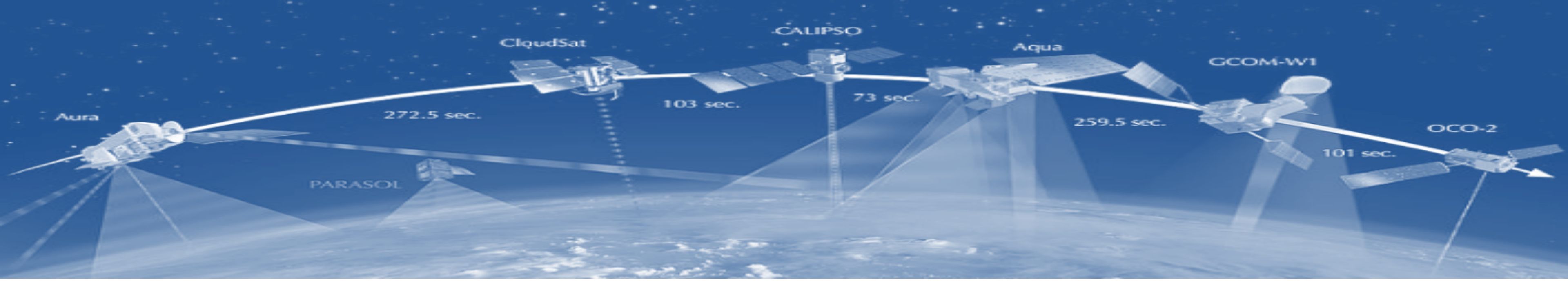


Magnitude and trend of NO_x and SO_2 emissions constrained by OMI observations

Zhen Qu and Daven K. Henze
University of Colorado Boulder

zhen.qu@colorado.edu

EIC 2019 , August 2, 2019

Two ways to estimate emissions

- Bottom-up estimates – prior emissions:

- Emission = emission factor x activity
- Large uncertainties, lag in time

- Top-down estimates – posterior emissions

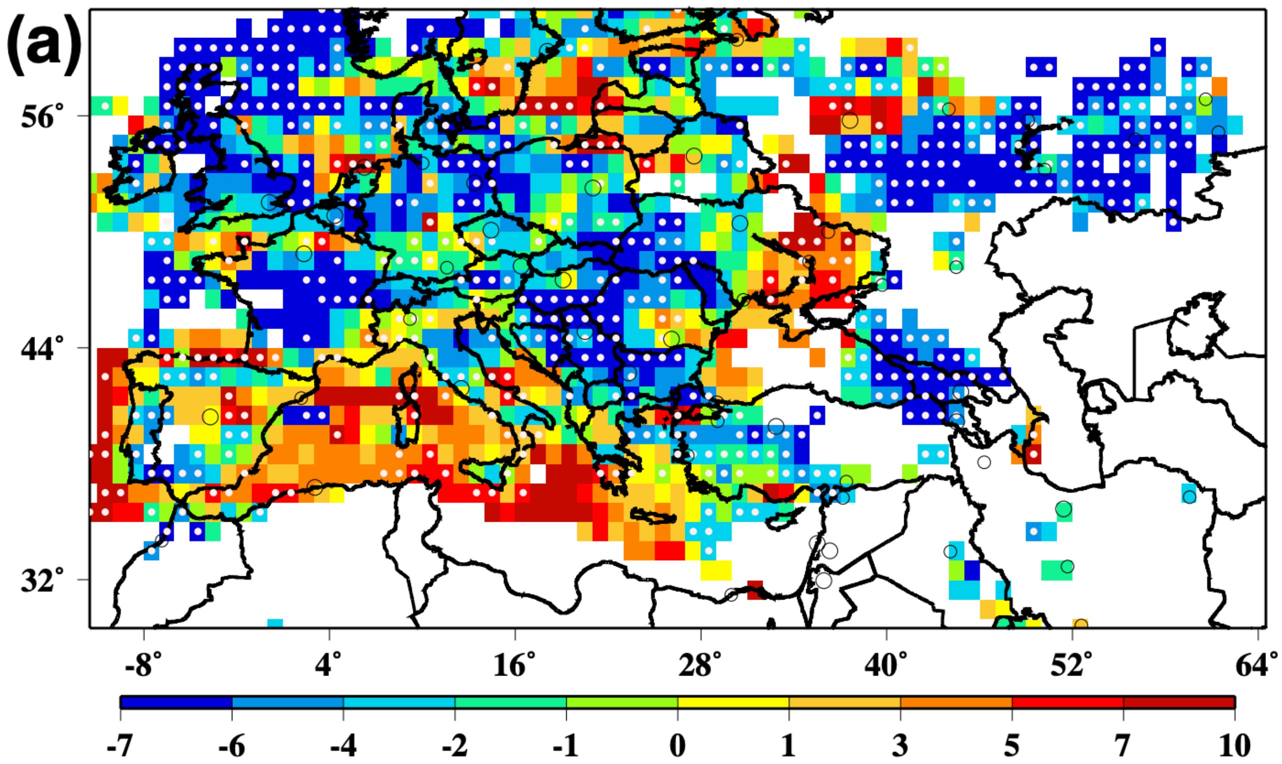
- Use observations and physical model to solve inverse problem which gives the maximum likelihood estimate of emissions

Top-down emission studies

- **Analytic inversion**
  - Expensive to compute the Jacobian matrix;
  - Approximated by linear relationships of NO_2 column to NO_x.

(*Konovalov et al.*, 2006, 2008)

Changes in European NO_x emissions



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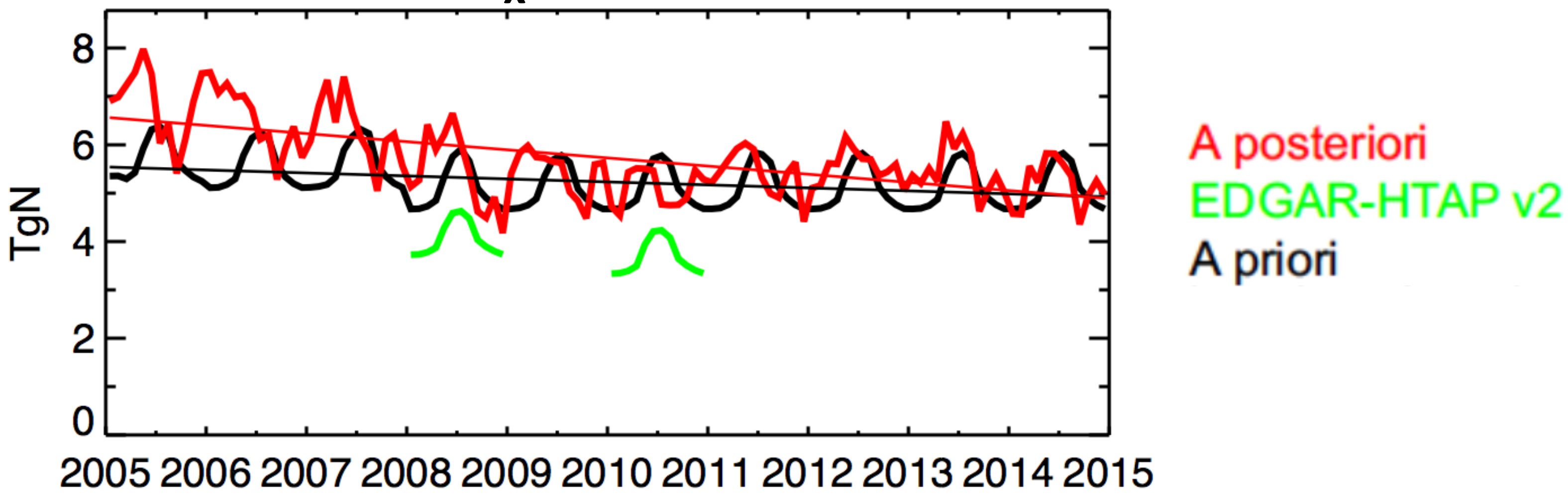
  - Expensive using large ensemble members;
  - Hard to implement realistic localization.

(*Miyazaki et al.*, 2015, 2017)

US NO_x emissions



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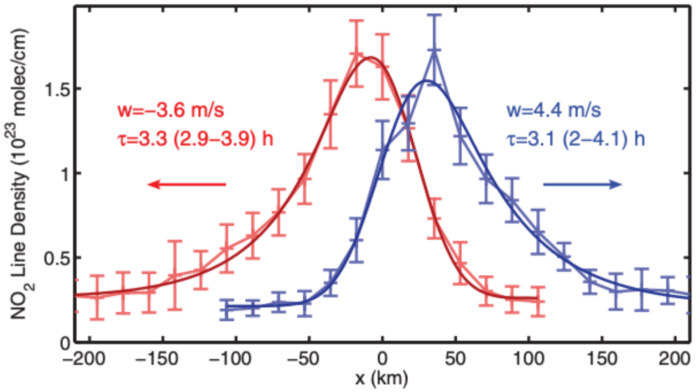
(*Beirle et al.*, 2011)



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  - Need average over multiple years.

(*Beirle et al.*, 2011)

- **Mass balance**
  - Fast;
  - Approximate transport & nonlinear chemistry.

(*Martin et al.*, 2003)

History of 4D-Var NO_x emission estimates

1999

Elbern & Schmidt
Full 4D-Var for 3D CTM

History of 4D-Var NO_x emission estimates

Elbern et al.
Pseudo observation

1999

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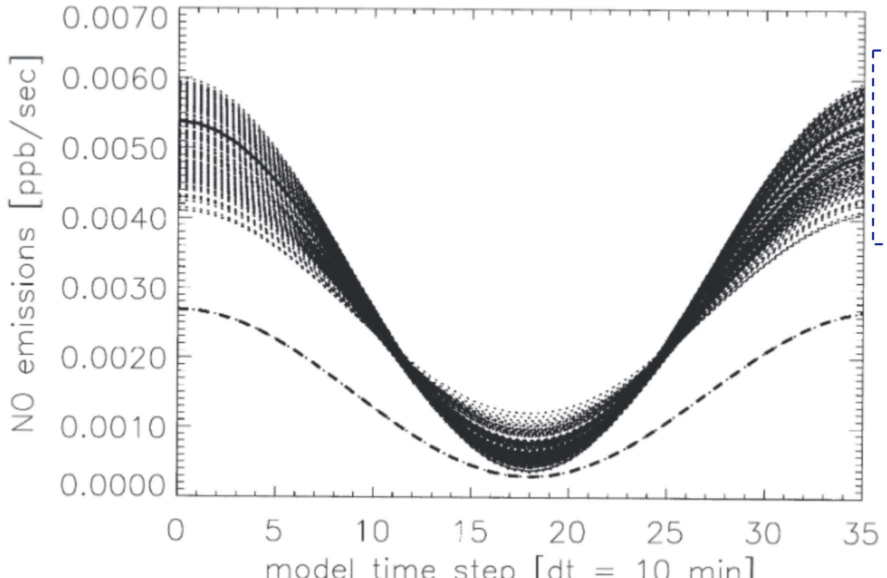

NO emission rates can be estimated using O_3 observations

History of 4D-Var NO_x emission estimates

Elbern et al.
Pseudo observation

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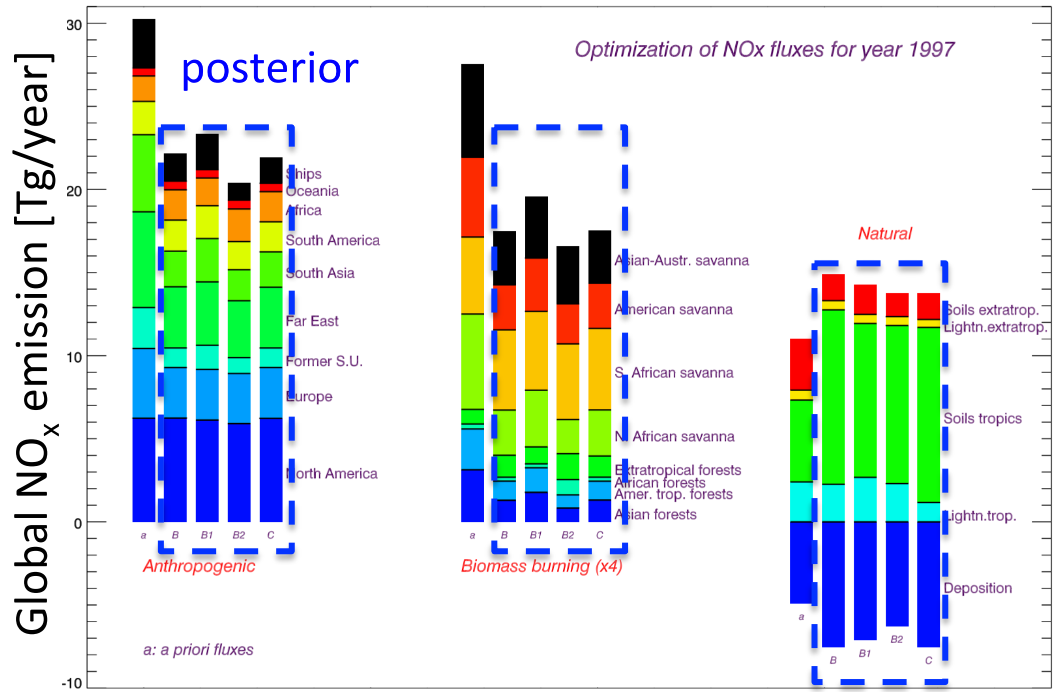

First use of NO_2 satellite observations

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Henze et al.
IMPROVE aerosol

First use of surface aerosol measurements

History of 4D-Var NO_x emission estimates

Elbern et al.
Pseudo observation

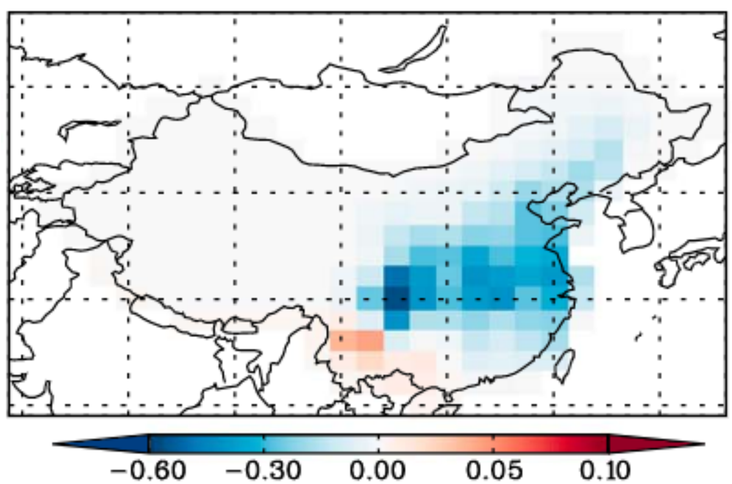
Henze et al.
IMPROVE aerosol

1999 **2005** **2013**

2000 **2009**

Elbern & Schmidt
Full 4D-Var for 3D CTM

NO_x emission (posterior − prior)/prior



Xu et al.
MODIS radiance

History of 4D-Var NO_x emission estimates

1999 — **Elbern & Schmidt**: Full 4D-Var for 3D CTM

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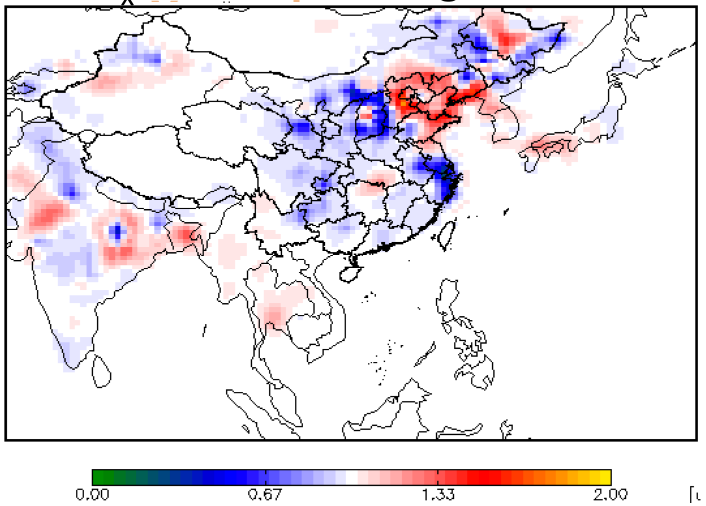
2009 — **Henze et al.**: IMPROVE aerosol

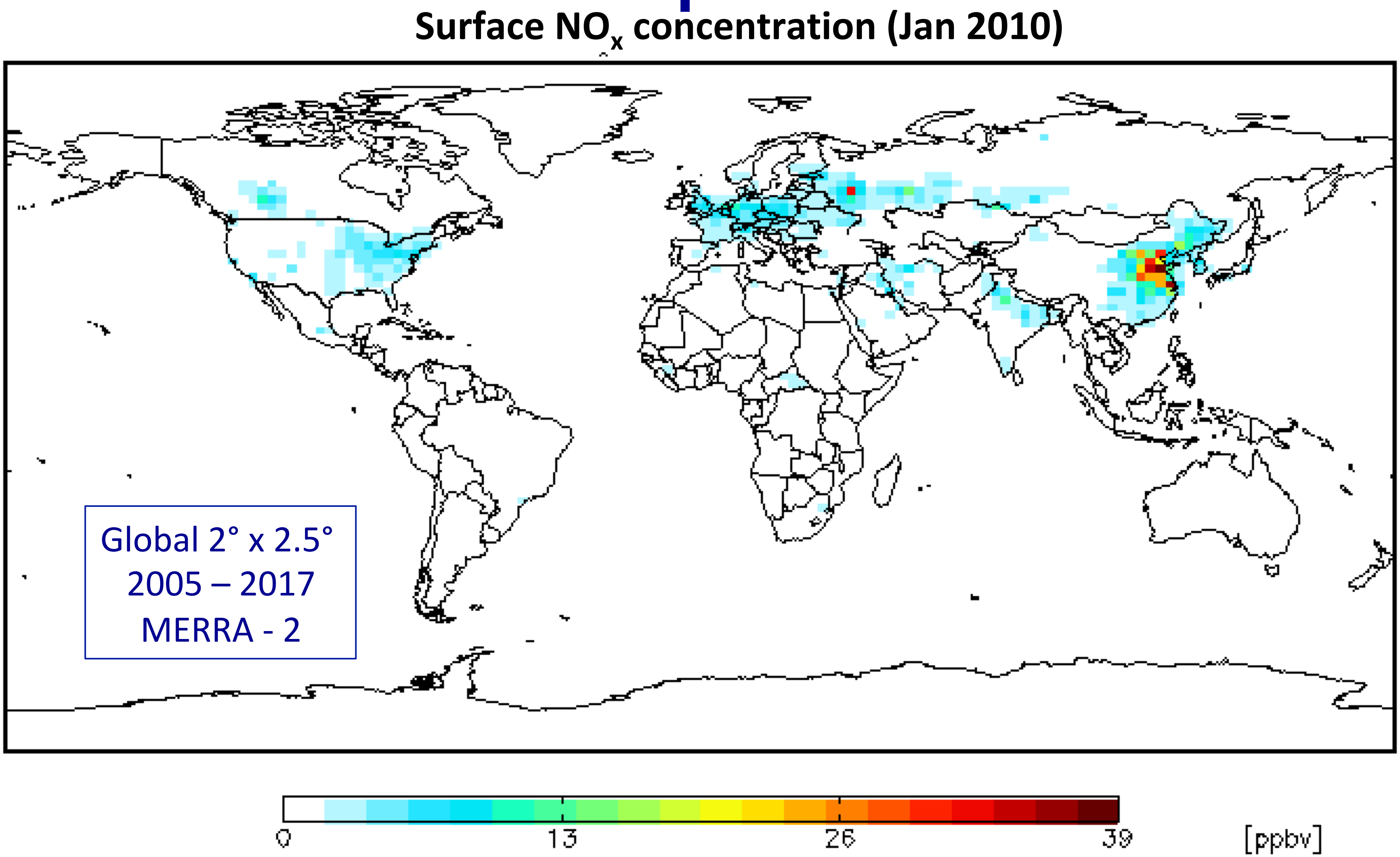
2013

2017 — **Qu et al.**: OMI NO_2

NO_x emission scaling factors

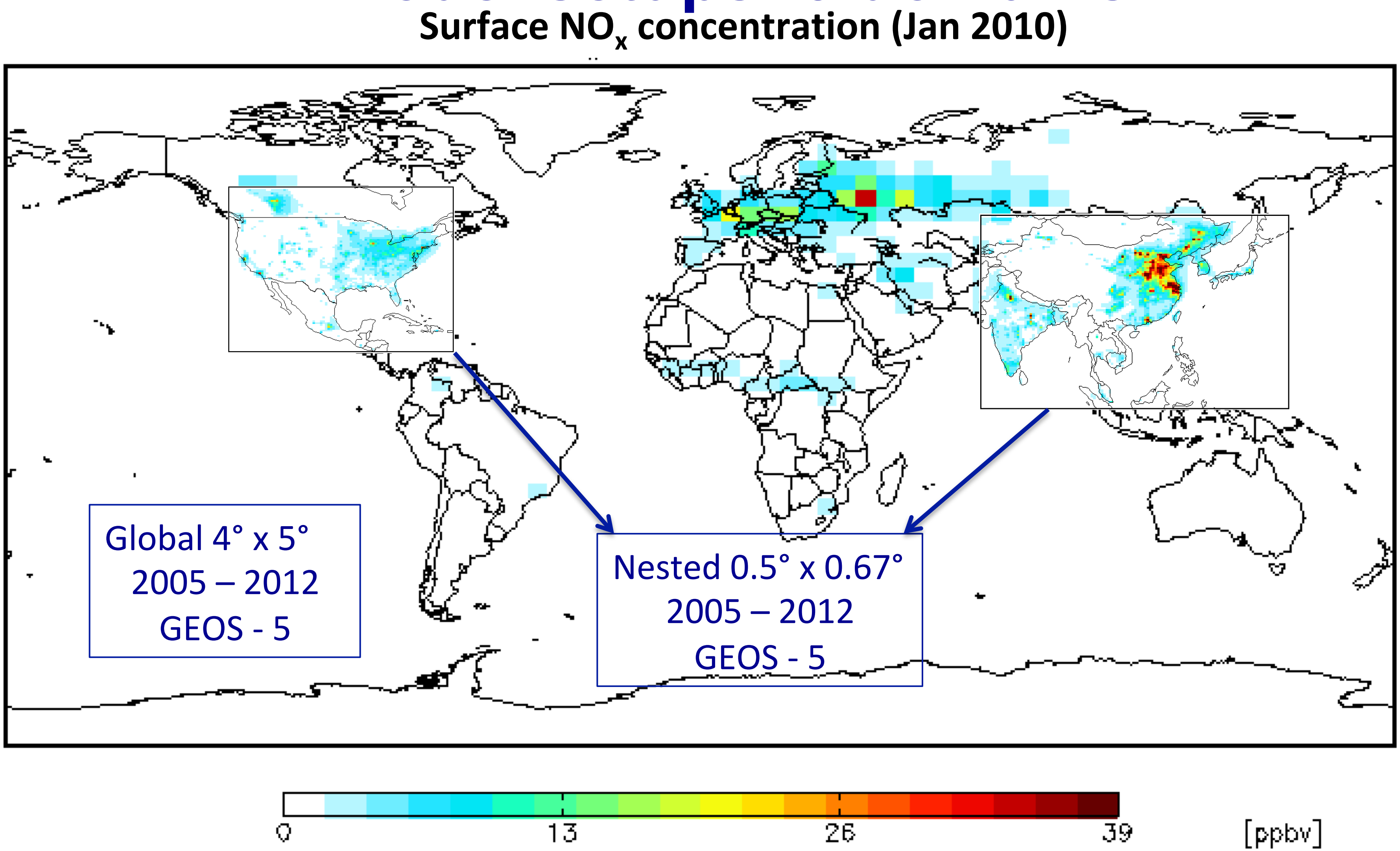


Model setups: 3 domains



- **Model:** GEOS-Chem chemical transport model and its adjoint
  - Meteorological input from Goddard Earth Observing System (GEOS)
  - Prior emissions: HTAP v2.1 bottom-up inventory (2010) for all years and domains
  - Global domain: 2° lat x 2.5° lon resolution for 2005 – 2017

Model setups: 3 domains



- Nested US and nested EA domain : BC from global 4° x 5° simulation, 2005 - 2012

Satellite observation

- Ozone Monitoring Instrument (OMI) onboard Aura: NO_2 and SO_2
- Overpass time : 13:45 local time, daily global coverage
- Footprint: 13 km x 24 km

- Use level 2 product for all work in the presentation
- Column density: total NO_2 and SO_2 molecules from surface to the top of the atmosphere within a model grid [molec cm^{-2}]

Methods

- **Inversion approaches:**

4D-Var:

- adjust emissions independently in each grid cell
- takes into account transport and chemical reactions
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Hybrid 4D-Var / Mass balance:

- blend of accuracy and efficiency

Outline

1. Top-down NO_x emissions
2. Top-down SO_2 emissions
3. Joint NO_x and SO_2 inversions
4. Sector-based inversion

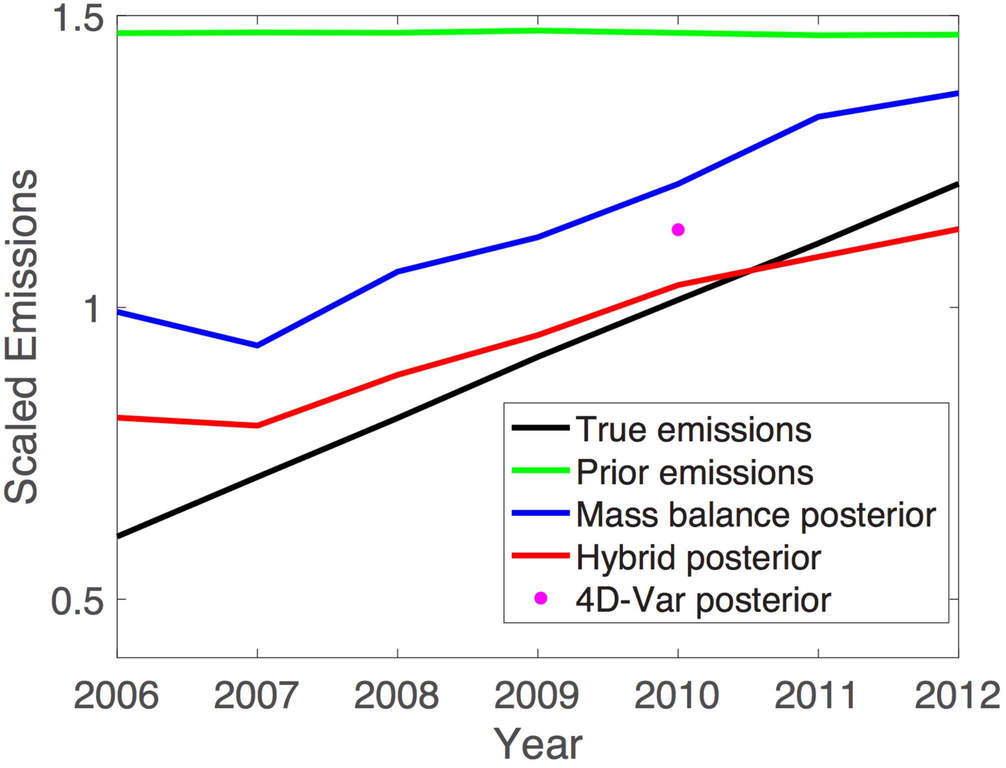
Hybrid inversion for NO_x

Hybrid method:

Base year (2010): 4D-Var

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

Scaled emissions in pseudo observation test

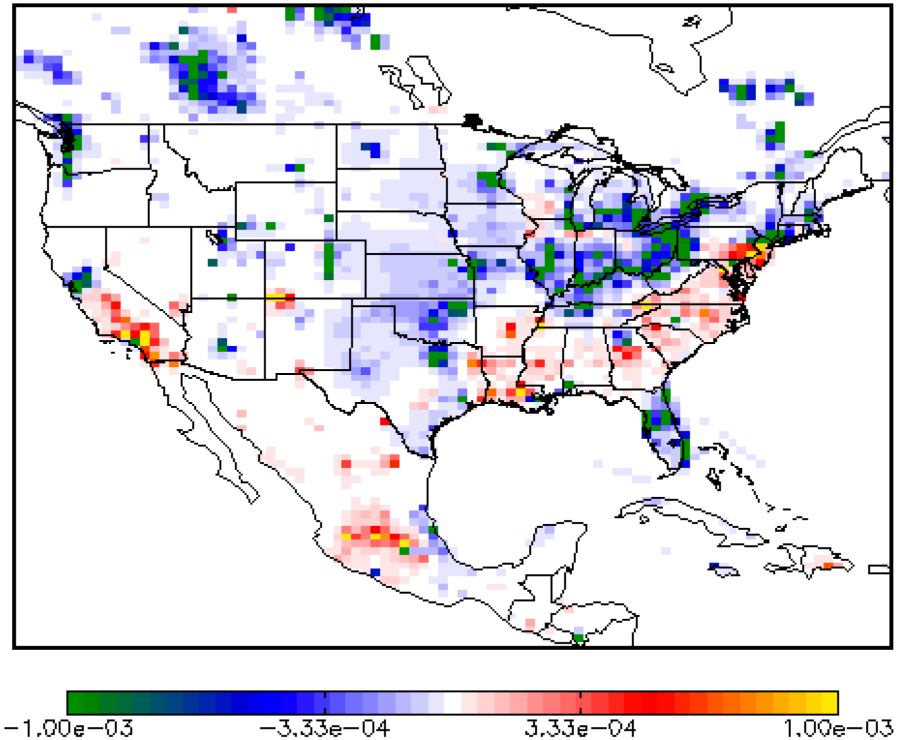


- Hybrid posterior has smaller NMSE (by 59% to 78%) and better correlation.

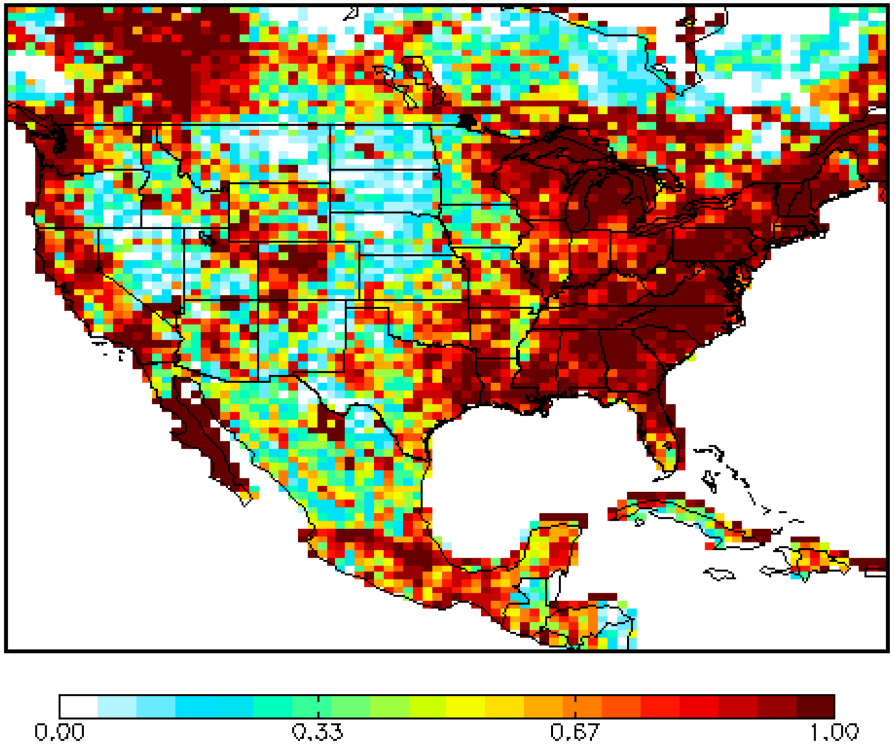
(Qu *et al.*, 2017)

Differences between bottom-up and top-down estimates

Top-down – bottom-up, 2010 [TgN/year]



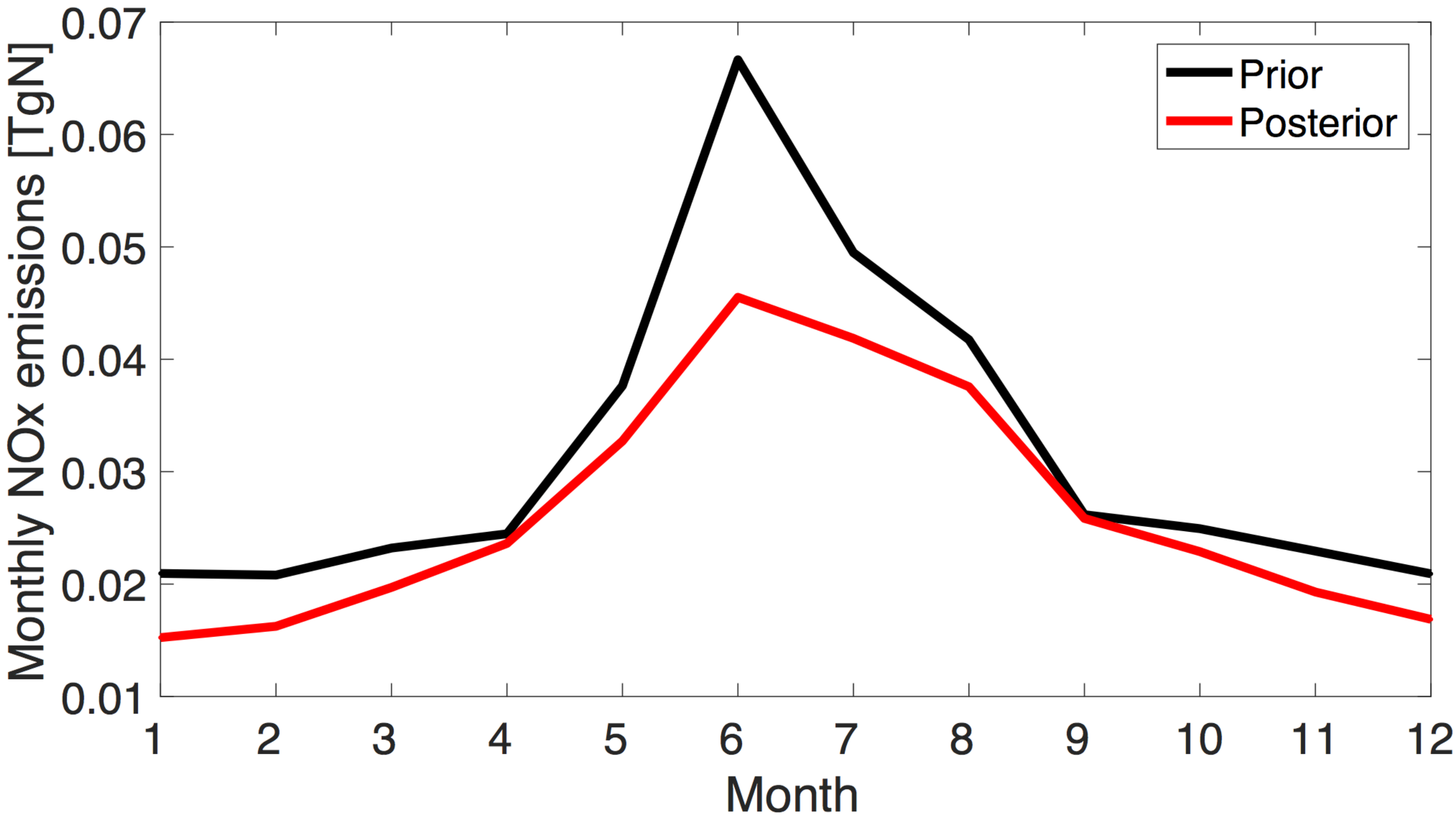
Anthropogenic / total NO_x emissions



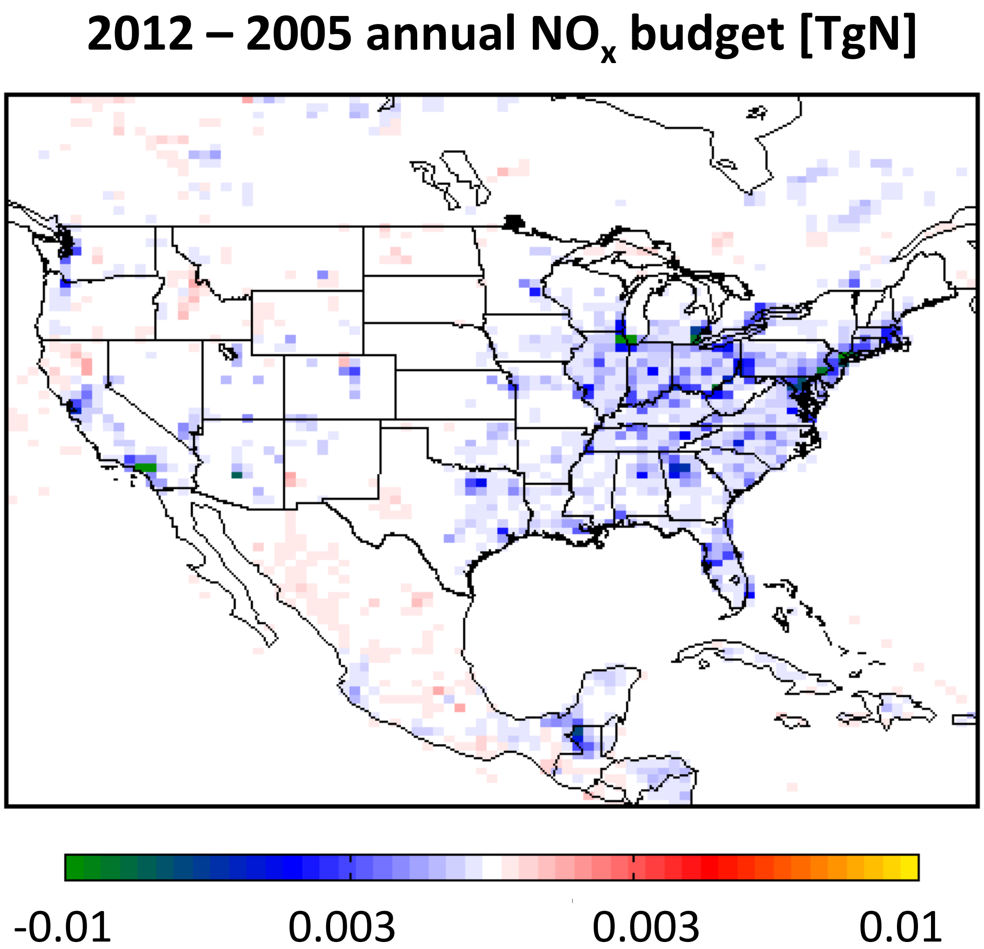
- Underestimates in HTAP at regions with large anthropogenic sources (East Coast of US & Mexico City)
- Overestimates in HTAP at regions with moderate anthropogenic sources (mid US)

Smaller seasonality of top-down NO_x emissions

US NO_x emissions in 2010



Inter-annual variation: Changes of NO_x emissions in NA



- Annual budget of top-down NO_x emissions decrease by 20% from 2005 to 2012 in the US
- NO_x emission changes in Mexico are less than 1% from 2005 to 2012

Large differences in OMI NO_2 column from two retrievals

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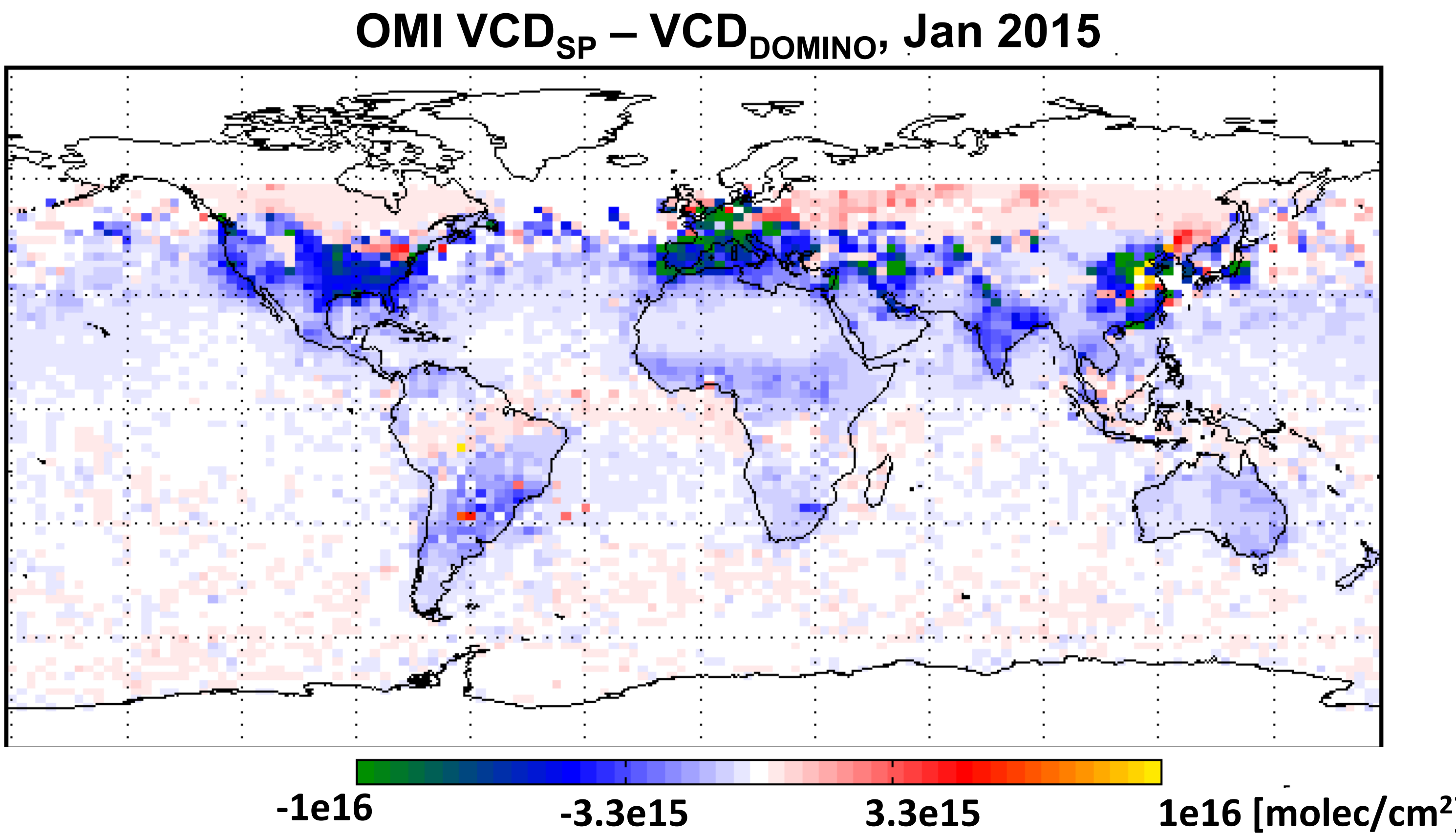


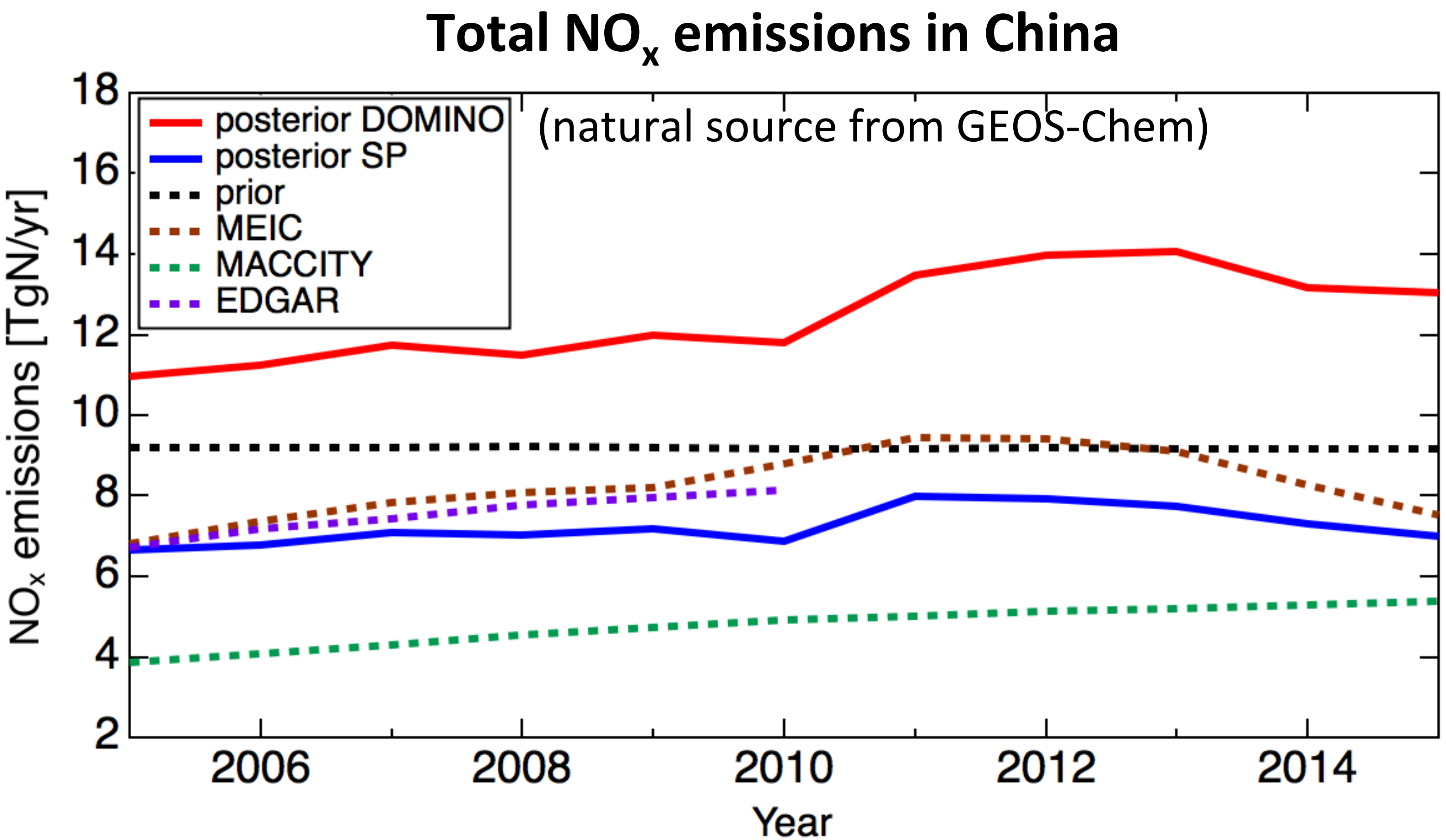
Vertical Column Density:

Standard Product (SP): $VCD_{SP} = VCD_{SP\_OMI} * AMF_{SP}/AMF_{GC\_SP}$

DOMINO Product: $VCD_{DOMINO} = VCD_{DOMINO\_OMI} * AMF_{DOMINO}/AMF_{GC\_DOMINO}$

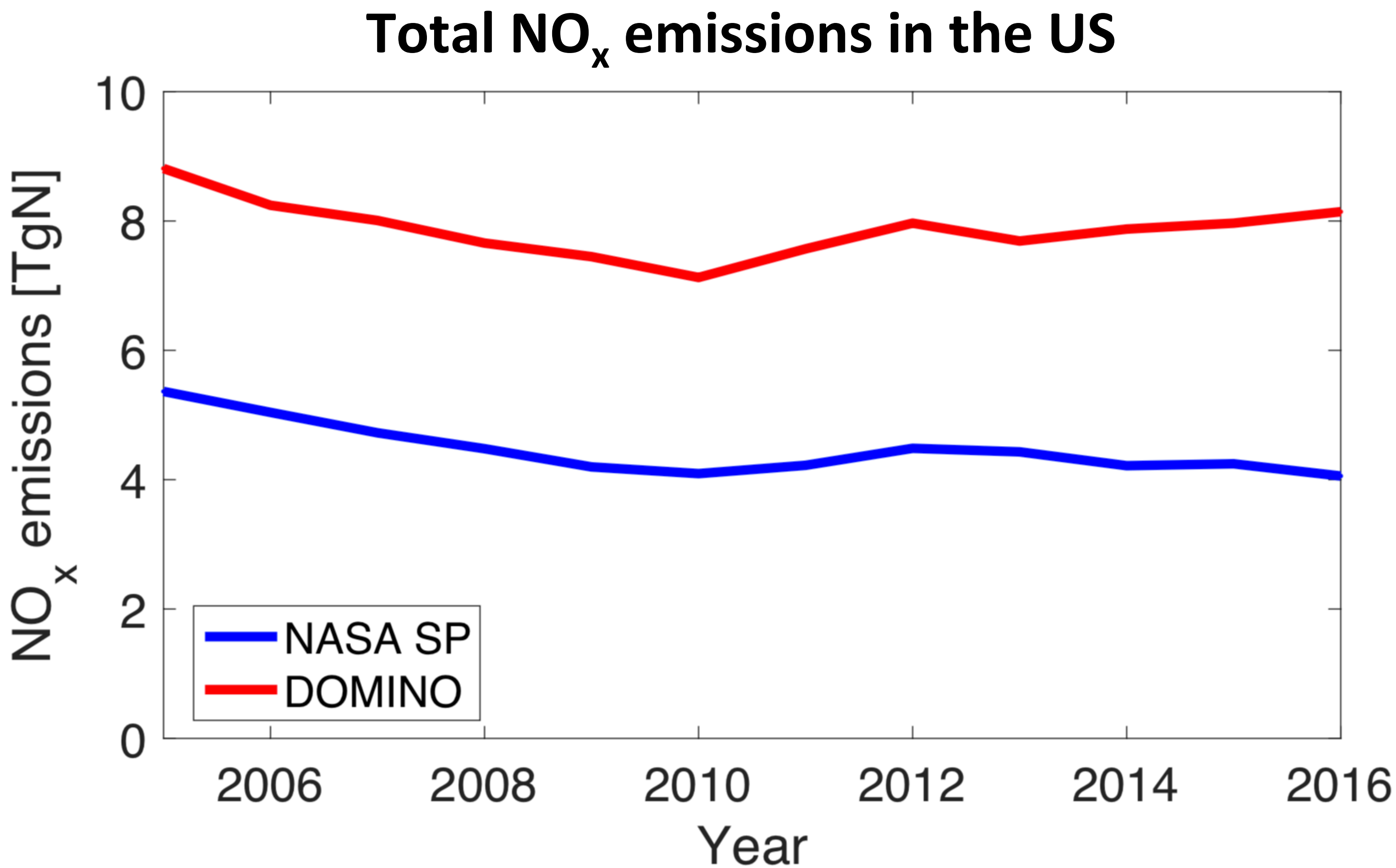
- NO_2 column densities from SP are ~ 50% smaller than that from DOMINO in densely populated and industrial regions. (*Qu et al.*, 2017; *Canty et al.*, 2015; *Zheng et al.*, 2014)

Different magnitude of NO_x emissions from NASA SP and DOMINO retrievals



- Posterior NO_x emissions from SP is smaller than that from DOMINO by 39-46%.

Different magnitude of NO_x emissions from NASA SP and DOMINO retrievals



- Posterior NO_x emissions from SP is smaller than that from DOMINO by 39-50%.
- The slowdown of NO_x emissions is not reflected in NEI inventory.

(*Jiang et al.*, 2018)

Different magnitude of NO_x emissions from NASA SP and DOMINO retrievals

Total NO_x emissions in Mexico

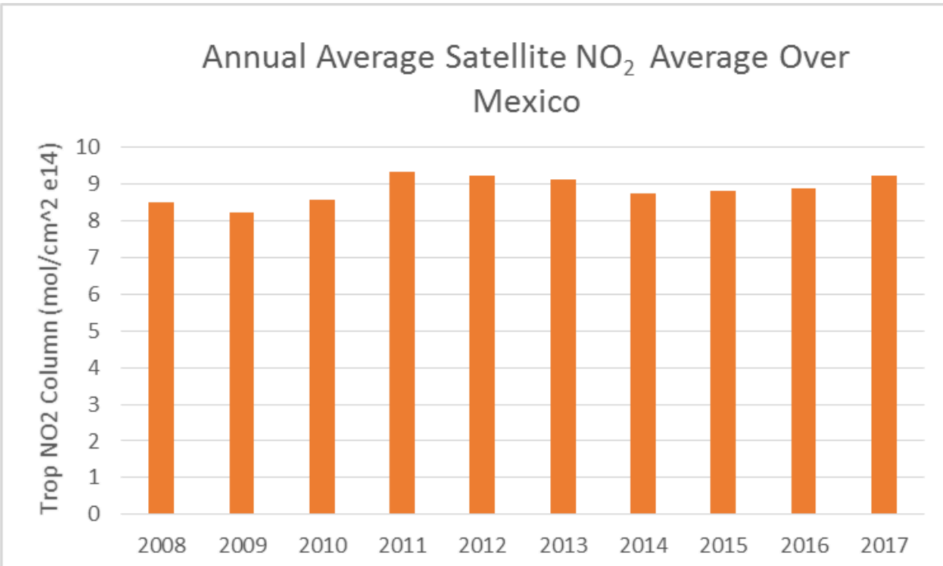


Figure 4-7. Annual Average Satellite NO_2 Columns over Mexico.

(RAMBOLL report)

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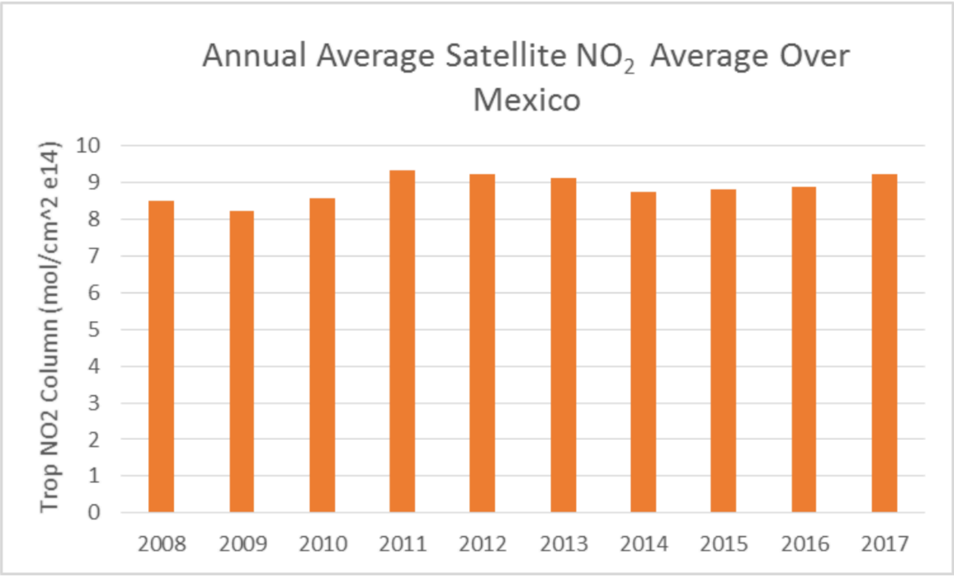

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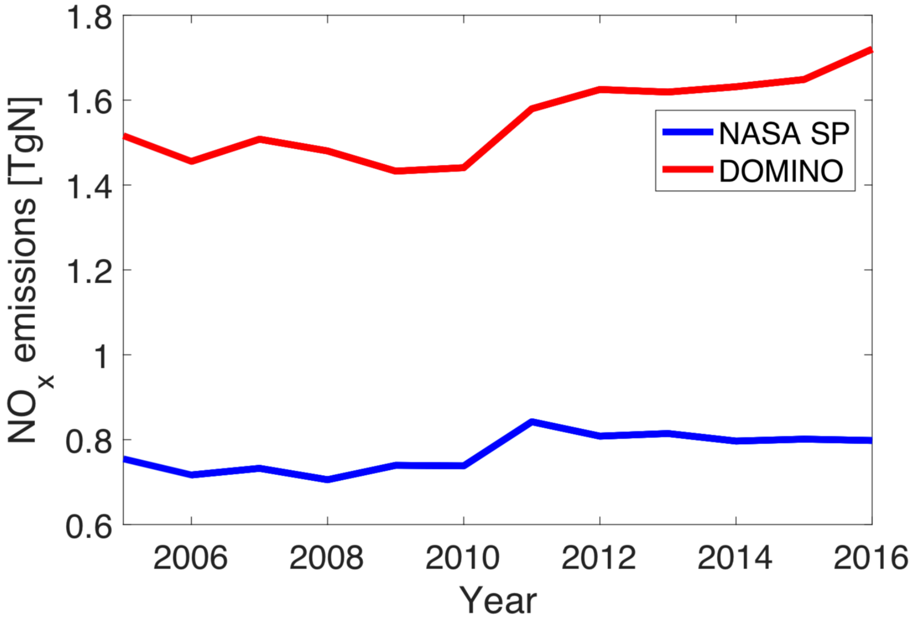
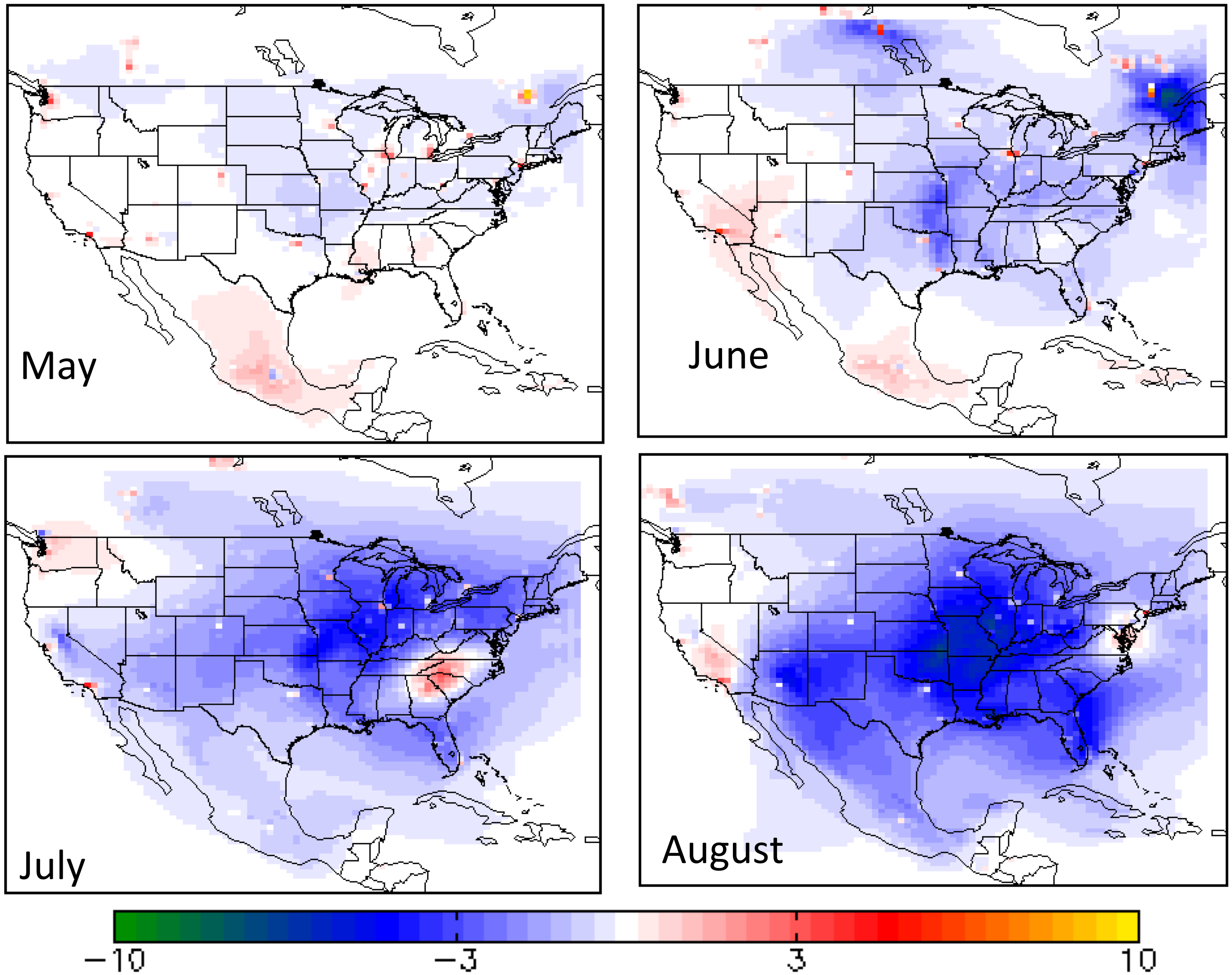


(RAMBOLL report)

- Posterior NO_x emissions from SP is smaller than that from DOMINO by 47-51%.

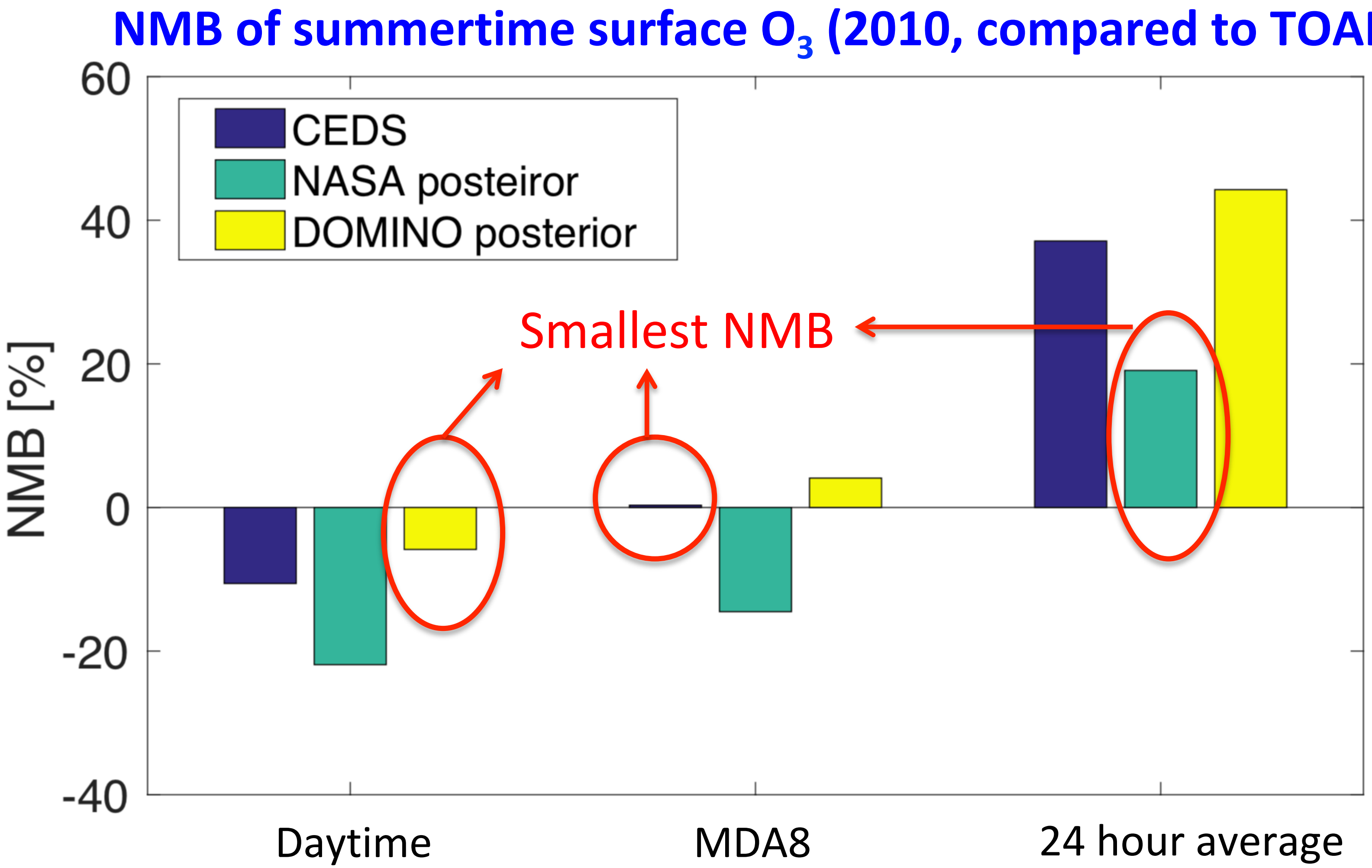
Impact of assimilating NO_2 observations on O_3 (2010)

Surface O_3 concentration (posterior NO_x – prior NO_x) [ppbv]



- NO_x emission is overestimated in US bottom-up inventory
- Simulated O_3 are generally overestimated in US using HTAP 2010 emissions

Impact of assimilation on improving estimates of surface O_3 depends upon the O_3 metric, emphasizing the importance of hourly NO_x constraints



- Posterior simulations have smaller NMB and NMSE and better seasonality and inter-annual variation in 24 hour O_3

SO_2 emissions constrained by OMI SO_2 NASA and BIRA products

- **3 OMI SO_2 products**: NASA standard (SP), NASA prototype, BIRA

  Treatment of clouds, radiative transfer model, and retrieval algorithm lead to differences in NASA and BIRA SO_2 retrievals, which are more consistent when VZA and SZA are small

(*Qu et al*., 2019a)

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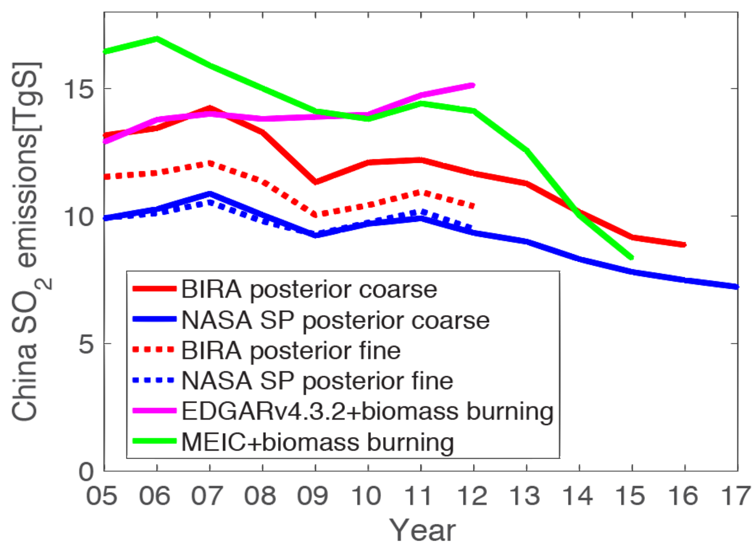

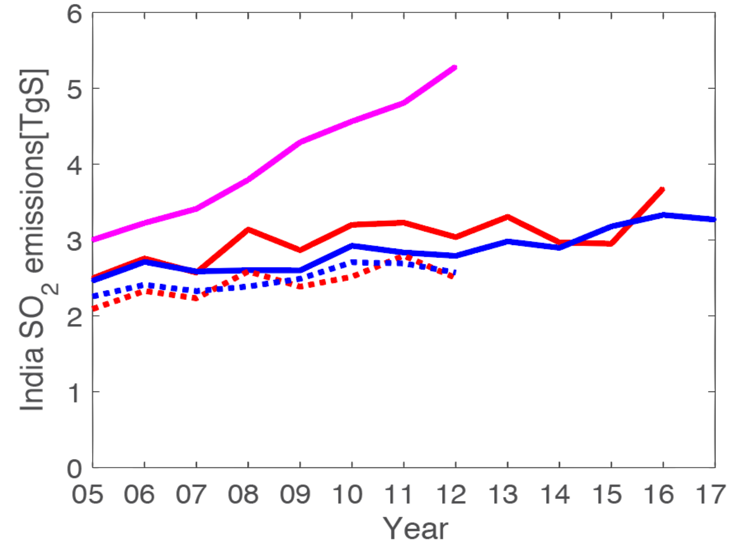

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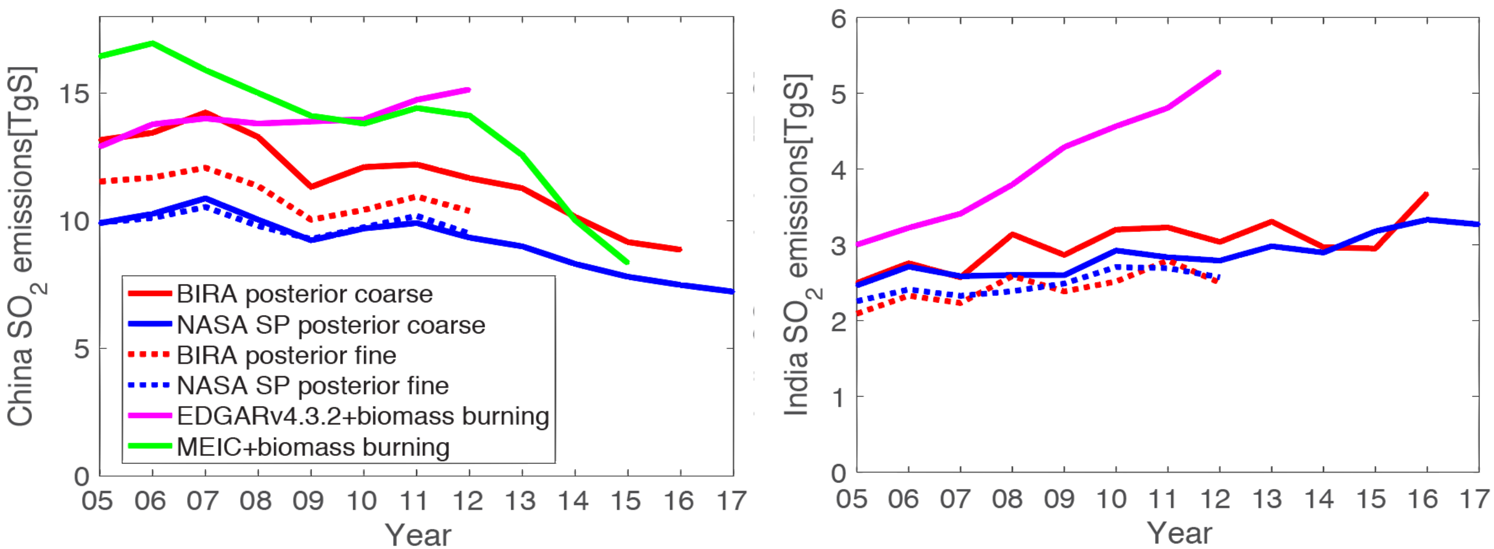


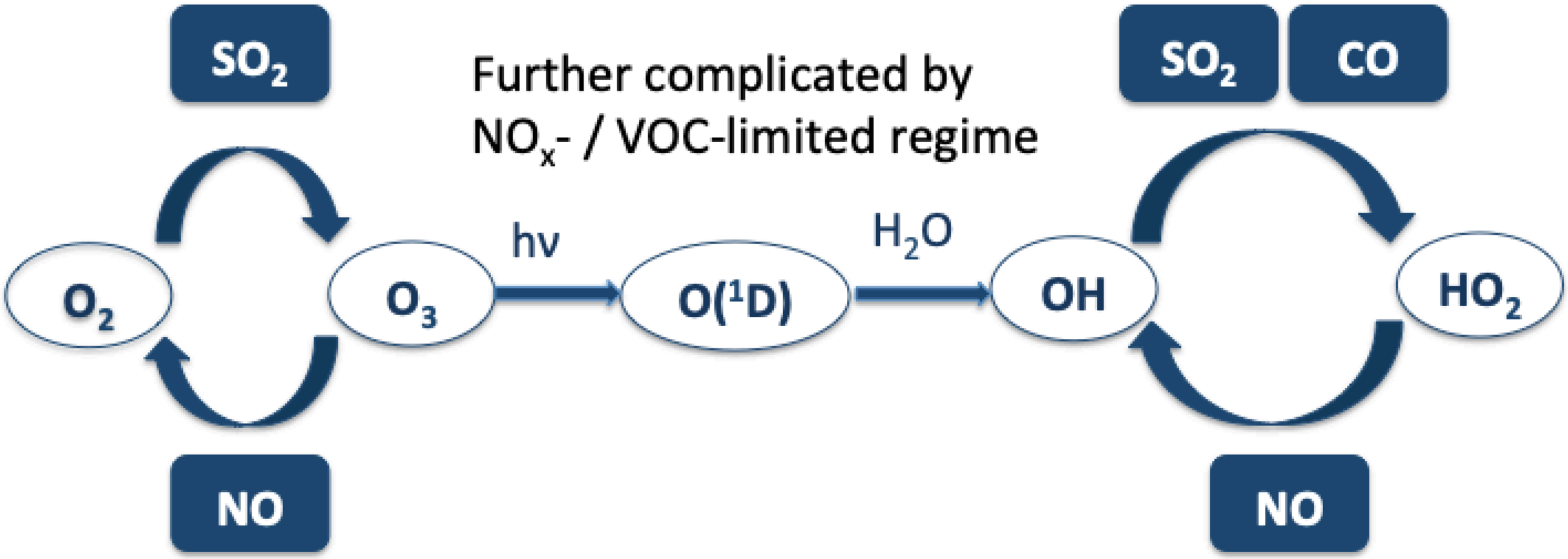
- **Evaluation with surface & aircraft measurements:** Reduced NMB in annual mean surface SO_2 in China, India and US but not in Korea possibly due to differences in SO_2 vertical profile in model and real atmosphere.

(*Qu et al*., 2019a)

Top-down emissions

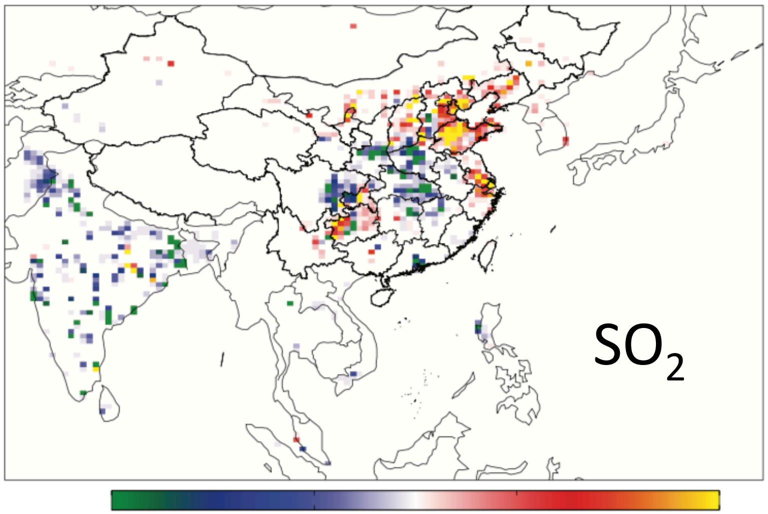
Still …

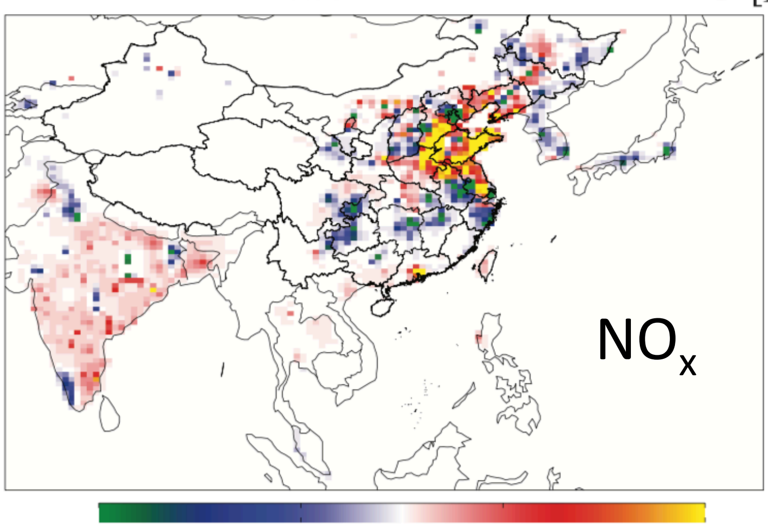
- Chemical interactions are not being considered so far
- Uncertainties in other species emissions are likely degrading the top-down emission of the constrained species



Joint NO_2 & SO_2 4D-Var inversion -- better match observations and surface measurements (January, 2010)

Joint − Single posterior emissions





Joint: assimilate NO_2 and SO_2 observations to optimize NO_x and SO_2 emissions simultaneously

Single: only assimilate NO_2 (SO_2) observations to optimize NO_x (SO_2) emissions

(*Qu et al.*, 2019b)

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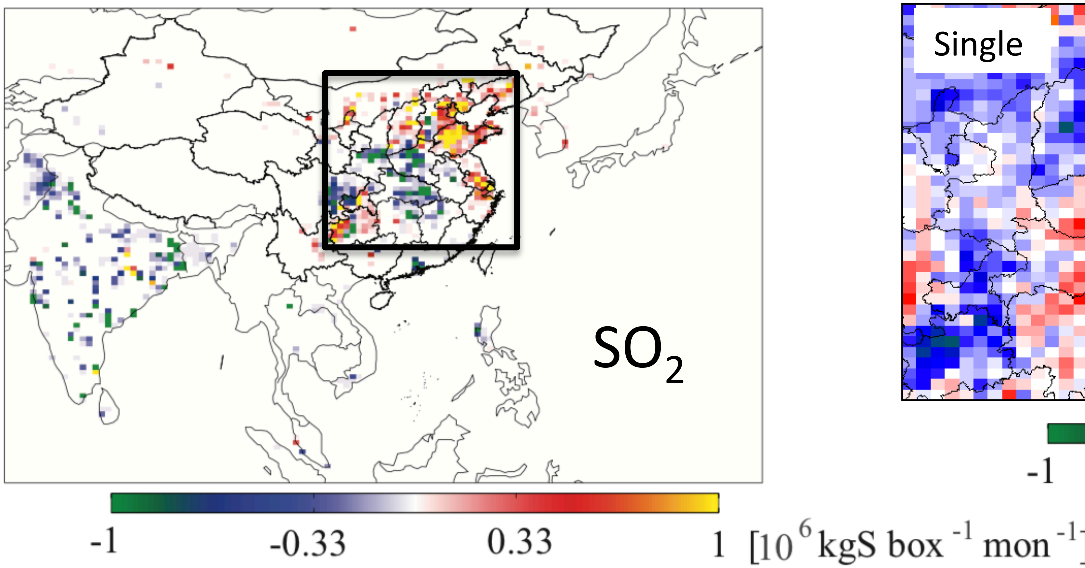
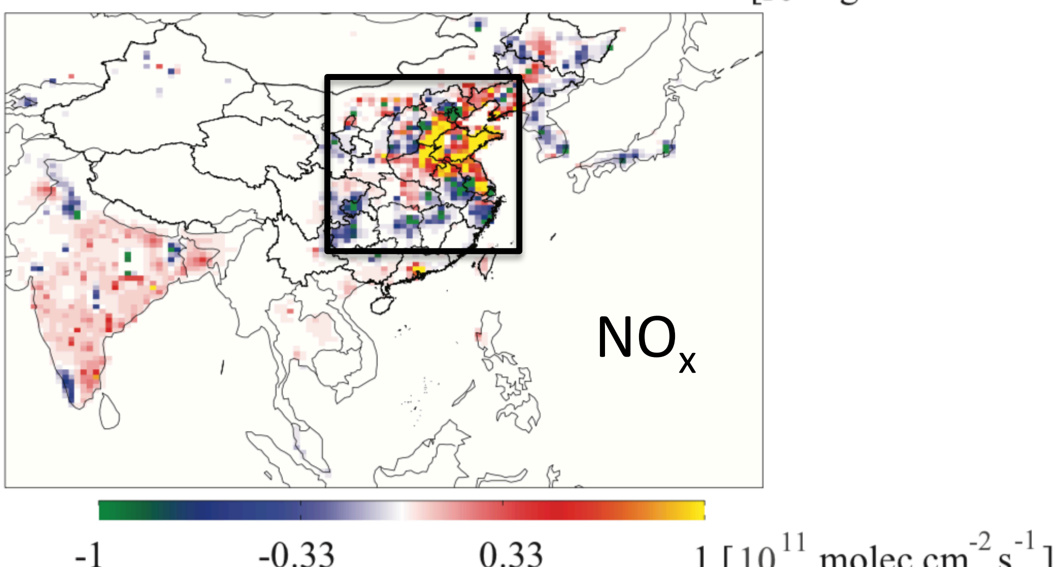
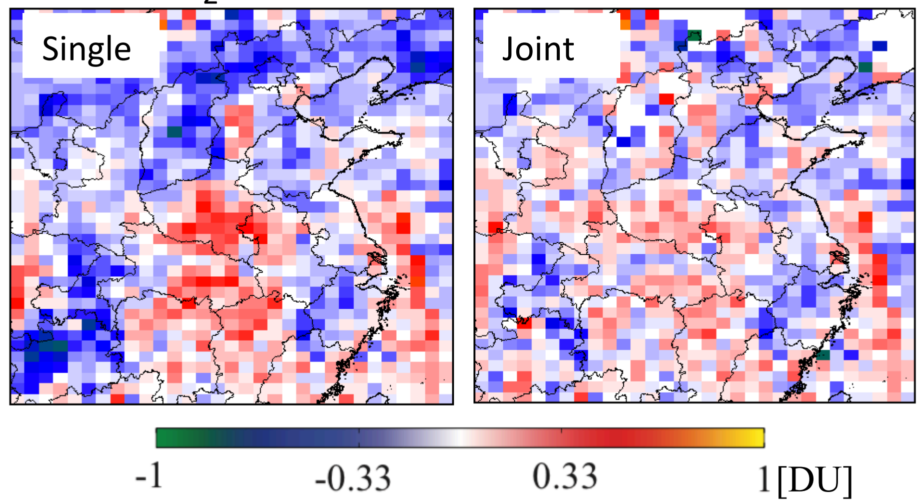




SO_2 columns (GEOS-Chem − OMI)



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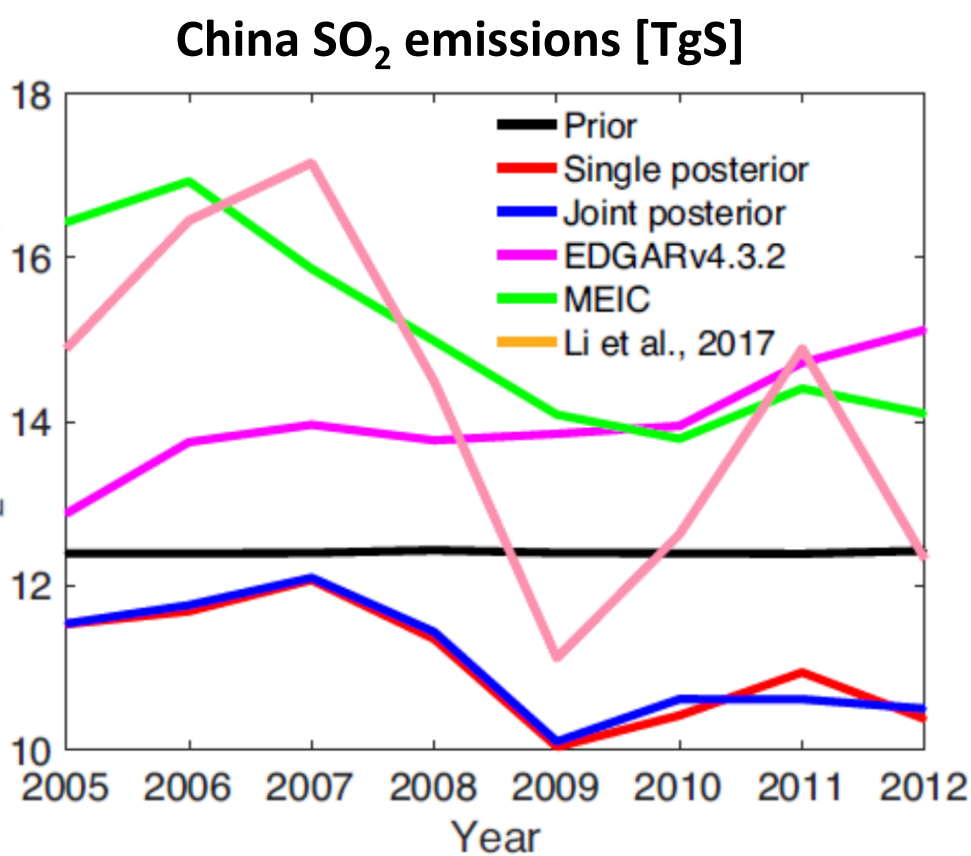
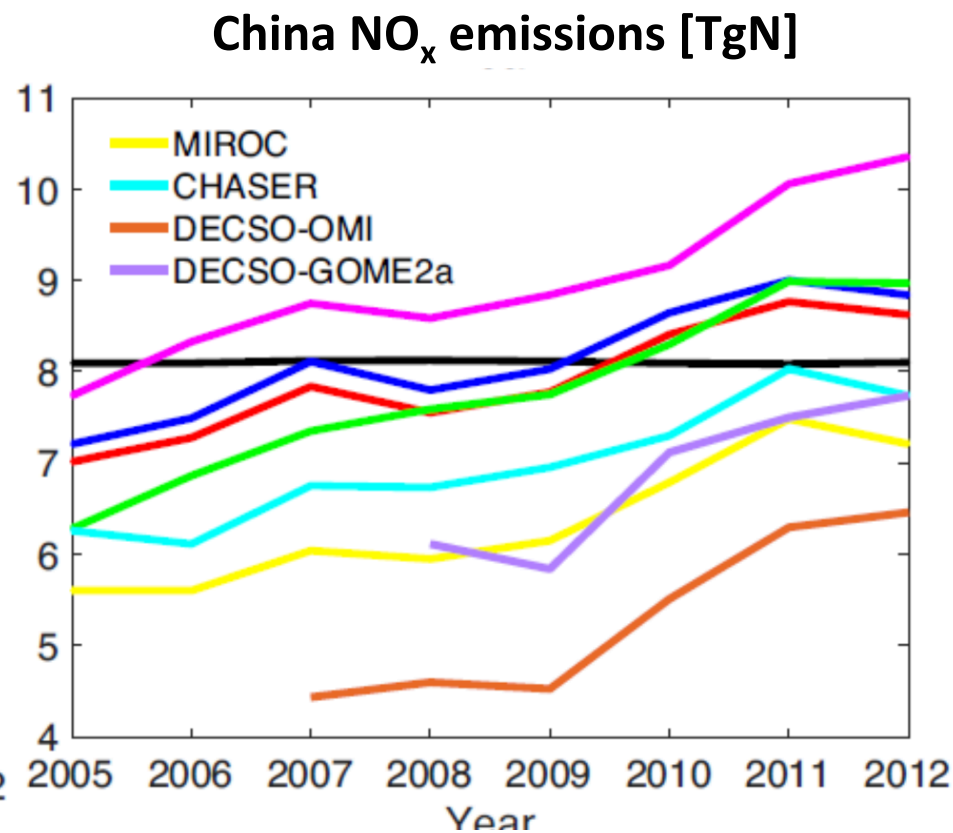
(*Qu et al.*, 2019b)

Similar magnitude and trend of single species and joint inversion posterior emissions





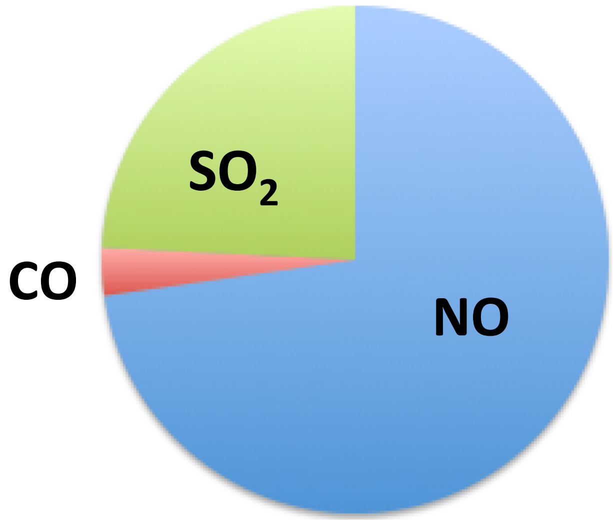
(*Qu et al.*, 2019b)

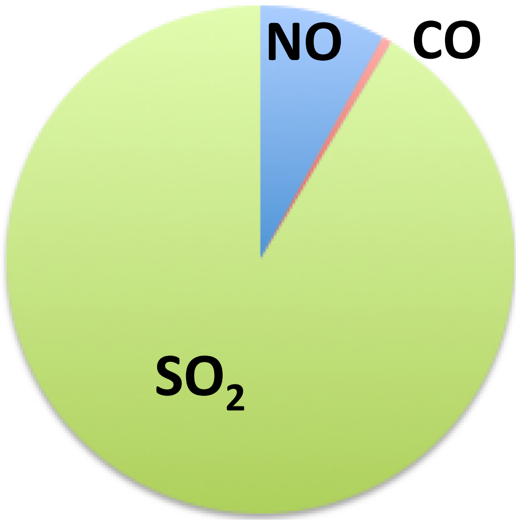
Accounting for correlated co-emitted pollutants in 4D-Var

Transportation

Energy

Sector-based emission scaling factor



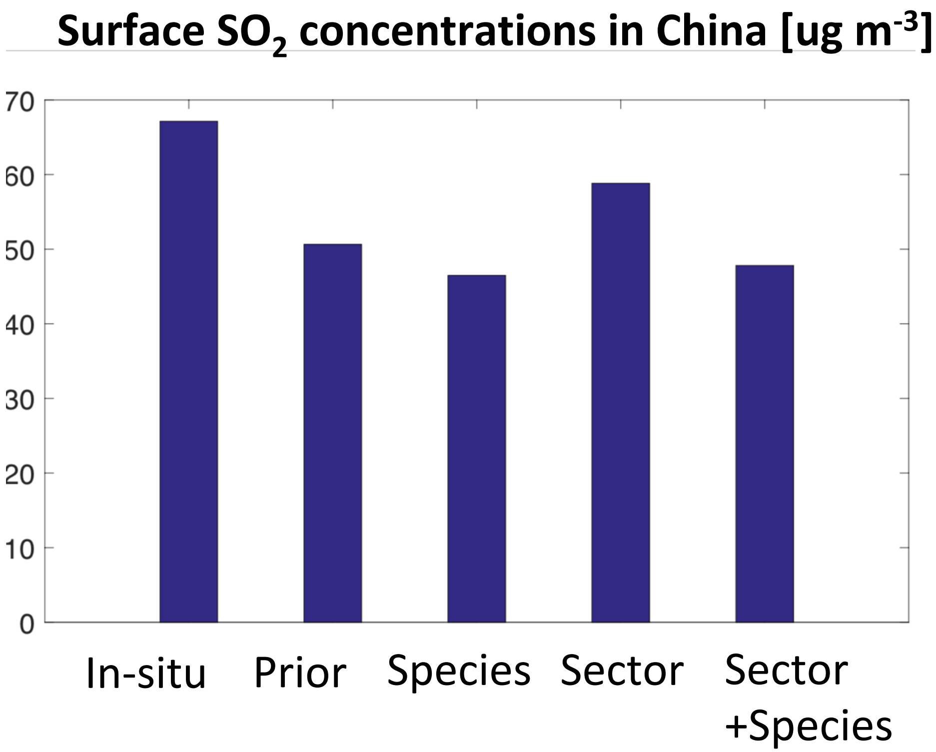


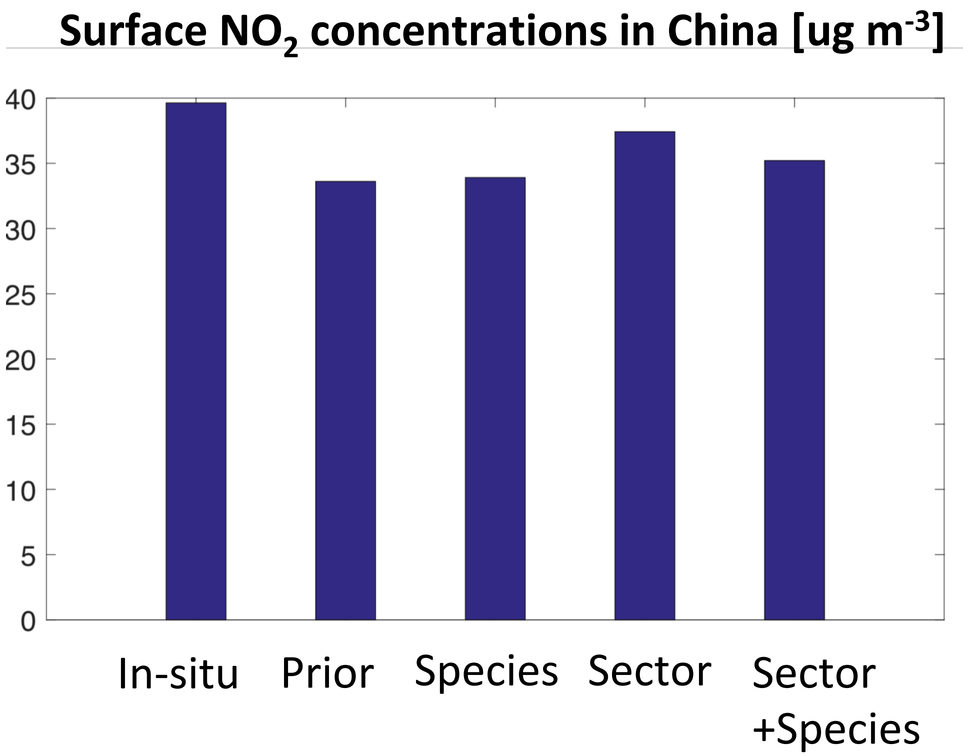
Assimilate:
MOPITT CO
OMI NO_2
OMI SO_2

Similar ratio of NO_x, SO_2 and CO emissions in the same sector, yet very different across sectors.

(*Qu et al.*, in prep)

Evaluations of posterior simulations with measurements





- NMB of posterior simulations from sector-based inversions are 59.8% (SO_2) and 61.4% (NO_2) smaller than the ones from species-based inversion.

Summary

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- Reduced error in NO_x and SO_2 top-down emissions using multiple species joint inversion, through correction of OH concentration in the model, at months when observation uncertainties of optimized species are large.
- A new sector-based inversion is developed to estimate emissions at process level using satellite observations.

Qu et al. (2019a), SO_2 emission estimates using OMI SO_2 retrievals for 2005 – 2017
Qu et al. (2019b), Hybrid mass balance / 4D-Var joint inversion of NO_x and SO_2 emissions in East Asia
Qu et al. (2017), Monthly top-down Nox emissions for China (2005-2012): A hybrid inversion method and trend analysis

Causes of slowdown

- The decreasing relative contributions of gasoline cars, due to the ongoing effectiveness of three-way catalytic converters
- The increasing relative emissions of NO_x from off-road vehicles and industrial, residential, and commercial boilers
- Slower-than expected reductions in emissions by heavy-duty diesel trucks that have newer (and still maturing) catalytic converter technologies

(*Jiang et al*., PNAS, 2018)