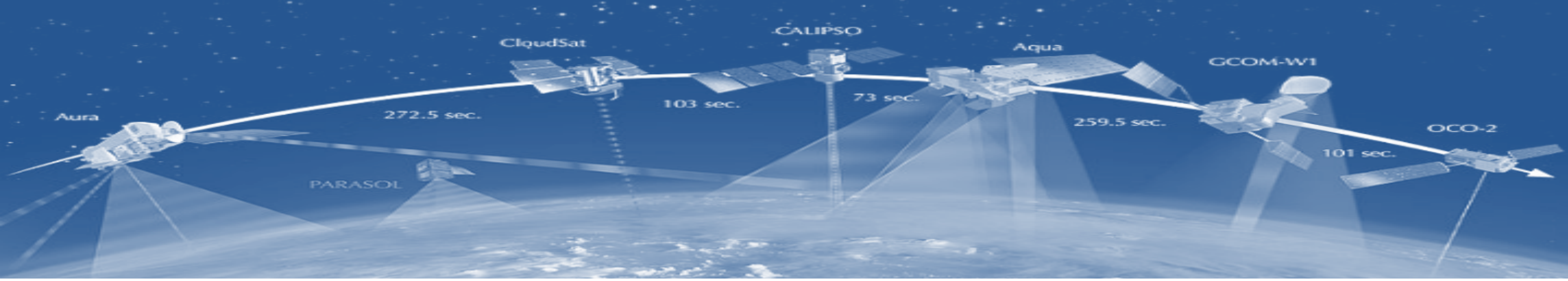

Regional and global long-term emissions constrained by NO_2 and SO_2 satellite observations

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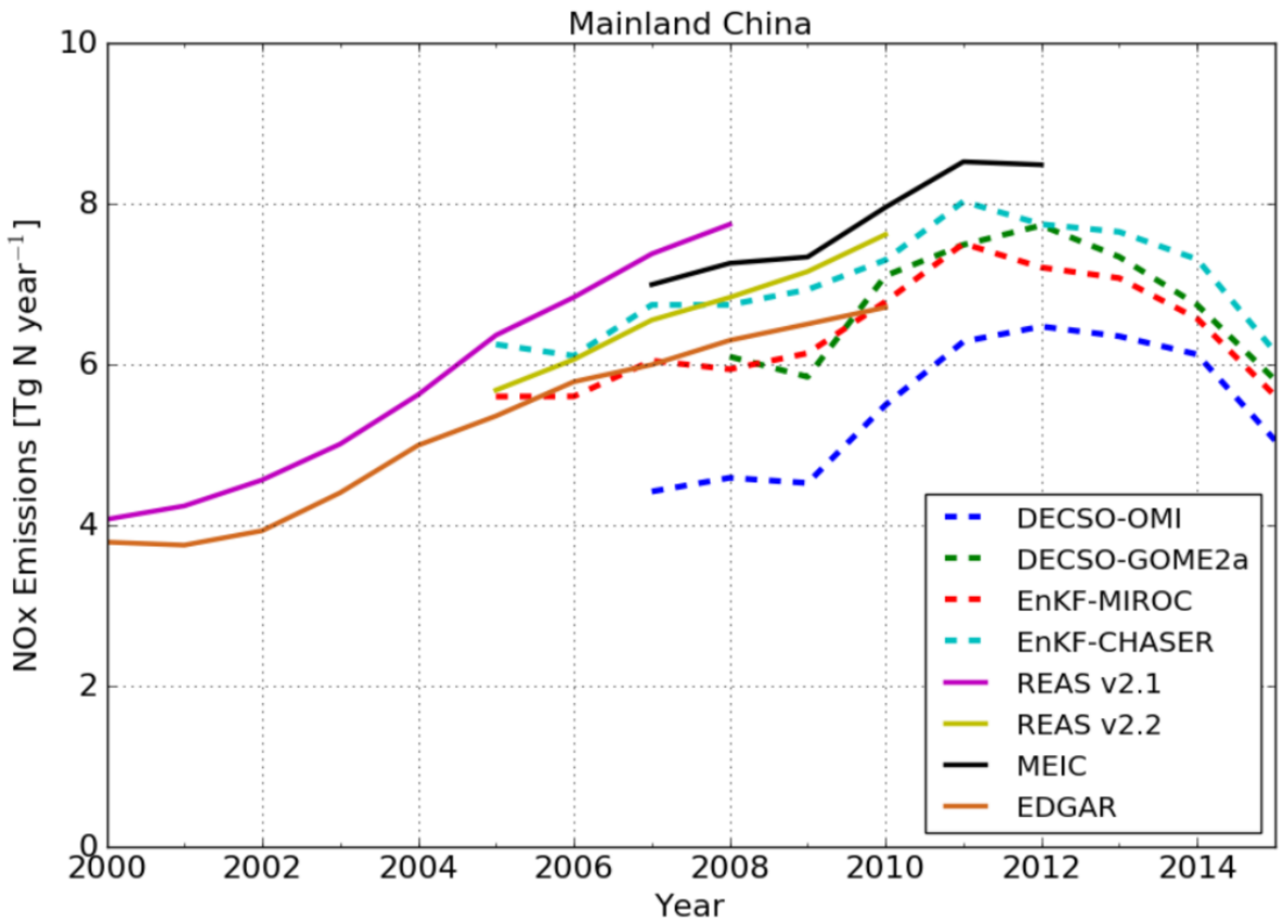
EPA EIC

University of Colorado Boulder

Motivations & Goals

- Long term & high resolution: effectiveness of control strategy
- Bottom-up estimates: uncertainties & long time to compile
- Top-down estimates: satellite & CTM

Recent NO_x trend studies for China



- Comparison of top-down (EKF & EnKF) and bottom up estimates (Ding *et al.*, 2017)

Motivations & Goals

- Long term & high resolution: effectiveness of control strategy
- Bottom-up estimates: uncertainties & long time to compile
- Top-down estimates: satellite & CTM

Kalman filter: large number of nodes and memory
4D-Var: time consuming
Mass balance: nonlinear chemistry & smearing from transport

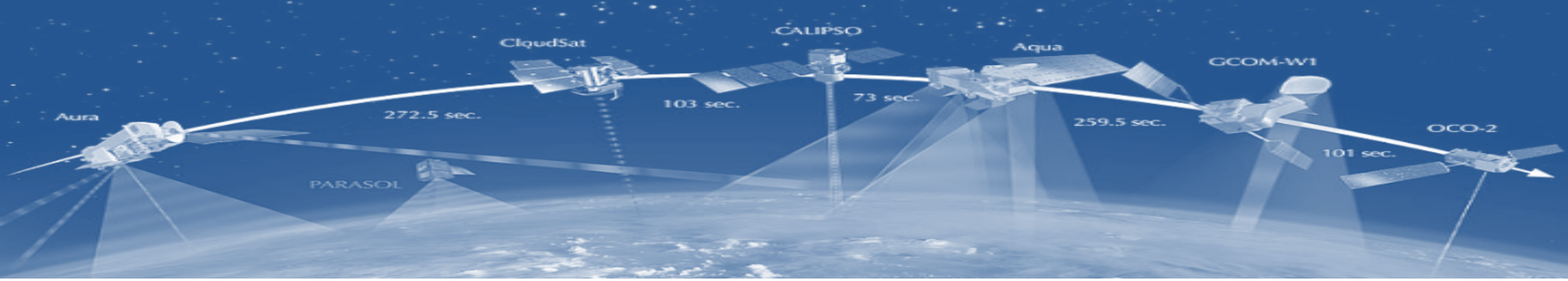
Goal 1: Facilitate long-term inversion of NO_x & SO_2 emissions.

- Chemical interactions

Goal 2: Assimilate multispecies observations and optimization

- Correlated emissions

Goal 3: Sector-based inversion

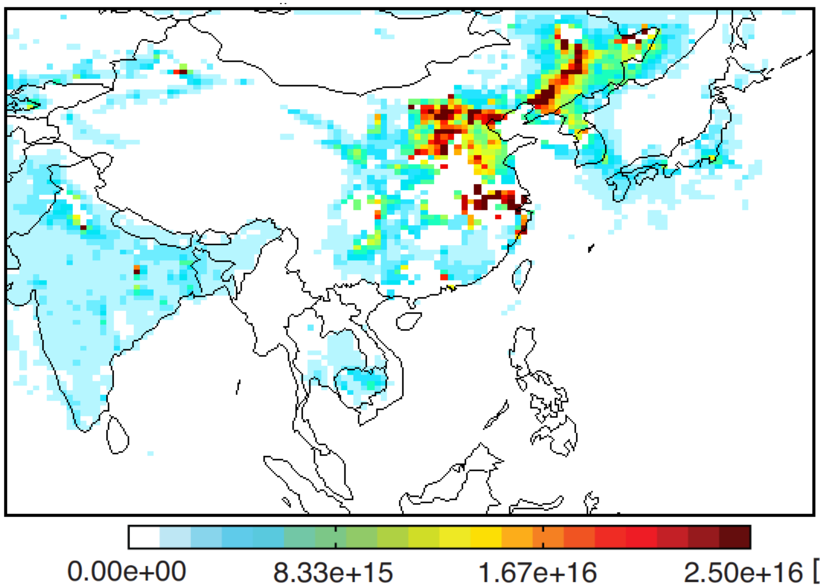


Goal 1: Hybrid inversion

- Method
- Evaluation using pseudo observation test
- Top-down NO_x emissions for China
- Global top-down NO_x emissions

Methods

- **Model:** GEOS-Chem chemical transport model and its adjoint



- Meteorological input from Goddard Earth Observing System (GEOS)
- Simulate chemical reactions, transport of species, convection and deposition
- Resolution: 0.5 latitude x 0.667 longitude, 47 vertical layers (from surface to P = 0.01hPa)
- Domain:0°N ~ 50°N, 70°E ~ 150°E

- Adjoint model adjust emissions at each model grid cell based on observations and bottom-up emissions

(http://www.geos-chem.org)

Methods

- **Observations:**

OMI NO_2

- NASA standard L2 product for China
- Compare standard product with DOMINO retrievals in global inversion

Methods

- **Inversion approaches:**

4D-Var:

- adjust emissions independently in each grid cell
- takes into account transport and chemical reactions
- computationally expansive

Mass balance:

- scale emissions by the ratio of observed & simulated column
- computationally cheap
- limited by nonlinear chemistry and smearing from transport

Hybrid 4D-Var / Mass balance:

- blend of accuracy and efficiency

Hybrid inversion for NO_x

Hybrid method:

Base year (2010): 4D-Var

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

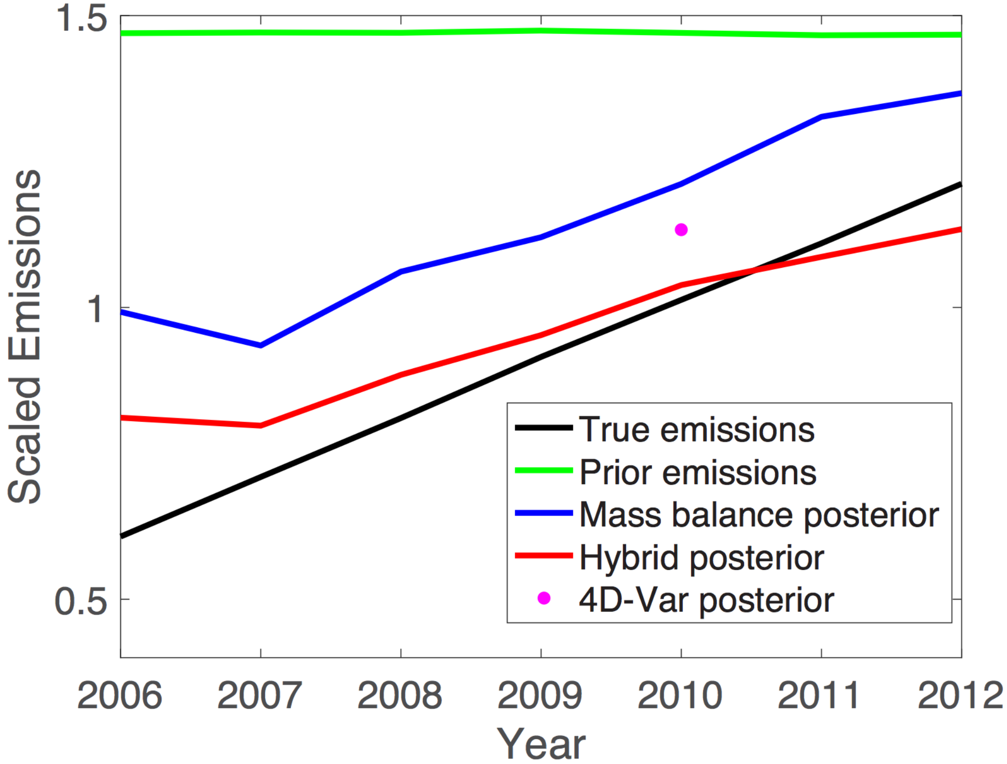
Hybrid inversion for NO_x

Hybrid method:

Base year (2010): 4D-Var

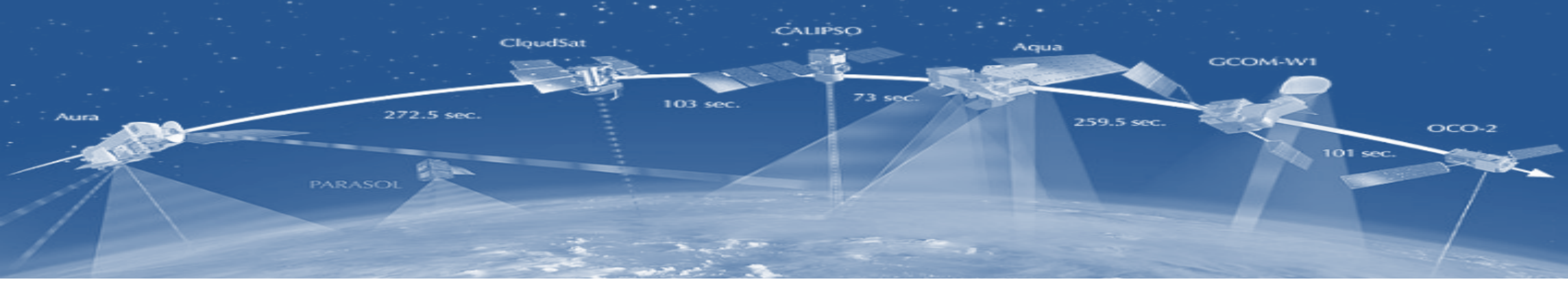
Other years (2005-2012): use 2010 4D-Var posterior for mass balance.

Scaled emissions in pseudo observation test



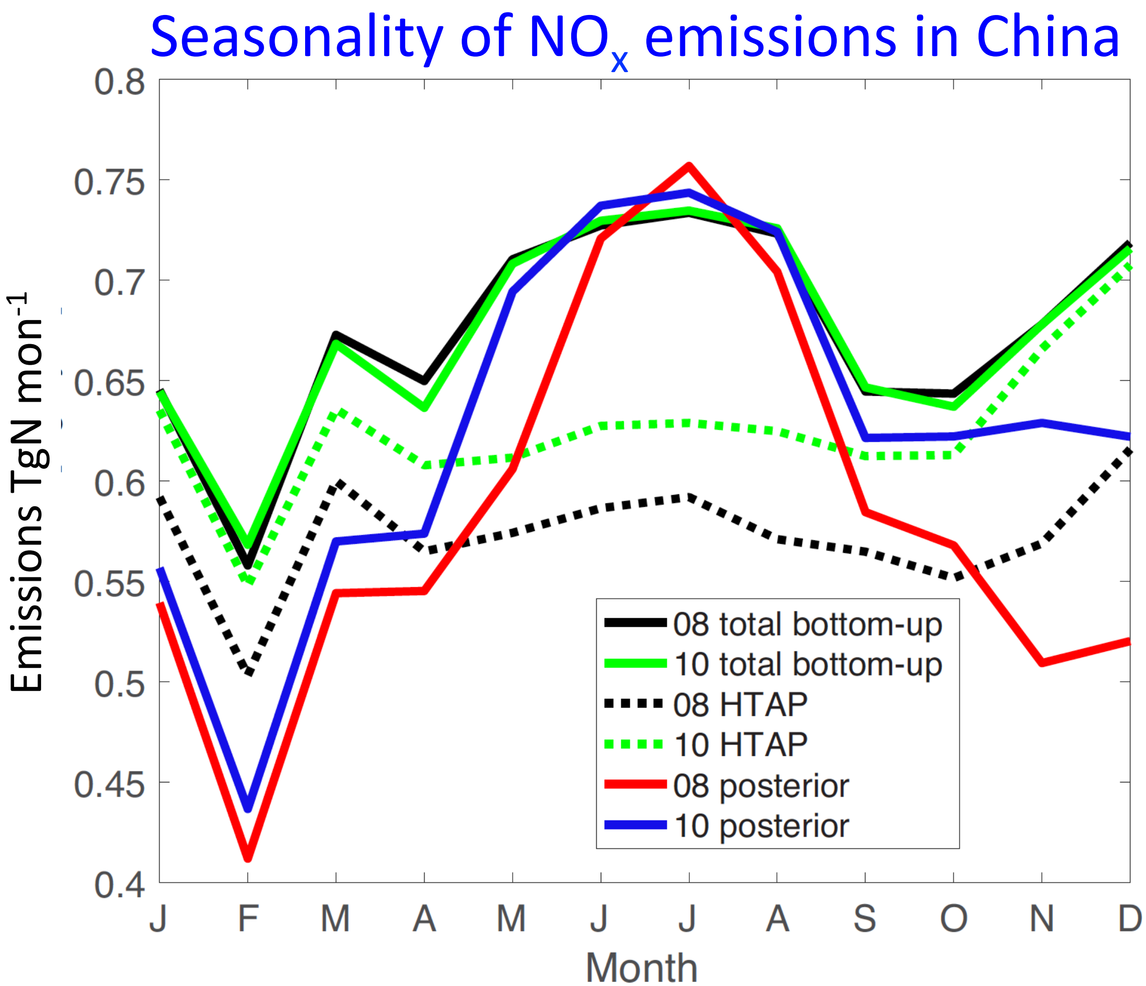
- Hybrid posterior has smaller NMSE (by 59% to 78%) and better correlation.

(Qu *et al.*, JGR, 2017)



Hybrid inversion for NO_x emission in China

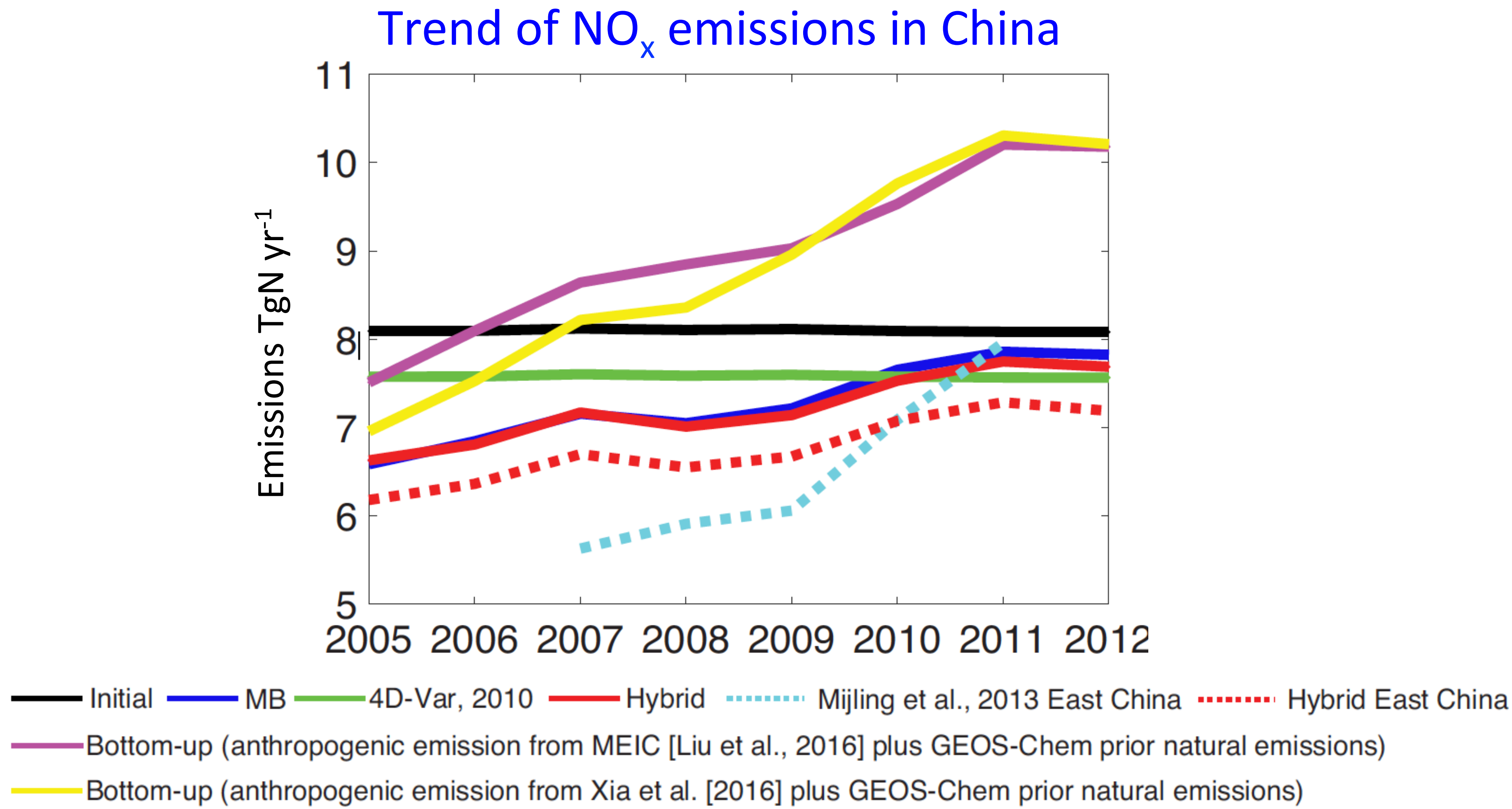
Comparison of hybrid posterior



- Larger seasonality and year-to-year variation in top-down emissions
- Top-down estimates are more consistent with bottom-up emissions in the summer.

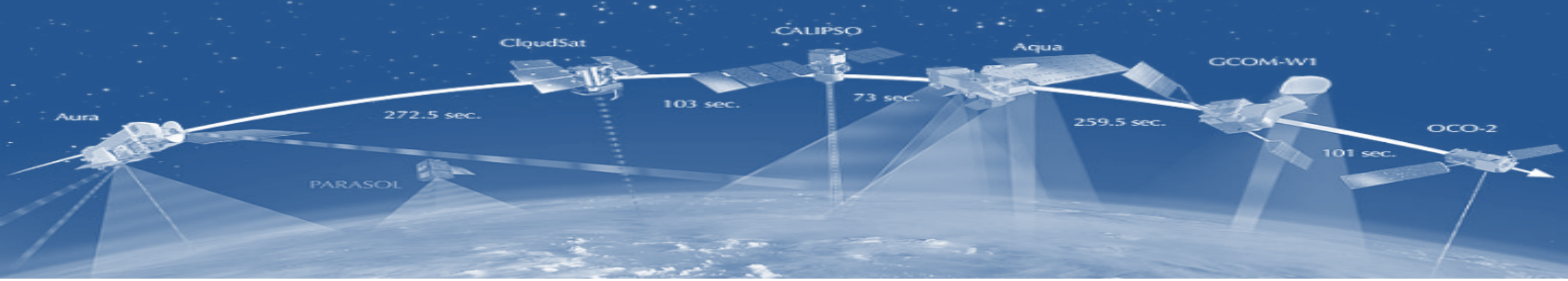
(Qu *et al.*, JGR, 2017)

Comparison of hybrid posterior



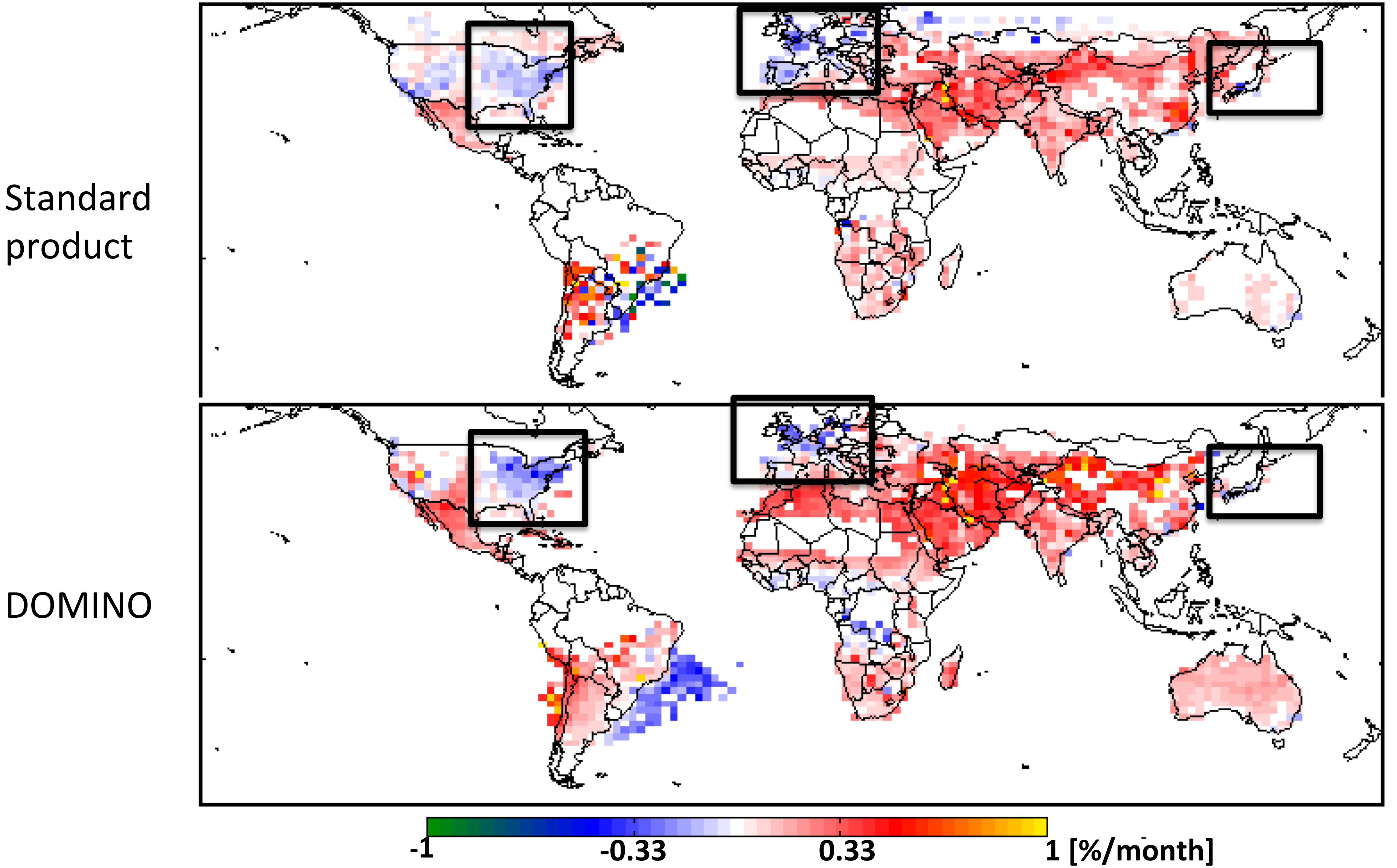
- Top-down estimate has smaller emissions and emission growth rate over China.

(Qu *et al.*, JGR, 2017)



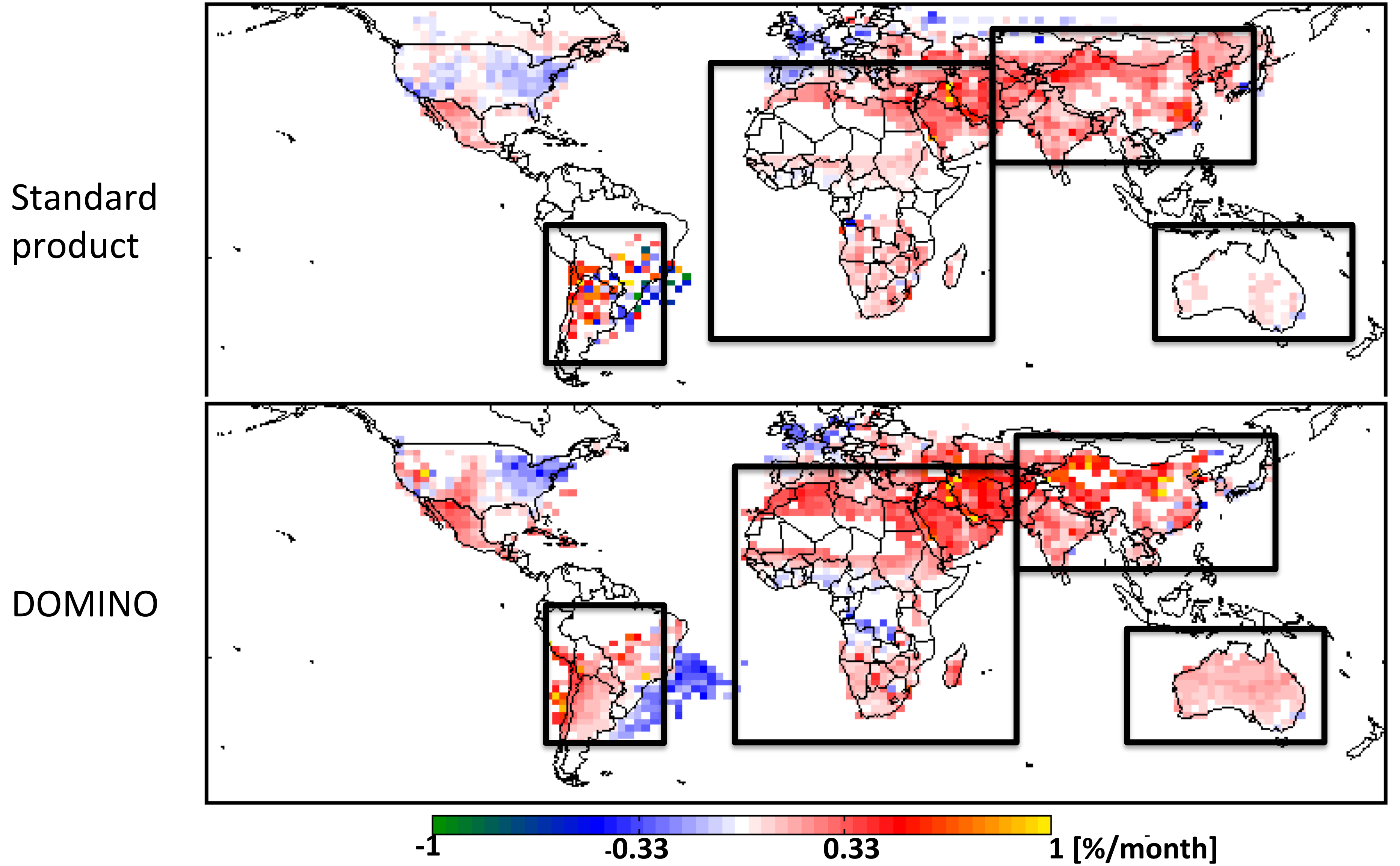
Hybrid inversion for global NO_x

Global NO_2 trend from OMI, 2005-2015, 95%CI



- Decreased NO_2 column density in East US, Europe and Japan

Global NO_2 trend from OMI, 2005-2015, 95%CI

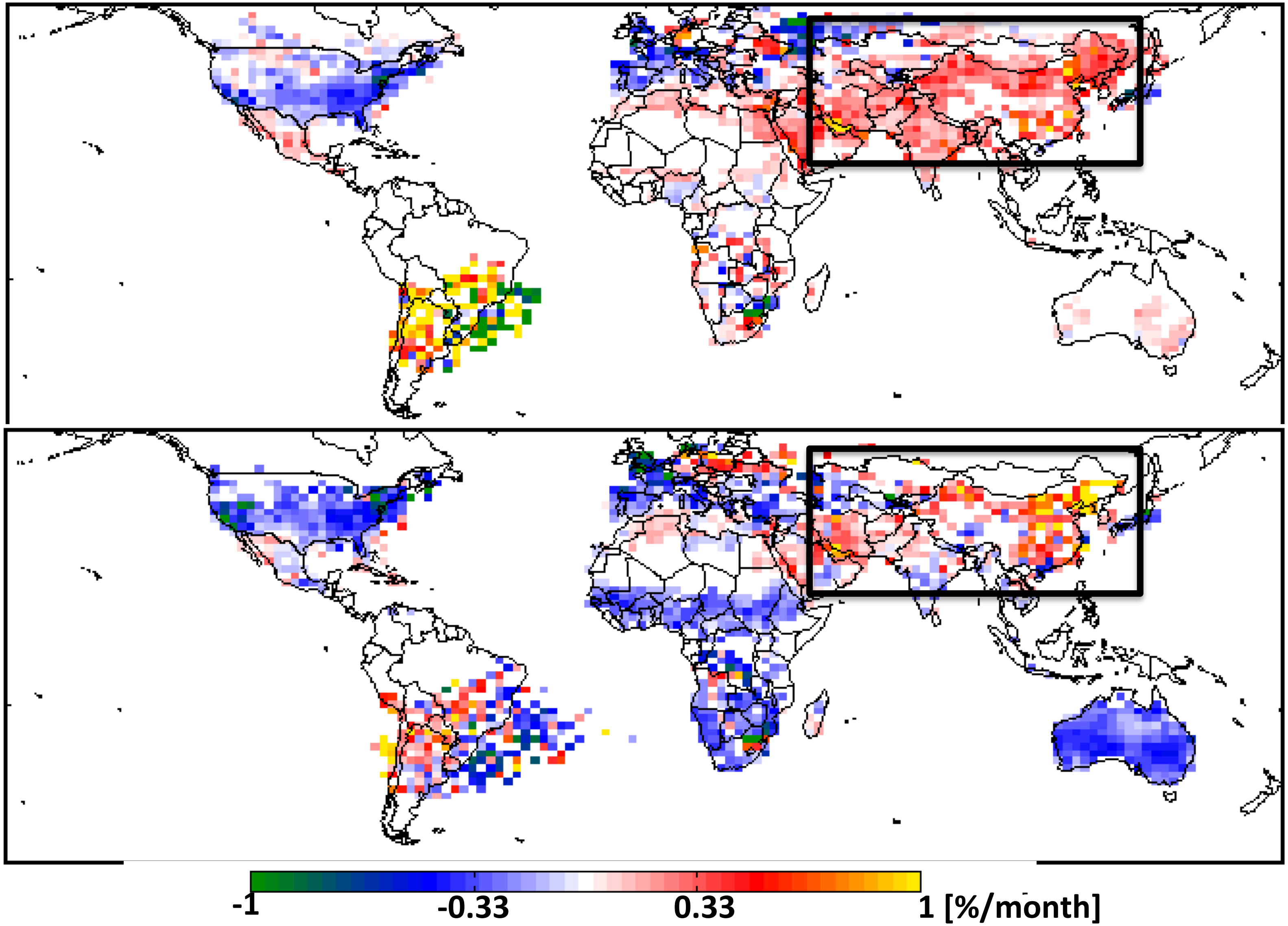


- Decreased NO_2 column density in East US, Europe and Japan
- Increased NO_2 column density in Asia, Australia, Africa and South America

Global NO_2 trend from OMI, 2005-2010, 95%CI

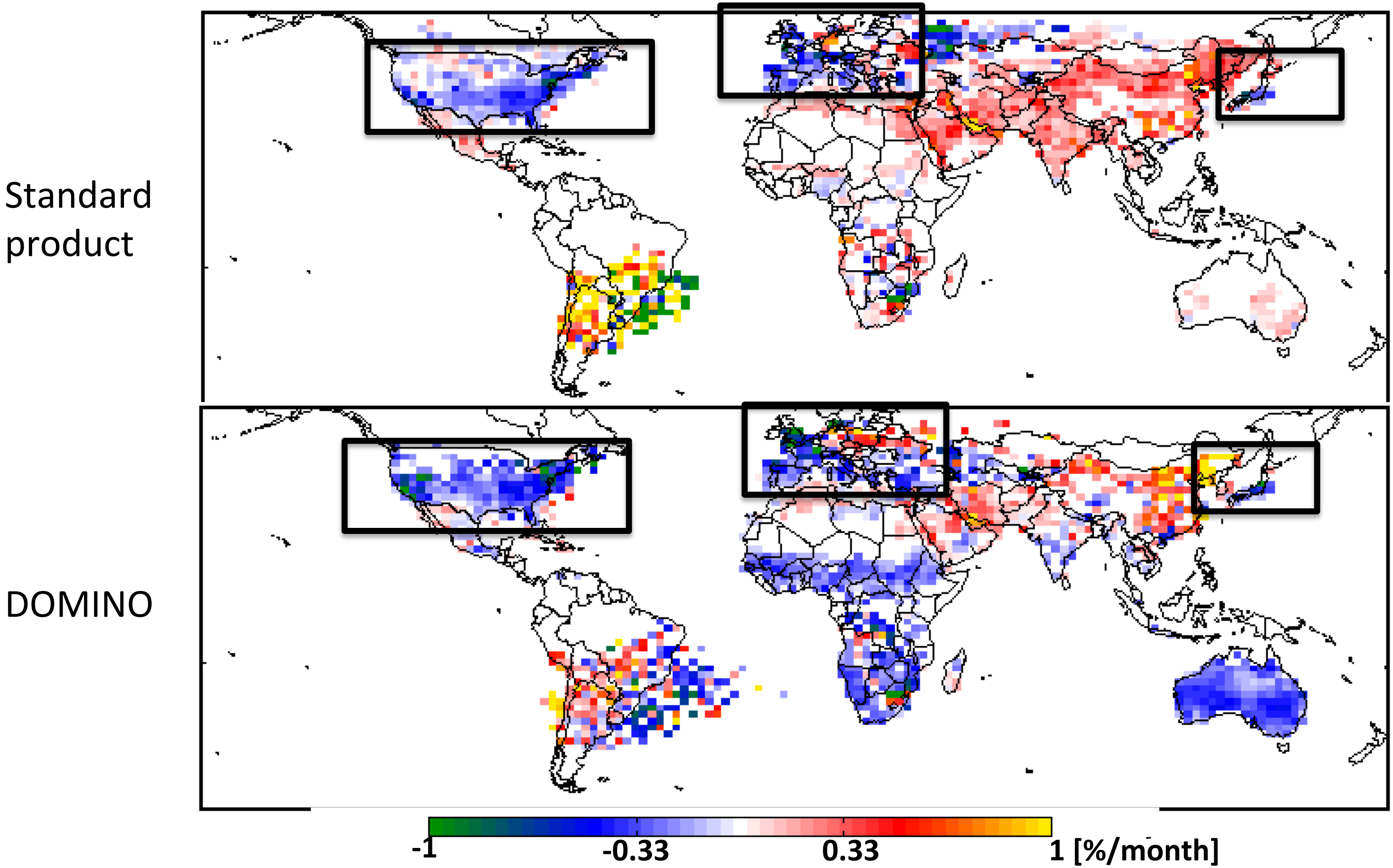
Standard product

DOMINO



- NO_2 column mainly increases in Asia;

Global NO_2 trend from OMI, 2005-2010, 95%CI



- NO_2 column mainly increases in Asia; decrease in US, Europe and Japan

Global NO_2 trend from OMI, 2005-2010, 95%CI

Standard product

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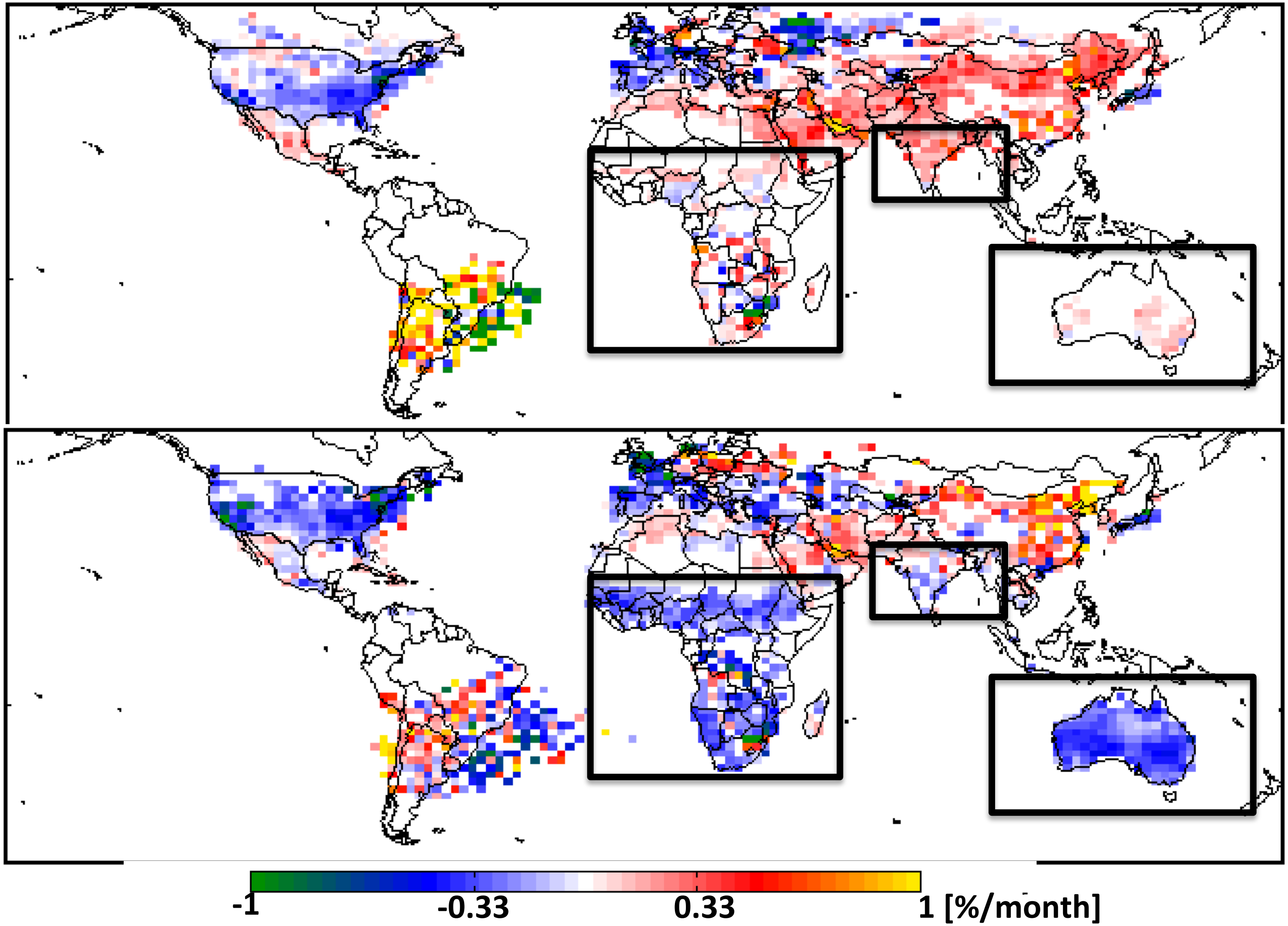
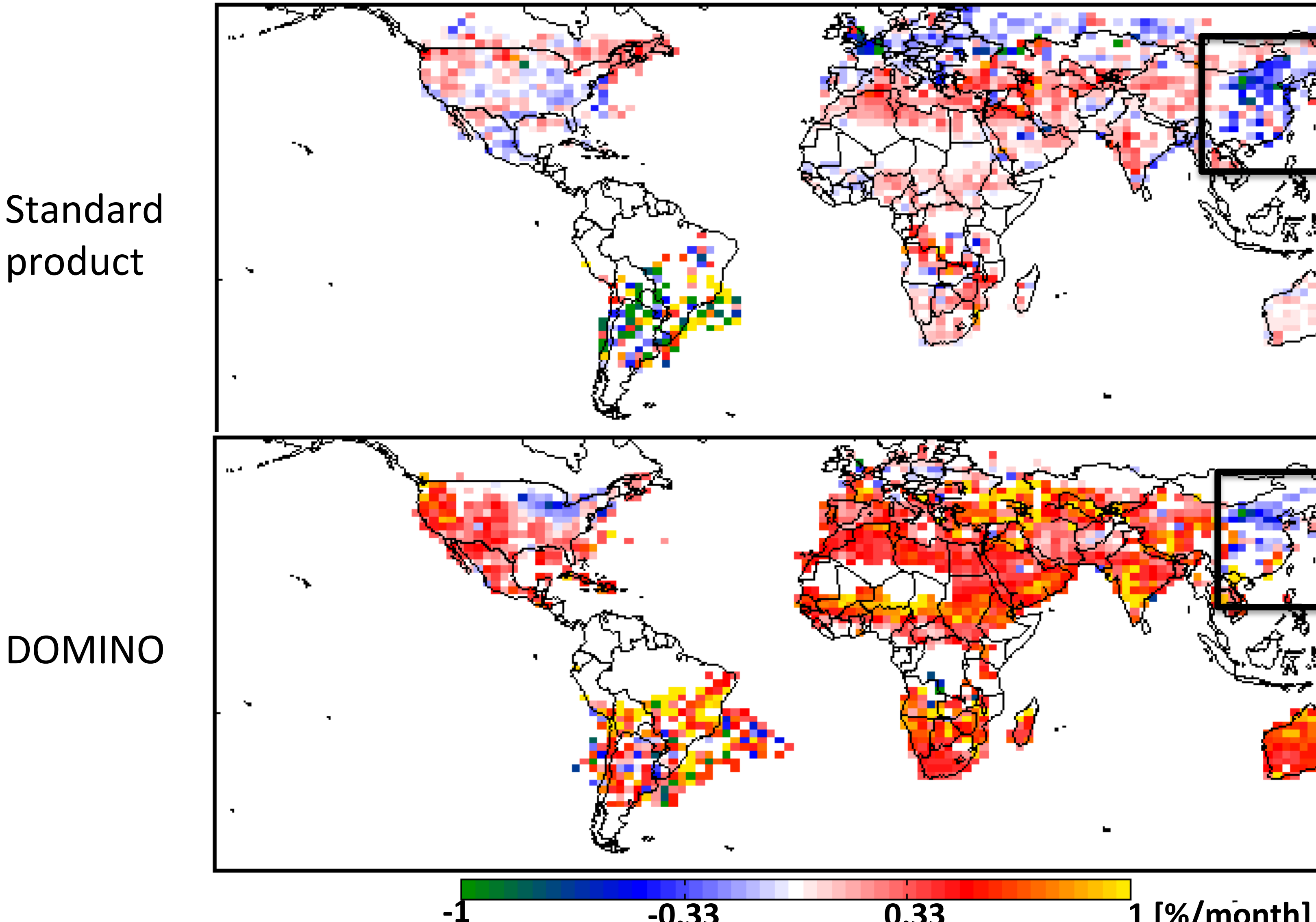


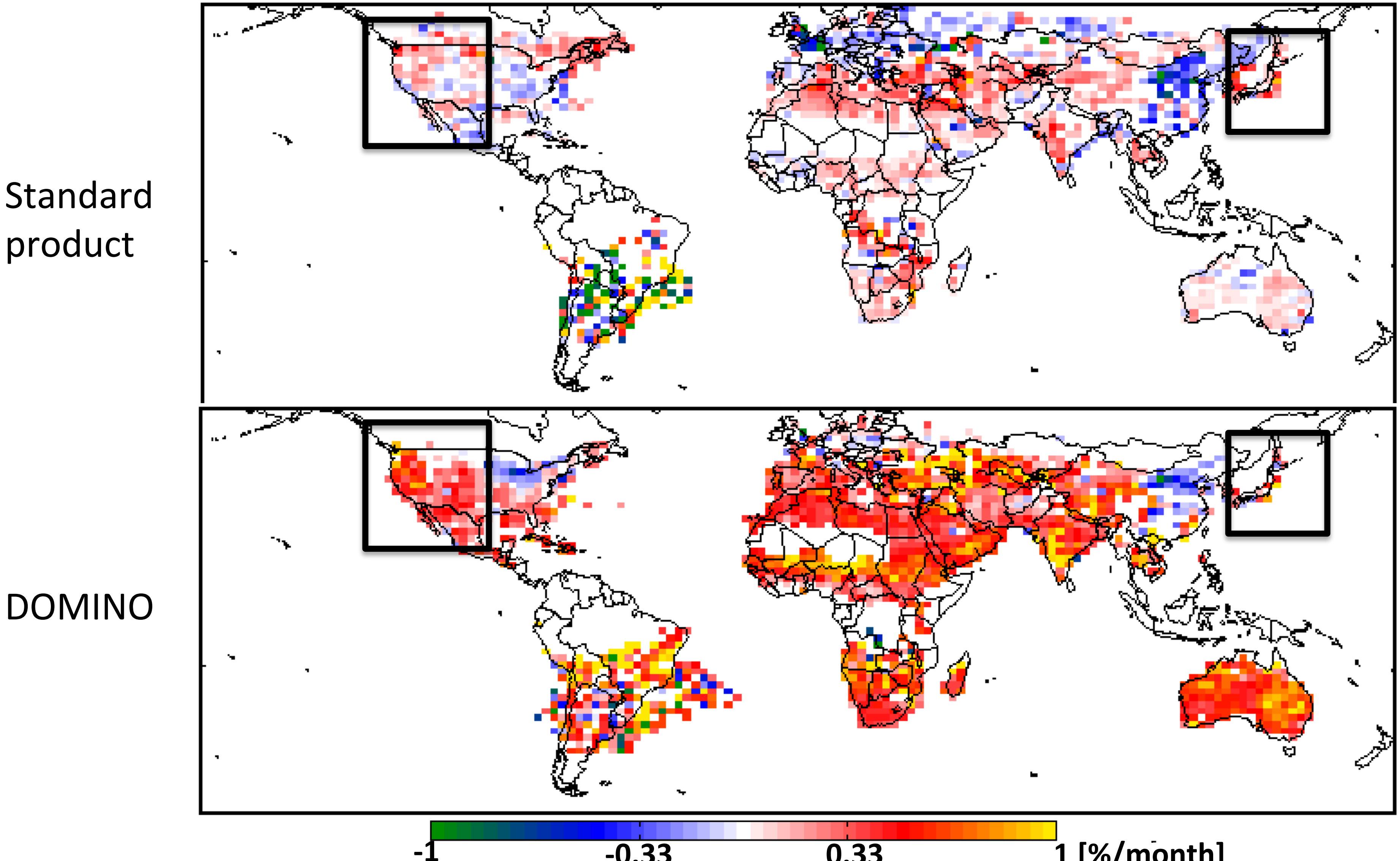
- NO_2 column mainly increases in Asia; decrease in US, Europe and Japan
- Different trend in Australia, south India and Africa

Global NO_2 trend from OMI, 2010-2015, 95%CI



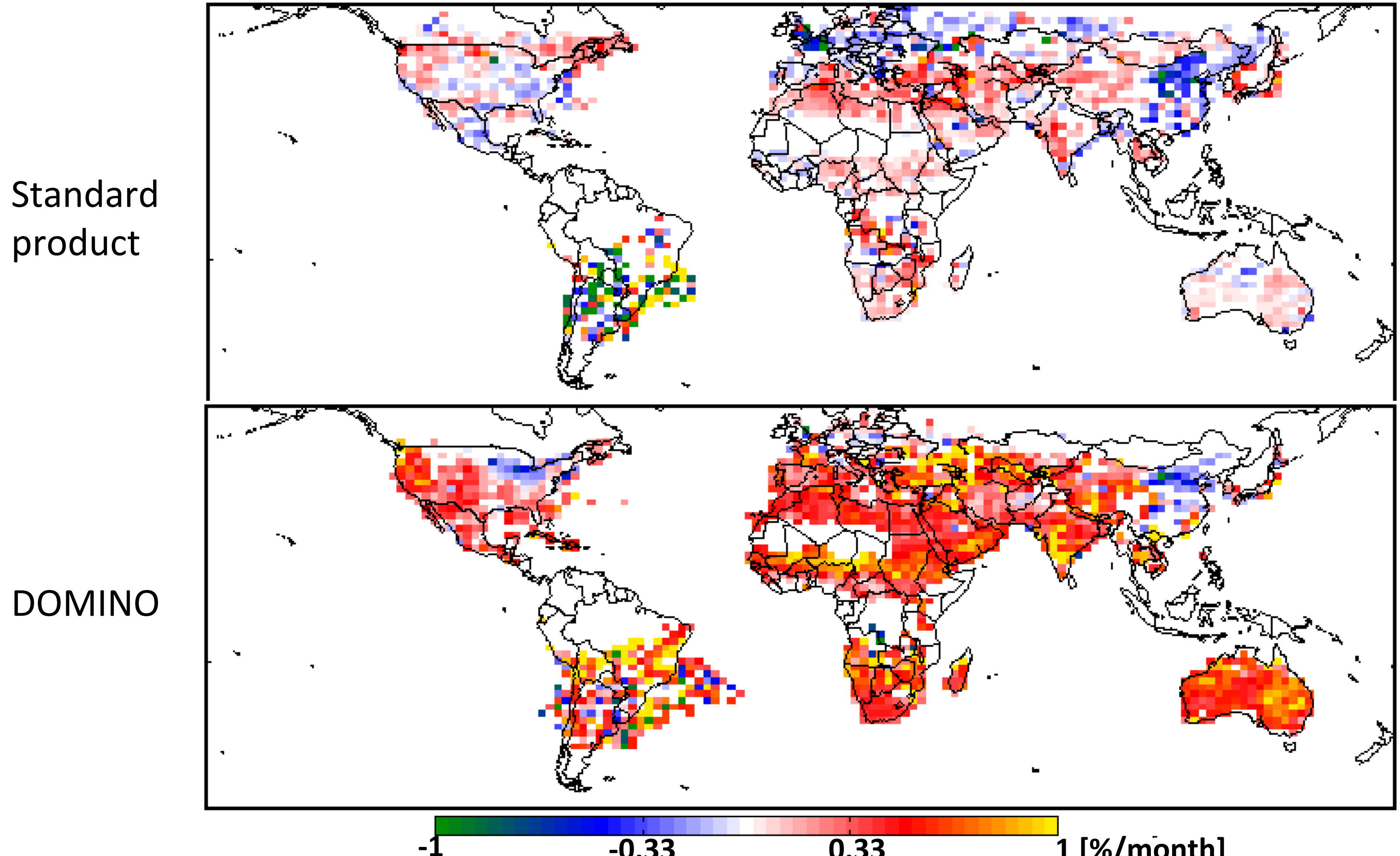
- NO_2 column decrease in East China;

Global NO_2 trend from OMI, 2010-2015, 95%CI



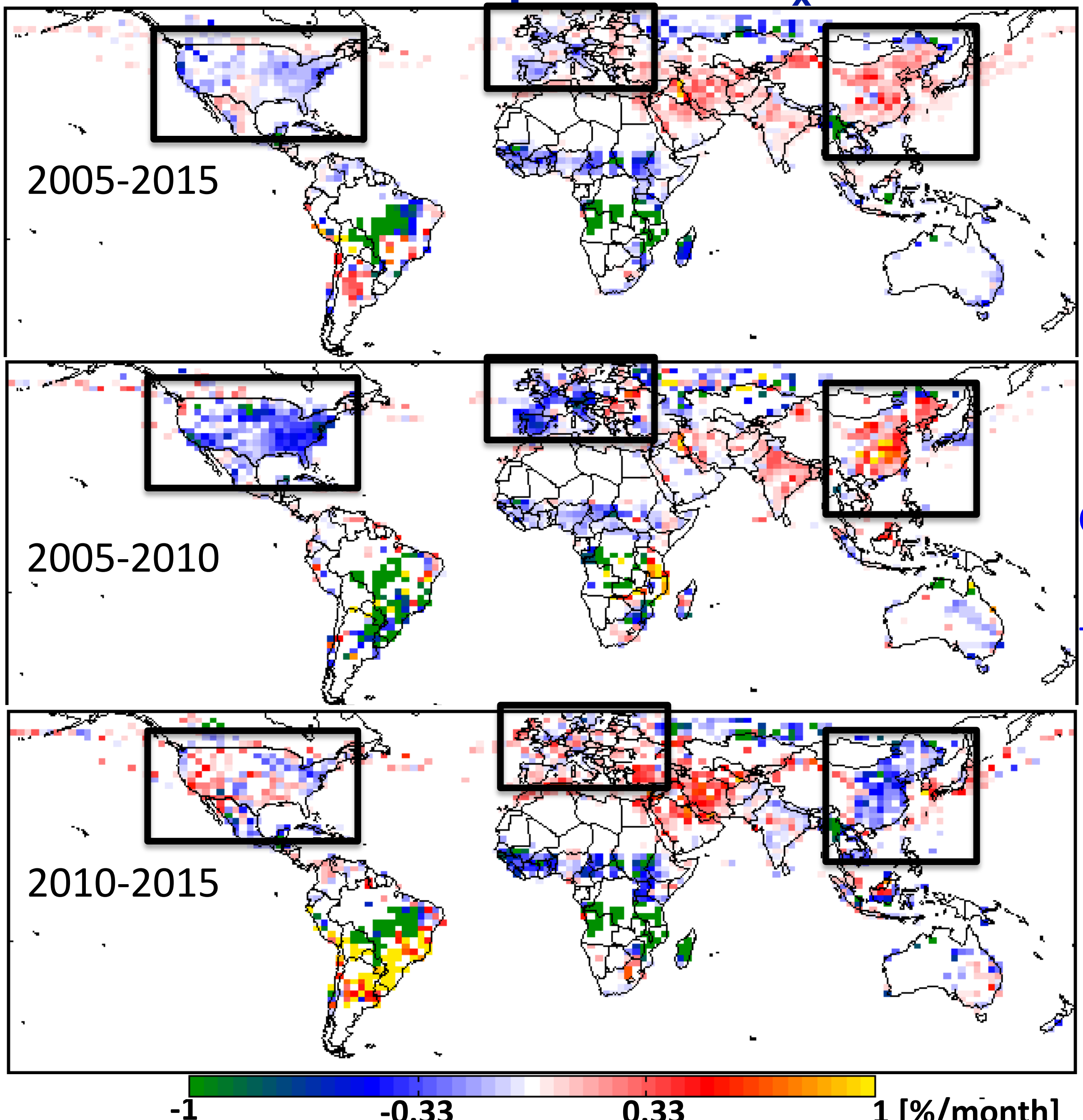
- NO_2 column decrease in East China; increase in west US, Japan

Global NO_2 trend from OMI, 2010-2015, 95%CI



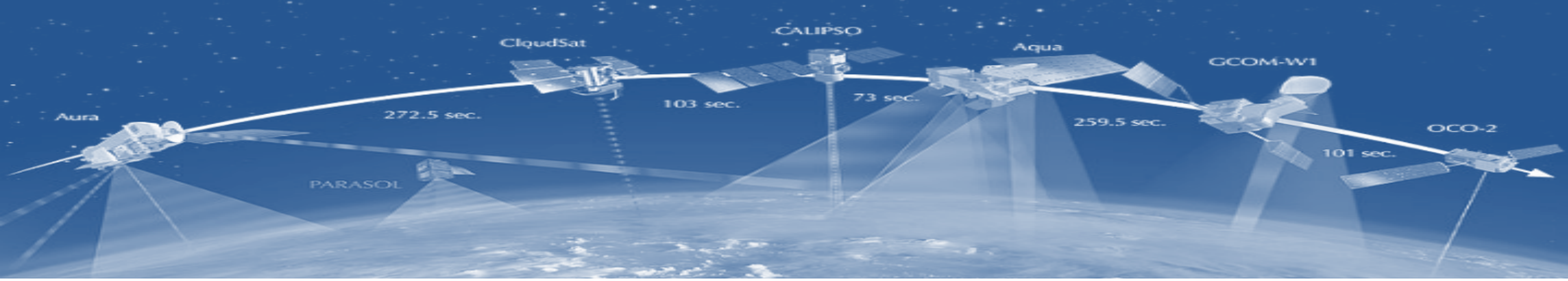
- NO_2 column decrease in East China; increase in west US, Japan
- Larger increasing trend from DOMINO retrieval
- Trends: robust versus retrieval specific

Trend of top-down NO_x emissions, SP, 95%CI



Compare w/ column trend:

-Similar in US, Europe, China

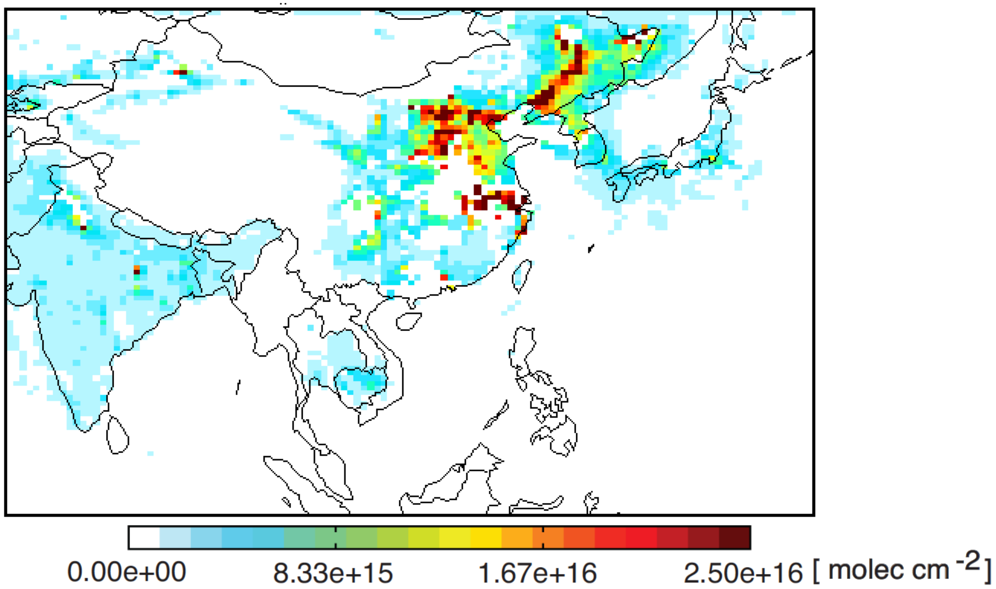


Goal 2: Assimilate & optimize multispecies

- Method
- Evaluation using pseudo observation test
- 4D-Var & hybrid inversion
- Comparison with in-situ measurements

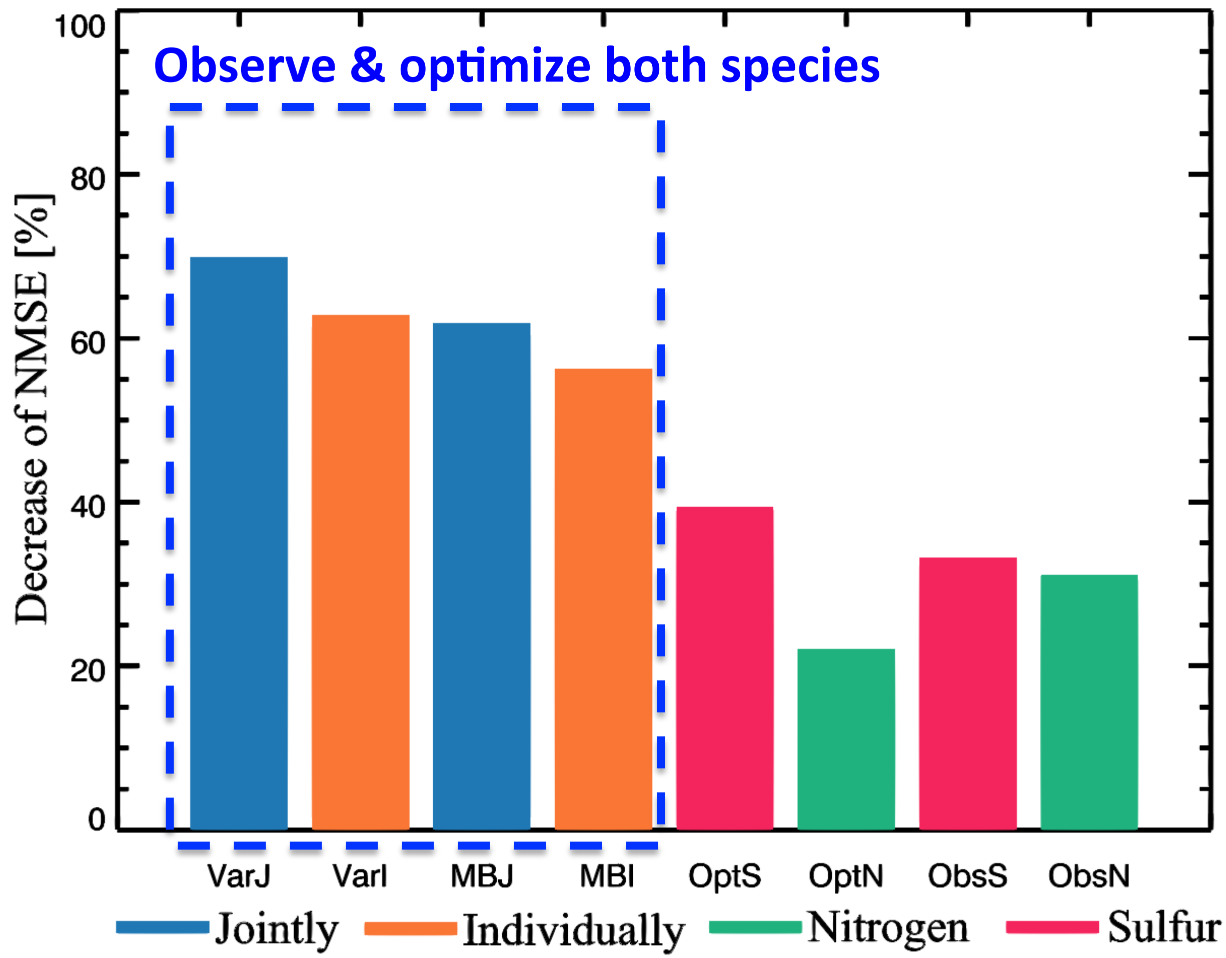
Model & Observations

- **Model:** GEOS-Chem adjoint, nested China, 0.5° x 0.667° .



- **Observations:**
  OMI NO_2 & SO_2, NASA standard L2 product

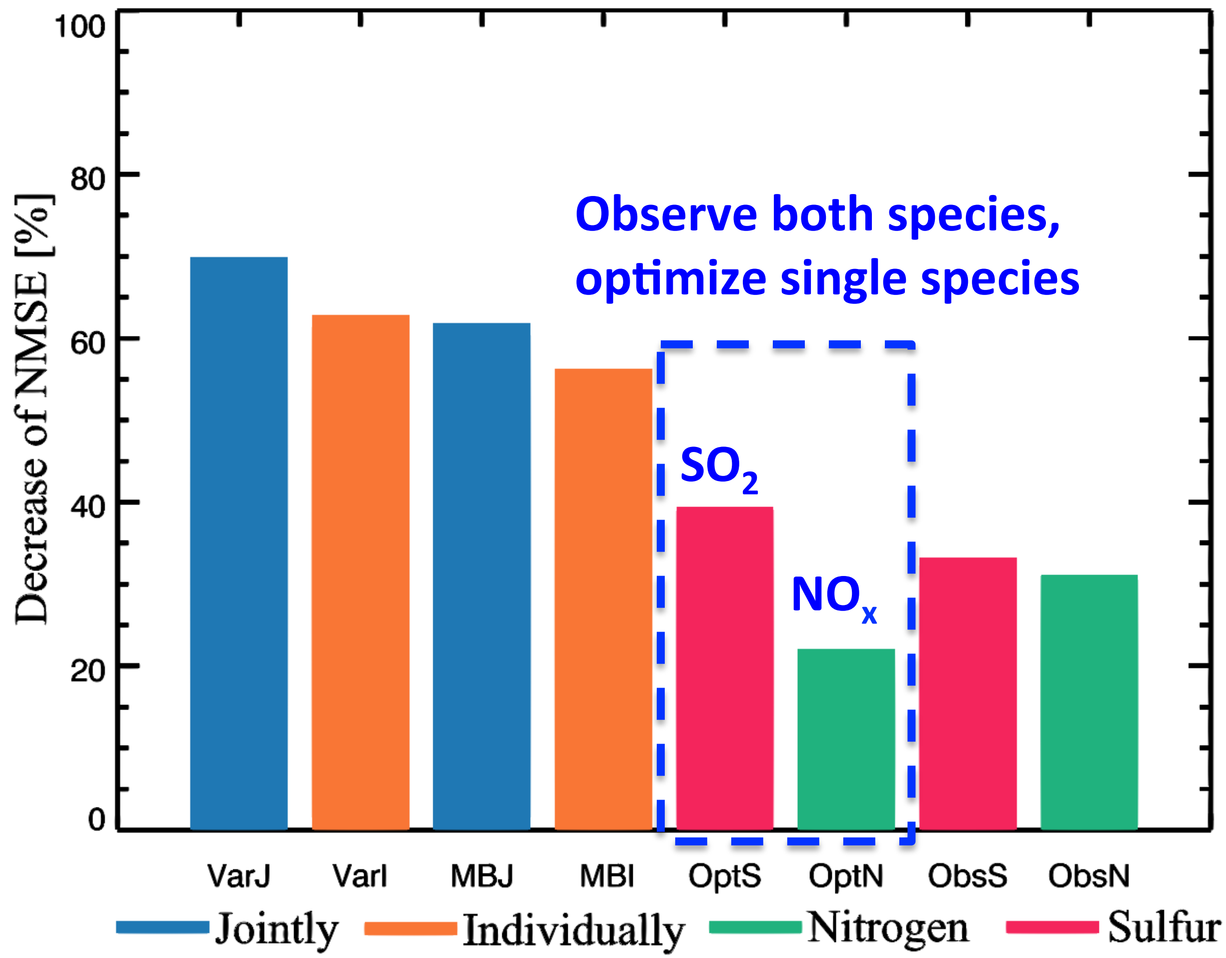
Pseudo observation test: multispecies optimization



VarJ: 4D-Var jointly
VarS: Sum of two individual 4D-Var
MBJ: Mass balance jointly
MBS: Sum of two individual mass balance
OptS: Optimize SO_2 emissions
OptN: Optimize NO_x emissions
ObsS: Observe SO_2 column
ObsN: Observe NO_2 column

- Better performance of joint 4D-Var (by 7.1%) and mass balance (by 5.6%) than single species inversion

Pseudo observation test: multispecies optimization



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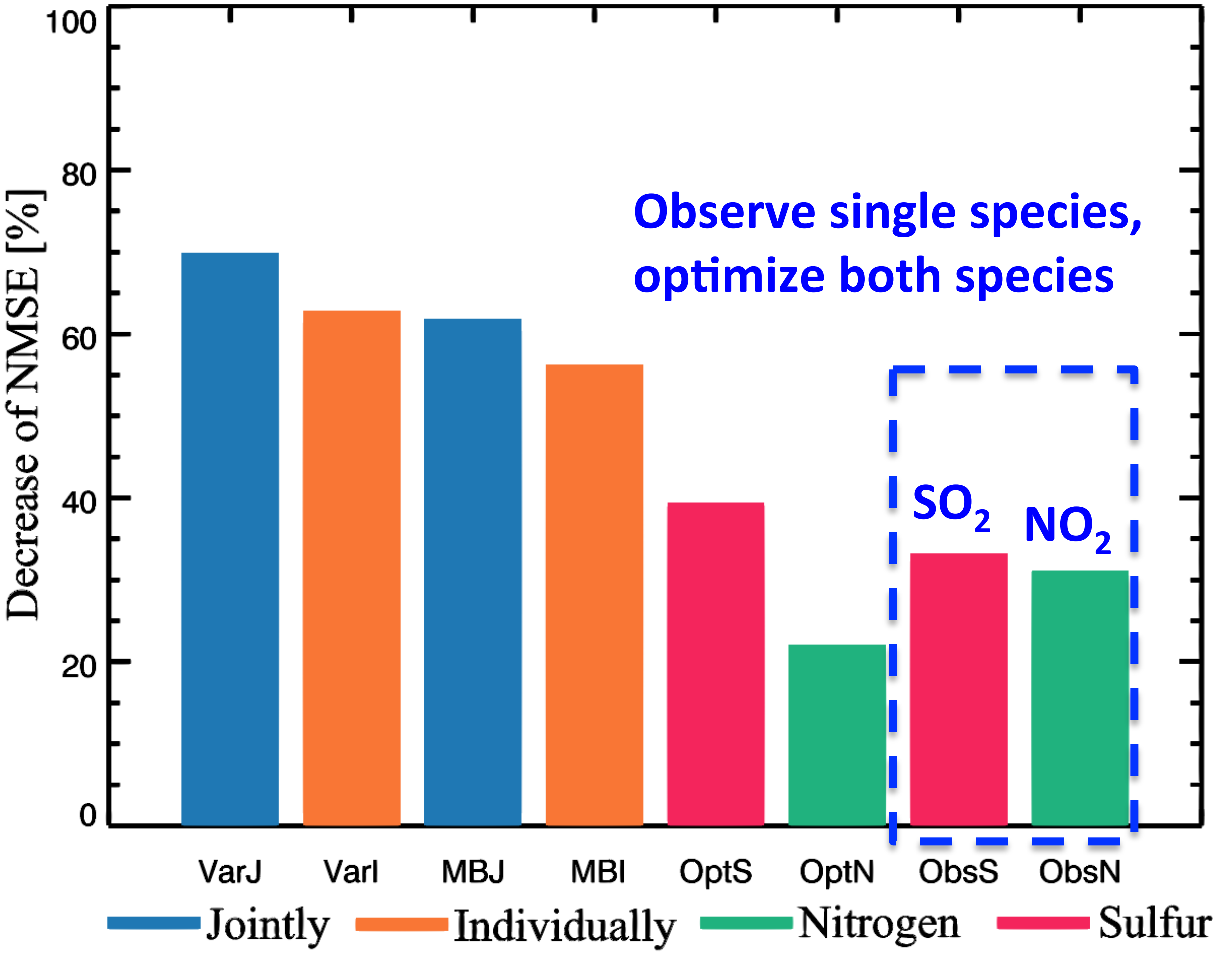

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ObsS: Observe SO_2 column
ObsN: Observe NO_2 column

- Better performance of joint 4D-Var (by 7.1%) and mass balance (by 5.6%) than single species inversion
- Largest decrease of NMSE if observe and optimize both species at the same time

Impact of multispecies observations on posterior emissions

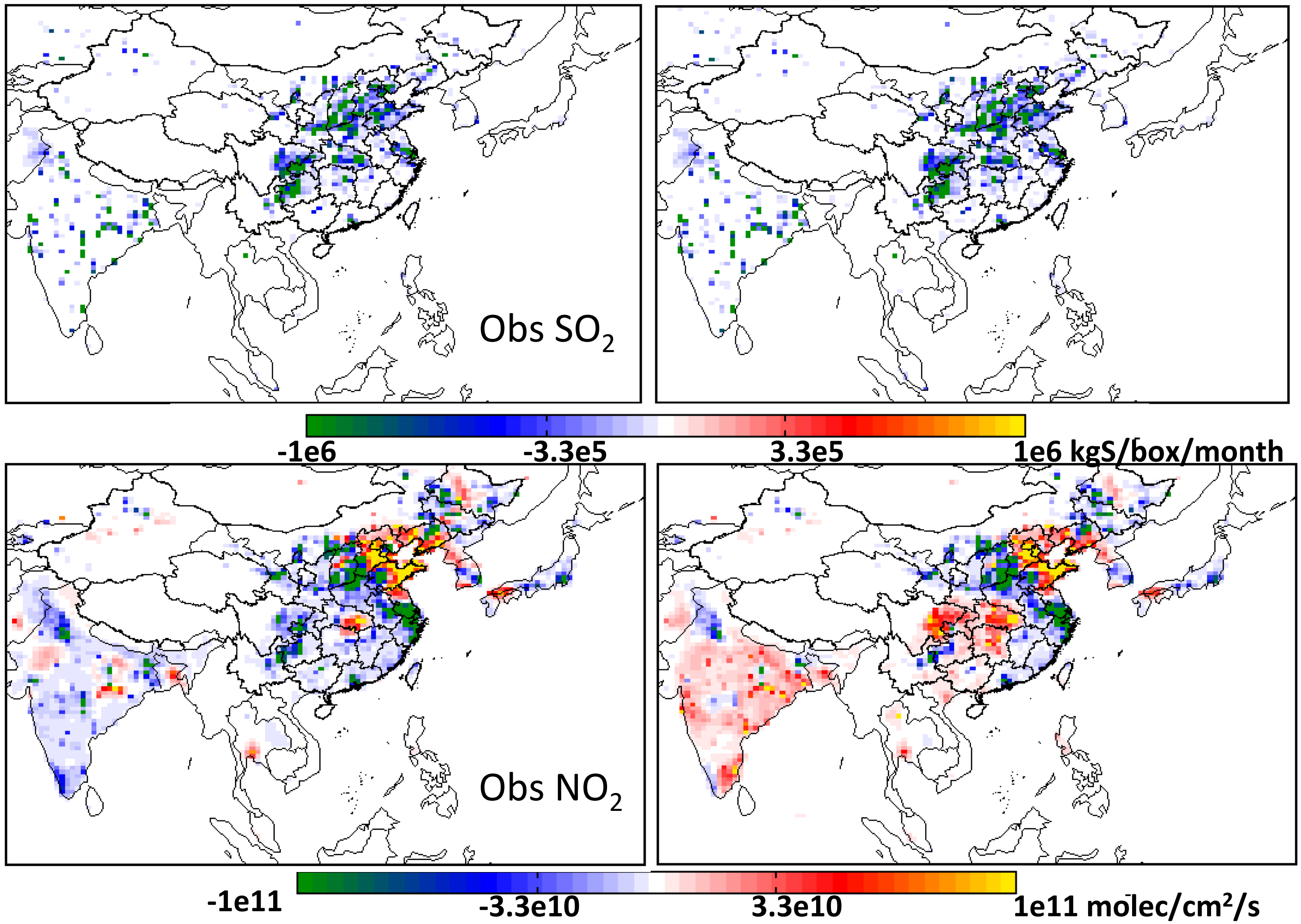
Step 1: 4D-Var

Jan, 2010

Single species observations | NO_2 & SO_2 observations

Posterior–prior SO_2

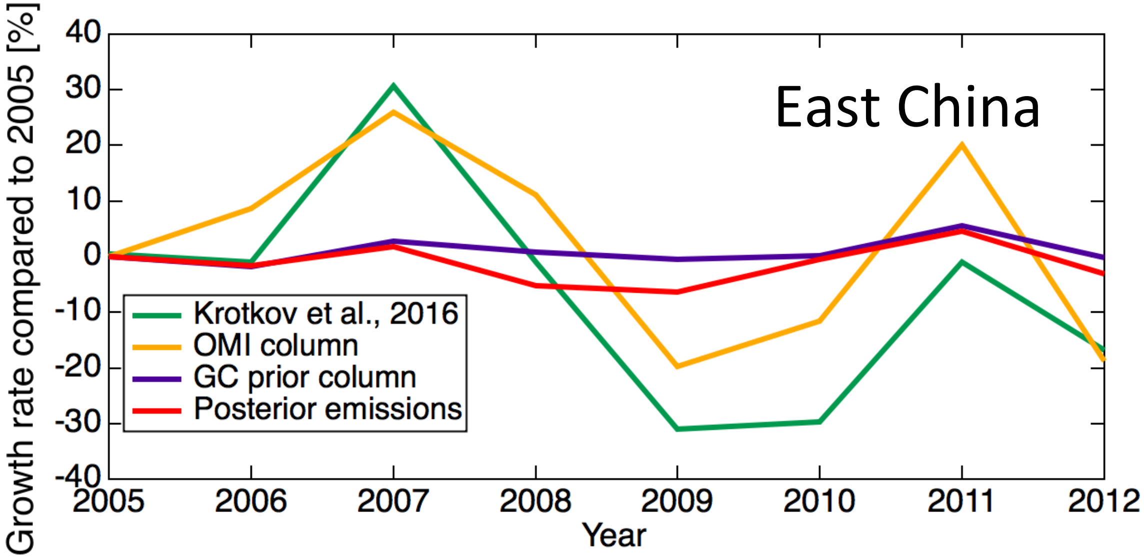
Posterior–prior NO_x



- Differences occur mostly in low emission regions.
- Changes of SO_2 emissions (70%) have little effect on NO_2 concentration (< 5%), so incorporating NO_2 observation does not change SO_2 emissions much.
- In remote region, increase of NO_x leads to increase of OH, and decrease of SO_2 column.

Top-down SO_2 emissions

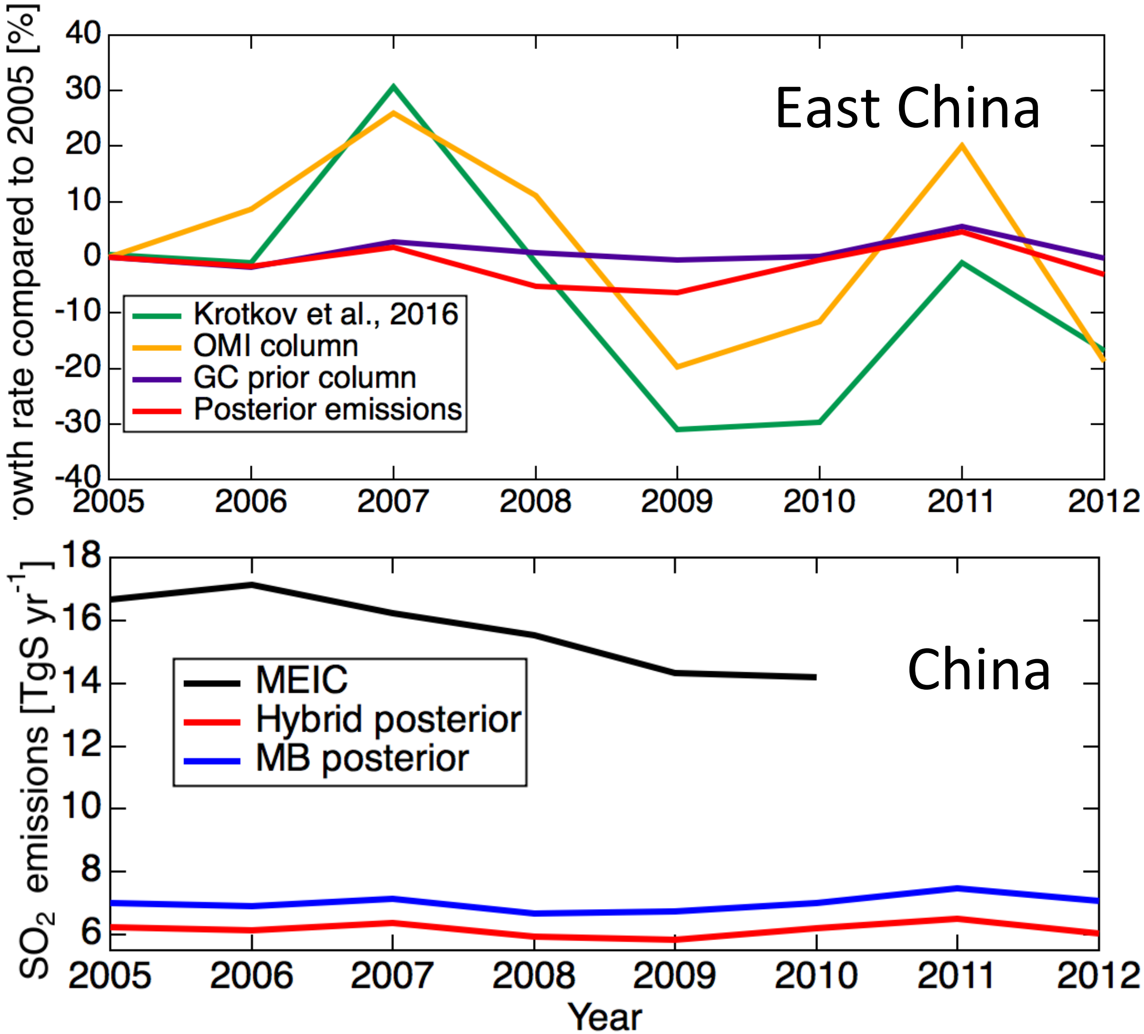
Step 2: MB for trend



- Trend of emissions and column densities are different; meteorology lead to 7% changes of SO_2 SCD at national level

Top-down SO_2 emissions

Step 2: MB for trend

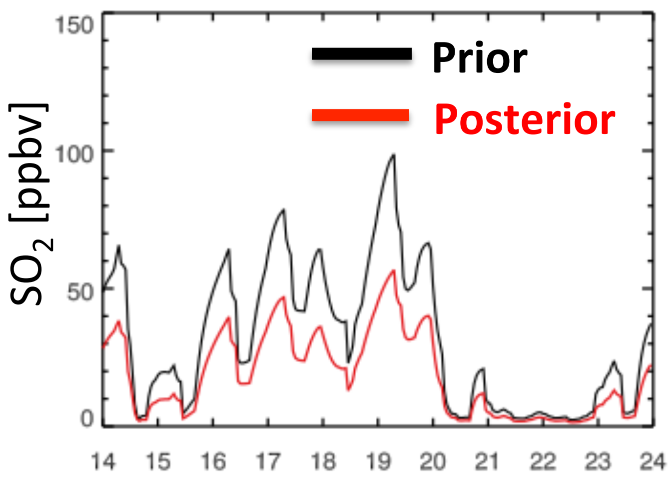


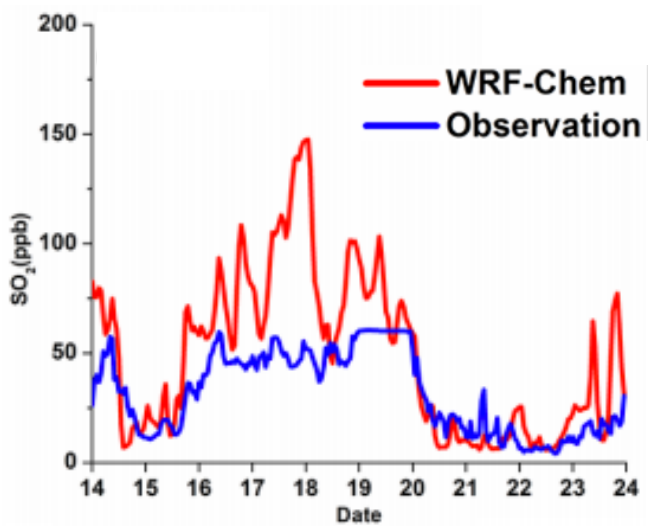
- Trend of emissions and column densities are different; meteorology lead to 7% changes of SO_2 SCD at national level
- Top-down emissions is lower than bottom-up estimate by ~100%

Evaluation of SO_2 emissions using in-situ measurements

China, Jan, 2010

Beijing



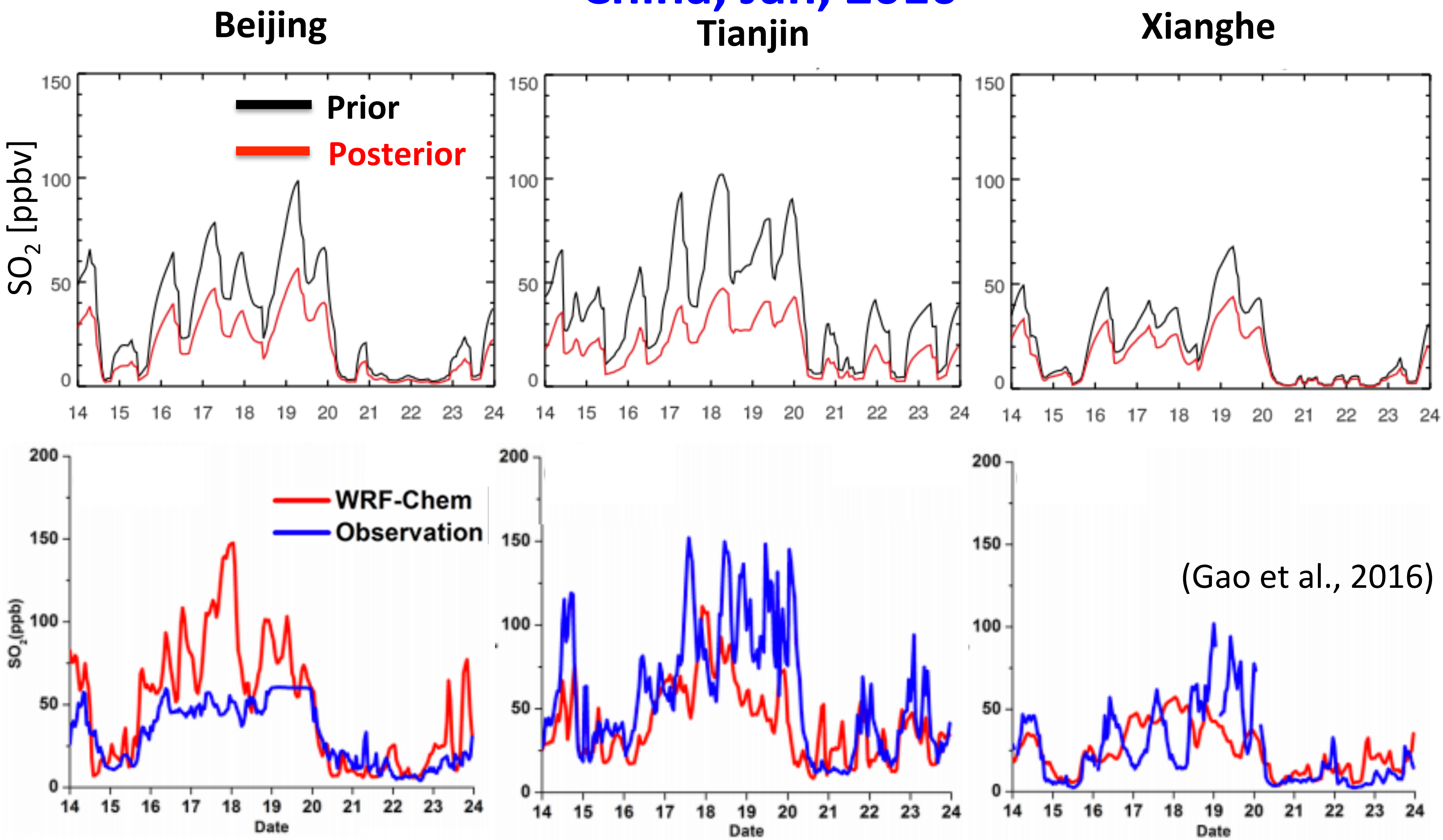


(Gao et al., 2016)

- GEOS-Chem timeseries is relatively consistent with observations

Evaluation of SO_2 emissions using in-situ measurements

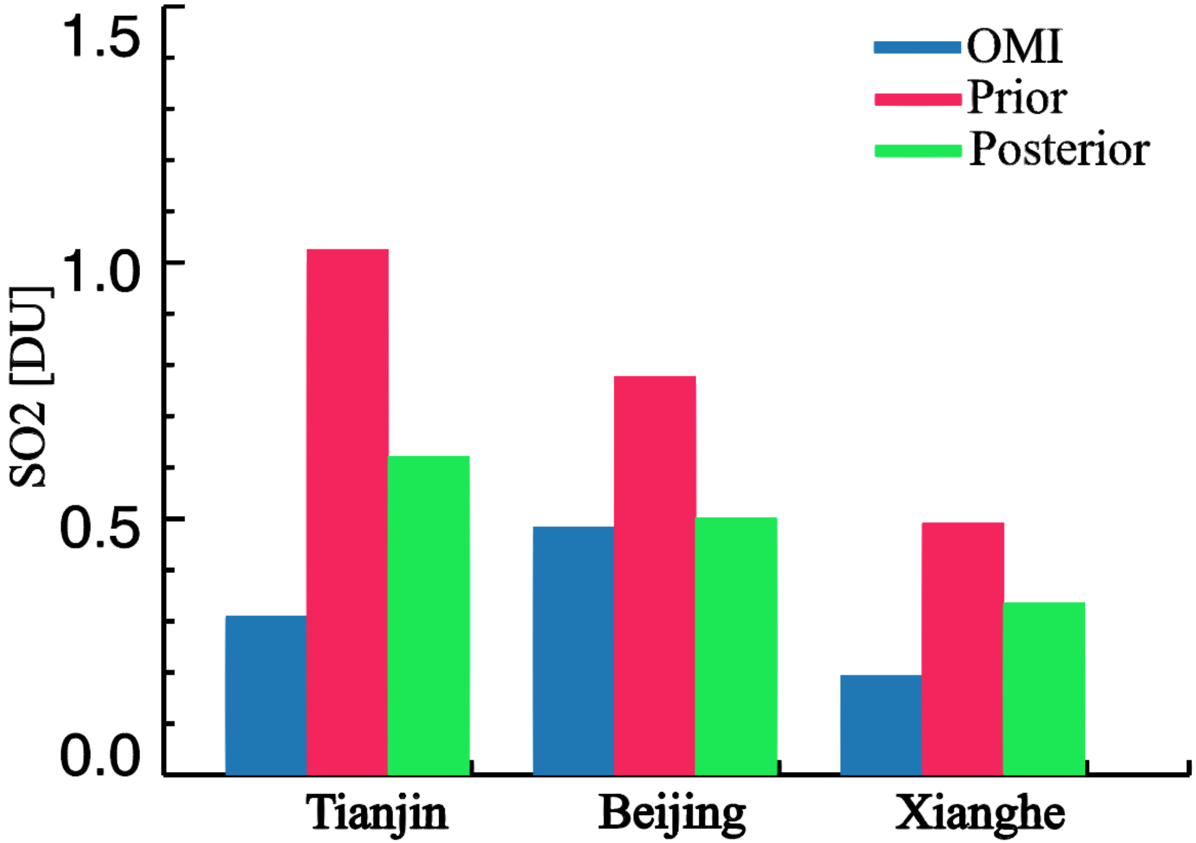
China, Jan, 2010



- GEOS-Chem timeseries is relatively consistent with observations
- GC SO_2 is lower in Tianjin & Xianghe, but similar with in-situ measurements in Beijing

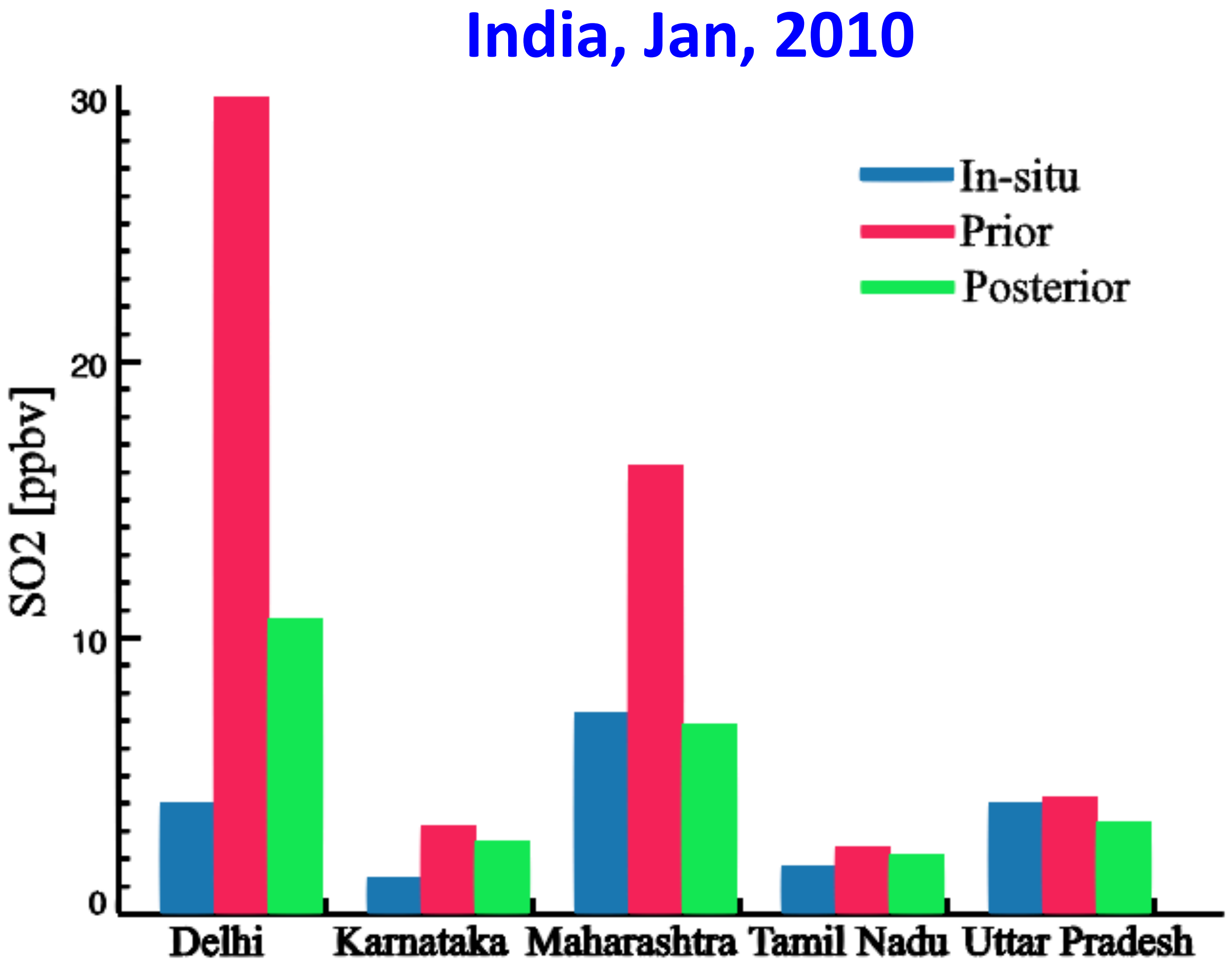
Evaluation of SO_2 emissions using observations

China, Jan, 2010



- GEOS-Chem SO_2 column bias high compared to OMI observation

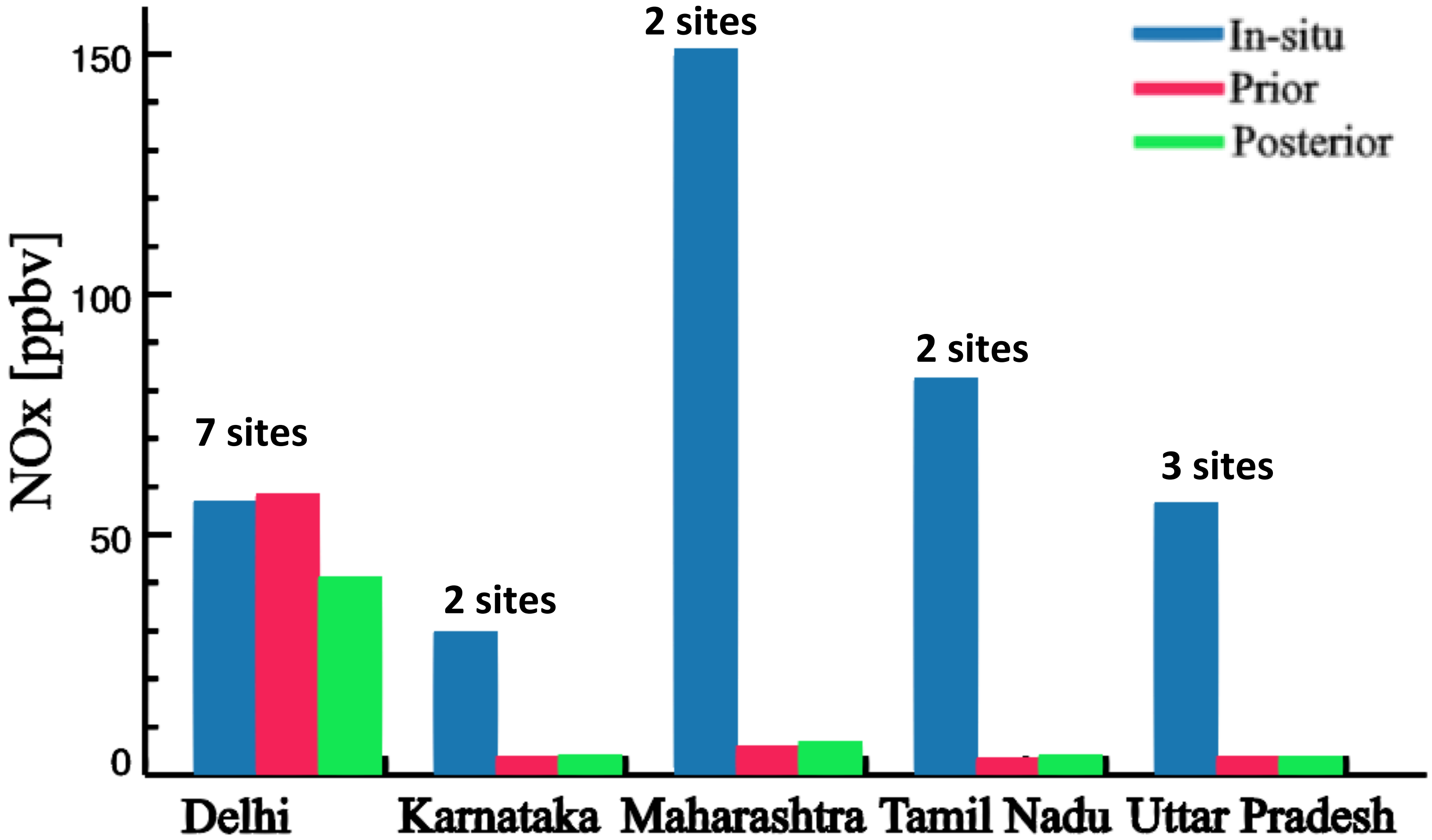
Evaluation of SO_2 emissions using in-situ measurements



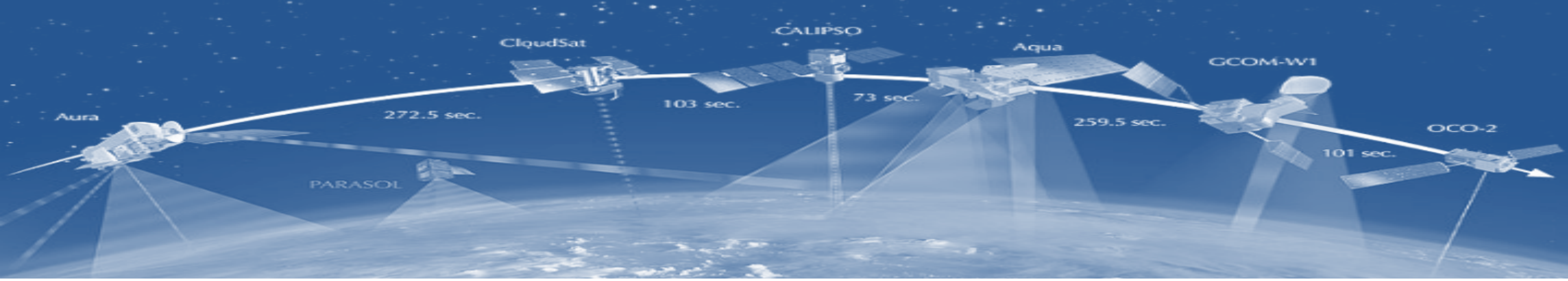
- SO_2 concentration in Delhi, Karnataka, Maharashtra and Tamil Nadu has been decreased and match better with in-situ measurements

Evaluation of NO_x emissions using in-situ measurements

India, Jan, 2010



- Lower NO_x concentration from GEOS-Chem probably due to coarse grid resolution and few measurements



Goal 3: Sector-based inversion

Sector-based inversion

- **Motivation:** Emissions are correlated through the amount of fuel combustions (Emission = species emission factor × fuel)
- Observe NO_2 and SO_2 column density.
- Optimize 7 sector-specific grid-cell emission scaling factors: industry, energy, residential, aviation, transportation, ship and agriculture.
- Apply scaling factors to NO_x, SO_2, CO, NH_3, BC, OC and NMVOC

Sector-based inversion

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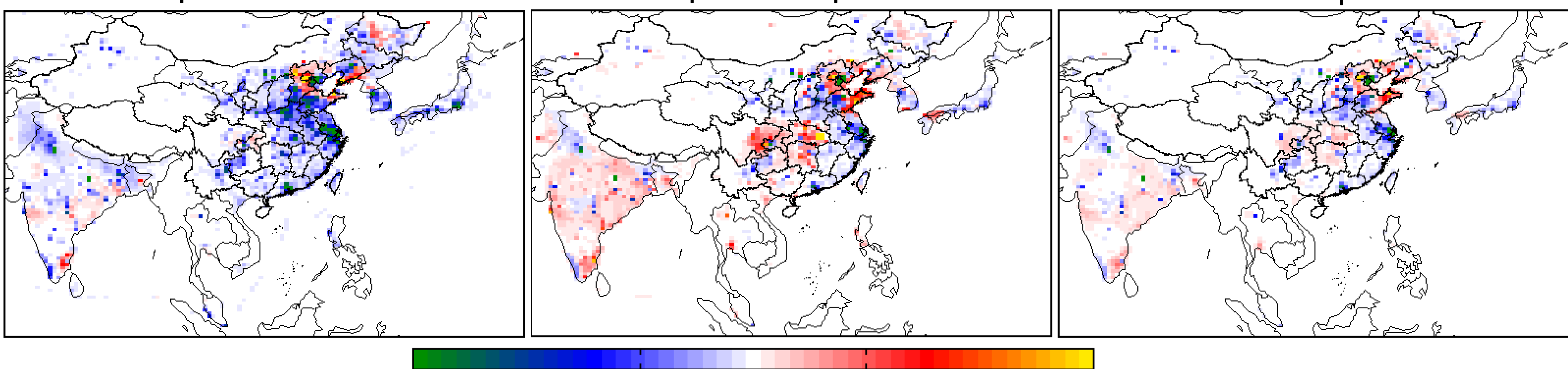
- Observe NO_2 and SO_2 column density.
- Optimize 7 sector-specific grid-cell emission scaling factors: industry, energy, residential, aviation, transportation, ship and agriculture.
- Apply scaling factors to NO_x, SO_2, CO, NH_3, BC, OC and NMVOC.
- No prior emissions constraints.

Changes of NO_x emissions in 4D-Var inversion (Jan, 2010, posterior - prior)

Optimize sector | Optimize species | Sector & species



- Sector-based and species-based inversions have similar decrease of model error (~70%), but converge to different solutions.
- Next step: Will evaluate through in-situ measurements.

Summary

- Develop a hybrid inversion that is more accurate than basic mass balance and less time-consuming than 4D-Var
- Different trend between column density and emissions
- Assimilating SO_2 observations affect NO_x posterior emissions
- Smaller posterior NO_x (1-18%) and SO_2 (~100%) emissions than bottom-up estimates in China
- Simulated NO_x concentrations are generally smaller than in-situ measurements; SO_2 simulation could be improved by the inversion depending on the location/bottom-up emissions
- Build up sector-based formulation to address the correlation among species

Qu et al., Monthly top-down NO_x emissions for China (2005-2012): a hybrid inversion method and trend analysis, JGR, 2017.

Methods

Hybrid 4D-Var / Mass balance

Base year (2010): 4D-Var

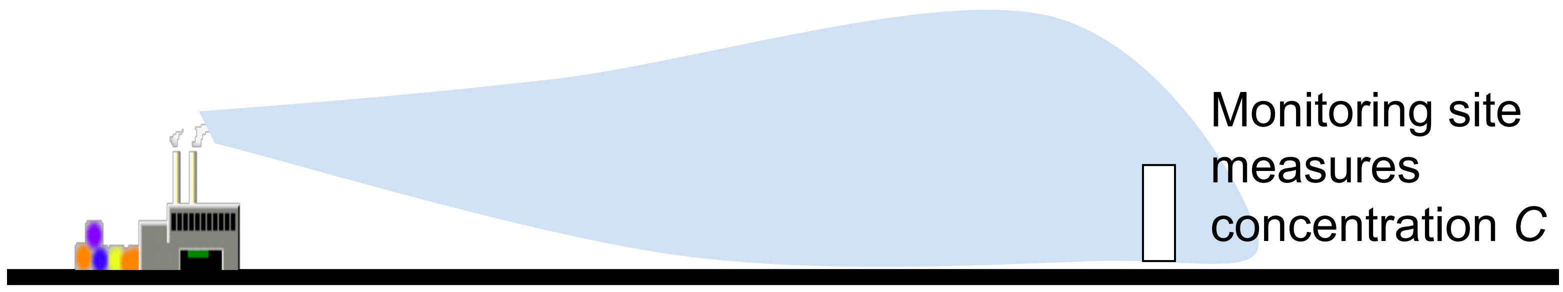
Other years (2005-2012): use 2010 4D-Var posterior for mass balance.

Motivation:

- Use 4D-Var to correct impact from transport
- Model / MB is sensitive to the prior emissions – more accurate than MB
- Computationally cheaper than 4D-Var

Inverse Modeling

Atmospheric "forward" model gives $C = kE$

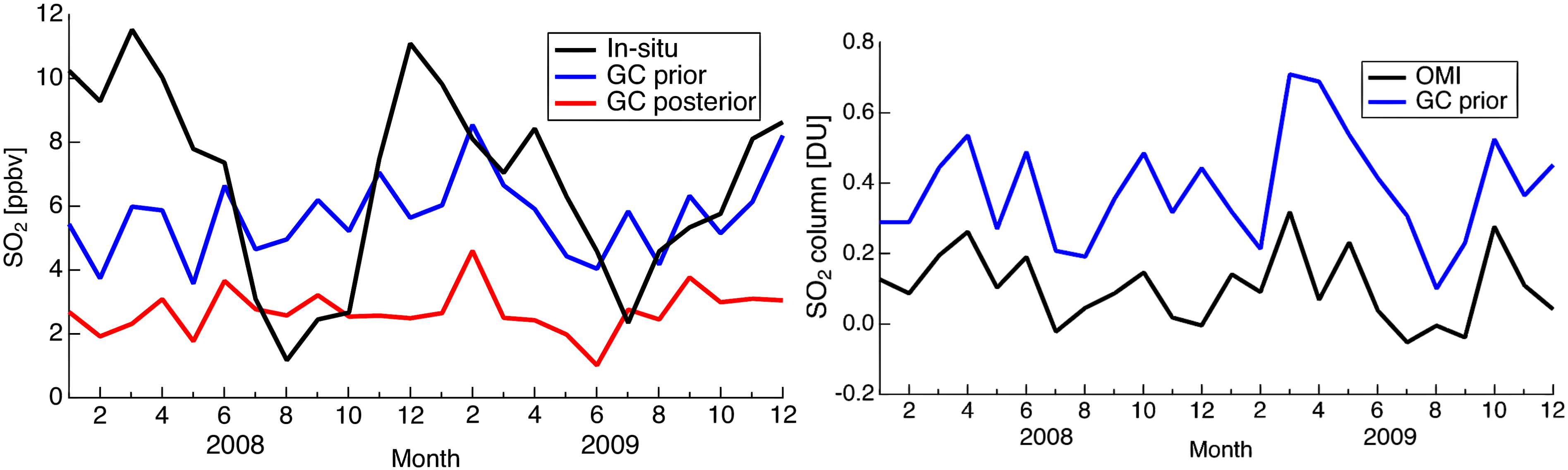


(fuel burned) X (emission factor)
⇨ *a priori* "bottom-up" estimate
$E_a \pm \sigma_a$

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

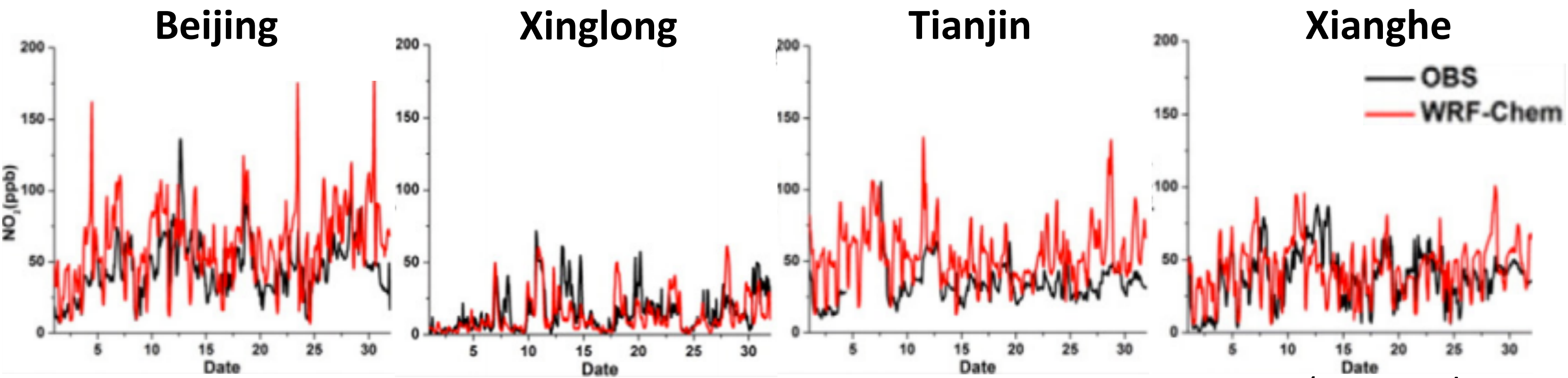
Evaluation of SO_2 emissions with observations

SO_2 at Beijing Miyun

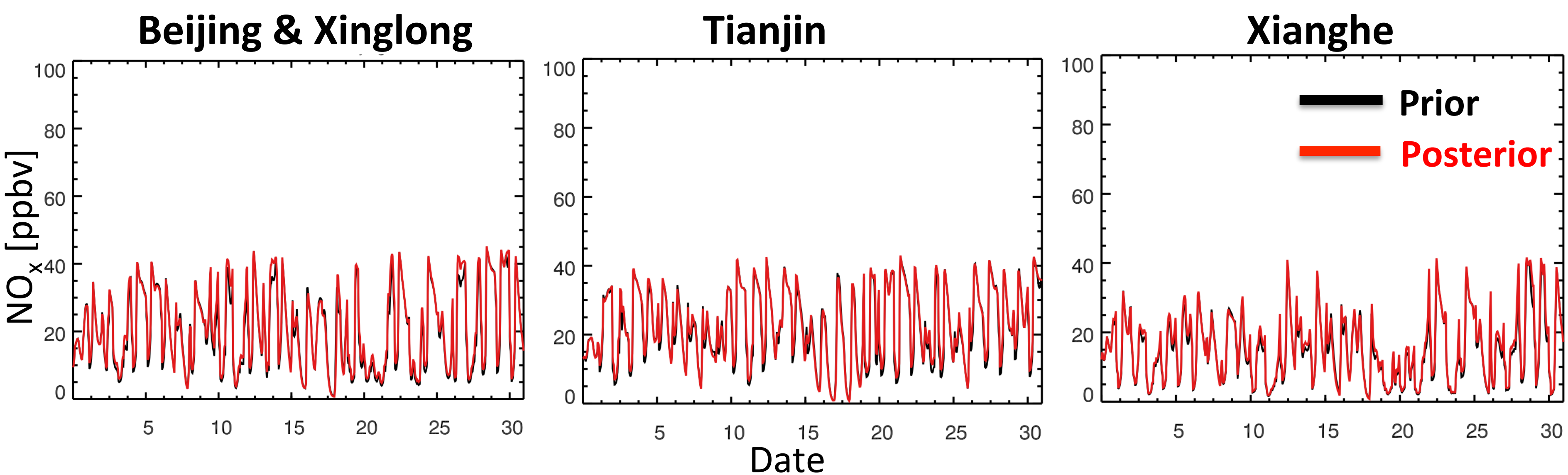


- Generally lower simulated SO2 concentration than in-situ measurements
- Higher simulated SO2 column compared to OMI observations
- Less SO2 in-situ measurements than satellite observations

Evaluation of NO_x emissions using in-situ NO_2 measurements
China, Jan, 2010



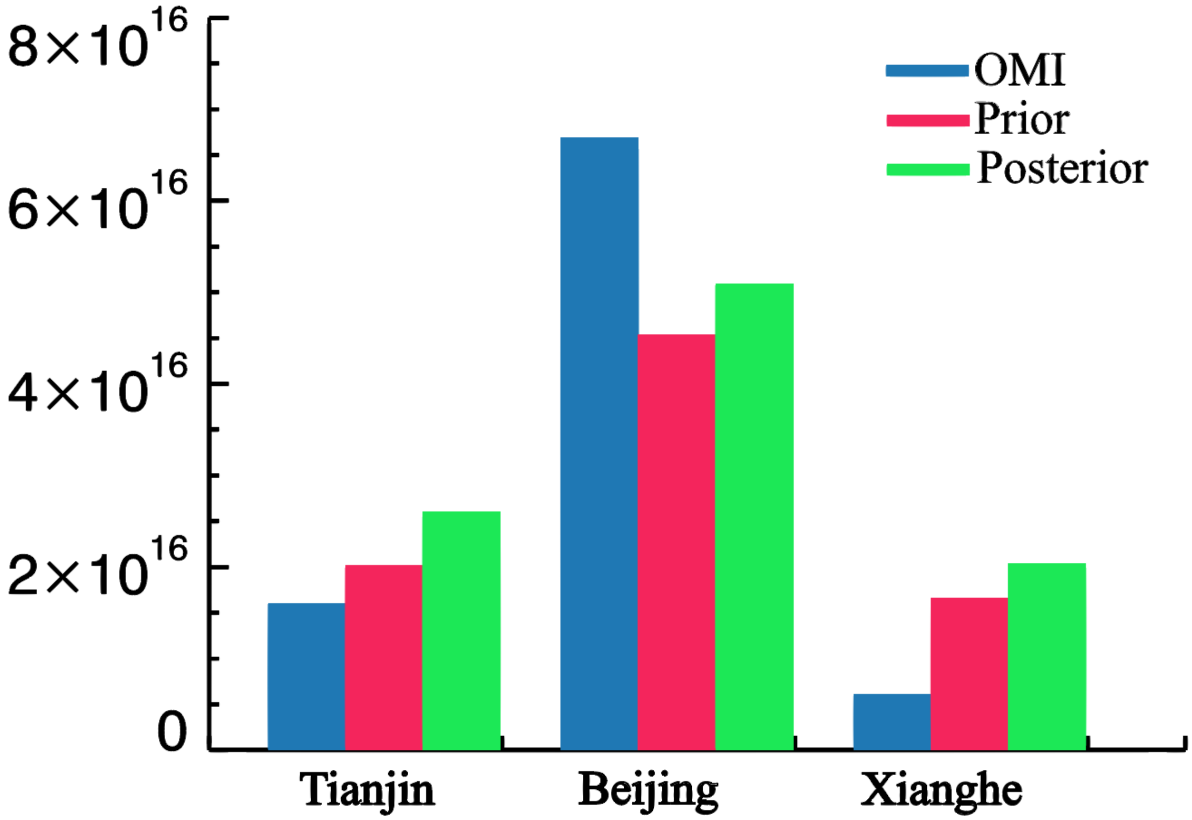
(Gao et al., 2015)



- GC simulation has lower NO_2 concentration and worse correlation with observations, probably due to coarse grid resolution

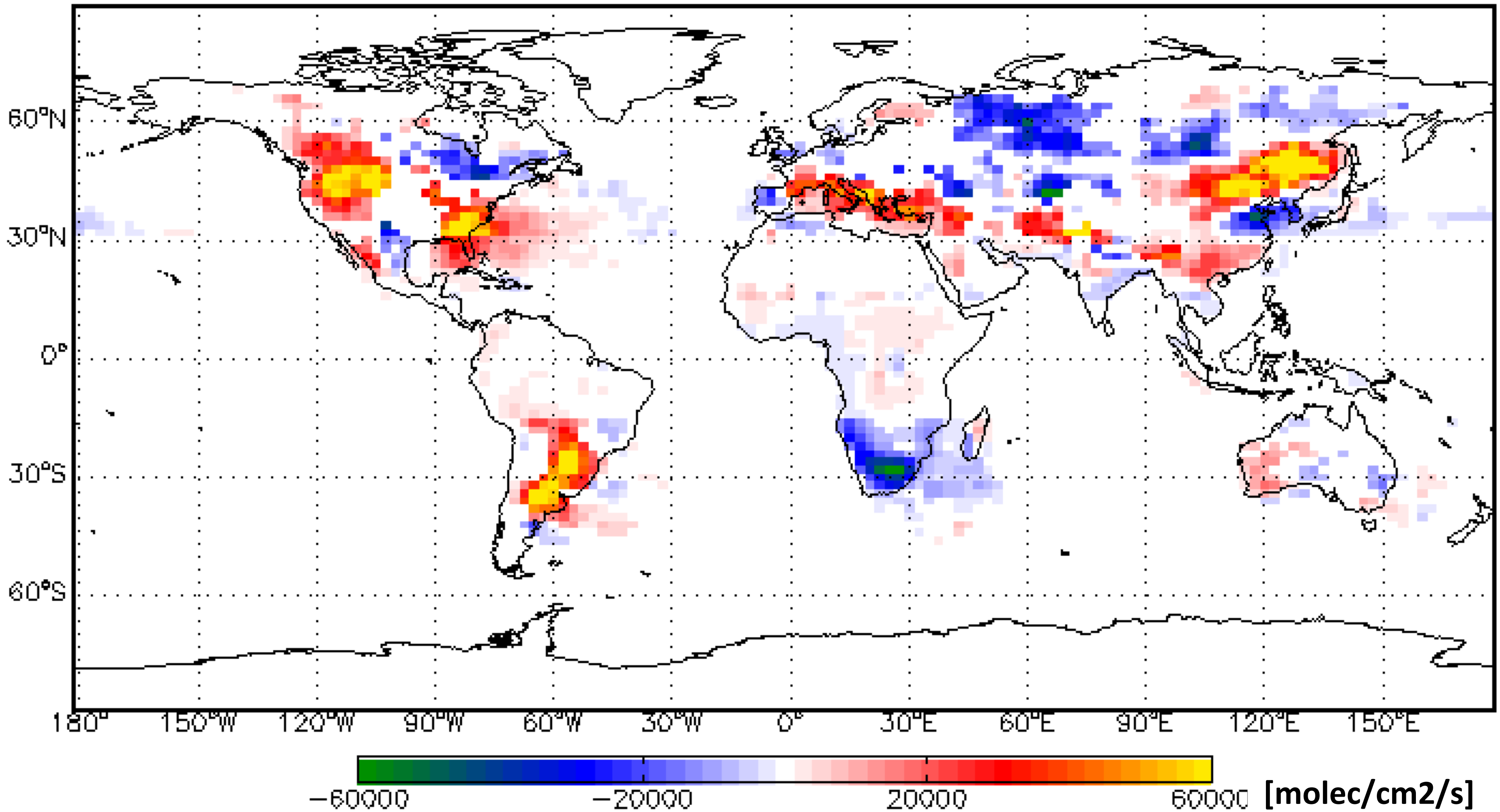
Evaluation of NO_x emissions using observations

China, Jan, 2010

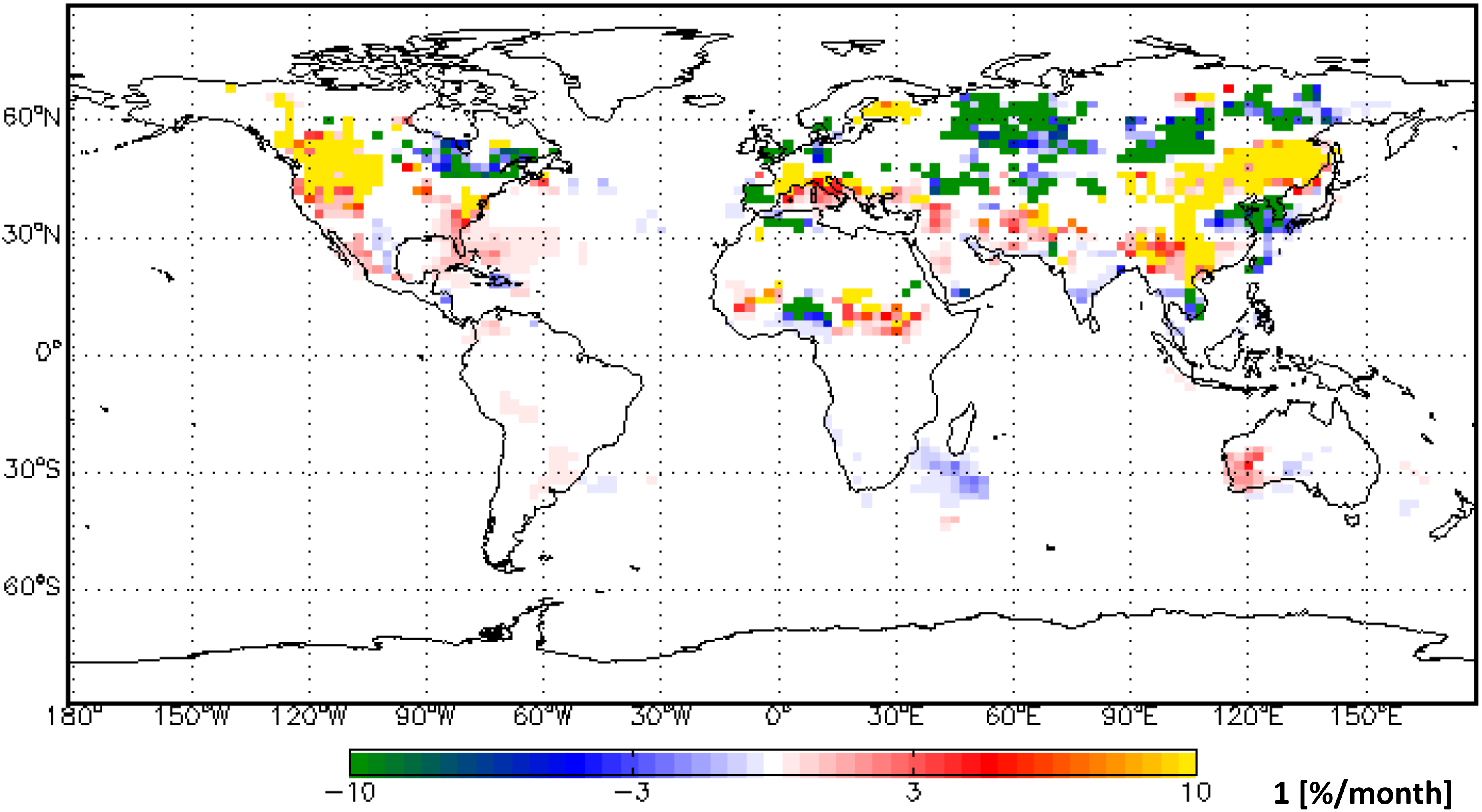


- Increased NO2 column due to spatially correlated error of emissions

Lightning NO_x growth rate, abs value



Lightning NO_x growth rate, relative value



Absolute growth rate NO_2 column