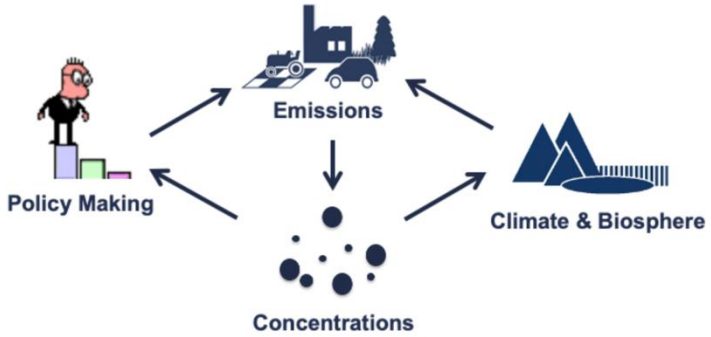
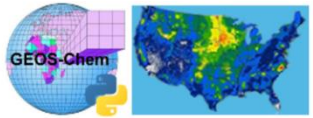
With Nana Wu and Carolina Nowlan

NC STATE UNIVERSITY

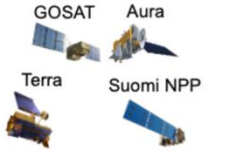
QU GROUP
ATMOSPHERIC CHEMISTRY MODELING AT NC STATE



Tools:



Air Quality and Atmospheric Chemistry Model



Satellite Observations

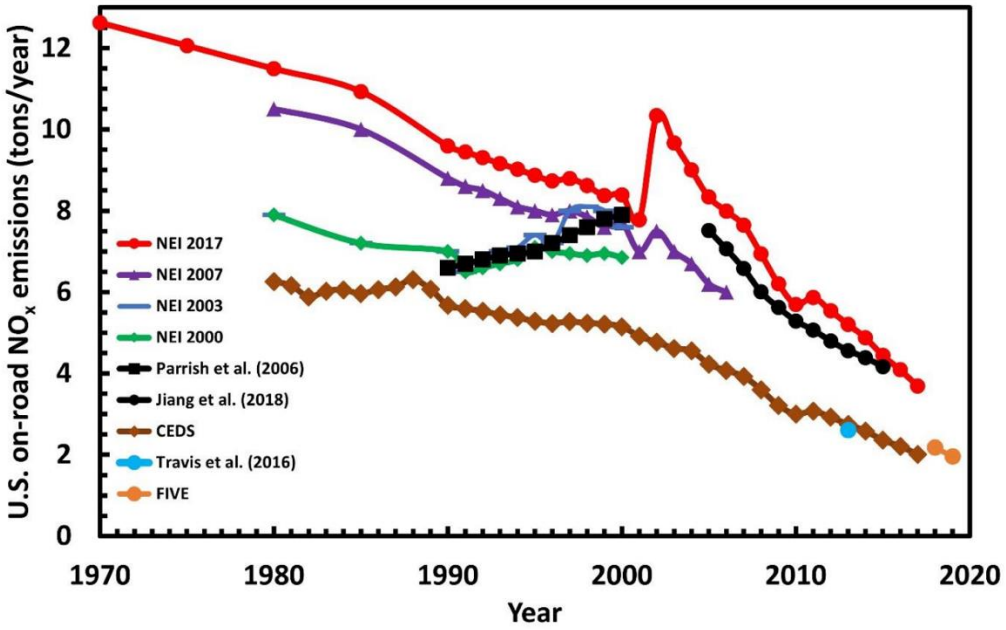
Data-driven Frameworks

Bottom-up Emissions Have Large Uncertainties & Lag in Time

Bottom-up estimate

Emission = species emission factor × activity

US on-road vehicle NO_x emissions



(*Christiansen et al.*, 2019)

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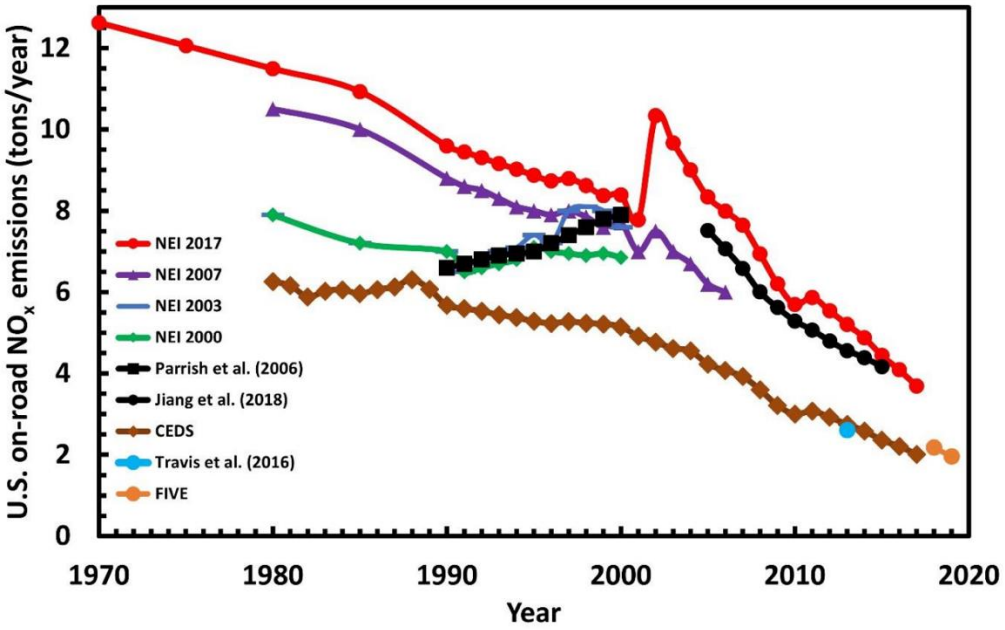
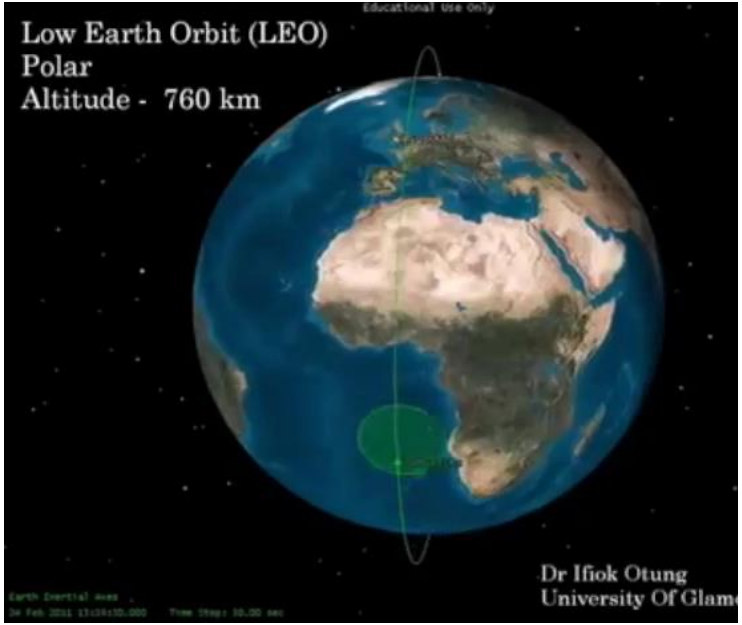


(*Christiansen et al.*, 2019)

Top-down estimate

Infers emissions from observations

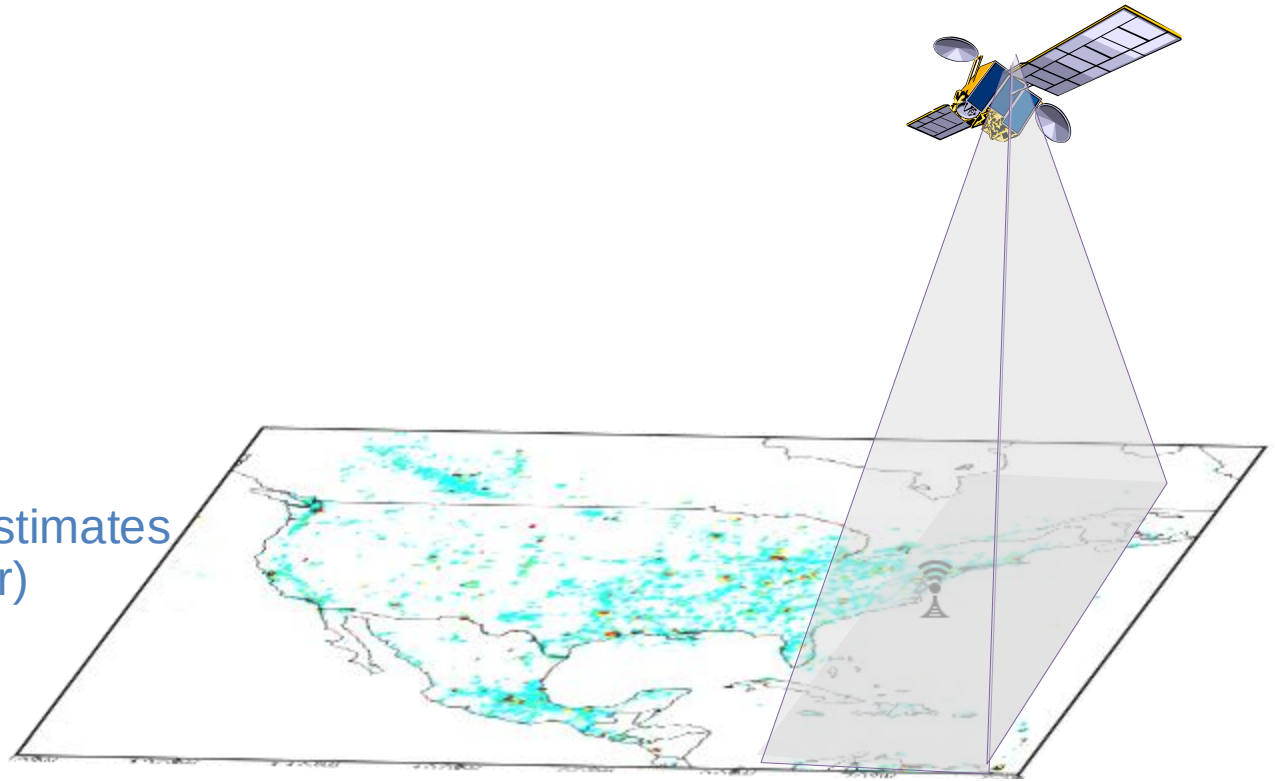
LEO Satellite



Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate

observed concentrations



bottom-up estimates
(prior)

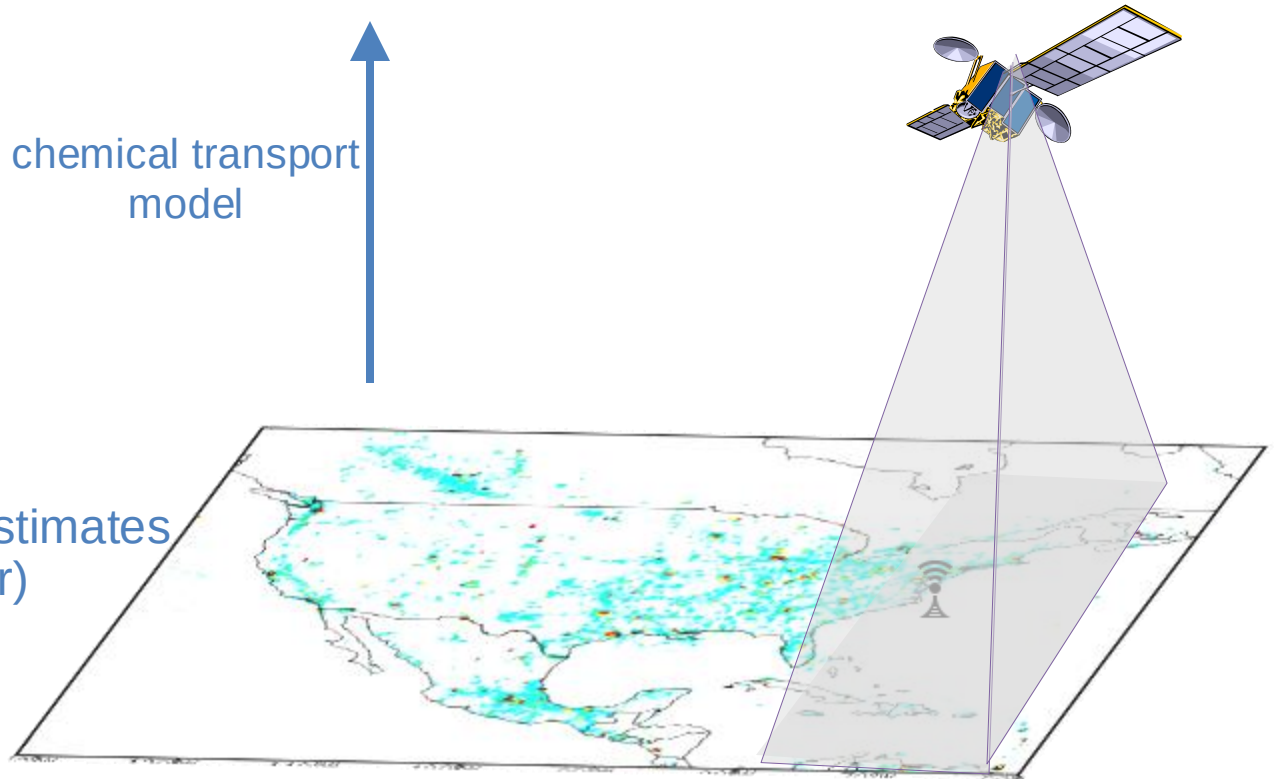
Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate

predicted concentrations ⟷ observed concentrations

chemical transport model

bottom-up estimates (prior)



Using Observations to Estimate Emissions Through Inverse Methods

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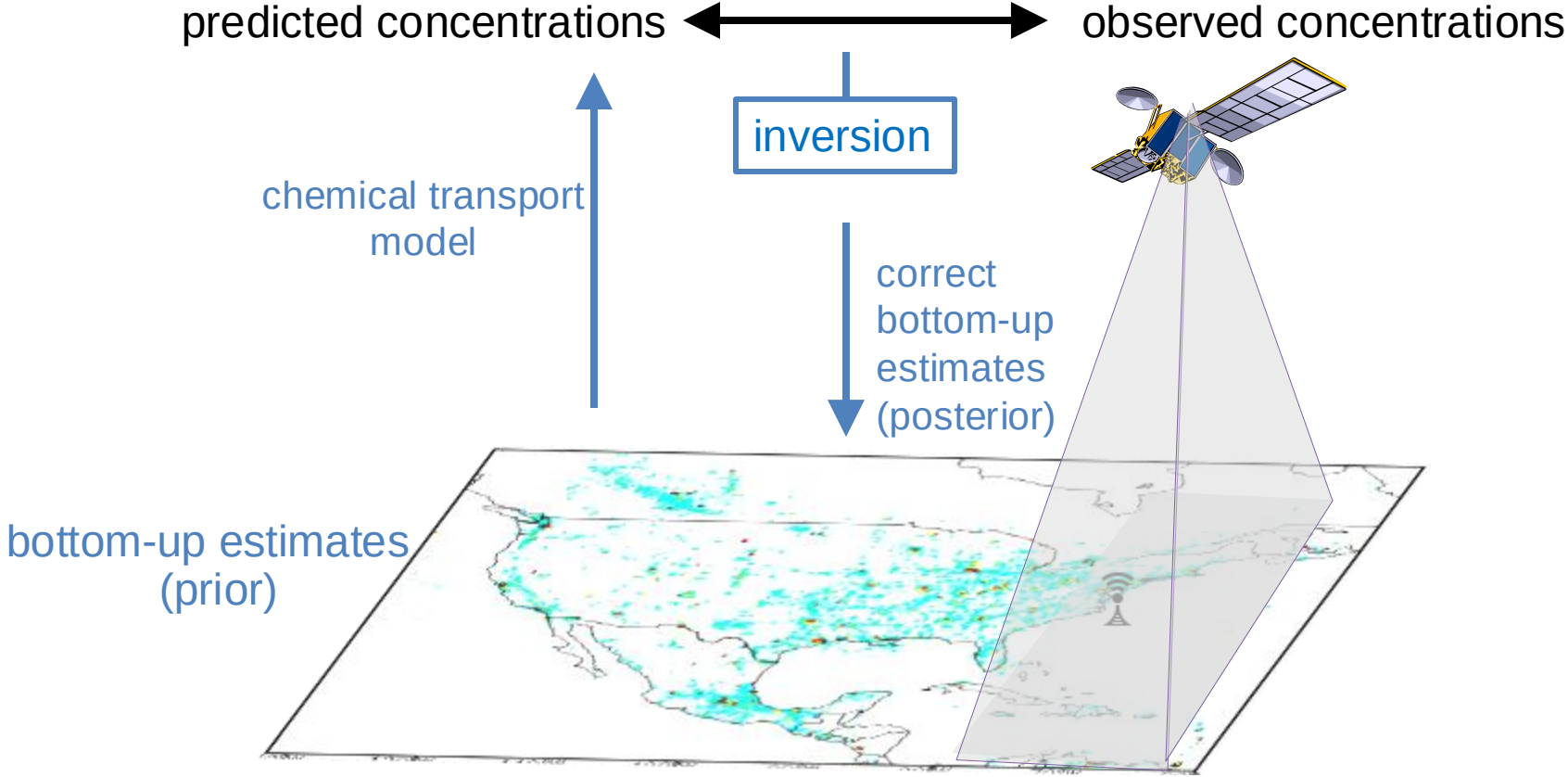


Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate

predicted concentrations ⟷ observed concentrations

inversion

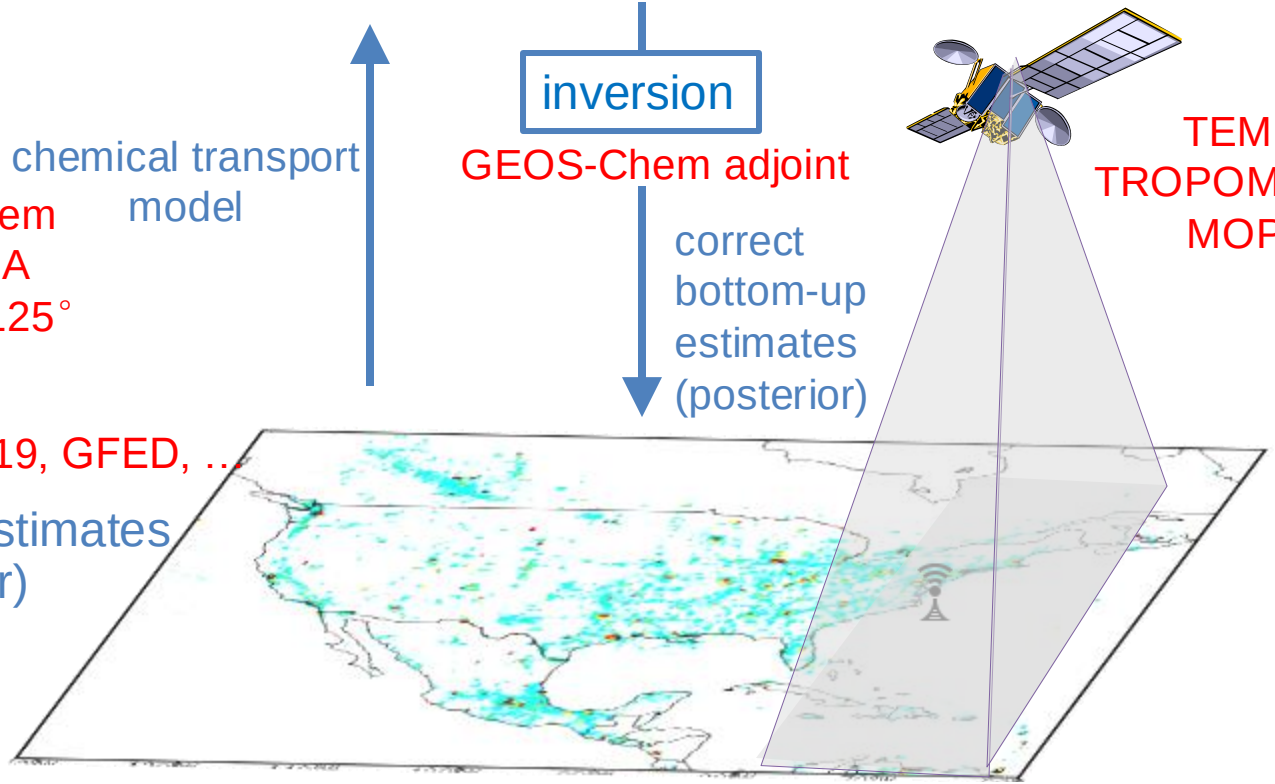
GEOS-Chem adjoint

chemical transport model

GEOS-Chem
nested NA
0.25° x 0.3125°

correct bottom-up estimates (posterior)

TEMPO NO_2
TROPOMI NO_2 & SO_2
MOPITT CO



EQUATES 2019, GFED, ..

bottom-up estimates (prior)

Bayesian Inference of Emissions: 4D-Var

Minimize cost Function:

Prior Term

Observational Term

$$J(x) = \frac{1}{2}(\boldsymbol{x} - \boldsymbol{x}_a)^T \mathbf{S_a}^{-1}(\boldsymbol{x} - \boldsymbol{x}_a) + \frac{1}{2}\gamma(\boldsymbol{y} - \mathcal{H}\boldsymbol{x})^T \mathbf{S_o}^{-1}(\boldsymbol{y} - \mathcal{H}\boldsymbol{x})$$

Bayesian Inference of Emissions: 4D-Var

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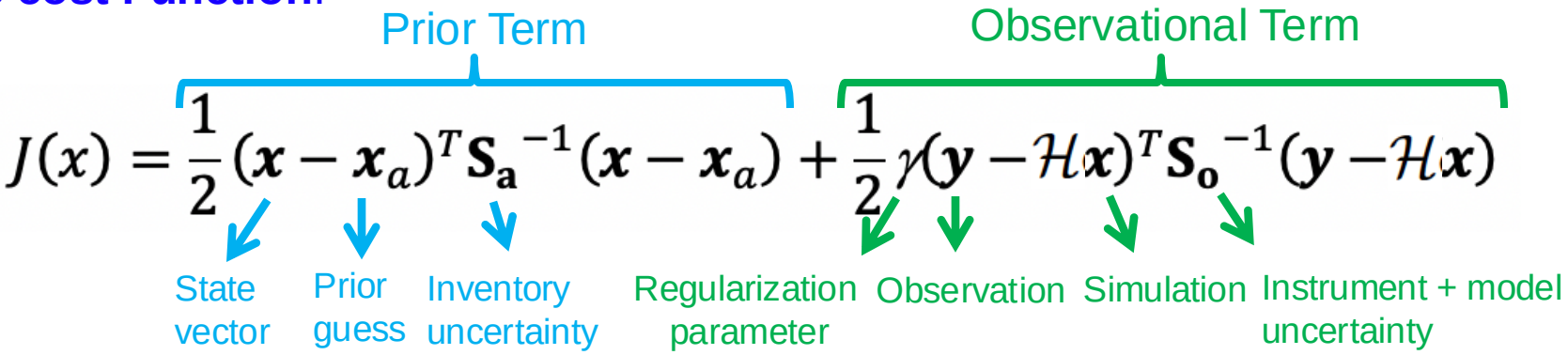

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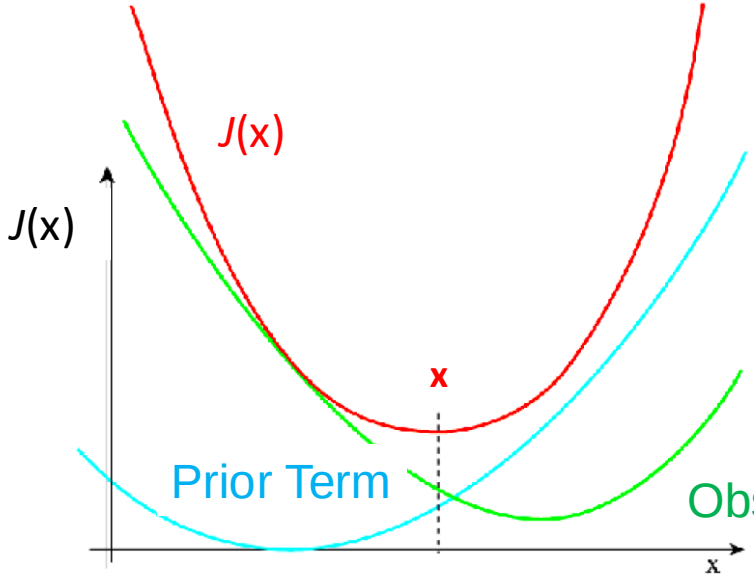
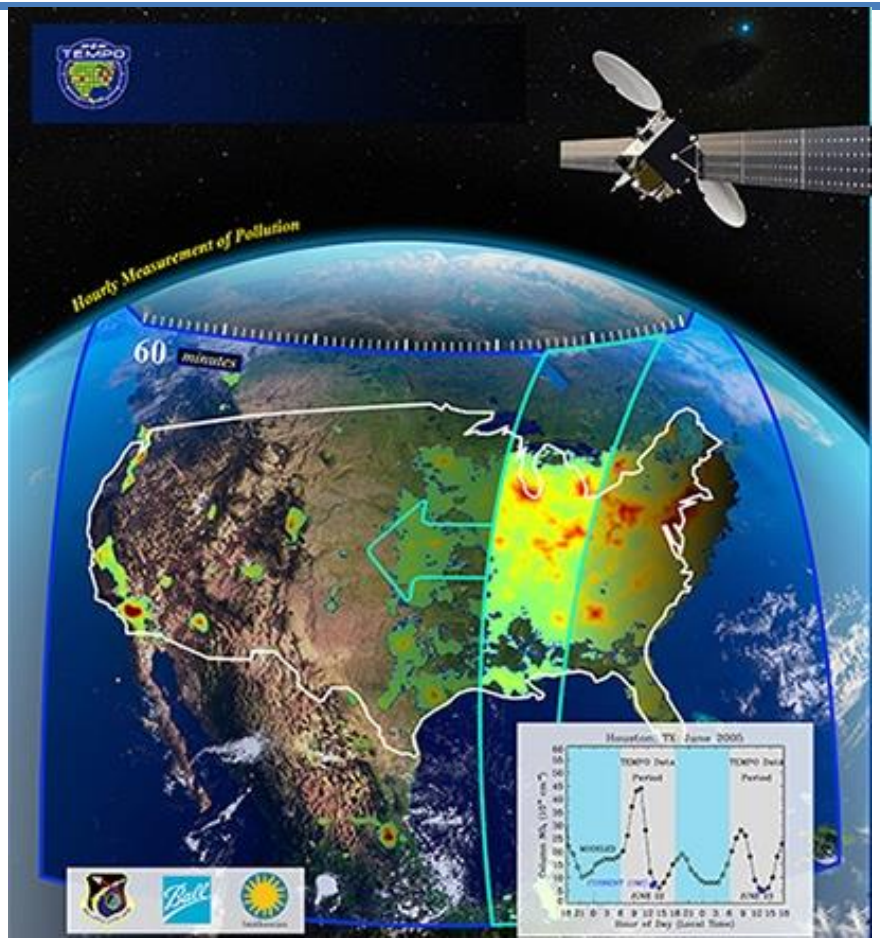


- Seeks solution iteratively
- Gradient based optimization
- Adjoint model calculates the sensitivity of cost function w.r.t. state vector

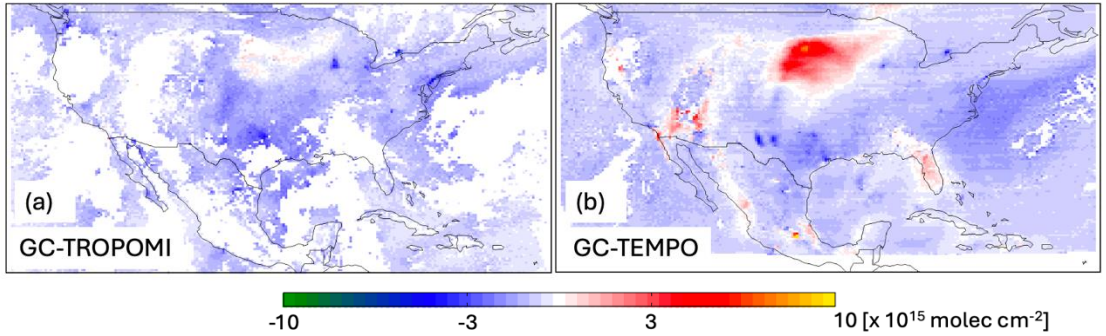
Geostationary Satellite TEMPO Enables Daytime Hourly Monitoring of NO_2



- NO_2 retrievals available from Aug 2023
- Daytime hourly data coverage
- Footprint: 2.1 km × 4.7 km
- TEMPO observations can better identify source sectors leveraging the different diurnal variations of each source

Observations Suggest Underestimate of EQUATES NO_x Emissions

NO_2 Slant Column Densities (Sep 1, 2023)



SCD = VCD x AMF

GEOS-Chem simulations with EQUATES inputs underestimate NO_2 compared to observations.

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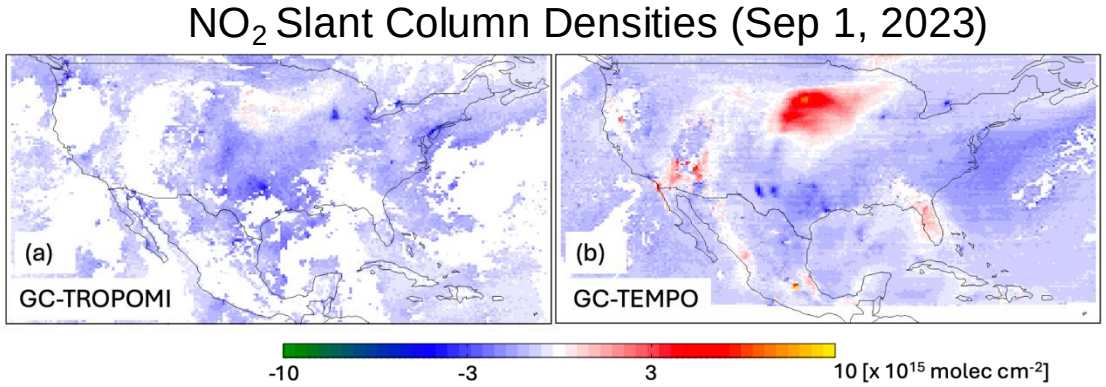

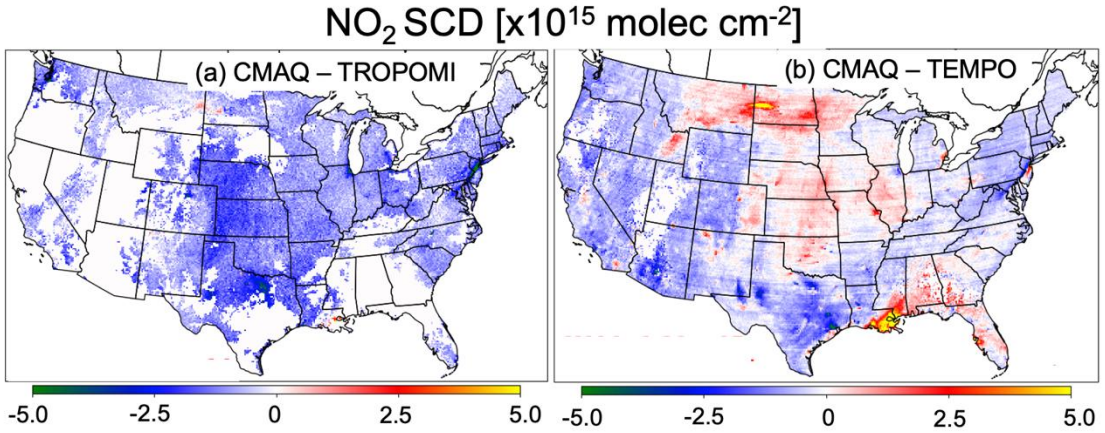

CMAQ simulations show consistent underestimates compared to observations.

(By Postdoc Nana Wu)

Observations Suggest Underestimate of EQUATES NO_x Emissions

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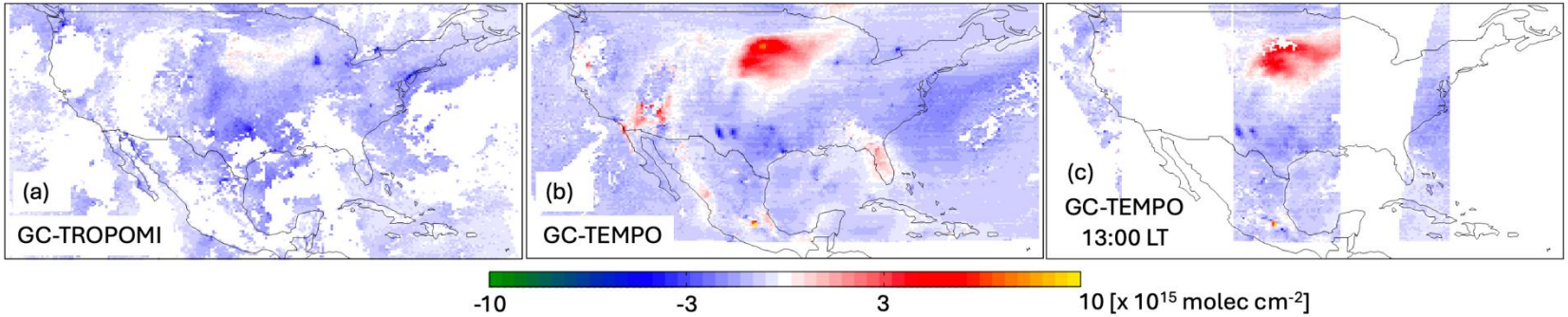

Consistent biases using (b) all TEMPO data & (c) only data at 13:00 LT
→ Discrepancies in (a) & (b) are due to retrieval differences, not data coverage

The differences are attributed to the different magnitudes of scattering weights in these two retrievals.

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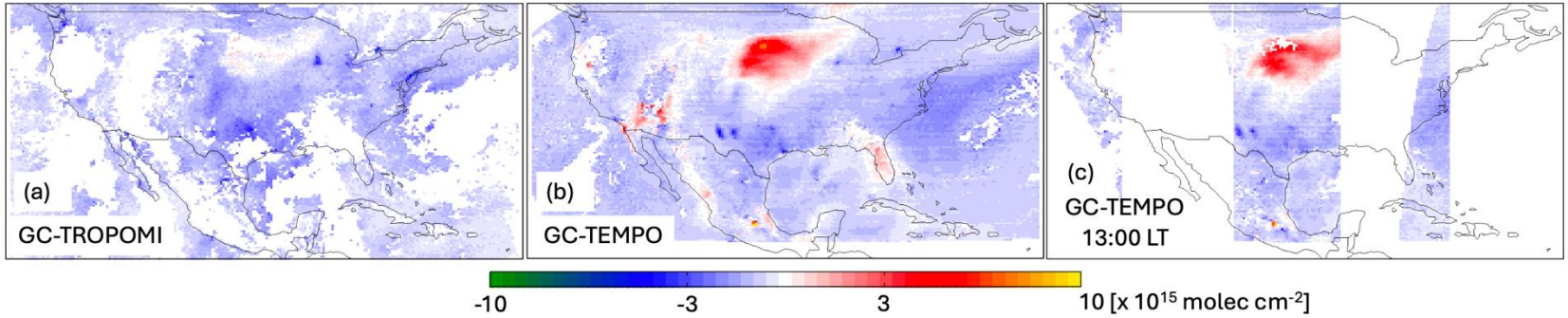
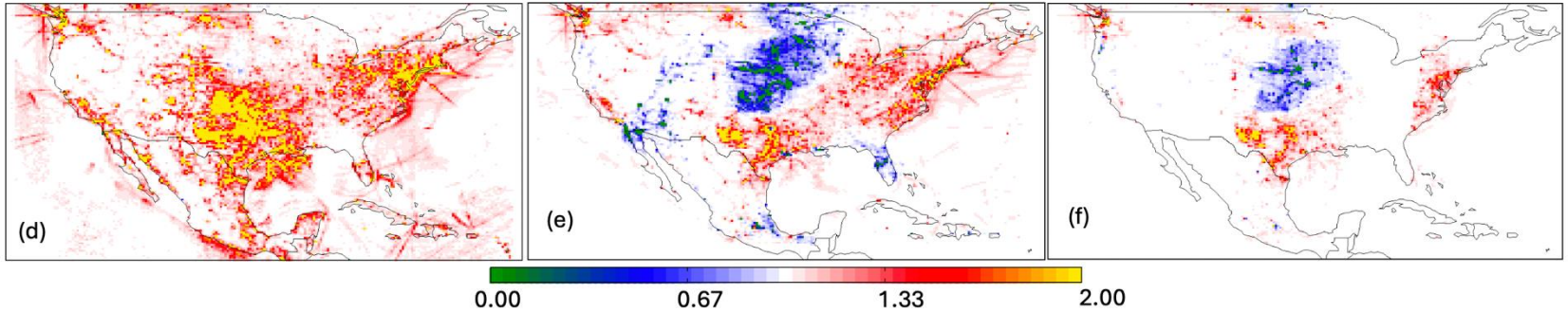
NO_2 Slant Column Densities (Sep 1, 2023)



Posterior / prior Ratio of Emissions (Sep 1, 2023)

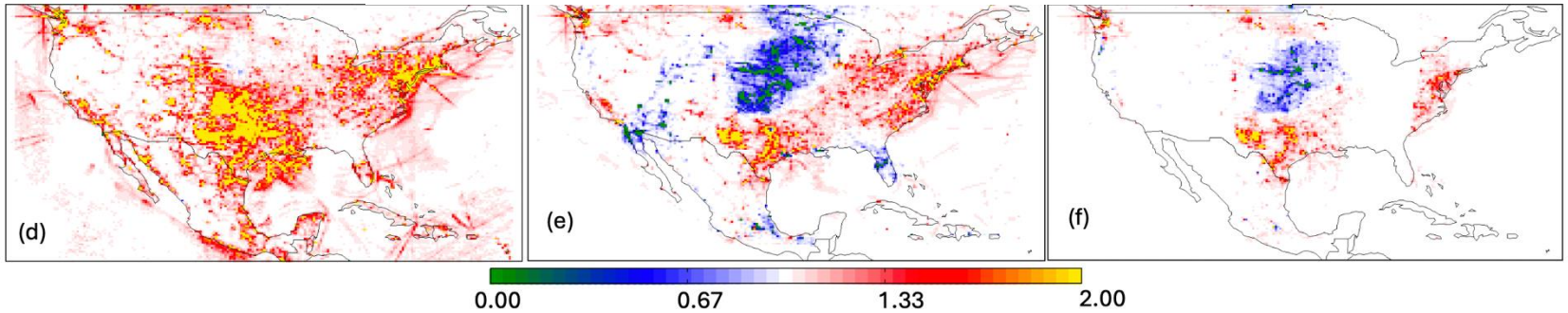


Observations Suggest Underestimate of EQUATES NO_x Emissions

NO_2 observations suggest underestimates in EQUATES emissions.

Is it true???

Posterior / prior Ratio of Emissions (Sep 1, 2023)

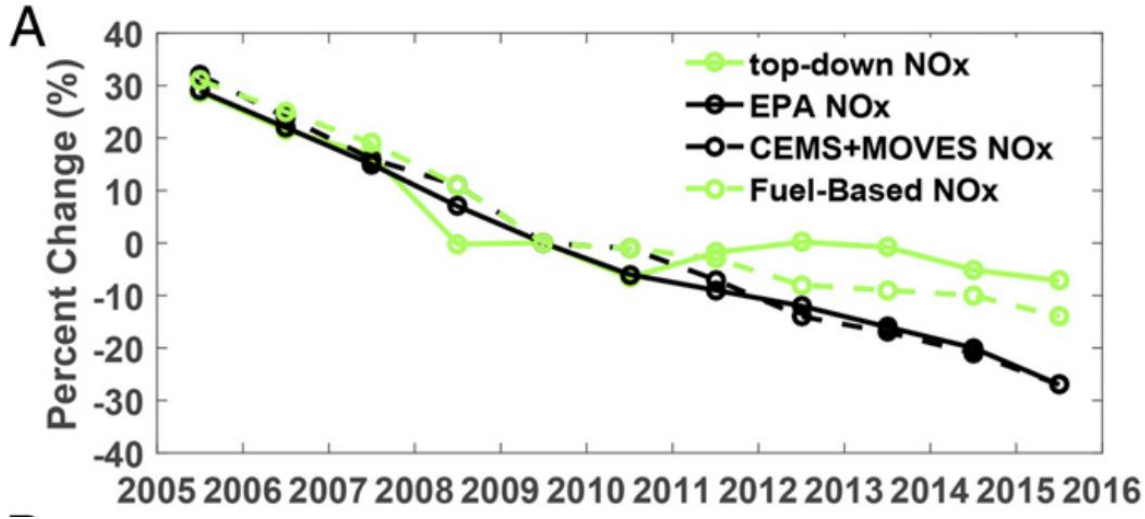


- TEMPO posterior NO_x emissions are 47% higher than the EQUATES emissions
- TROPOMI posterior NO_x emissions are 56% higher than TEMPO posterior over CONUS

Unexpected slowdown of US pollutant emission reduction in the past decade

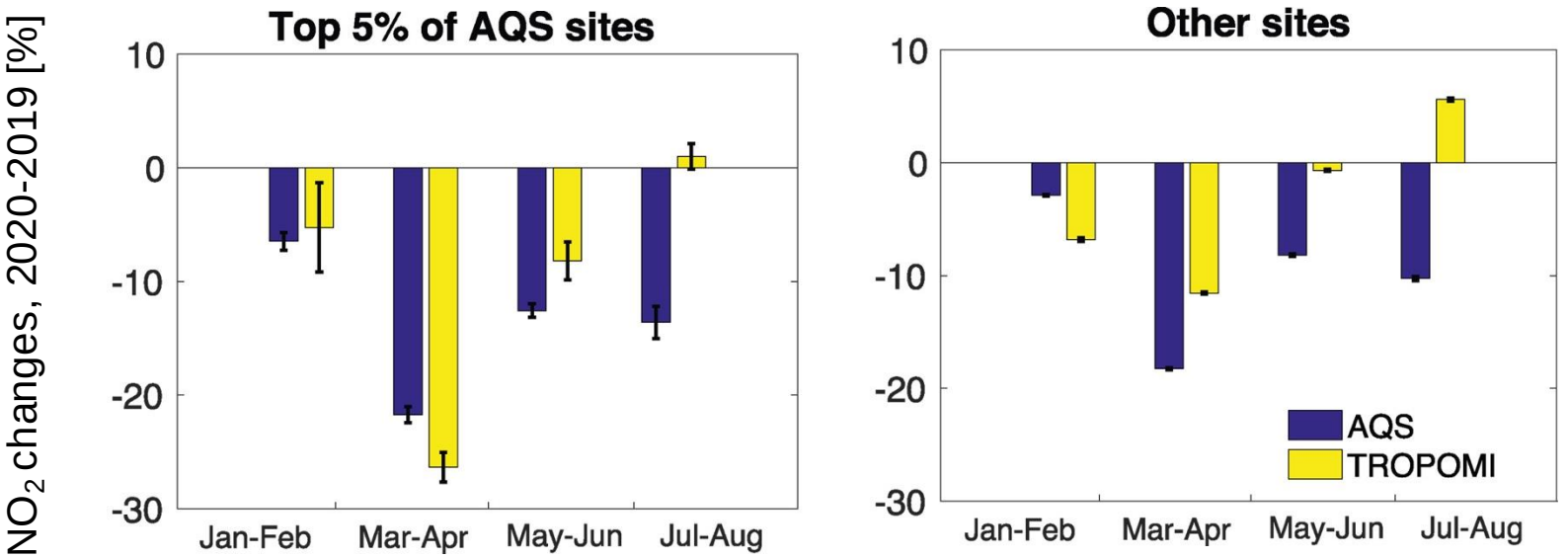
Zhe Jiang[a,b,1], Brian C. McDonald[c,d], Helen Worden[a], John R. Worden[e], Kazuyuki Miyazaki[f], Zhen Qu[g], Daven K. Henze[g], Dylan B. A. Jones[h], Avelino F. Arellano[i], Emily V. Fischer[j], Liye Zhu[j,2], and K. Folkert Boersma[k,l]

[a]Atmospheric Chemistry Observations and Modeling Laboratory, National Center for Atmospheric Research, Boulder, CO 80301; [b]School of Earth and Space Sciences, University of Science and Technology of China, Hefei, Anhui, 230026, China; [c]Chemical Sciences Division, National Oceanic and Atmospheric Administration Earth System Research Laboratory, Boulder, CO 80305; [d]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO 80309; [e]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91009; [f]Research and Development Center for Global Change, Japan Agency for Marine-Earth Science and Technology, Yokohama, 236-0001, Japan; [g]Department of Mechanical Engineering, University of Colorado Boulder, Boulder, CO 80309; [h]Department of Physics, University of Toronto, Toronto, ON M5S 1A7, Canada; [i]Department of Hydrology and



(*Jiang et al.*, 2018)

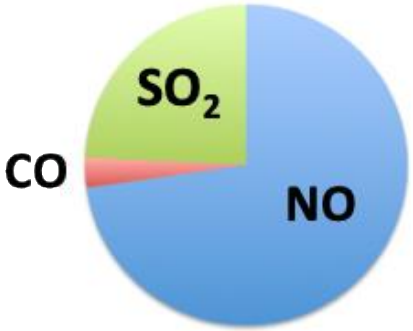
US COVID-19 Shutdown Demonstrates Importance of Background NO_2 in Inferring NO_x Emissions from Satellite NO_2 Observations

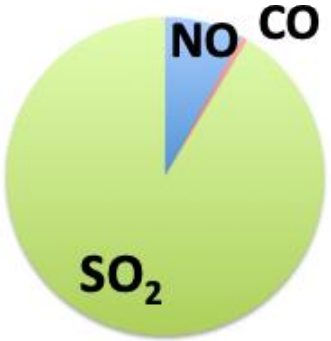


- TROPOMI tracks the change in Mar-Jun at low background sites but only reductions in Mar-Apr at other sites.

(*Qu et al.*, 2021)

Unique Emission Profile for Each Source





Sector-based Inversion: Independent Adjustments for Each Source

Species-based inversion: optimize total NO_x, SO_2, and CO emissions.

Sector-based inversion: optimize activity rates of each sector.

E = species emission factor × activity

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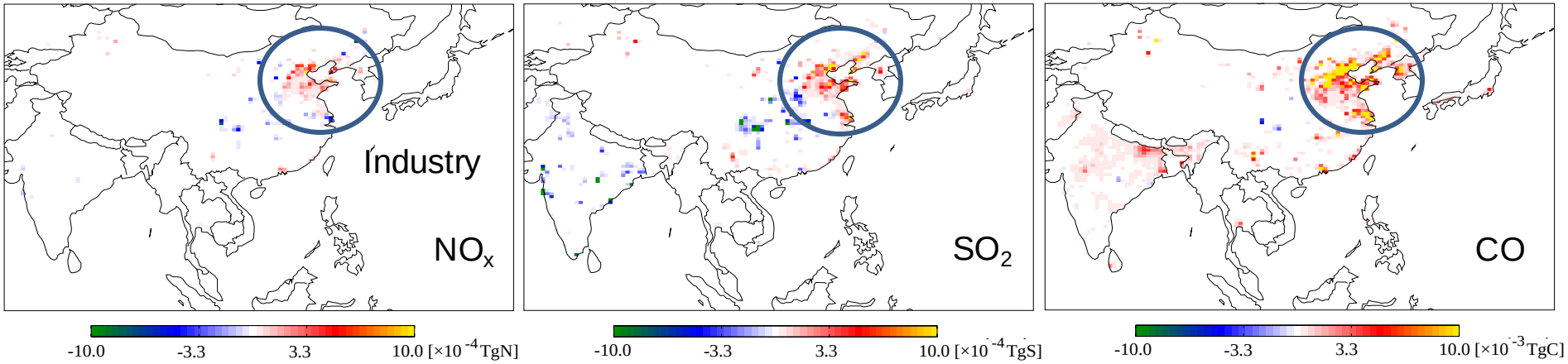
Emission adjustments (Top-down – bottom-up, Jan, 2010)

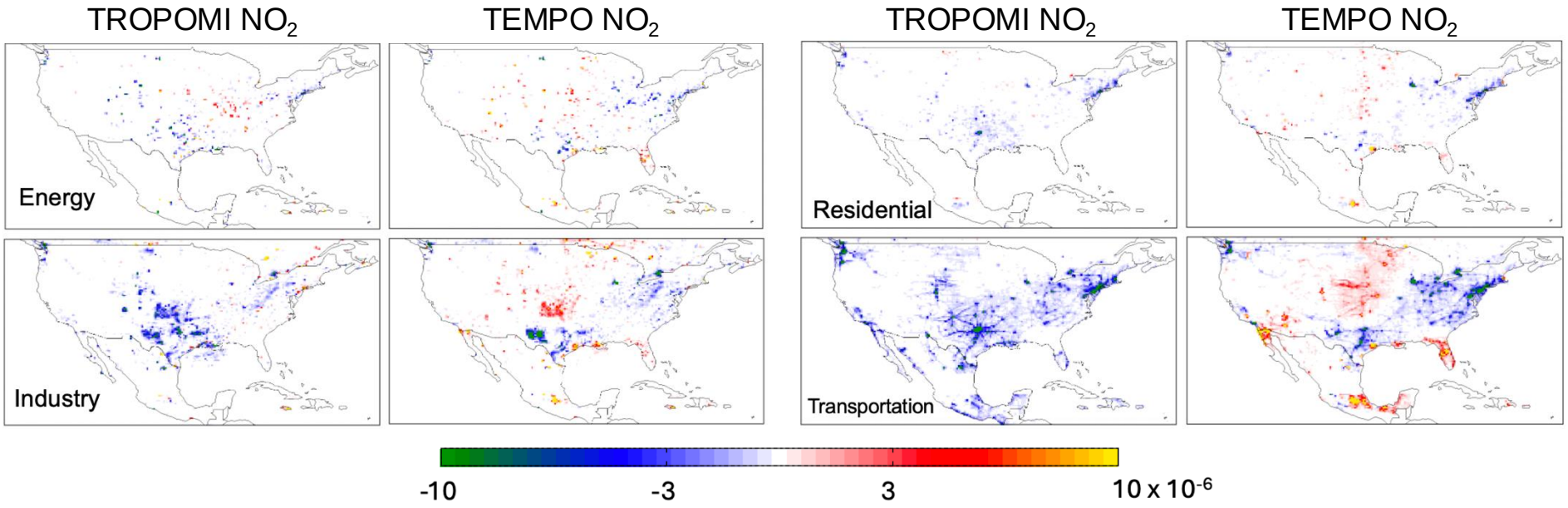


(*Qu et al.*, 2022)

Bottom-up emissions: overestimate underestimate

Sector-based Inversion: Independent Adjustments for Each Source

Normalized Adjoint Sensitivities (Sep 1, 2023)



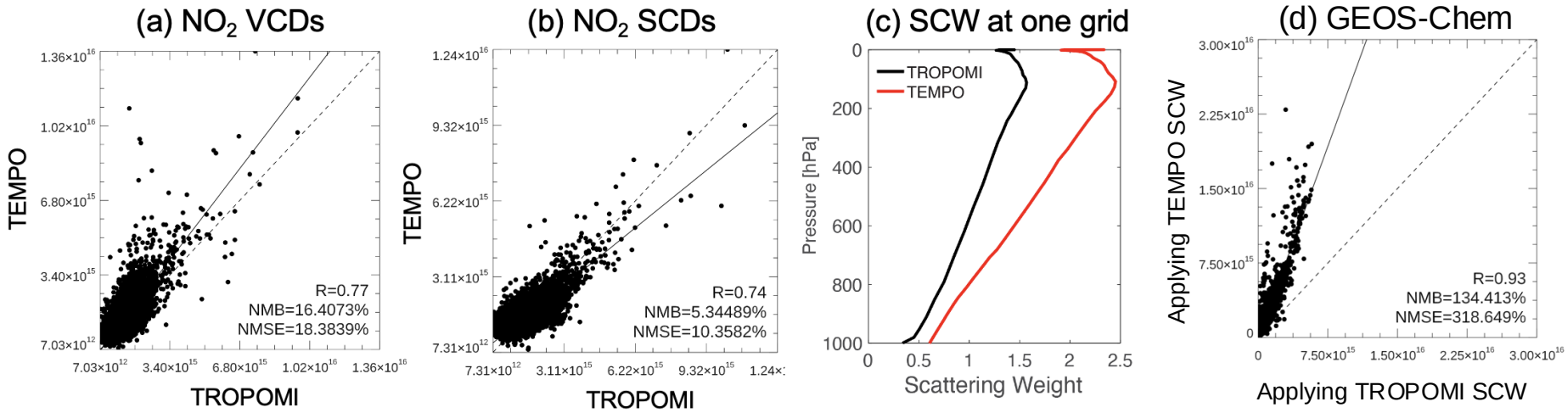
Positive adjoint sensitivity: emissions should decrease to match observations

Summary

Integrating daytime hourly TEMPO NO2 observations into a sector-based inversion framework have the potential to better inform emission adjustments from each source sector and evaluate bottom-up inventories at the process level.

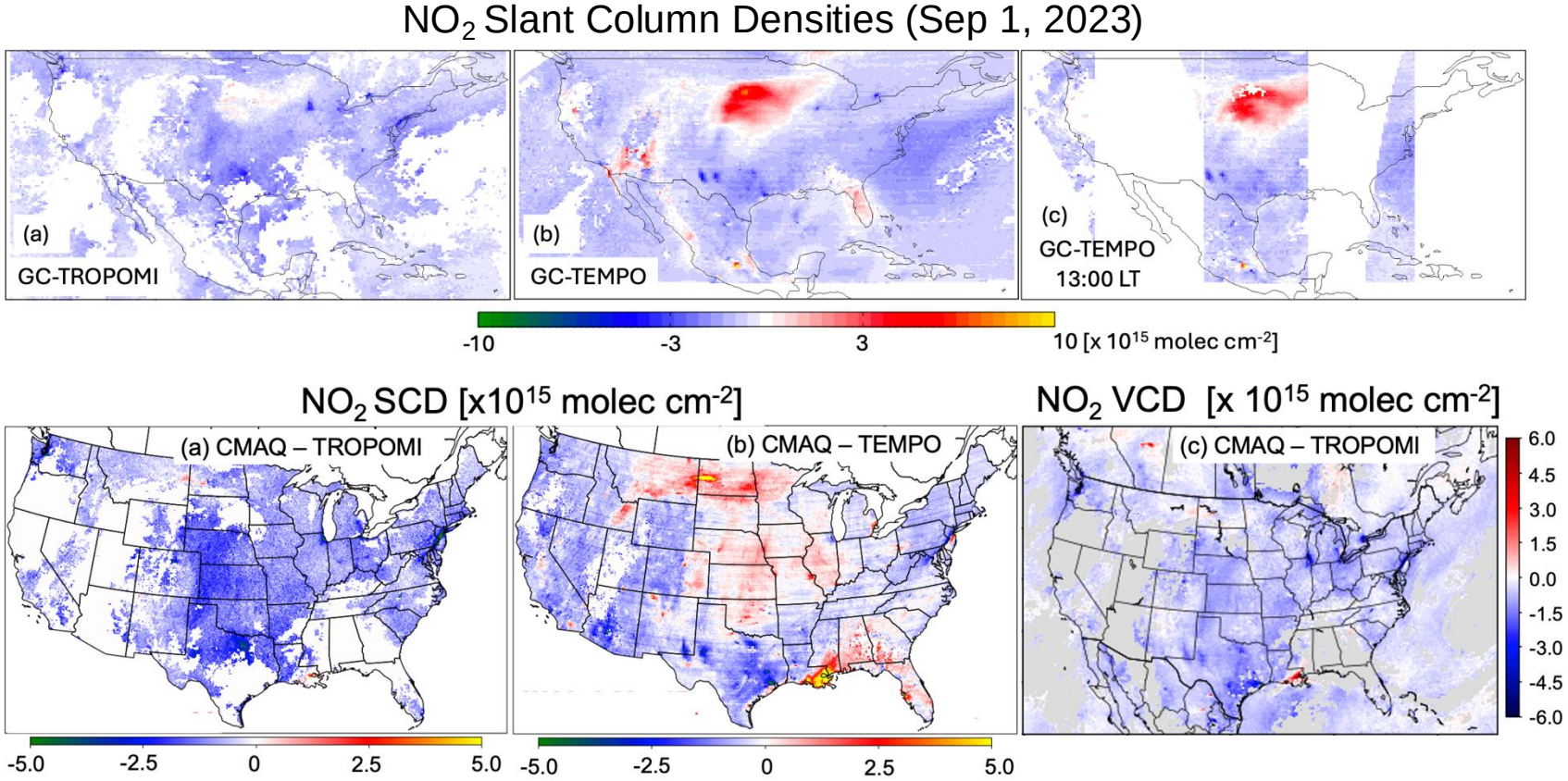
- TROPOMI-constrained NOx emissions show upward adjustments compared to EQUATES, whereas TEMPO constraints show mixed adjustments. The differences are attributed to different magnitudes of scattering weights in these two retrievals.
- A sector-based inversion framework attributes the underestimates of emissions mostly to the transportation and energy sectors.

NO_2 Discrepancies Are Caused by Scattering Weight



Larger scattering weight (SCW) from TEMPO leads to larger simulated NO2 SCDs and high biases compared to observations.

Observations Suggests Underestimate of EQUATES NO_x Emissions



CMAQ simulations show consistent comparisons with GEOS-Chem.

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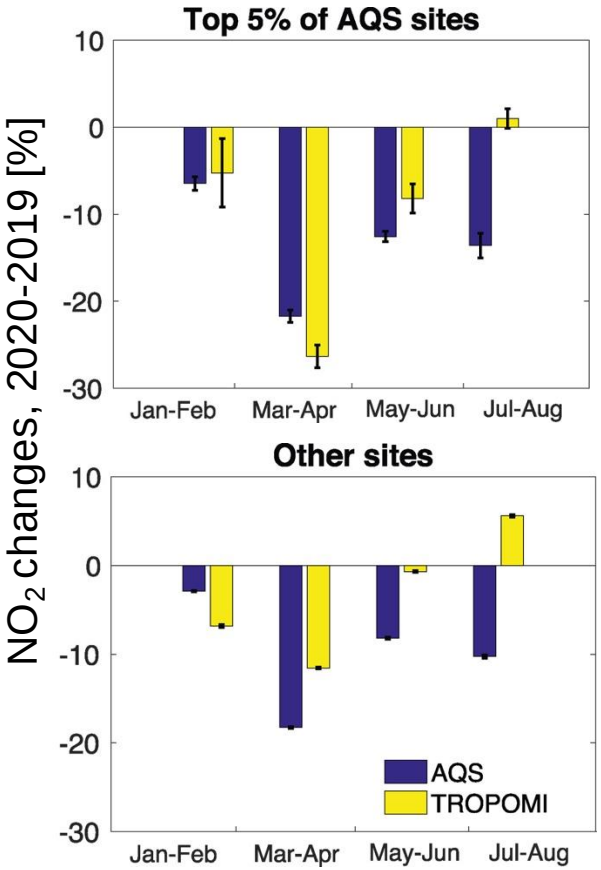

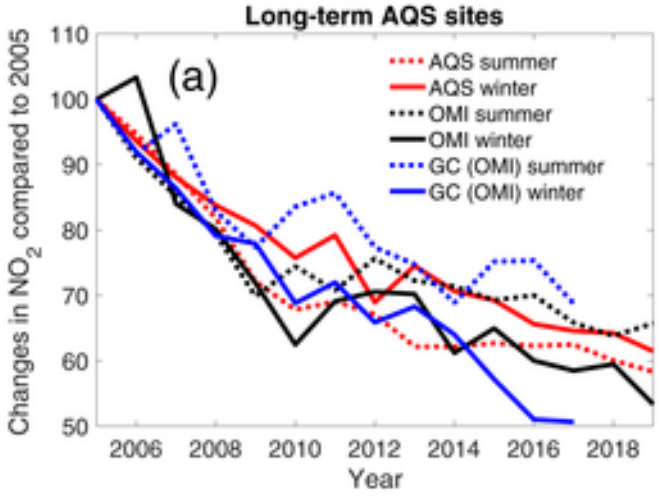

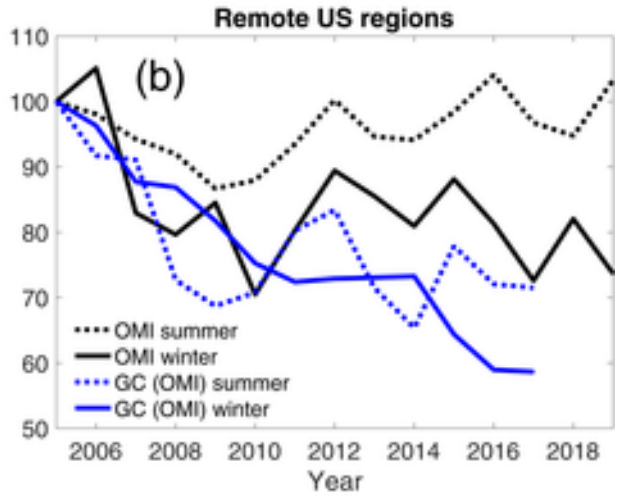

- TROPOMI tracks the change in Mar-Jun at low background sites but only reductions in Mar-Apr at other sites.
- The OMI NO_2 post-2010 observations show a weaker trend in summer than winter attributed to an increasing relative contribution of background NO2 as NOx emission decreases.

(*Qu et al.*, 2021)