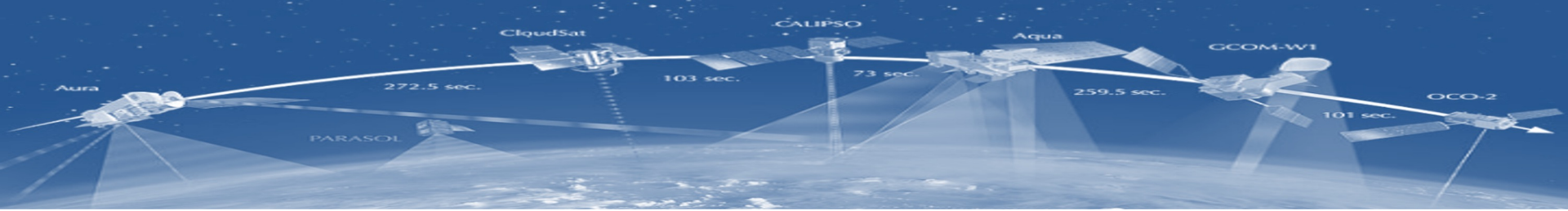


Advancing sectoral emission estimates of NO_x, SO_2, and CO using satellite observations

Zhen Qu
Assistant Professor
North Carolina State University
zqu5@ncsu.edu
CMAS, 10/17/2023

With Daven Henze, Helen Worden, Nana Wu, Zhe Jiang, Benjamin Gaubert, Nicholas Theys, Wei Wang, Can Li, Yi Wang, Jun Wang

Sources of SO_2, NO_x, and CO

Power Plant

Fossil Fuel Combustion

Soil

Biomass Burning

Lightning

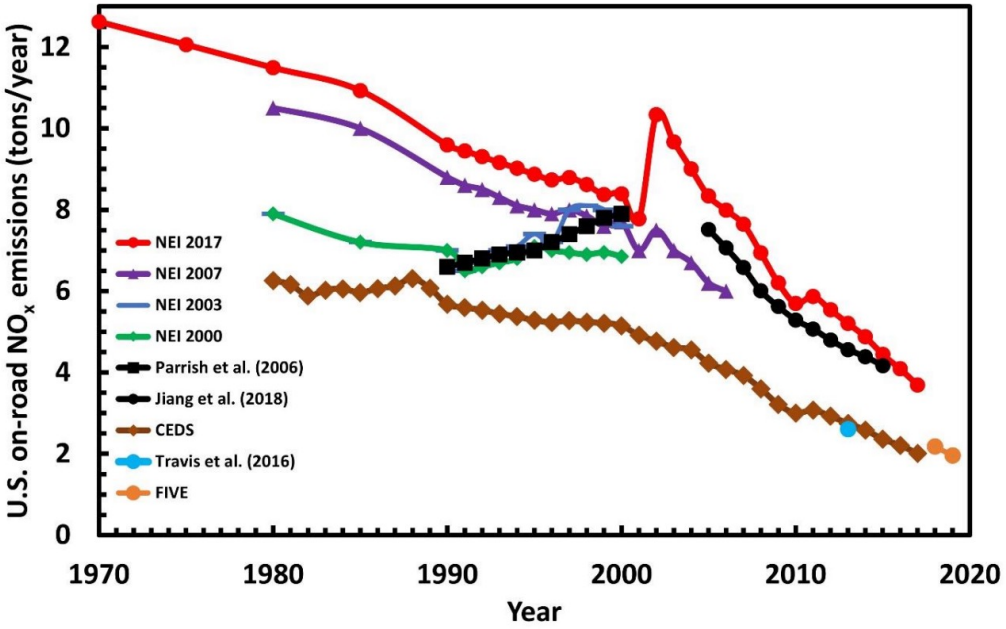
Volcano

Bottom-up Emissions Have Large Uncertainties & Lag in Time

Bottom-up estimate

Emission = species emission factor × activity

US on-road vehicle NO_x emissions

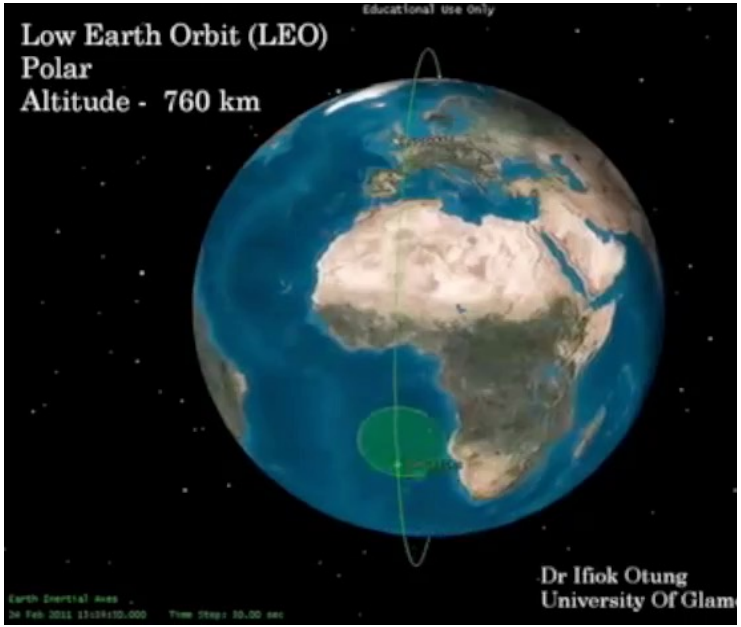


(*Christiansen et al.*, 2019)

Top-down estimate

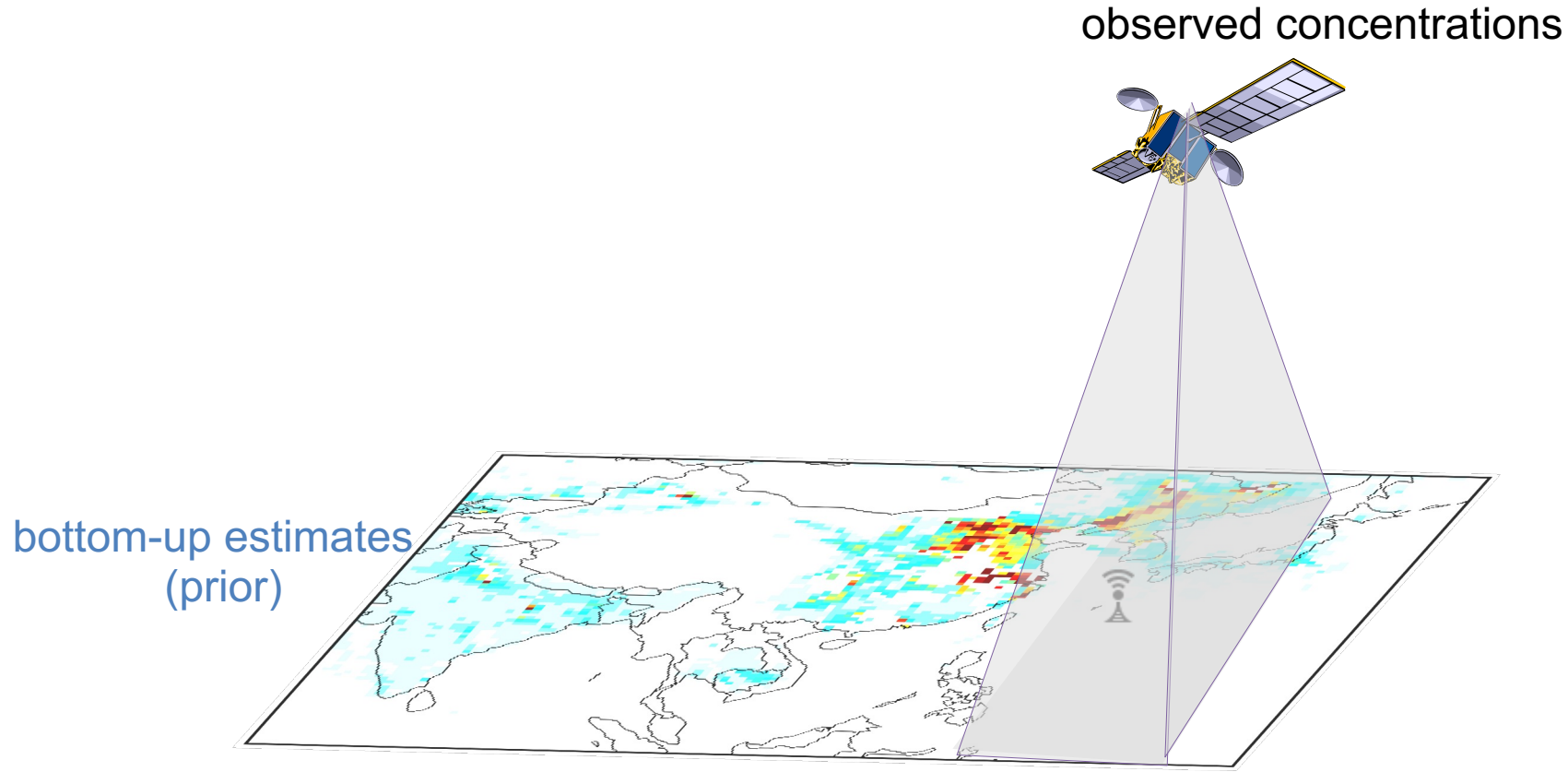
Infers emissions from observations

LEO Satellite



Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate



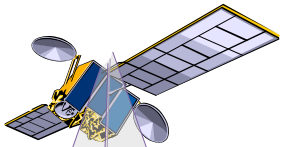
Using Observations to Estimate Emissions Through Inverse Methods

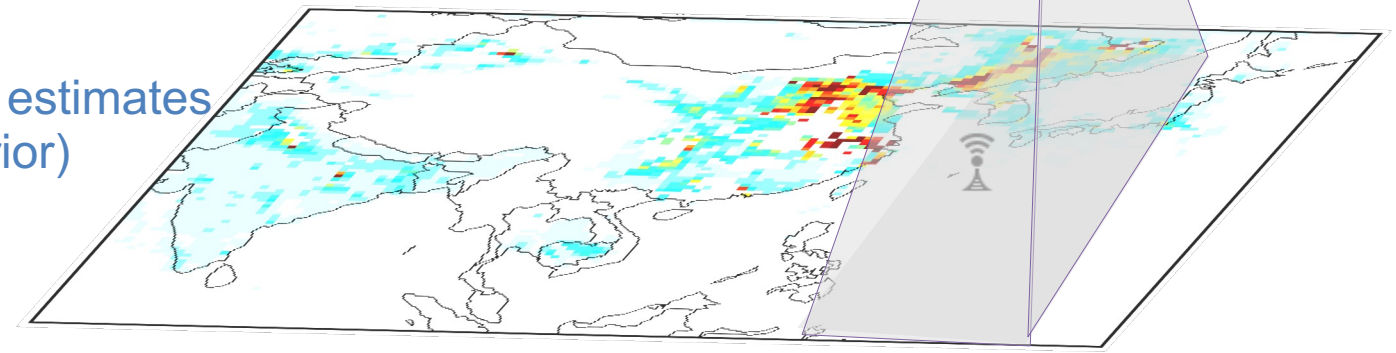
Top-down estimate

predicted concentrations ⟷ observed concentrations

chemical transport model

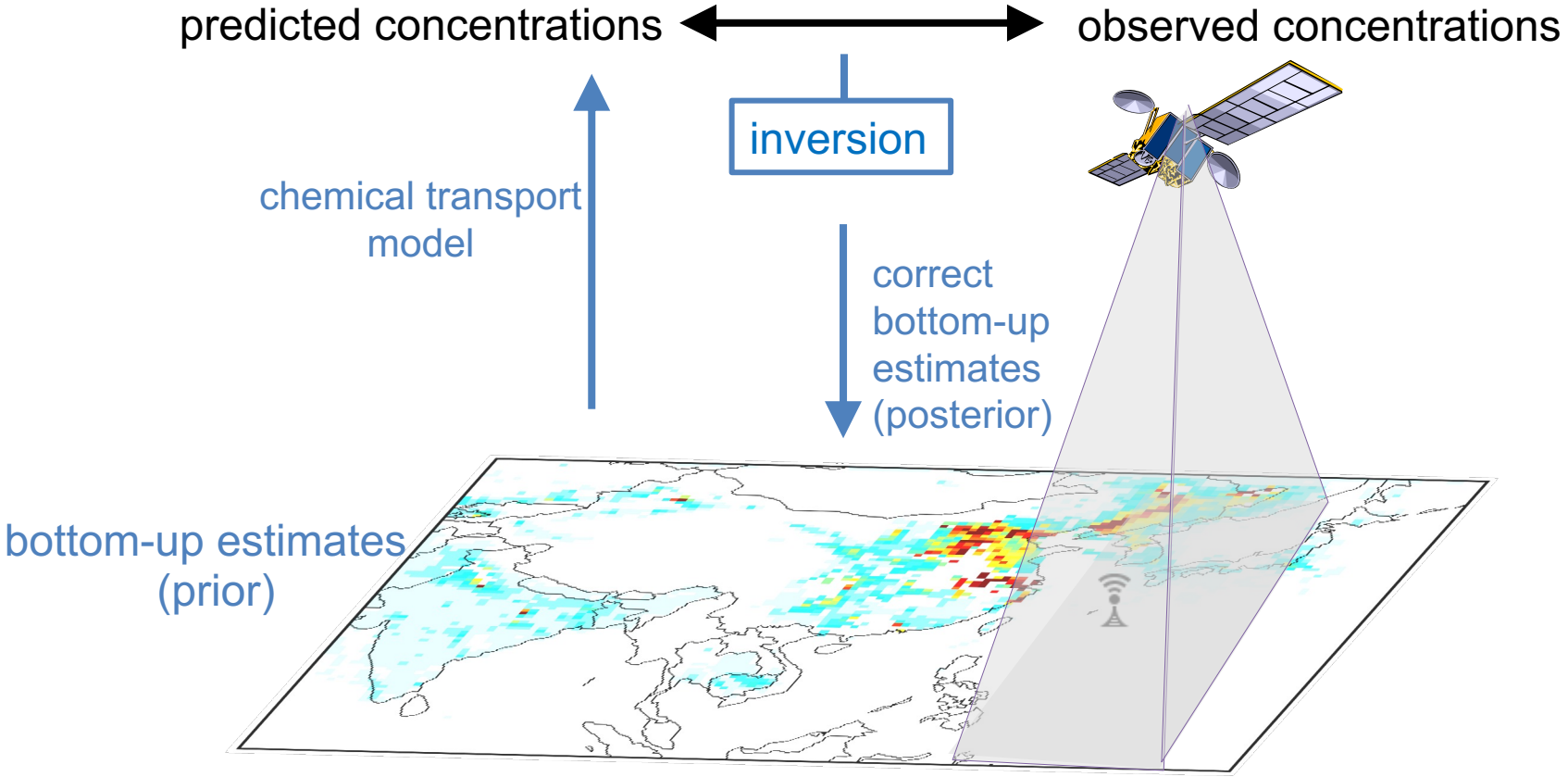
bottom-up estimates (prior)





Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate



Using Observations to Estimate Emissions Through Inverse Methods

Top-down estimate

predicted concentrations ⟷ observed concentrations

inversion

GEOS-Chem adjoint

GEOS-Chem chemical transport model

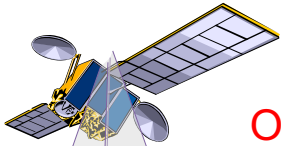
Nested EA

0.5° x 0.667°

correct bottom-up estimates (posterior)

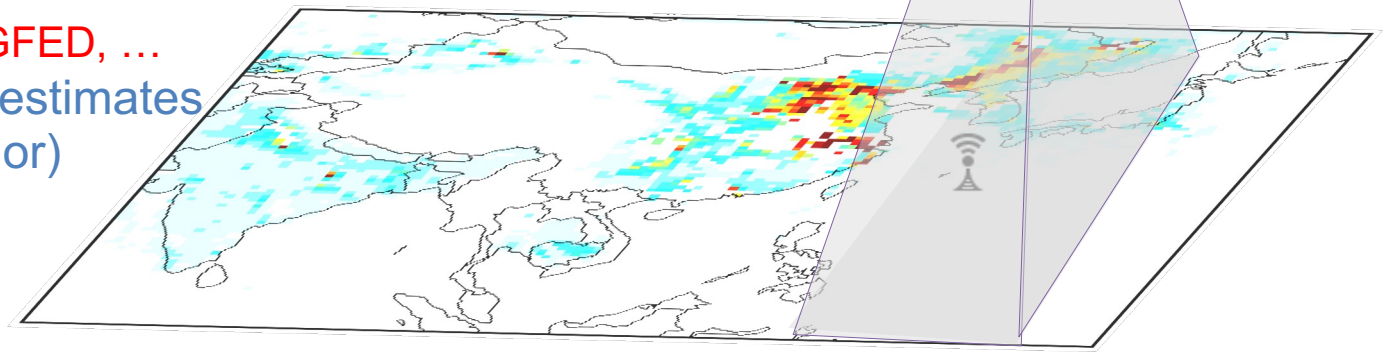
HTAP, GFED, …

bottom-up estimates (prior)



OMI NO_2 & SO_2

MOPITT CO



Bayesian Inference of Emissions: 4D-Var

Minimize cost Function:

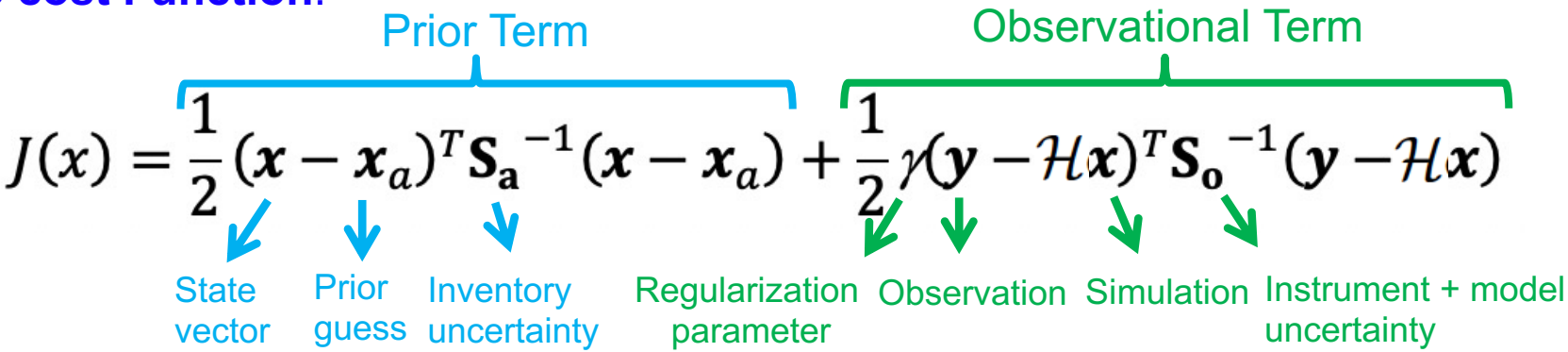
Prior Term

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

Minimize cost Function:



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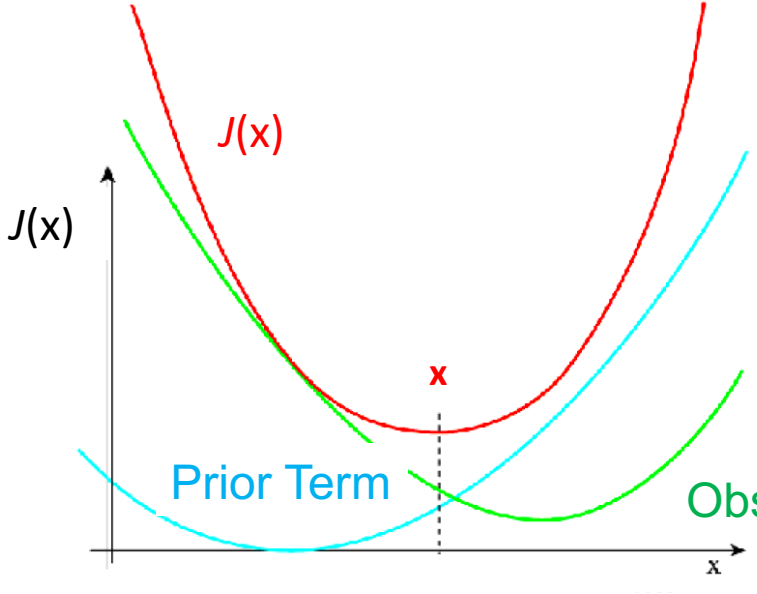


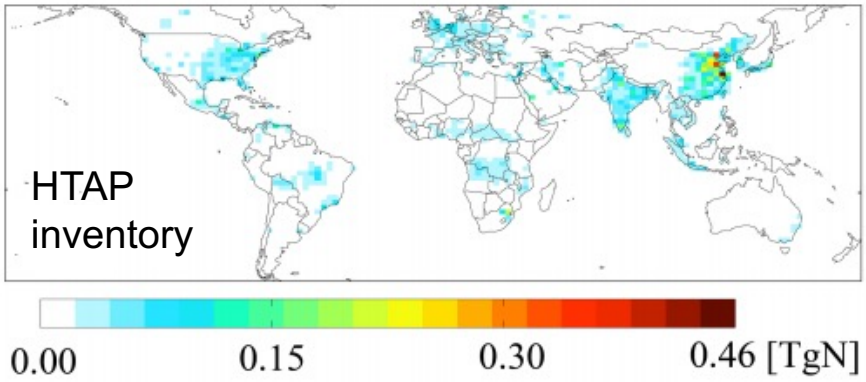
- Seeks solution iteratively
- Gradient based optimization
- Adjoint model calculates the sensitivity of cost function w.r.t. state vector

Top-down Estimate Suggests Underestimate of Bottom-up NO_x

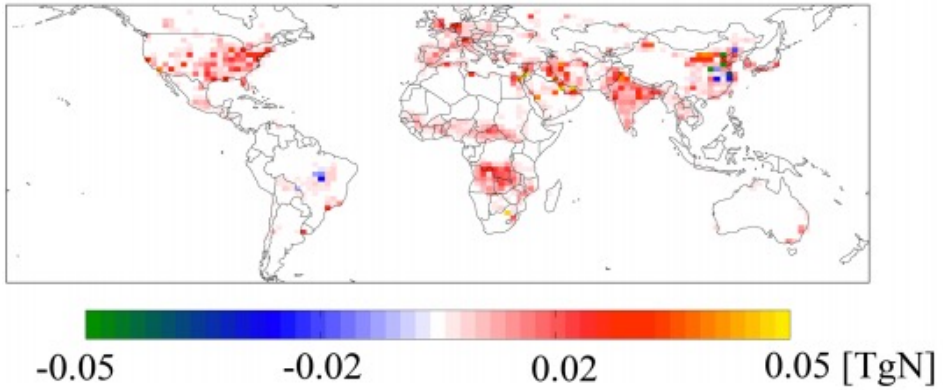
Spatial Distribution

NO_x emissions (2010)

Bottom-up



Top-down – bottom-up



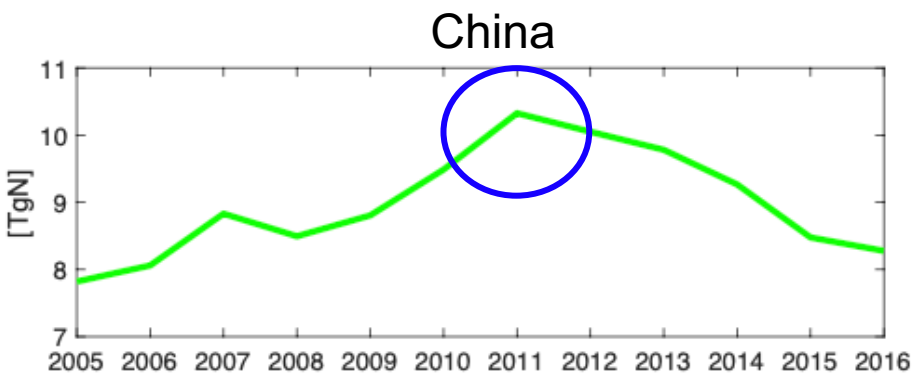
(*Qu et al.*, 2020)

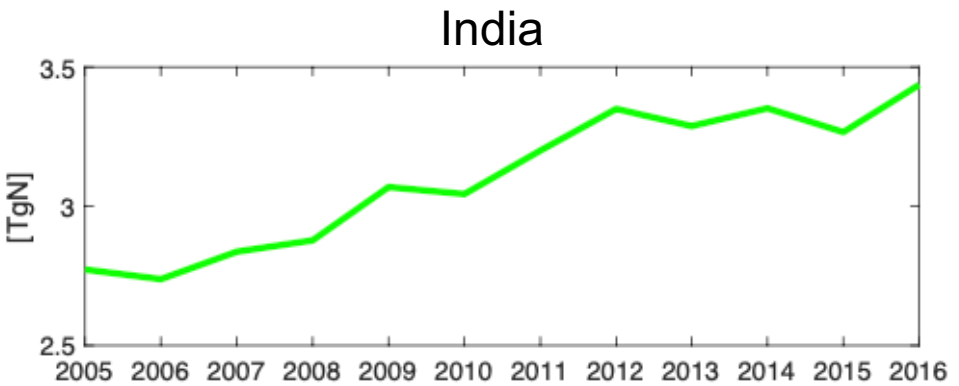
Top-down estimates correct bottom-up emissions and assist in interpreting simulations with these emission inputs.

Top-down NO_x Estimates Reflect Emission Regulations

Trend

Top-down NO_x emissions (2005-2016)

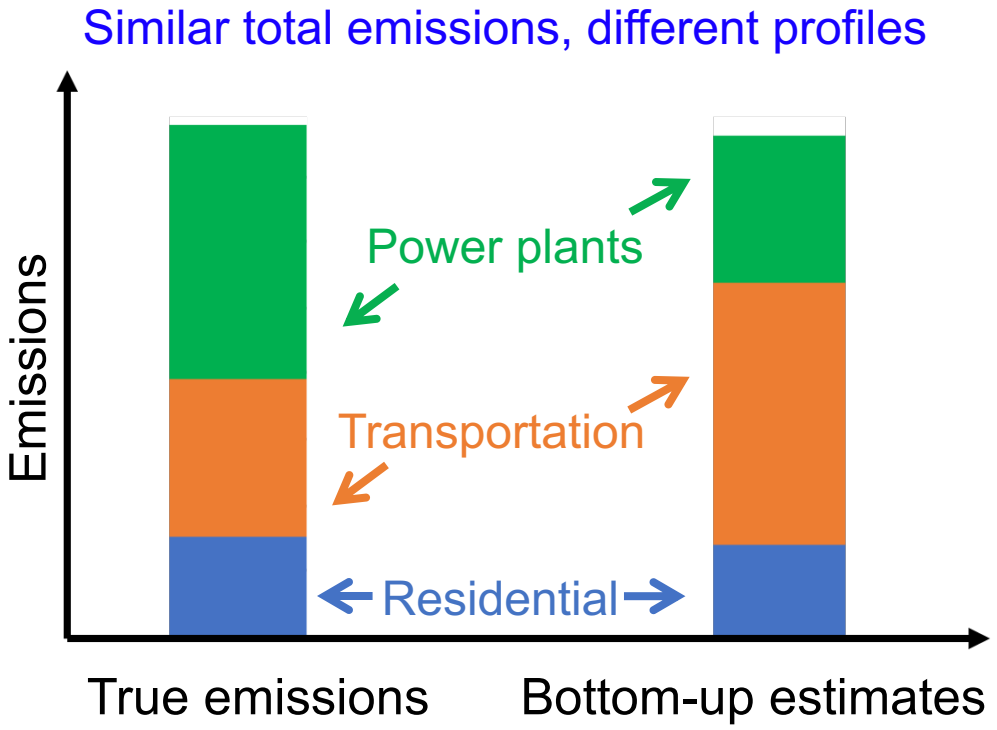




(*Qu et al.*, 2020)

China: peak in 2011 reflects regulations since the 12th Five Year Plan

Optimize Sectoral Profiles of Emissions



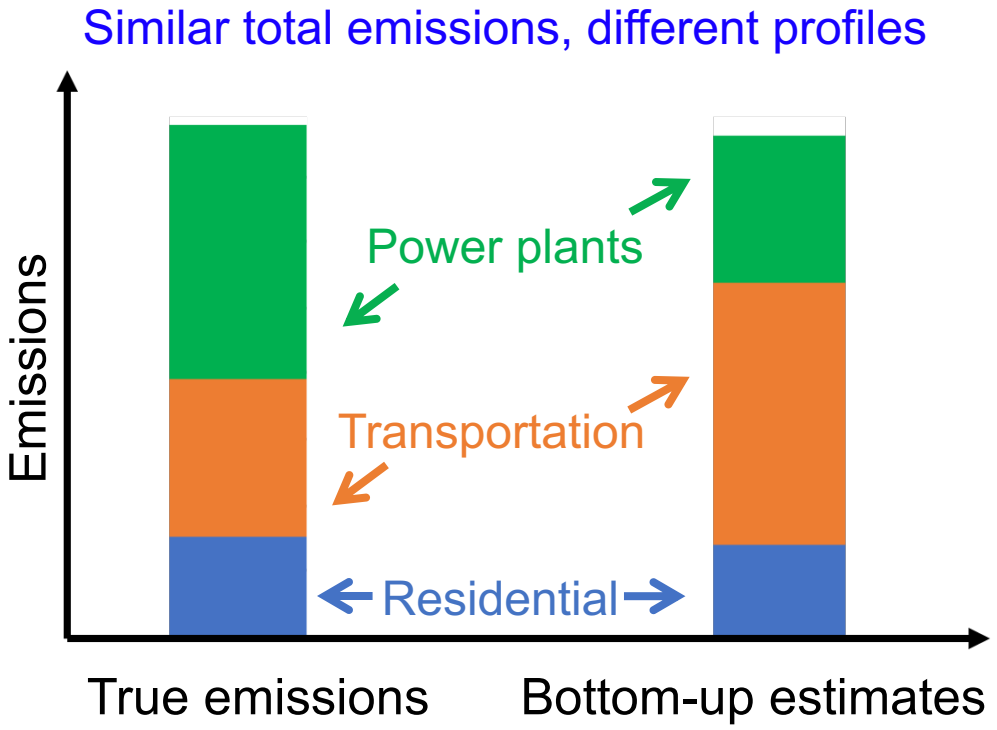
Optimize Sectoral Profiles of Emissions

Separate errors from emission factor & activity rates:

E = species emission factor × activity

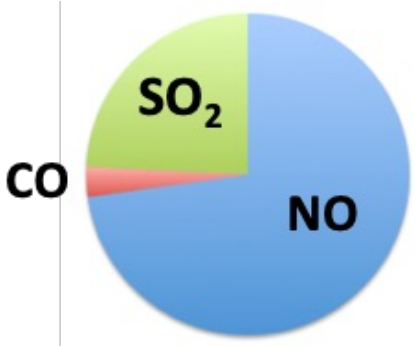
optimize

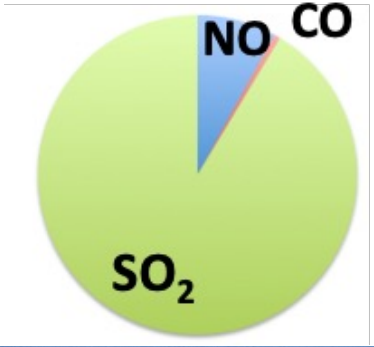
New sector-based inversion



Unique Emission Profile for Each Source

Sectoral Contribution





Need Observations of Multiple Species

TERRA

MOPITT CO

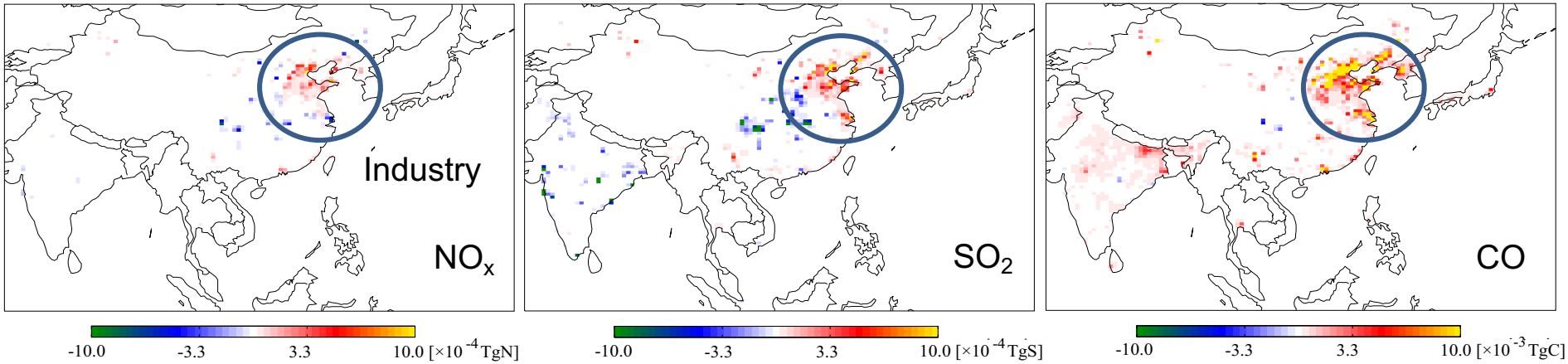
Aura

OMI NO_2 & SO_2

Sector-based Inversion: Independent Adjustments for Each Source

Sectoral Contribution

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



(*Qu et al.*, 2022)

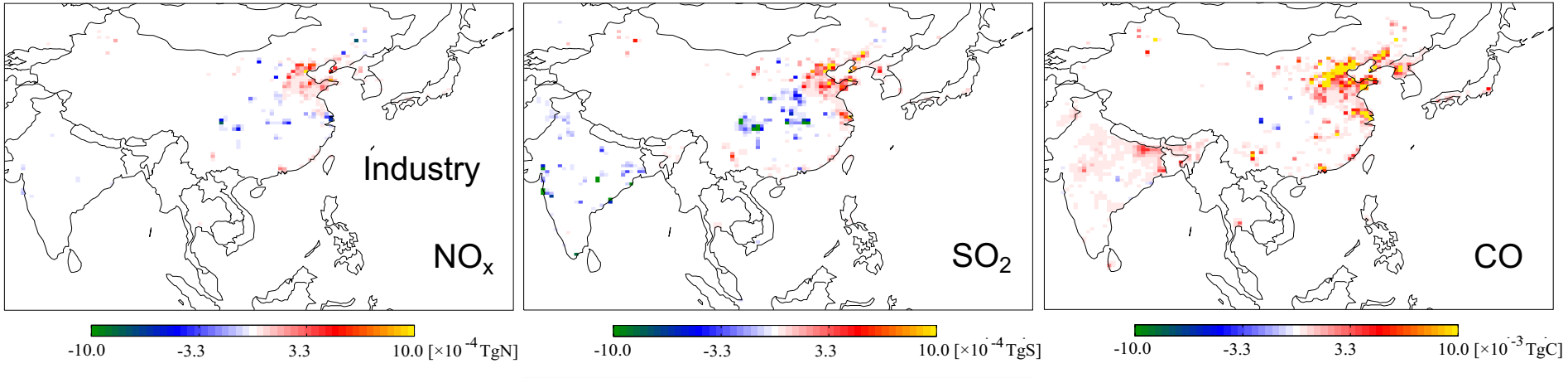
Bottom-up emissions: overestimate underestimate

E = species emission factor × activity

Sector-based Inversion: Independent Adjustments for Each Source

Sectoral Contribution

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

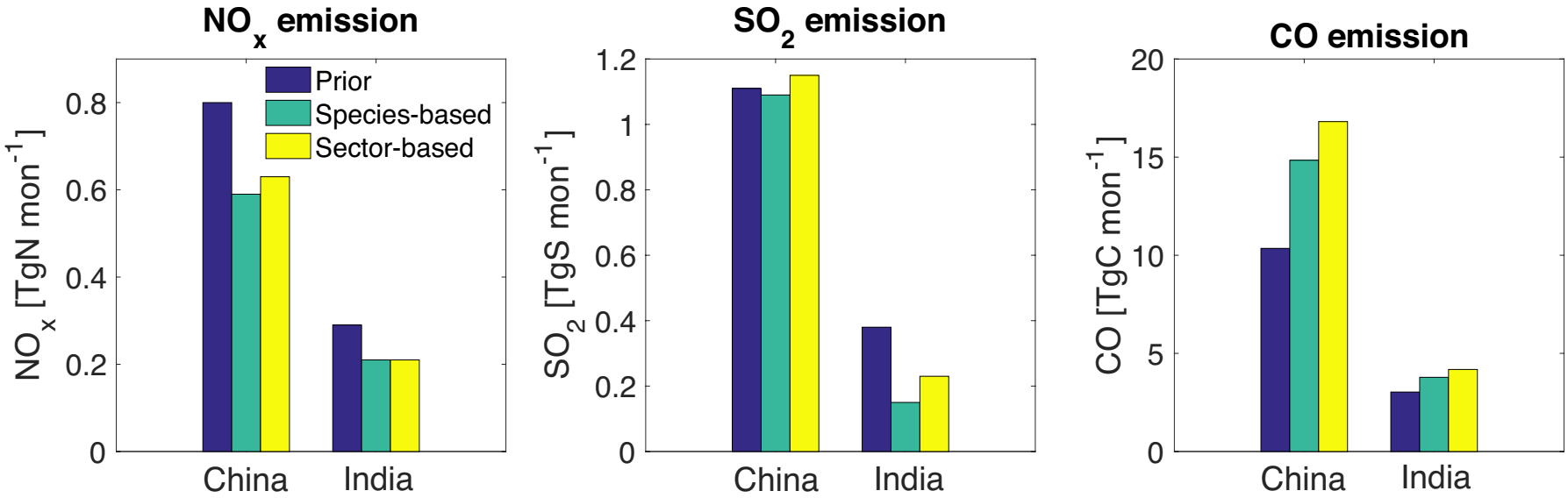


(*Qu et al.*, 2022)

Bottom-up emissions: overestimate underestimate

E = species emission factor × activity

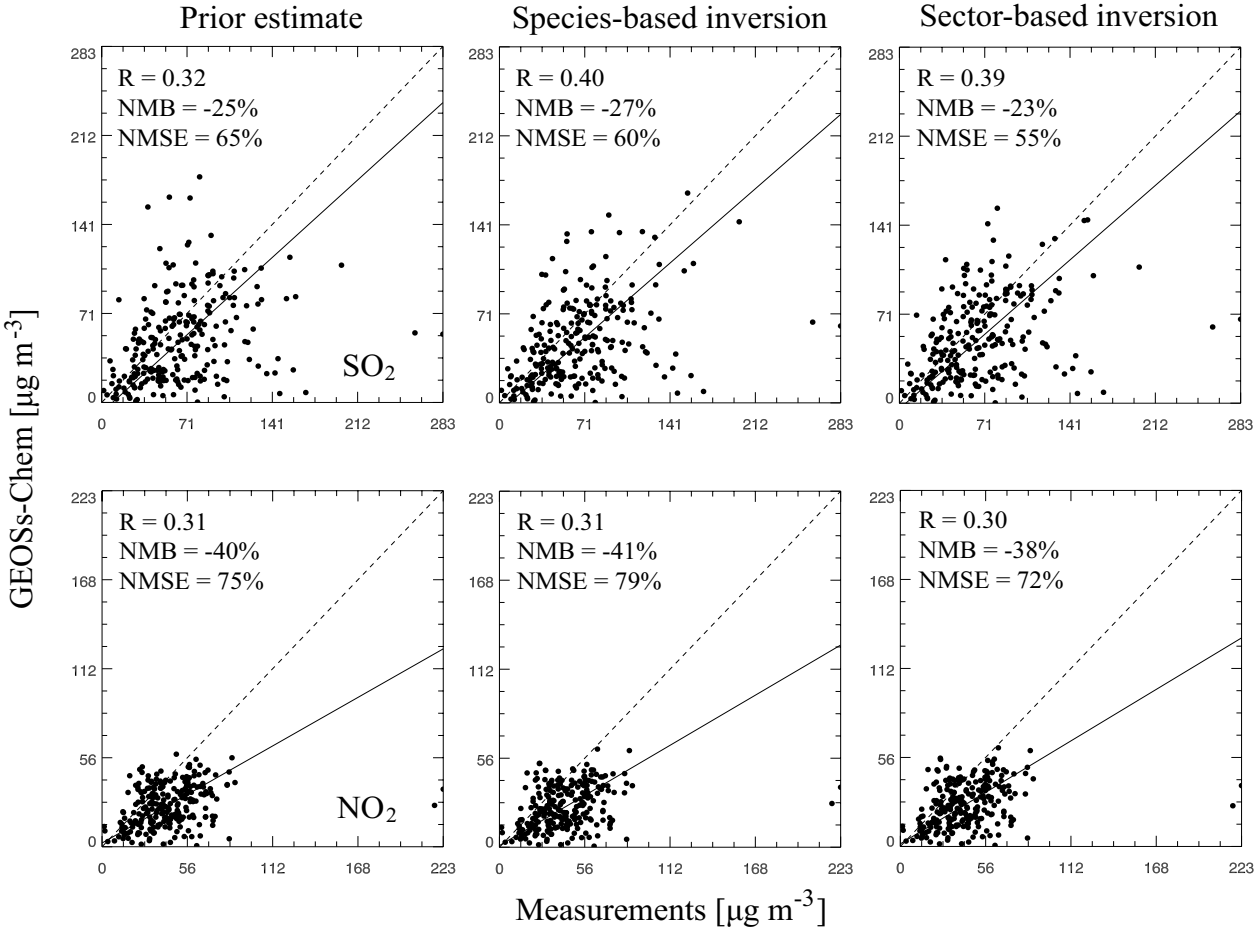
National Budget (Jan, 2010)



- NO_x: < HTAP emissions by 20-30%
- SO_2: HTAP emissions are overestimated in India
- CO: >HTAP estimates by 43-62% in China and 25-38% in India

(*Qu et al.*, 2022)

Sector-based Posterior Show the Best Agreement with Measurements

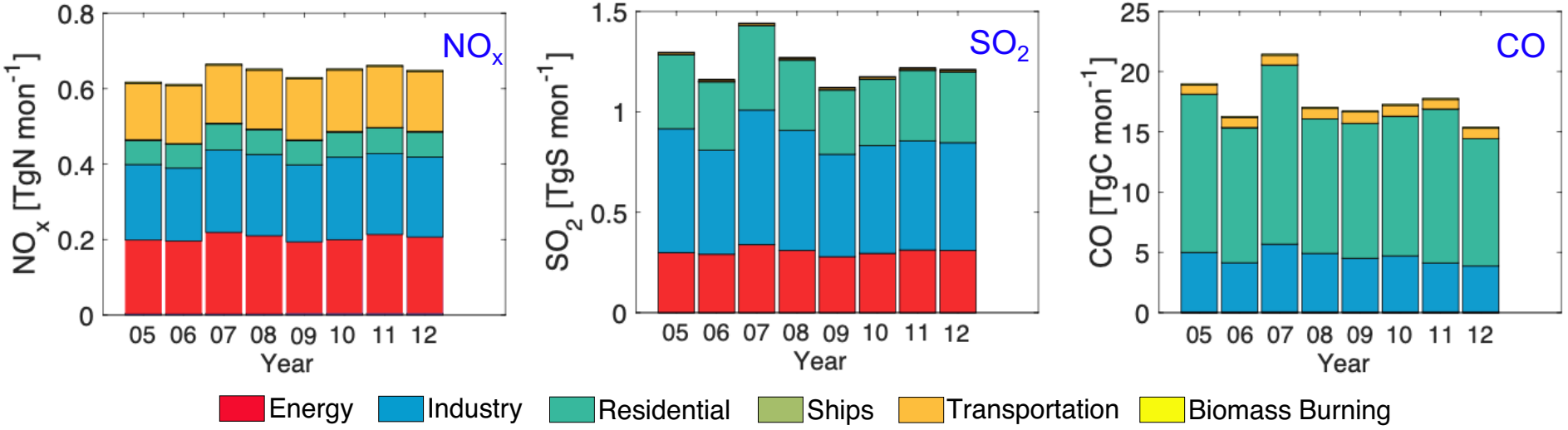


(*Qu et al.*, 2022)

How Different Sources Respond to Regulations in China?

Sectoral Contribution

Top-down emissions in China (Jan, 2005-2012)



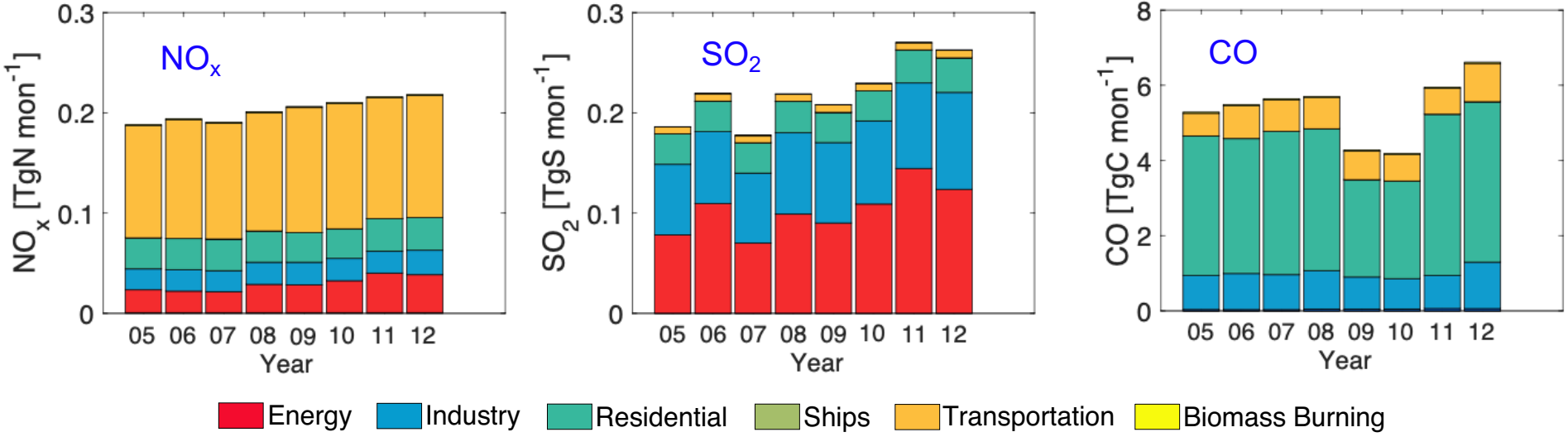
- Industry and energy sectors drive NO_x & SO_2 trends
- Residential and industry sectors drive CO trends

(*Qu et al.*, 2022)

Emissions Continuously Increase in India

Sectoral Contribution

Top-down emissions in India (Jan, 2005-2012)



- Energy sector drives NO_x & SO_2 trends
- Residential sector drives CO trends

(*Qu et al.*, 2022)

Observing System Simulation Experiments (OSSEs) over CONUS

Mimic the real atmosphere using CMAQ

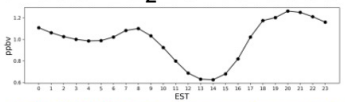
WRFv4.5

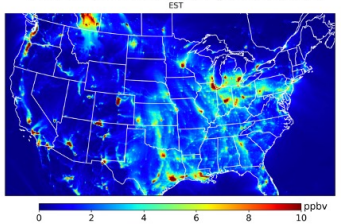
EQUATES Emissions

CMAQv5.4

Hourly, CONUS, 12 km

Surface NO_2 concentration





Mobile
- On-road Vehicles
- Non-road Equipment
- C1/C2 Commercial Marine Vessels
- C3 Commercial MarineVessels
- Rail

Point
- EGU
- Oil & Gas
- Non IPM (not EGU or oil & gas)
- Agricultural Field Burning
- Wild/Prescribed Fire
- Airports

Nonpoint
- Agriculture
- Area Fugitive Dust
- Oil & Gas
- Residential Wood Combustion
- Other

Inline
- biomass burning
- lightning

https://www.epa.gov/cmaq/equates

Bottom-up emissions based on NEI

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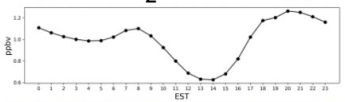

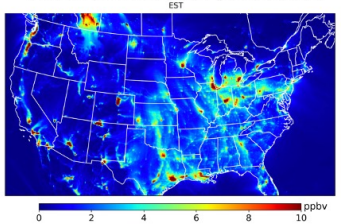

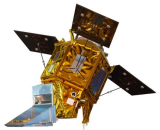

Column densities (NO_2, SO_2, CO, …)

Averaging kernel & sampling pattern from LEO & GEO satellite

Mobile
- On-road Vehicles
- Non-road Equipment
- C1/C2 Commercial Marine Vessels
- C3 Commercial MarineVessels
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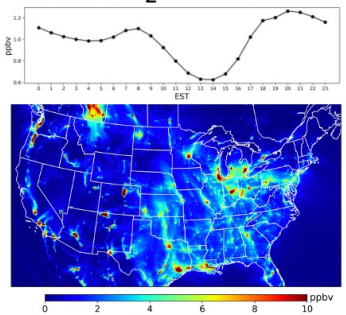

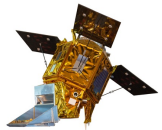

Column densities (NO_2, SO_2, CO, …)

Averaging kernel & sampling pattern from LEO & GEO satellite

Sectoral Inversion

Hourly, CONUS 0.25° × 0.3125°

- Energy
- Industry
- Residential
- Ships
- Transportation
- Aviation
- Biomass Burning

Posterior Emissions

Mobile
- On-road Vehicles
- Non-road Equipment
- C1/C2 Commercial Marine Vessels
- C3 Commercial MarineVessels
- Rail

Point
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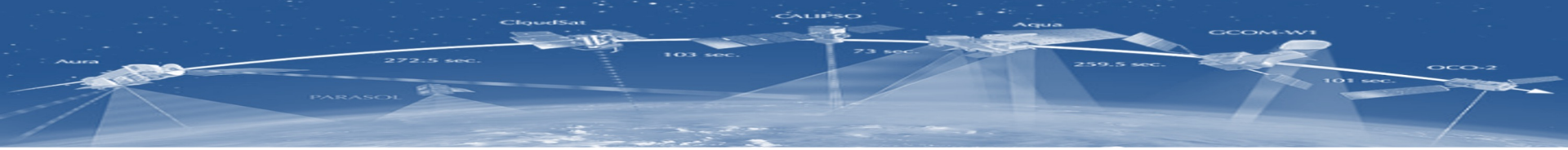

Summary

Top-down emissions from a newly developed sector-based inversion framework lead to the best agreement with independent surface measurements and provide a new perspective to evaluate bottom-up estimates by activities.

- This new inversion attributes the drivers of the peak of Chinese SO_2 (2007) and NO_x (2011) emissions to industry and energy activities, and CO (2007) to residential and industry emissions.
- In India, the inversion attributes NO_x and SO_2 trends mostly to energy and CO trend to residential emissions.
- OSSEs are designed to evaluate how much this new framework can improve sectoral emission estimates in the US.

NC STATE

We are recruiting Ph.D. students!

Marine, Earth and Atmospheric Sciences

North Carolina State University

zqu5@ncsu.edu