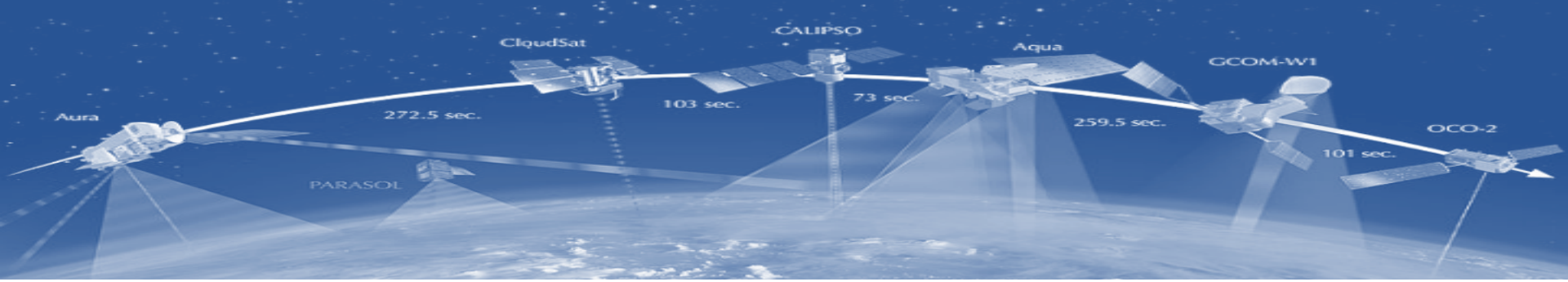

4D-Var and hybrid inversion of NO_x emissions & the impact of satellite retrievals on emission estimates

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4D-Var method

Description	Variable	
Model simulation	c	
Observation operator	$\mathcal{H}$	Maps c to obs space
Satellite observation	SCD_{obs}	vector
Prior emission scaling factor	σ_a	= 1
Emission scaling factor	σ	= Emis / Emis_prior
Observational error covariance matrix	S_{obs}	diagonal
Prior error covariance matrix	S_a	

$$J(\boldsymbol{\sigma}) = \frac{1}{2}\Sigma_{\mathbf{c}\in\Omega}(\mathcal{H}\mathbf{c} - \mathbf{SCD}_{obs})^T\mathbf{S}_{\mathbf{obs}}^{-\mathbf{1}}(\mathcal{H}\mathbf{c} - \mathbf{SCD}_{obs}) + \frac{1}{2}\gamma_r(\boldsymbol{\sigma} - \boldsymbol{\sigma_a})^T\mathbf{S}_{\mathbf{a}}^{-\mathbf{1}}(\boldsymbol{\sigma} - \boldsymbol{\sigma_a})$$

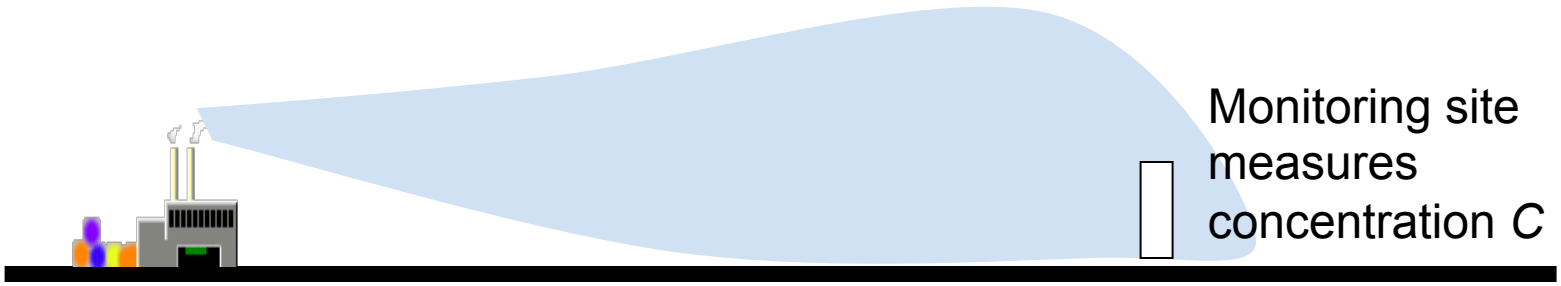
- 4D-Var:
  - Summed over time (4D)
  - Seeks σ iteratively (Var)
  - Requires adjoint model calculates the sensitivity of cost function w.r.t. scaling factor σ

(*Sandu et al.*, 2005)

4D-Var method

Inverse Modeling

Atmospheric "forward" model gives $C = kE$

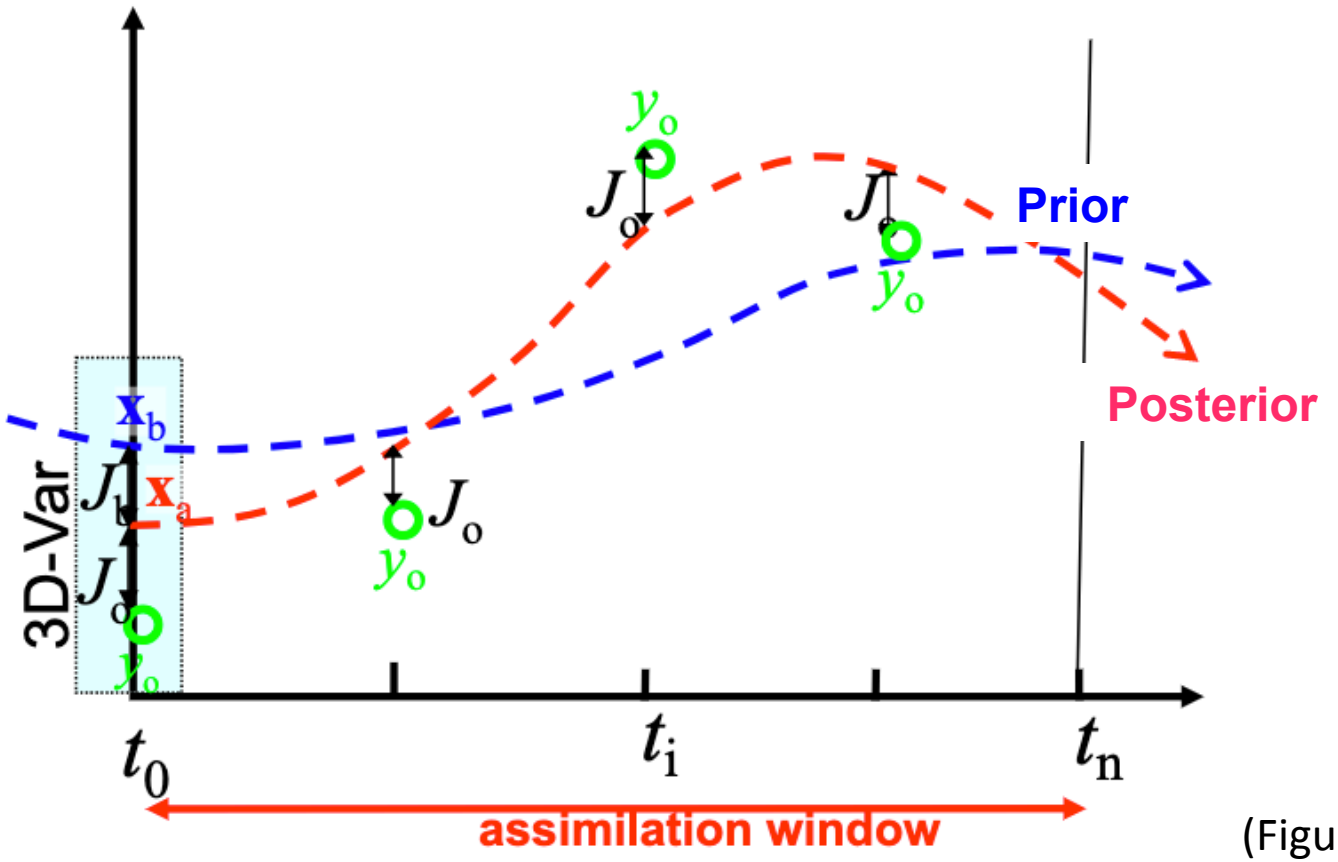


(fuel burned) X (emission factor)
⇨ *a priori* "bottom-up" estimate
$E_a \pm \sigma_a$

Inverse model $E = k^{-1}C$
⇨ "top-down" estimate
$E_\varepsilon \pm \sigma_\varepsilon$

- Forward model: CMAQ, WRF-Chem, GEOS-Chem
- Adjoint code is required for each process in the forward model

4D-Var v.s. 3D-Var



(Figure from ECMWF)

- 3D-Var finds the best estimate using an observation and a state at **one specific time point**.
- 4D-Var adjusts the initial state (NO_x emissions) to optimize the model trajectory to better match the observations distributed **over the assimilation window**, needs adjoint.

4D-Var v.s. Mass balance

Shortcoming of 4D-Var:

- Time consuming (search minimum iteratively)
- 17 hours/ iteration, more than 20 iterations
- ~ 2 - 3 weeks to finish monthly inversion (1 node, 24 threads)

Mass balance:

- Scale prior emission E_a by the ratio of observed and simulated column density

$$E_t(i,j) = E_a(i,j)\frac{SCD_{obs}(i,j)}{SCD_{GC}(i,j)}.$$

(*Martin et. al*, 2003)

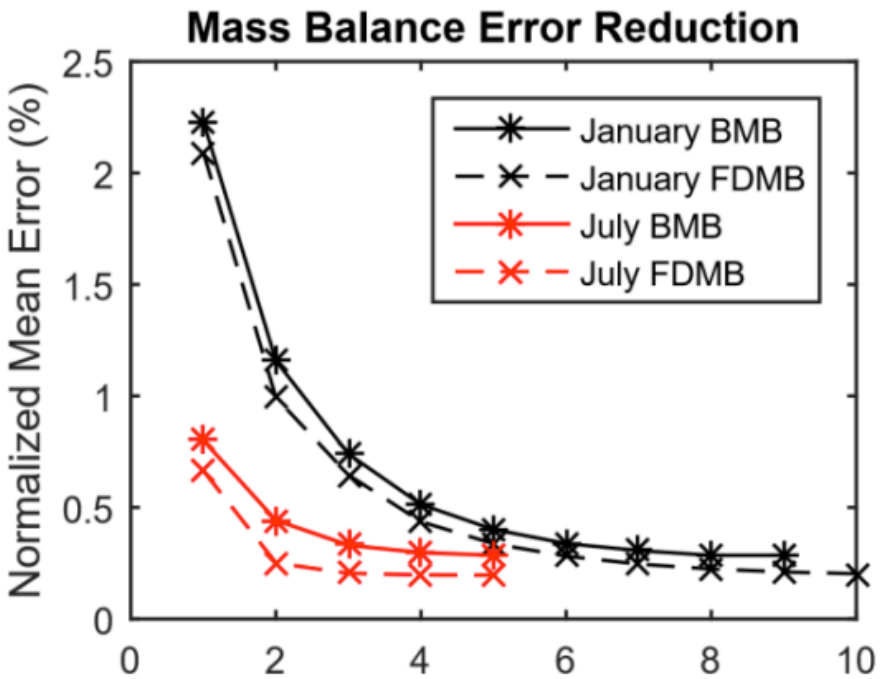
- Computational cost: ~ 8 hours for monthly regional estimates.

Hybrid 4D-Var / Mass balance

Computational cost of 4D-Var is especially challenging when applying to TROPOMI and geostationary satellite data

- 4D-Var (base year) + mass balance (trend) (*Qu et. al*, 2017)

- Iterative mass balance



(*Cooper et. al*, 2017; *Li* et al., 2019)

- Iterative mass balance (global) + 4D-Var (regional) (*Choi et. al*, in prep)

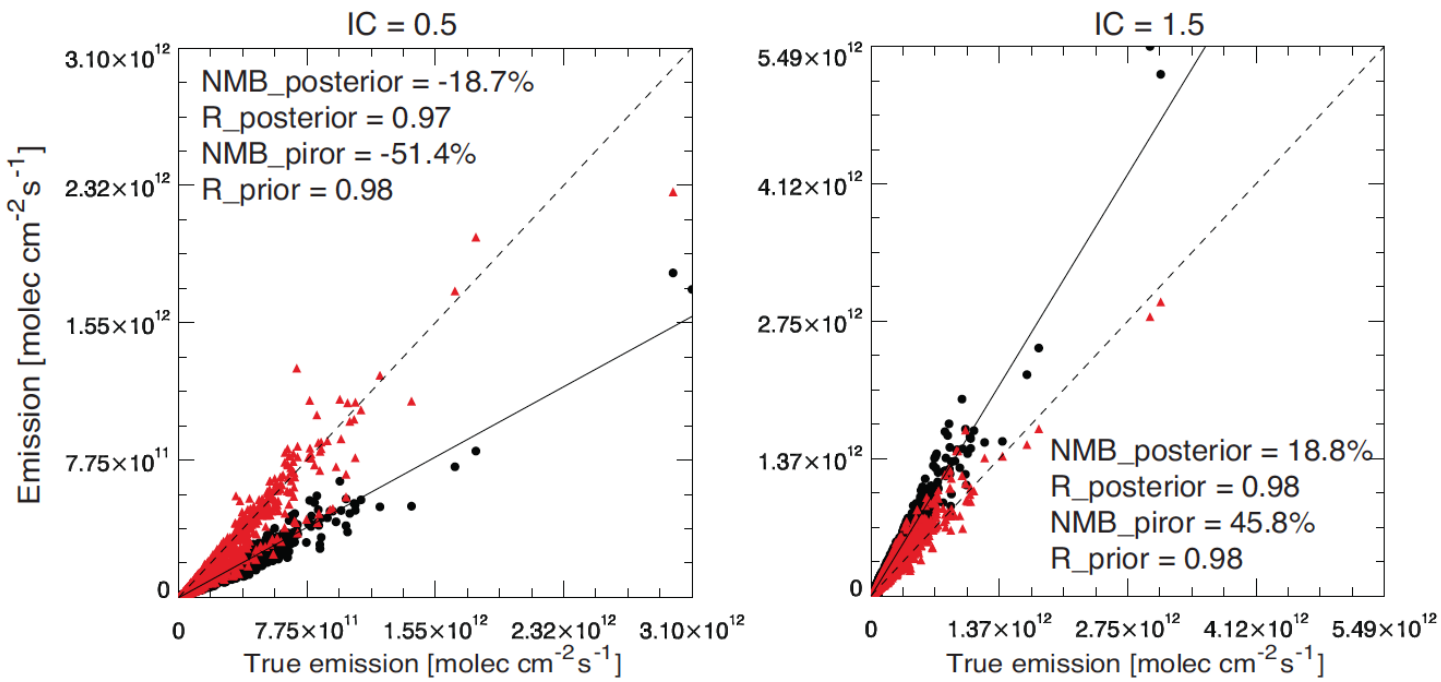
Evaluation: pseudo observation tests (simplified OSSE)

- Design inverse problem with known solution (true emission) to evaluate the performance of the new inversion framework

- Pseudo observations: NO_2 column densities from model simulation at satellite overpass time and locations using “true emissions” (random noise applied to observations)

Pseudo observation tests: 4D-Var

- 135915 pseudo observations generated for Jan, 2010 (random noises applied in observations)
- Initial guesses : 0.5 x true emission for IC = 0.5; 1.5 x for IC = 1.5; normally distributed random noises applied in emissions



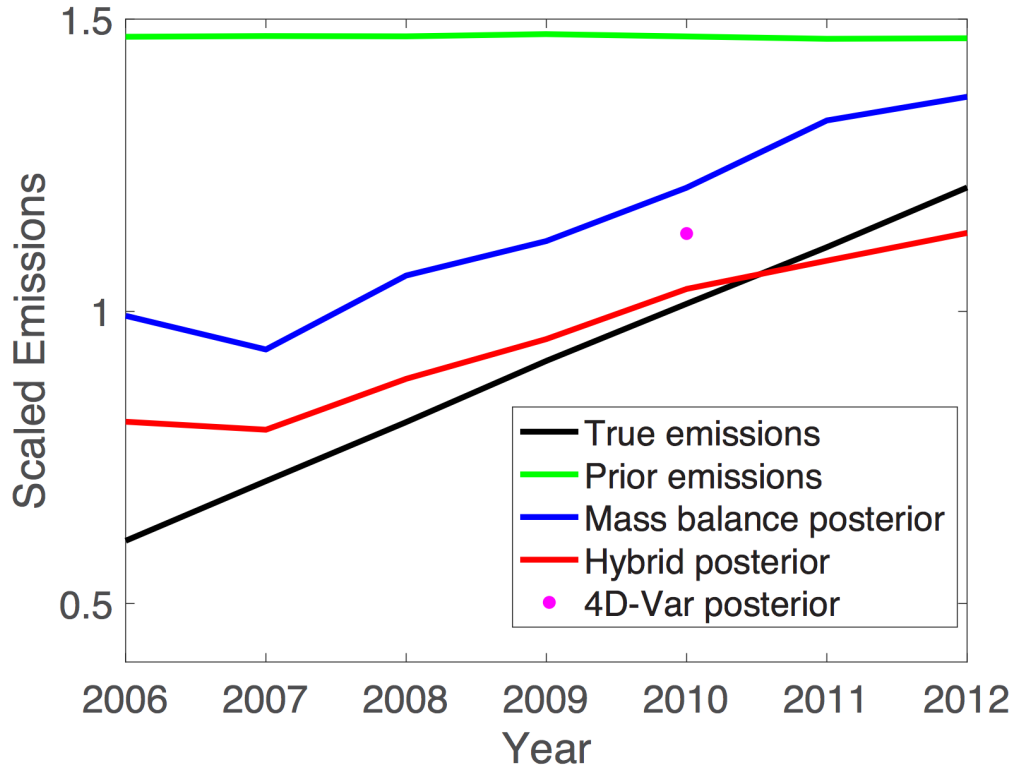
- Smaller NMSE (by 61%, IC=0.5 and 41%, IC=1.5) in posterior emissions

(*Qu et al.*, 2017)

Pseudo observation tests: Hybrid 4D-Var + mass balance

Base year (2010): 4D-Var
Other years (2005-2012): use 2010 4D-Var posterior for mass balance

Scaled emissions in pseudo observation test



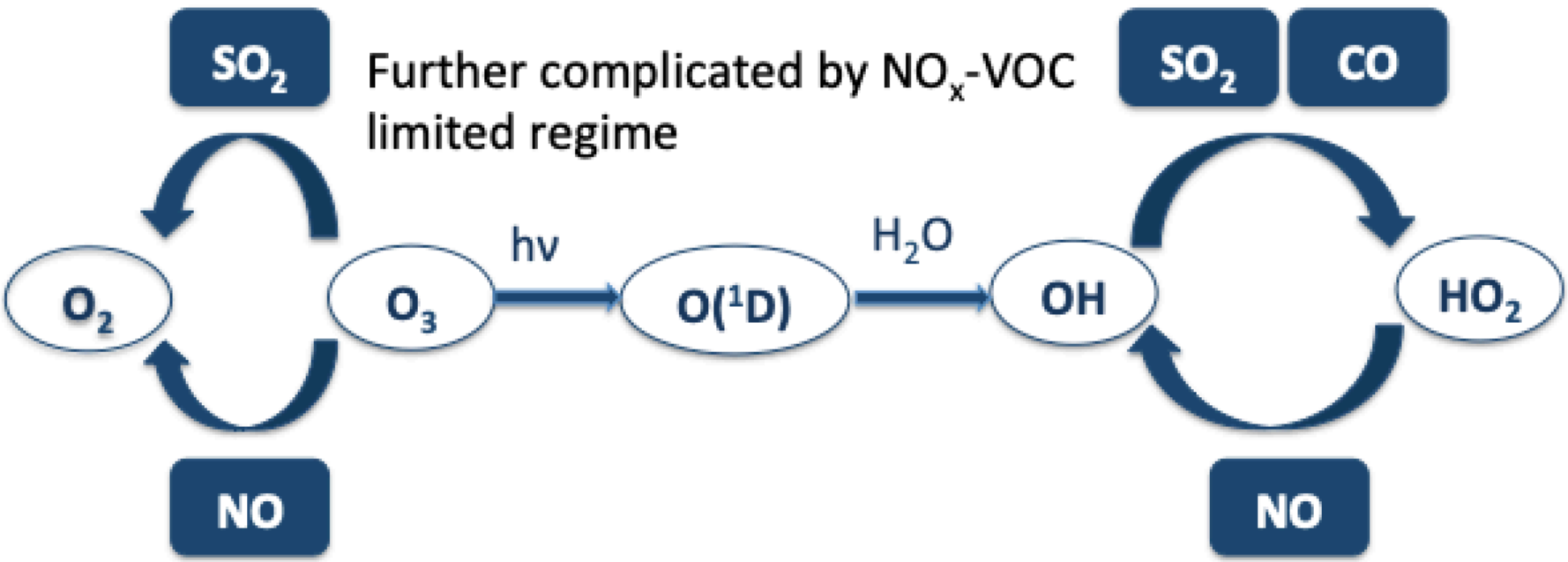
- Hybrid posterior has smaller NMSE (by 59% to 78%) and better correlation compared to mass balance.

(*Qu et. al*, 2017)

Top-down emission estimates

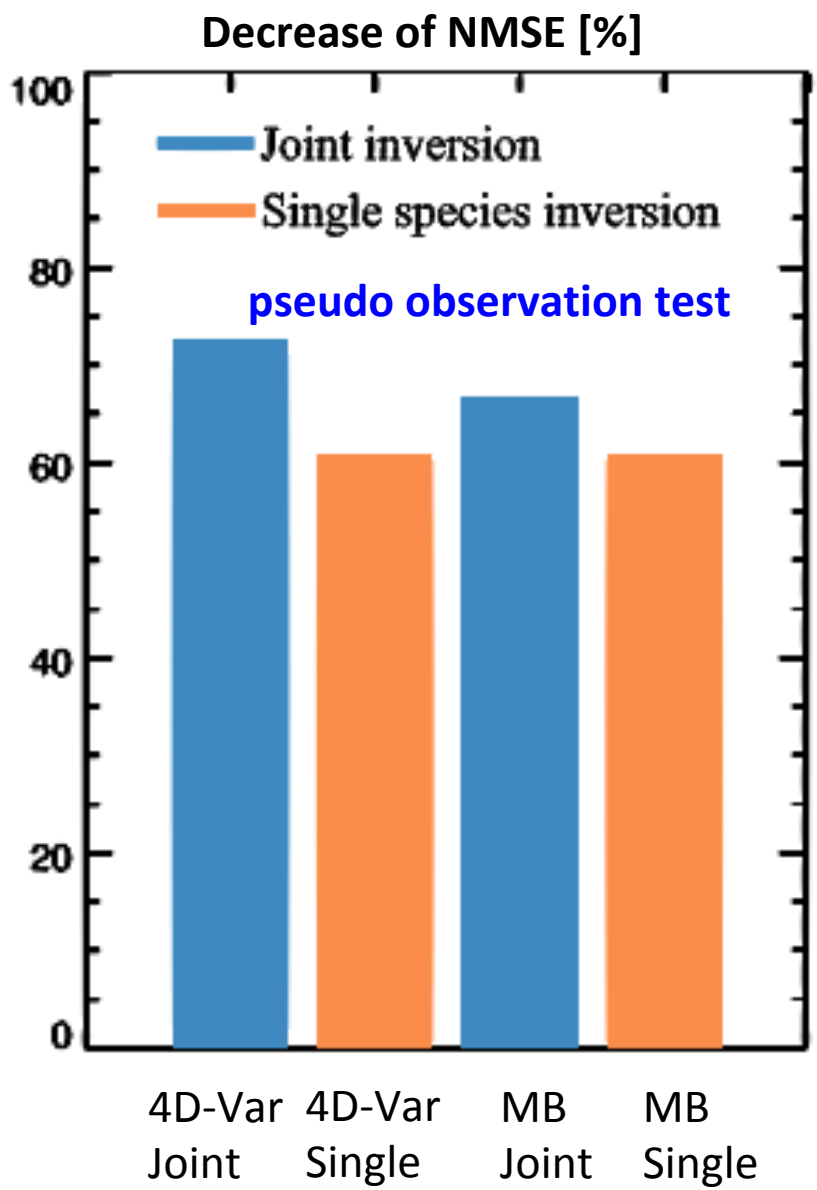
Still …

- The impacts of adjusting emission of one species on changes in concentrations of other species are not being considered so far
- Errors in other species emissions are likely biasing the top-down emission of the constrained species



Joint NO_x & SO_2 inversion

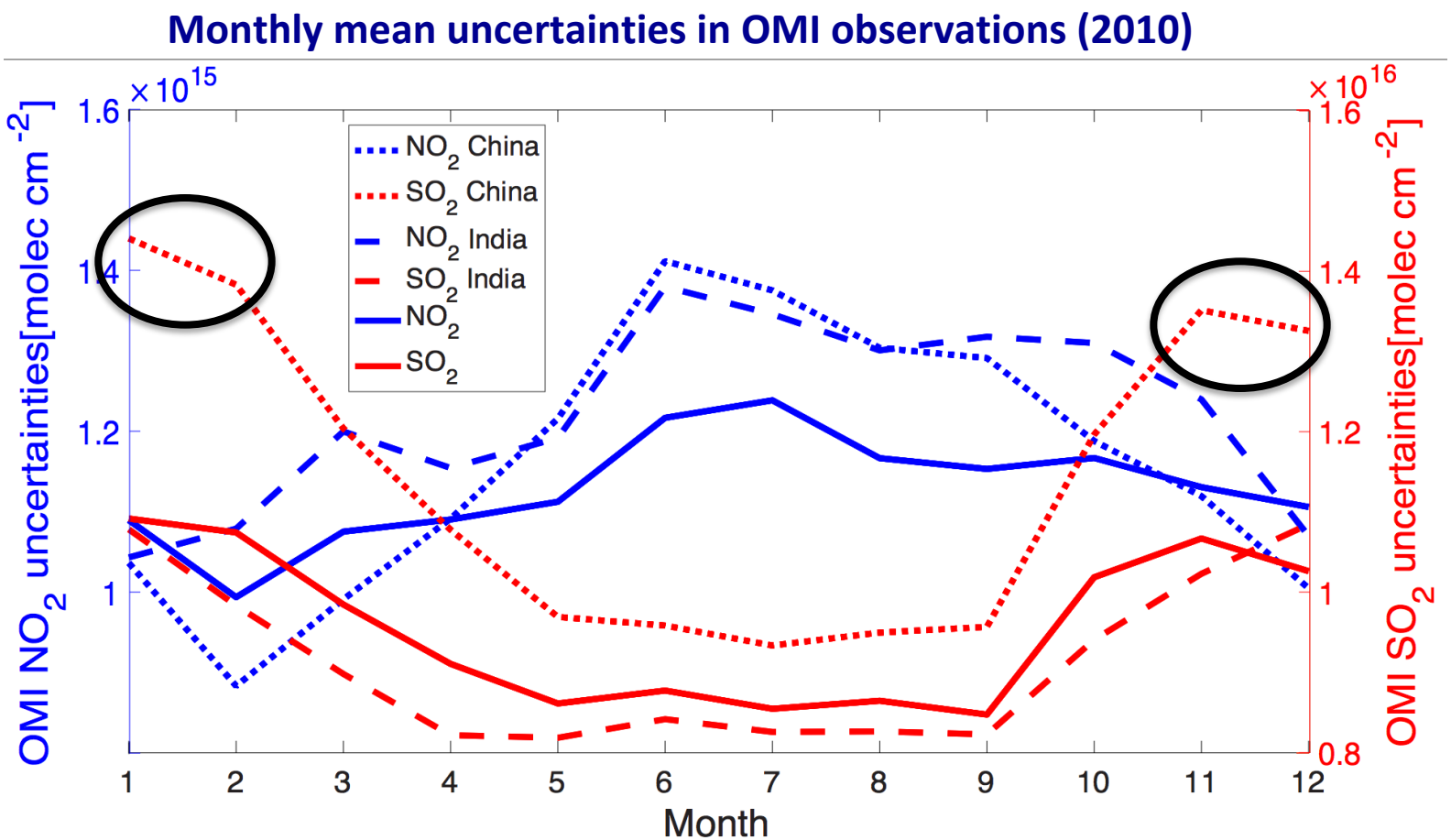
- Use both NO_2 & SO_2 observations to optimize NO_x and SO_2 emissions



- Joint inversion decreases NMSE in pseudo observation test

(*Qu et al.*, 2019)

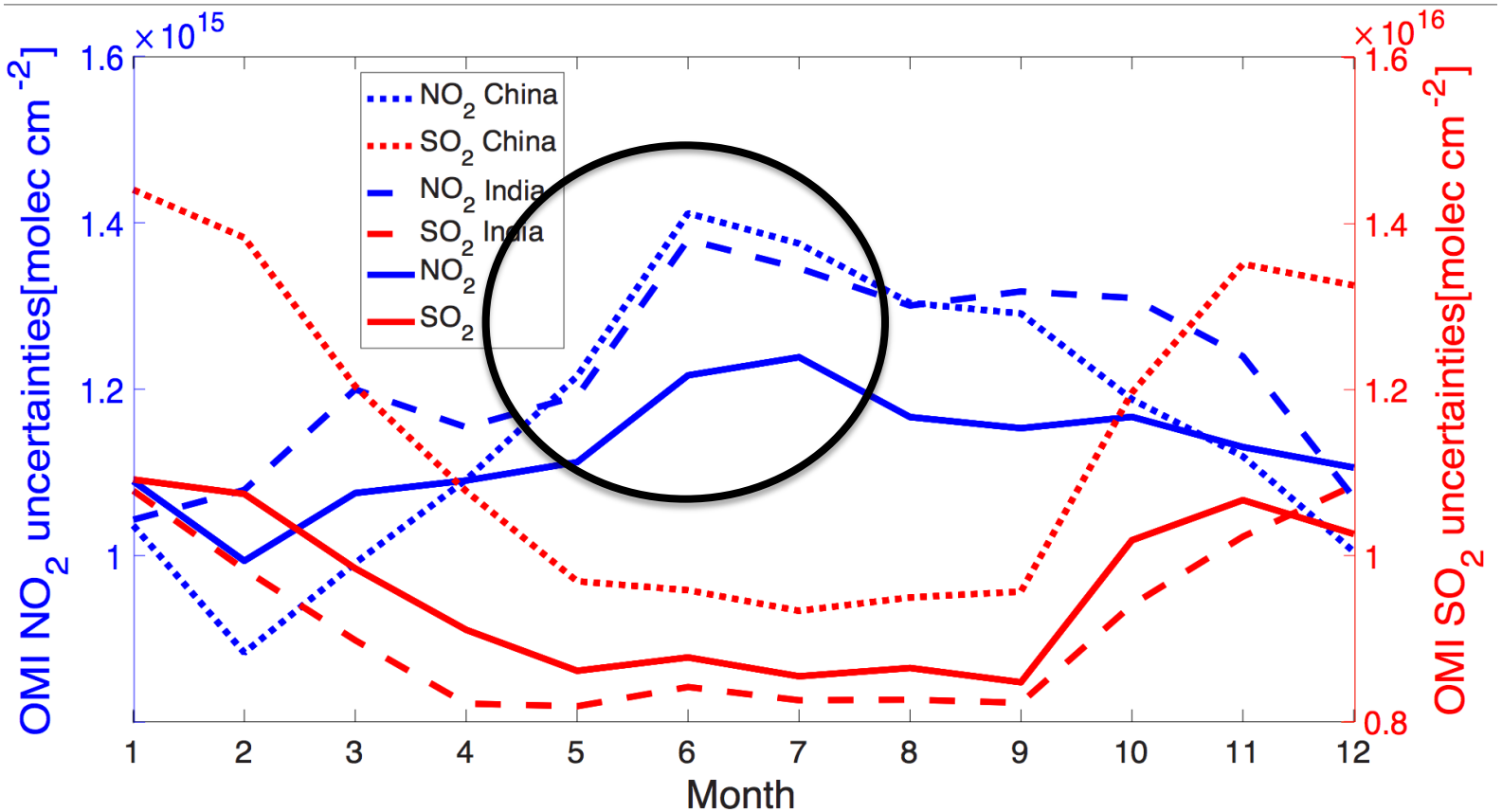
Impact of observation uncertainties



- In China, NMB and NMSE of SO_2 joint posterior reduce by up to 26% and 18% compared to single species posterior in most months between November and February;

Impact of observation uncertainties

Monthly mean uncertainties in OMI observations (2010)



- In China, NMB and NMSE of SO_2 joint posterior reduce by up to 26% and 18% compared to single species posterior in most months between November and February;
- For NO_x, joint posterior has reduced NMSE from May to October and reduced absolute value of NMB from May to July

Sector-based inversion

Bottom-up emission inventory:

E = species emission factor × activity

New sector-based inversion:

E_NO_x_pos = SF_industry x E_NO_x_industry + SF_energy x E_NO_x_energy + …

E_SO_2_pos = SF_industry x E_SO_2_industry + SF_energy x E_SO_2_energy + …

E_CO_pos = SF_industry x E_CO_industry + SF_energy x E_CO_energy + …

Sector-based inversion

Bottom-up emission inventory:

E = species emission factor × activity

New sector-based inversion:

$E\_NO_x\_pos = SF\_industry \times E\_NO_x\_industry + SF\_energy \times E\_NO_x\_energy + \ldots$

$E\_SO_2\_pos = SF\_industry \times E\_SO_2\_industry + SF\_energy \times E\_SO_2\_energy + \ldots$

$E\_CO\_pos = SF\_industry \times E\_CO\_industry + SF\_energy \times E\_CO\_energy + \ldots$

Adjust sector and species emissions simultaneously

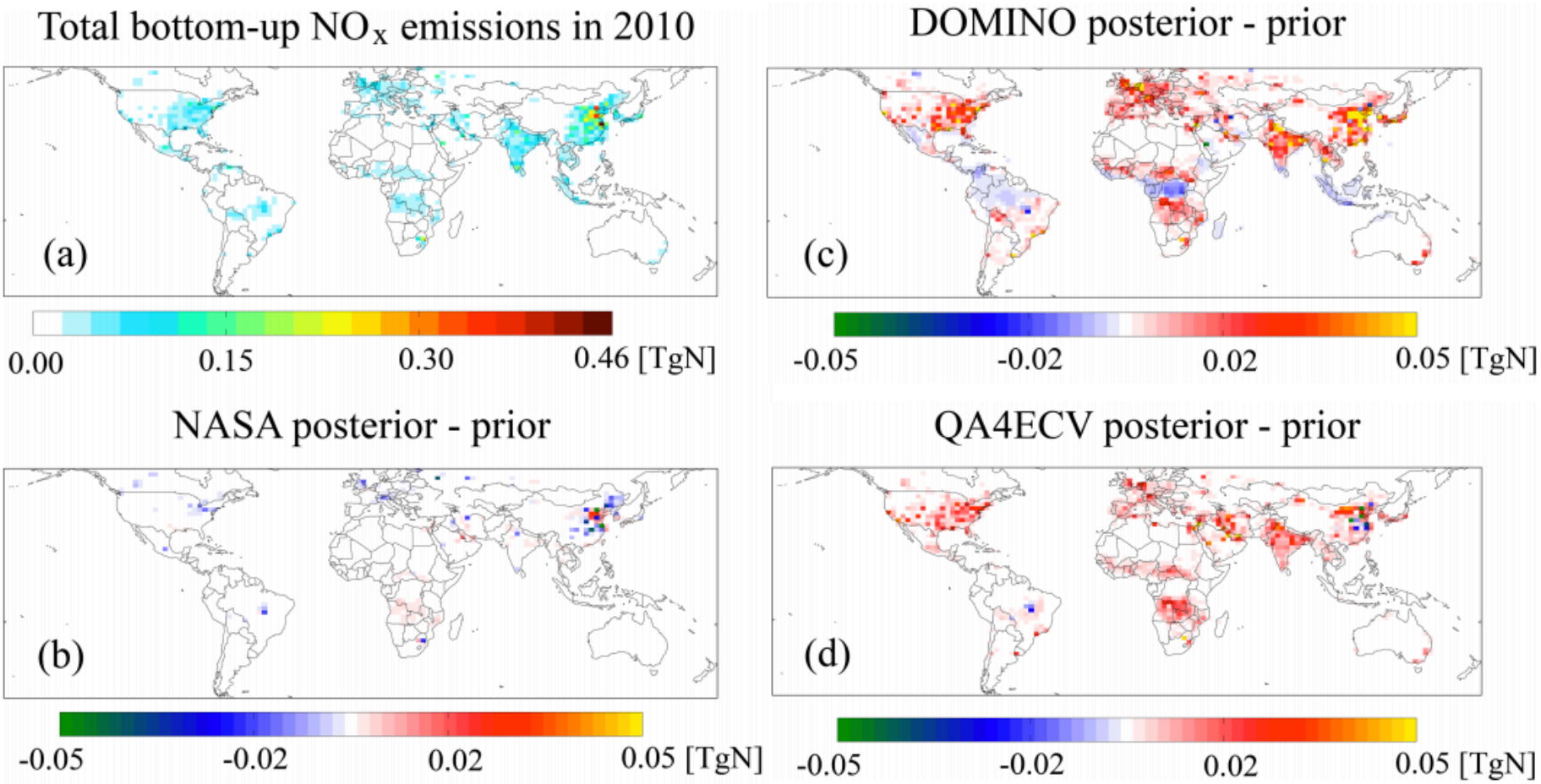
$E\_NO_x\_pos = SF\_industry \times E\_NO_x\_industry \times SF\_NO_x\_industry$

$E\_SO_2\_pos = SF\_industry \times E\_SO_2\_industry \times SF\_SO_2\_industry$

$E\_CO\_pos = SF\_industry \times E\_CO\_industry \times SF\_CO\_industry$

(*Qu et al.*, in prep)

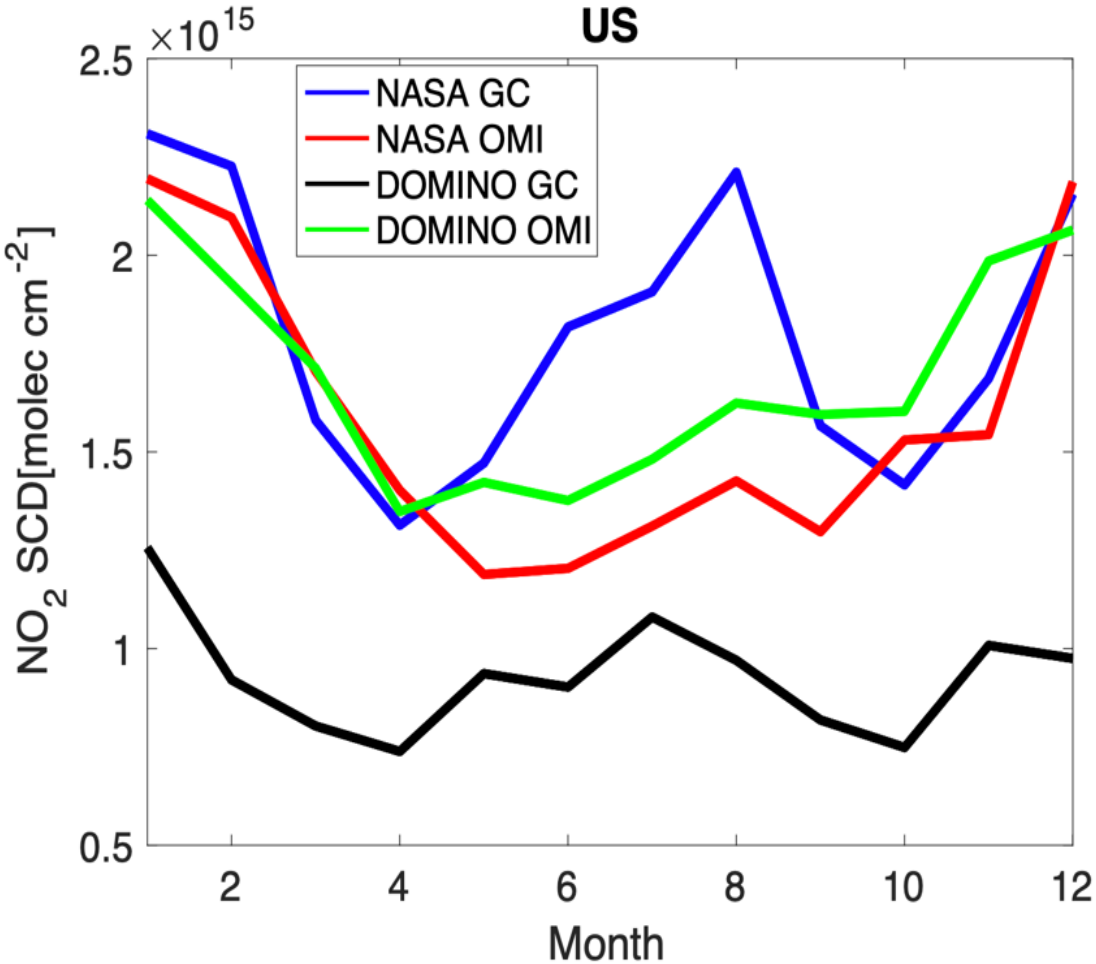
Impact of NO_2 retrievals on top-down NO_x emissions



(*Qu et al.*, 2020)

- NASA SP version 4 is up to 50% higher in urban regions
- Expect more similar results between newest NASA and KNMI retrievals

Impact of NO_2 retrievals on top-down NO_x emissions

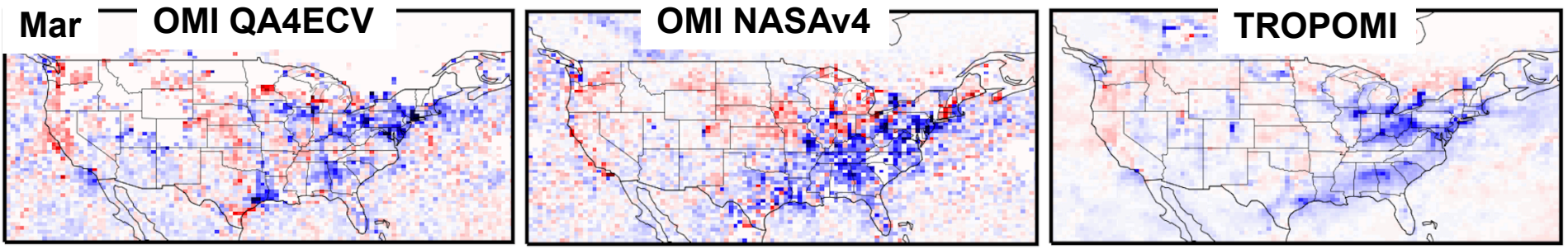


- The different magnitude and seasonality of top-down NO_x emissions reflect the AMF structural uncertainties when retrieving NO_2 columns using different ancillary data

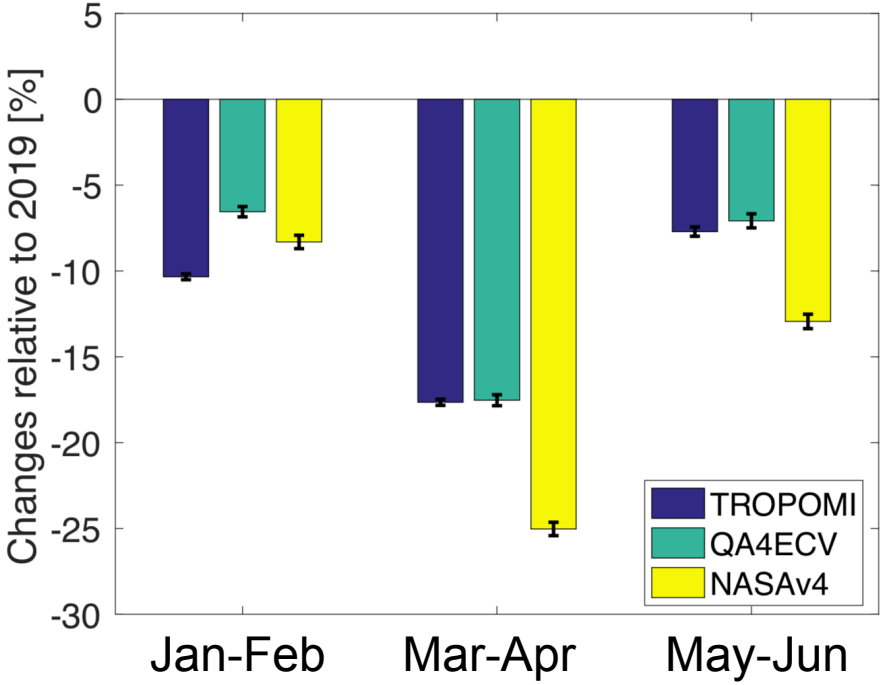
(*Qu et al.*, 2020)

Impact of NO_2 retrievals on top-down NO_x emissions

Changes in NO_2 column densities (2020-2019) [x10^{15} molec cm^{-2}]

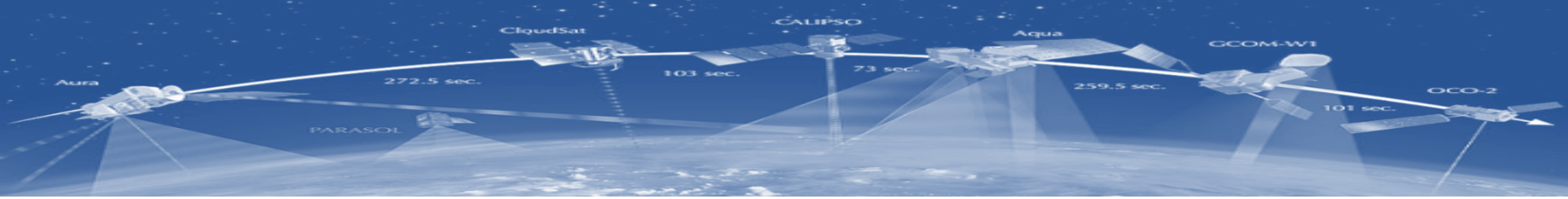


NO_2 reductions in CONUS (2020-2019)



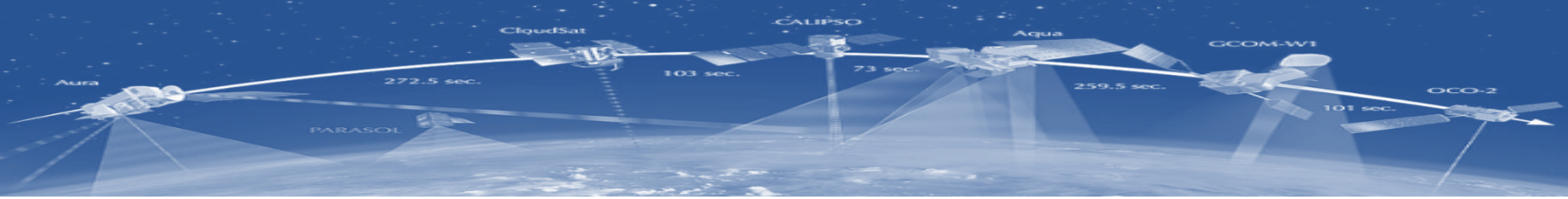
- TROPOMI NO_2 is less noisier than OMI NO_2
- More consistent reductions between TROPOMI and QA4ECV

(*Qu et al.*, in prep)



Summary

- 4D-Var is relatively accurate in emission estimates but is time-consuming and requires adjoint model.
- 4D-Var can be combined with other methods (mass balance) to reduce computational costs − important for TROPOMI and geostationary satellite data.
- Addressing chemical interactions and co-emissions can reduce errors in top-down emission estimates.
- Uncertainties from satellite retrievals also impact top-down emission estimates.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CloudSat
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CALIPSO
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Aqua
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GCOM-W1
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OCO-2
PARASOL

Thanks!