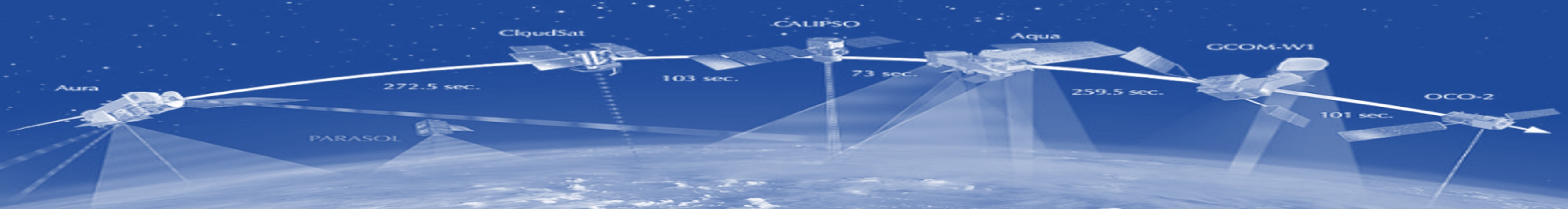

Applying analytical inversion and 4D-Var to estimate sources of methane and air pollutants

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BIRS workshop
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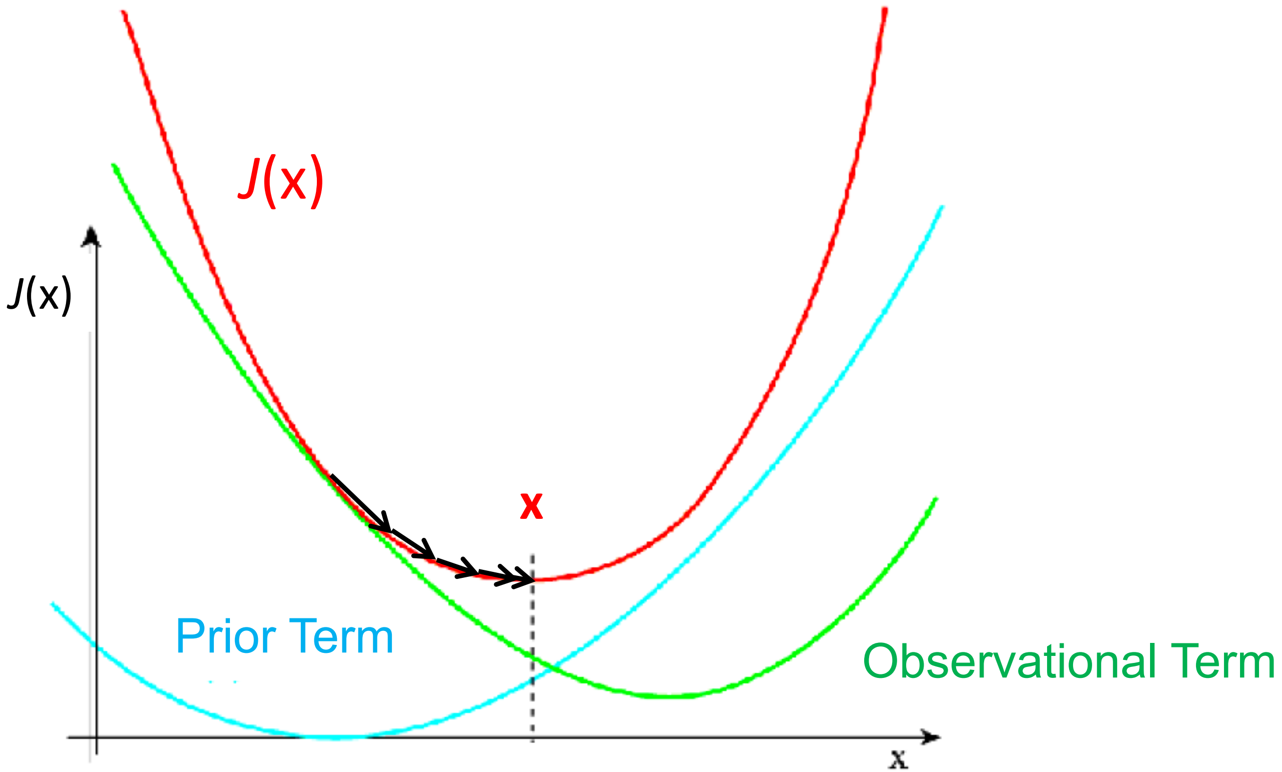
Bayesian inference of emissions

Minimize cost Function:

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



Solutions:

1. Analytical solution (linear model, $\mathcal{H} \rightarrow$ **K**):

$$\hat{\boldsymbol{x}} = \boldsymbol{x}_a + \left(\gamma \mathbf{K}^T \mathbf{S_o}^{-1} \mathbf{K} + \mathbf{S_a}^{-1}\right)^{-1} \gamma \mathbf{K}^T \mathbf{S_o}^{-1} (\boldsymbol{y} - \mathbf{K}\mathbf{x})$$

$$\hat{\mathbf{S}} = (\gamma \mathbf{K}^T \mathbf{S_o}^{-1} \mathbf{K} + \mathbf{S_a}^{-1})^{-1}$$

$$\mathbf{A} = \mathbf{I} - \hat{\mathbf{S}} \mathbf{S_a}^{-1}$$

2. 4D variational method (4D-Var):

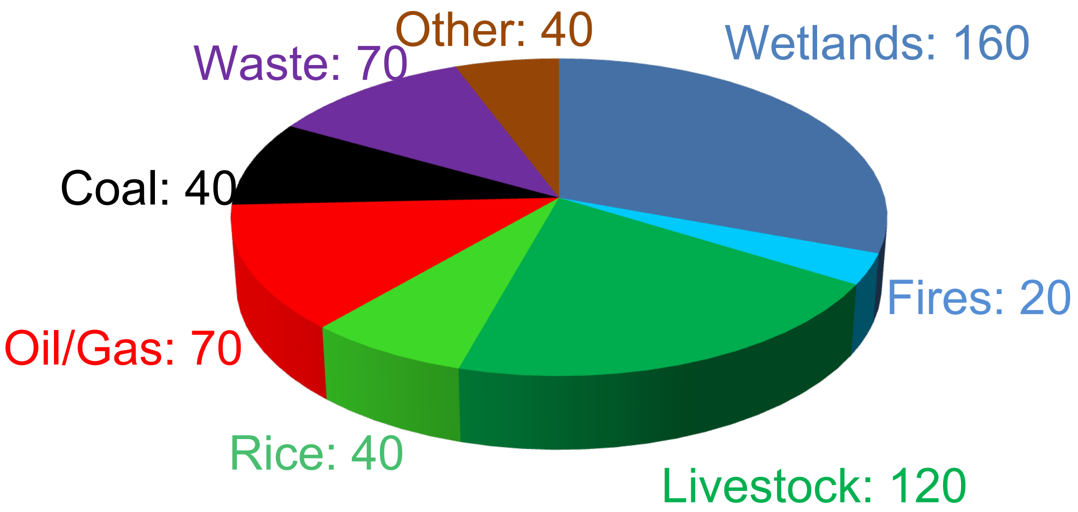
- Non-linear model – seeks a solution iteratively
- Adjoint model calculates the sensitivity of cost function with respect to state vector

1. Analytical Inversion of Methane

Global Methane Burden Is Balanced by the Sources and Sinks

Sources

Emission
560 ± 60 Tg a^{-1}



Sinks

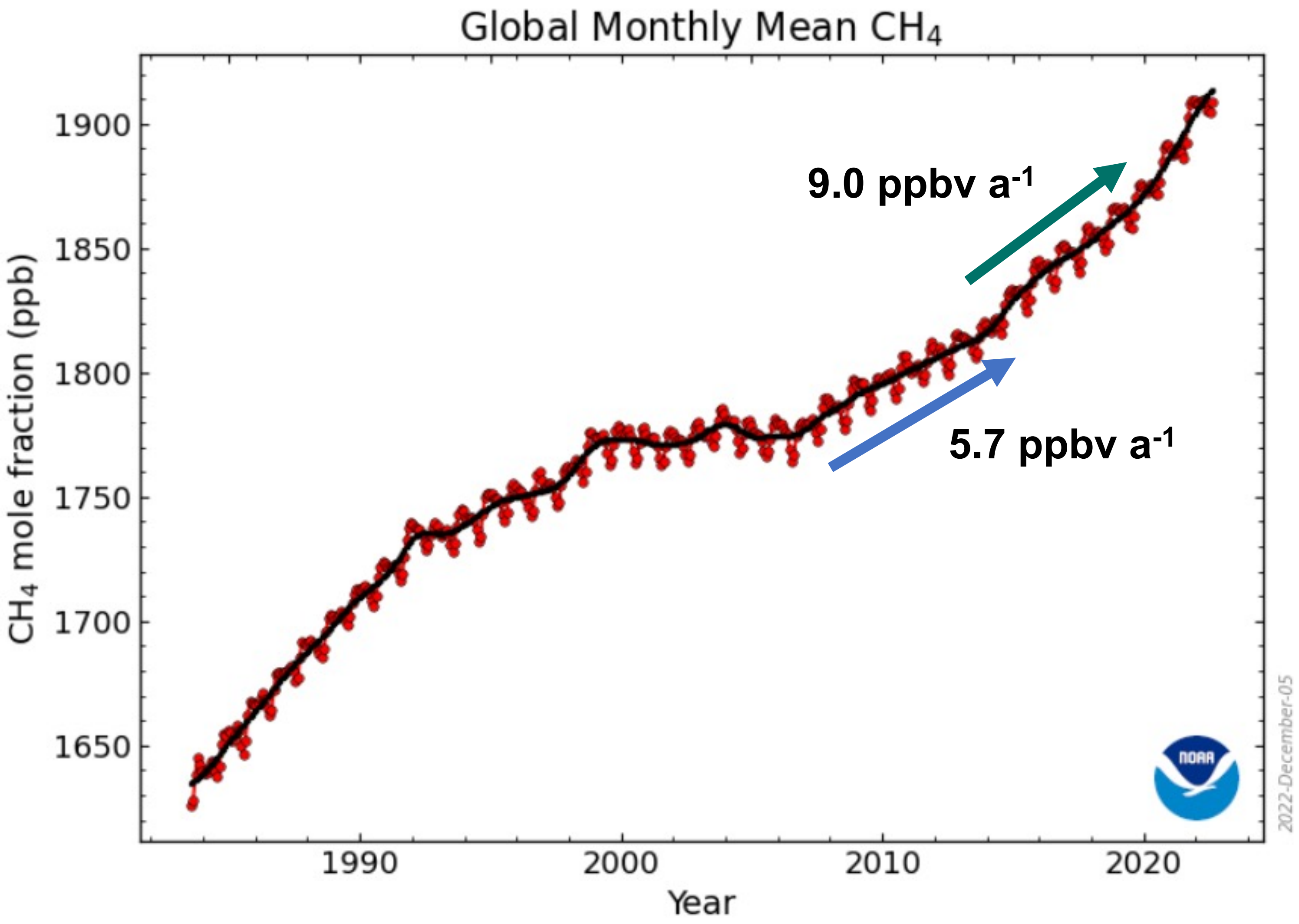
Mainly through reactions with OH:

$$CH_4 + OH \longrightarrow CH_3 + H_2O$$

From prescribed full-chemistry simulation

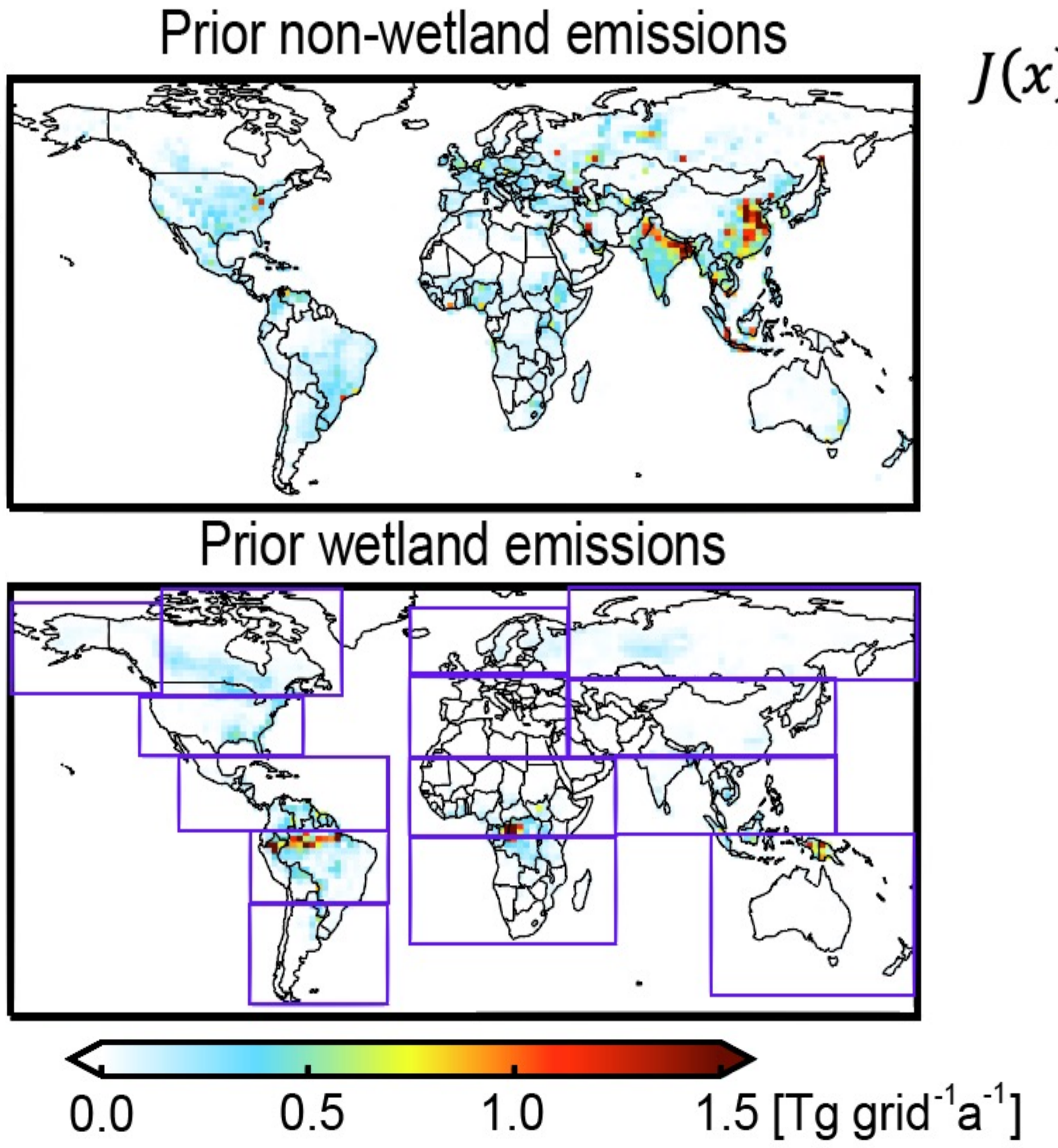
Sources – Sinks = Imbalance → Growth of CH_4 concentration

Global Methane Concentration Surge in 2020 & 2021



Record high annual increase of 14.7 ppbv in 2020 and 16.0 ppbv in 2021.

Analytical Inversion Simultaneously Estimates the Sources and Sinks of CH_4



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

- Prior estimates:
  EDGARv4.3.2 as global default;
  EPA greenhouse inventory for CONUS;
  oil, gas, and coal from GFEI;
  wetland from WetCHARTs
- x: 4020 non-wetland emission, 12 x 14 wetland emissions, 2 hemispheric OH concentrations
- K: sensitivity of each observation to 4190 state vector elements
- S_o include both satellite and model errors

Changes in CH_4 Sources and Sinks in 2019-2020

Methane growth rate: $$\frac{dm}{dt} = E - km - L$$

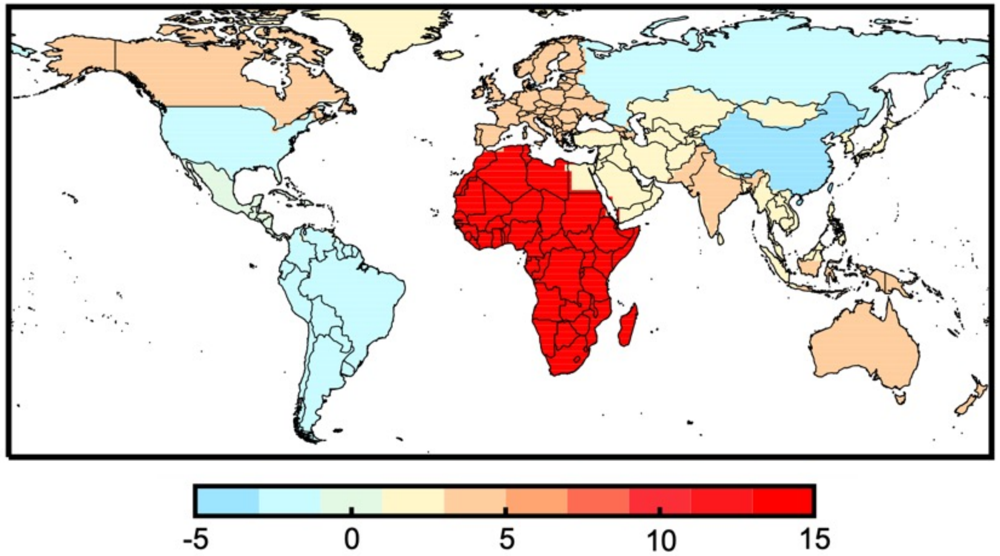
Acceleration of growth rate: $$\frac{d^2m}{dt^2} = \frac{dE}{dt} - k\frac{dm}{dt} - m\frac{dk}{dt} - \frac{dL}{dt}$$

m = methane mass
E = emission
k = OH loss rate constant
L = minor losses (strat, Cl. Dep)

Forcing away from steady state: $$F = \frac{dE}{dt} - m\frac{dk}{dt}$$ = 36 Tg a^{-1}

Sources + 31 Tg a^{-1} (86%)

2020-2019 change in emissions [Tg a^{-1}]



Sinks - 5 Tg a^{-1} (14%)

Global mean OH reduce by 1.2%

African wetlands drive 50% of the surge; independent evidence from inundation data

9-member inversion ensemble: + 30 ± 5.5 Tg a^{-1} (82% ± 18%)

(*Qu et al.*, 2022)

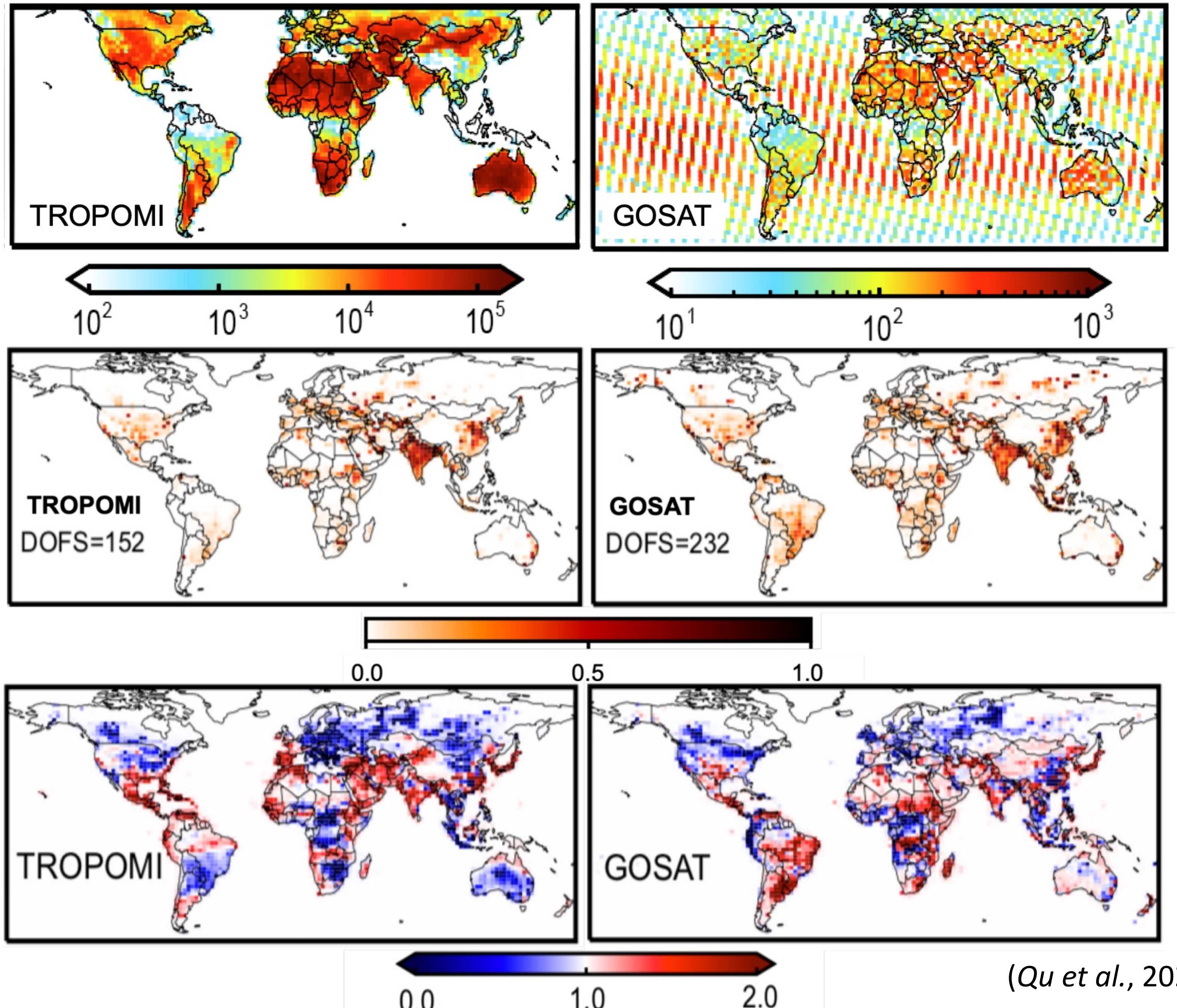
Analytical Inversion: Compare Observations from Different Instruments

of observations

Averaging kernel (A)

Degrees of freedom for signal (DOFS): trace(A)

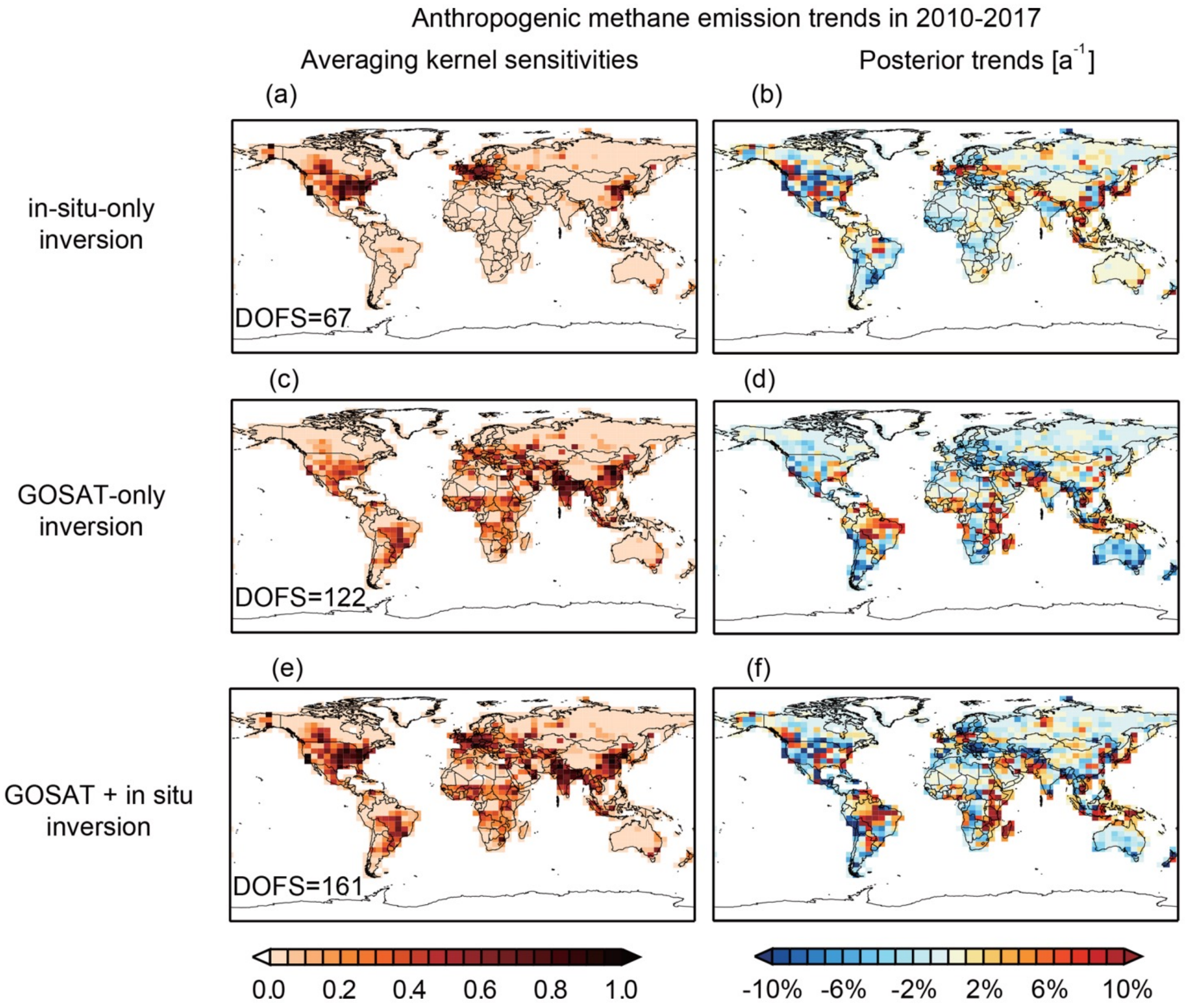
Posterior / prior



(*Qu et al.*, 2021)

Other Applications of CH_4 Analytical Inversions

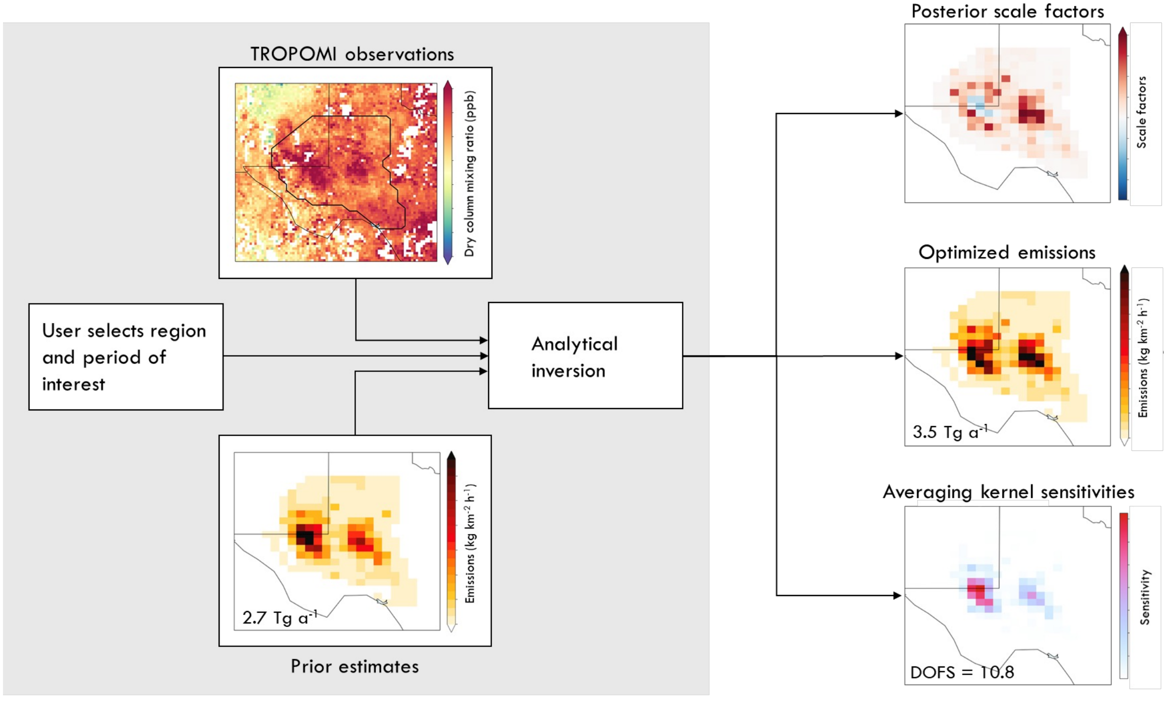
Complementarity of GOSAT and in-situ observations



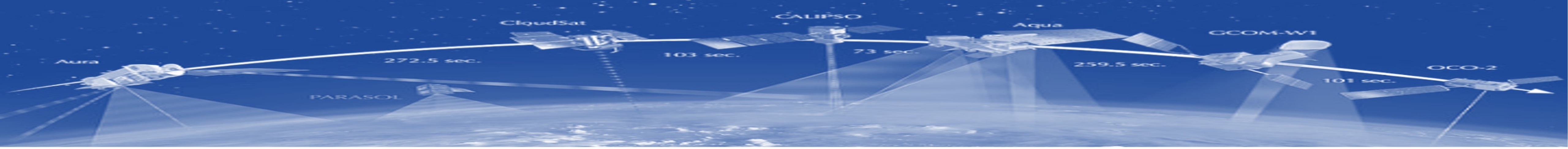
(*Lu et al.*, 2021)

Integrated Methane Inversion cloud-based facility

- 0.25° or 0.5° resolution
- TROPOMI data and GEOS-Chem on AWS



(*Varon et al.*, 2022)



Summary

- Analytical inversion: forward model is linear; can easily quantify information content and generate an ensemble of inversions

- The largest increases of methane emissions over 2010-2021 are from Africa and South America.

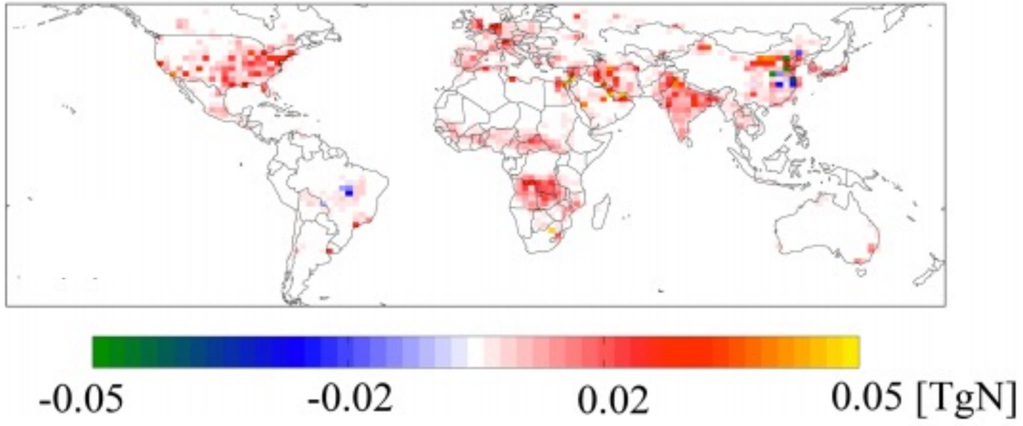
- Emissions from Africa, South America, and Equatorial Asia drives the methane surge in 2020 and 2021.

2. Quantifying NO_x, SO_2, and CO Emissions using 4D-Var

Previous Applications of 4D-Var

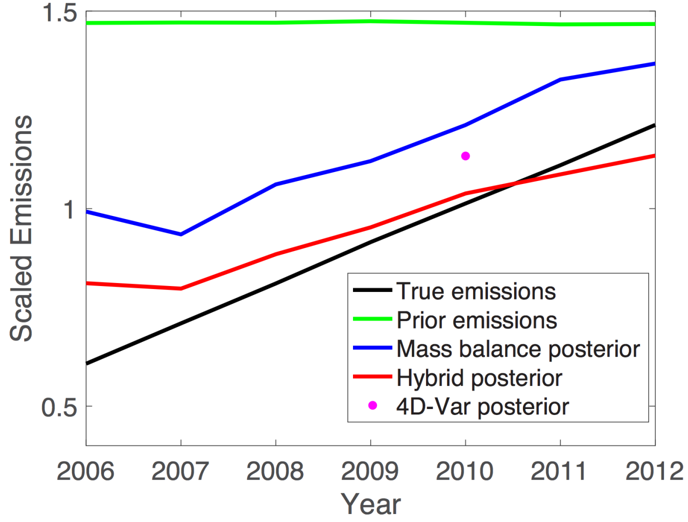
Improve spatial distribution:

NO$_x$ emissions (Top-down – HTAP, 2010)



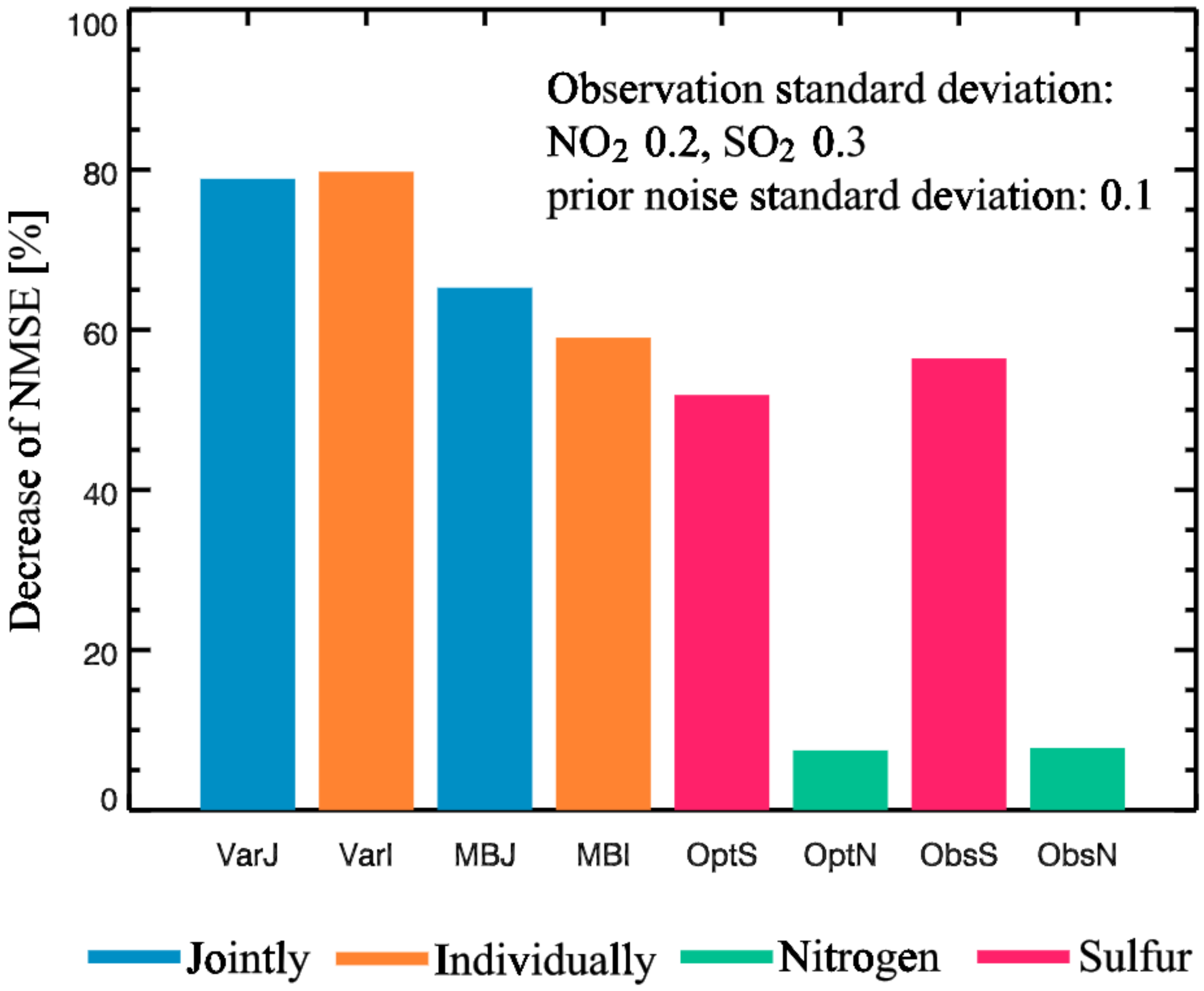
(*Qu et al.*, 2020)

Combined w/ mass balance for trend:



(*Qu et al.*, 2017)

Address the impact of chemical interactions:



(*Qu et al.*, 2019)

Limitations in Previous Top-down Estimates

Cannot…

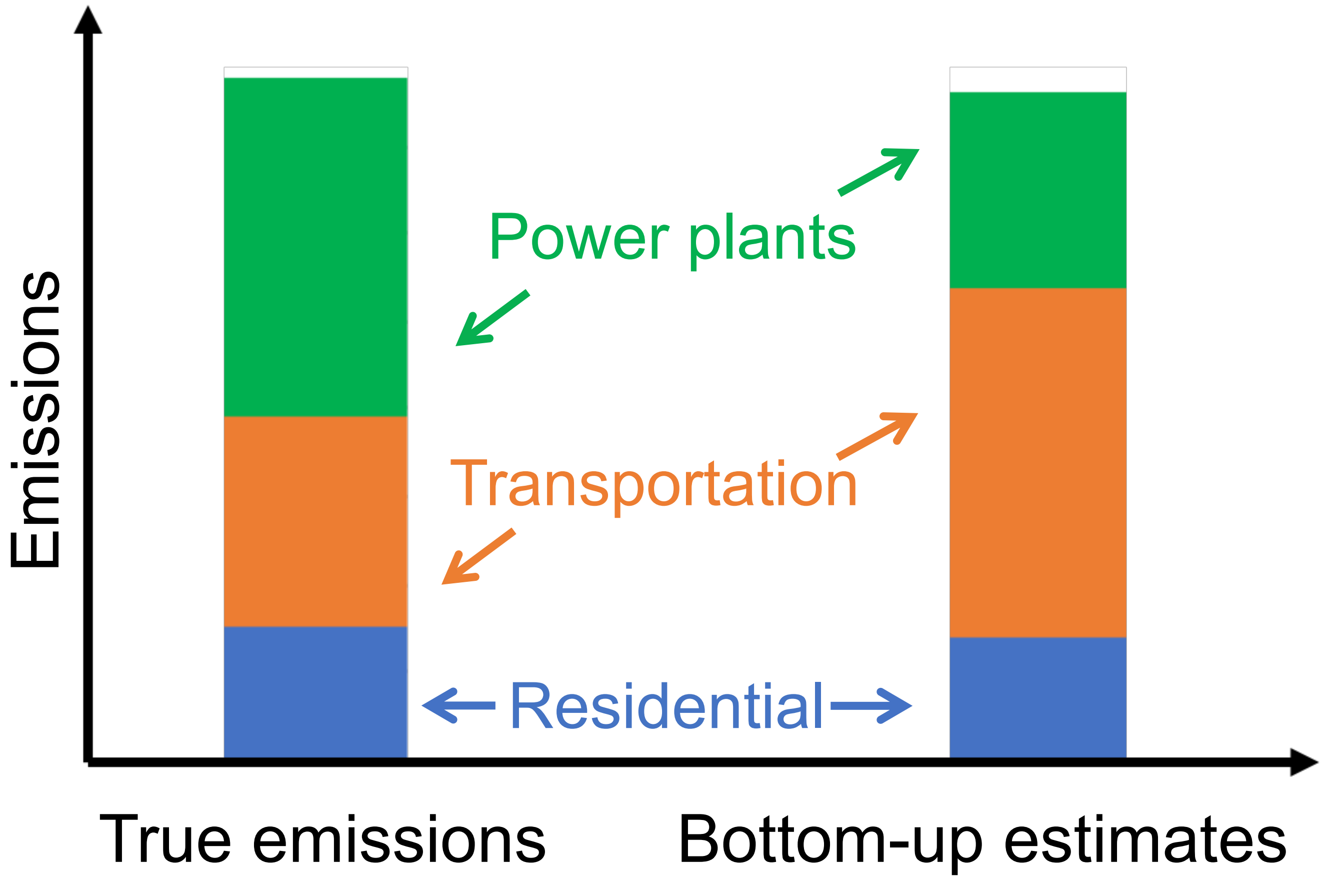
1. optimize sectoral profiles
2. separate errors from emission factor & activity rates

E = species emission factor × activity

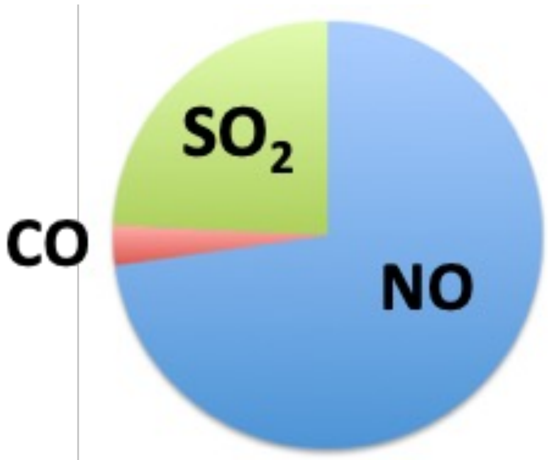
optimize

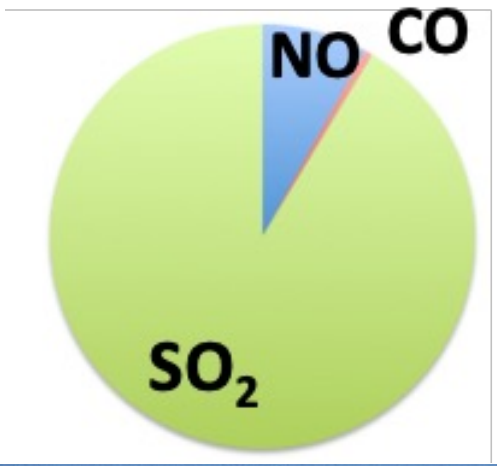
New sector-based inversion

Similar total emissions, different profiles



Unique Emission Profile for Each Source





Formulation of a sector-based inversion

Species-based inversion:

$$E_l = \sigma_l \sum_{k=1}^{7} A_{a,k} F_{a,k,l} = \sigma_l E_{a,l}$$

Emission Scaling Factor Activity Emission Factor

l : species
k: sector

$$J_1(\sigma_{k,l}) = J_o + \frac{1}{2}\gamma_{r1} \sum_{l=1}^{3} (\boldsymbol{\sigma}_l - \boldsymbol{\sigma}_{a,l})^{\mathrm{T}} \mathbf{S}_{a,l}^{-1} (\boldsymbol{\sigma}_l - \boldsymbol{\sigma}_{a,l})$$

Sector-based inversion:

$$E_{k,l} = \sigma_k A_{a,k} \sigma_{m,l} F_{a,m,l}$$

Scaling Factor for Activity Scaling Factor for Emission Factor

$$J_2(\boldsymbol{\sigma}_k, \boldsymbol{\sigma}_{m,l}) = J_o + \frac{1}{2}\gamma_{r2} \sum_{k=1}^{7} (\boldsymbol{\sigma}_k - \boldsymbol{\sigma}_{a,k})^{\mathrm{T}} \mathbf{S}_{a,k}^{-1} (\boldsymbol{\sigma}_k - \boldsymbol{\sigma}_{a,k})$$

activity rates in sector k

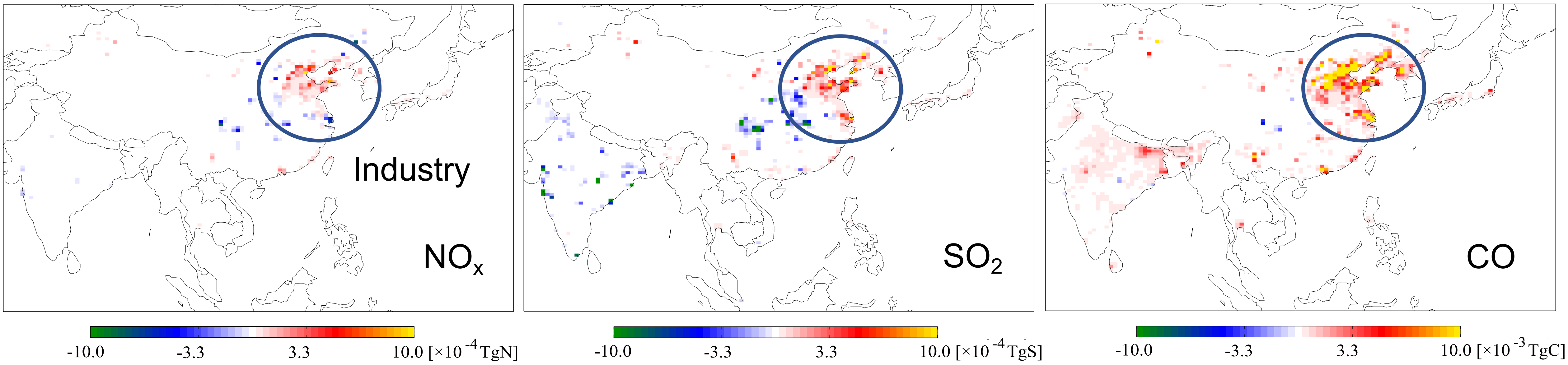
emission factor for species l in sector m

$$+\frac{1}{2}\gamma_{r2} \sum_{m=1}^{3} \sum_{l=1}^{3} (\boldsymbol{\sigma}_{m,l} - \boldsymbol{\sigma}_{a,m,l})^{\mathrm{T}} \mathbf{S}_{a,m,l}^{-1} (\boldsymbol{\sigma}_{m,l} - \boldsymbol{\sigma}_{a,m,l}).$$

(*Qu et al.*, 2022)

Sector-based Inversion: Independent Adjustments for Each Source

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



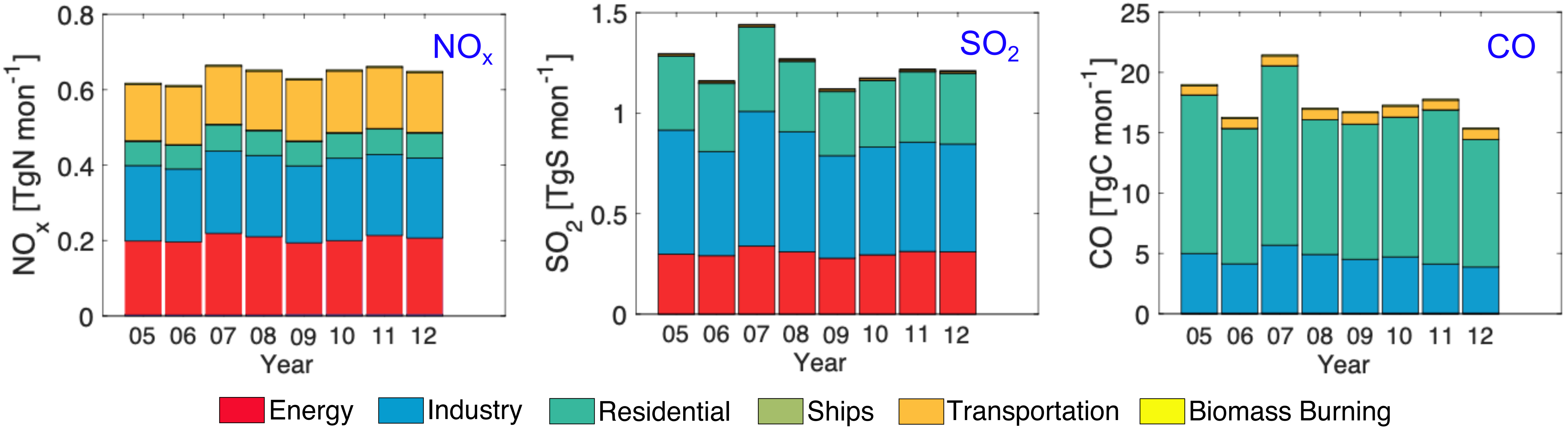
(*Qu et al.*, 2022)

Bottom-up emissions: overestimate underestimate

E = species emission factor × activity

How Different Sources Respond to Regulations in China?

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

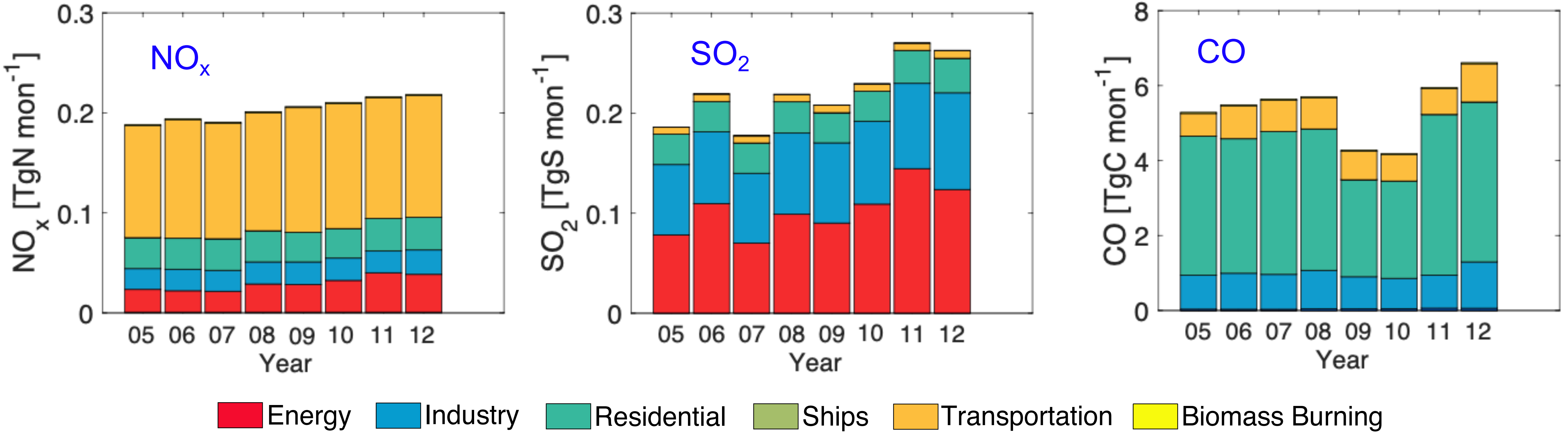


(*Qu et al.*, 2022)

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

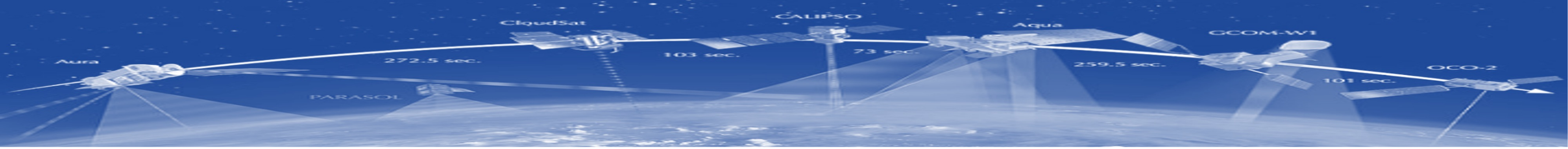
Emissions Continuously Increase in India

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)



Summary

- 4D-Var: good for non-linear problems, computational expensive, but can be combined with other methods (e.g., mass balance) to reduce the computational cost.

- 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.