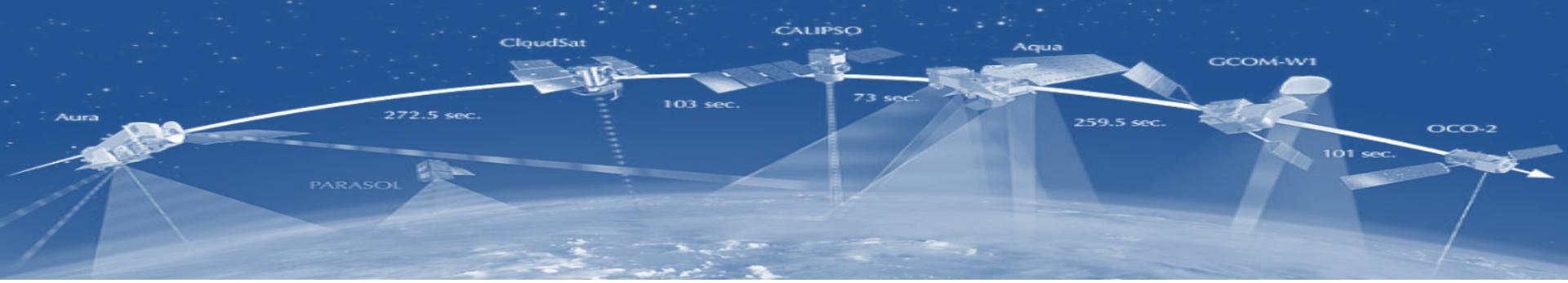


Incorporating chemical interactions and co-emissions in top-down constraints on sources of NO_x, SO_2 and CO

Zhen Qu[1], Daven K. Henze[1], Nicolas Theys[2], Jun Wang[3], Wei Wang[4], Helen Worden[5]

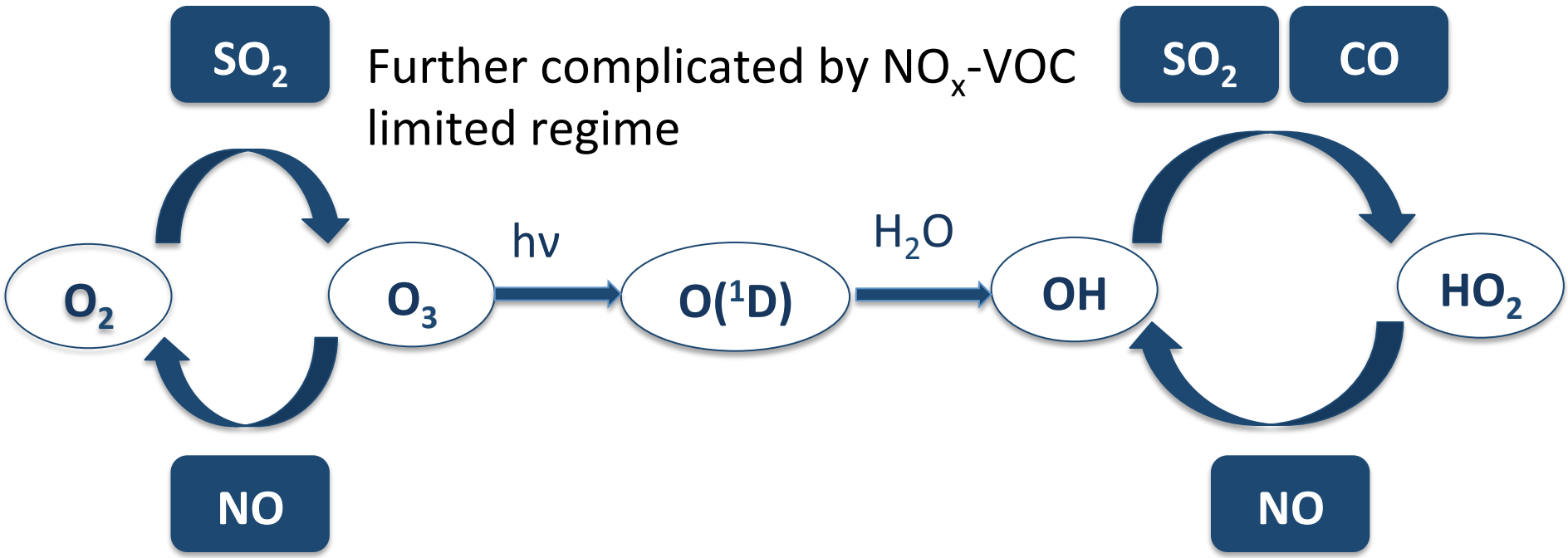
zhen.qu@colorado.edu

IGC 9, May 7, 2019

Funding: NASA ACMAP & HAQAST

[1]University of Colorado Boulder, [2]Belgian Institute for Space Aeronomy, [3]University of Iowa, [4]China National Environmental Monitoring Center, [5]National Center of Atmosphere Research.

NO_x, SO_2 and CO interact through reactions with OH and O_3



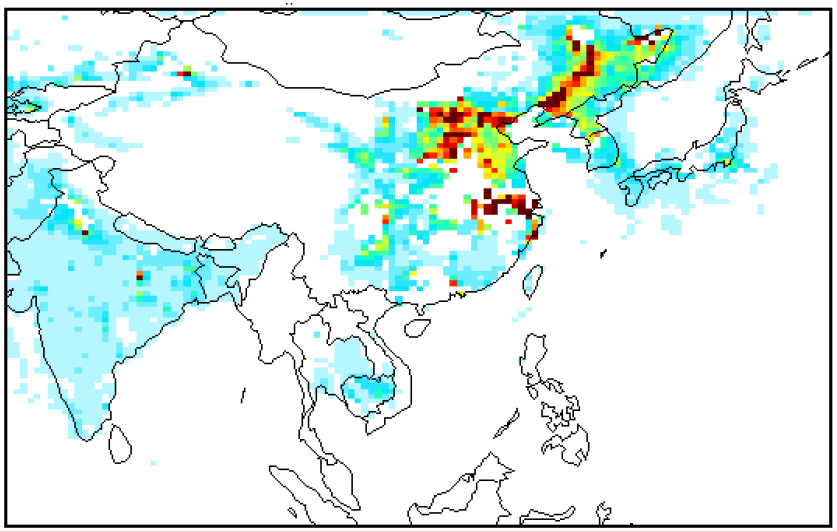
Neglecting these interactions could project errors in emissions of other species onto the ones being optimized.

Model & Observations

- **Method:** 4D-Var
- **Model:** GEOS-Chem adjoint, nested China, 0.5°x 0.667°
  Prior emissions from HTAP inventory
- **Observations:**

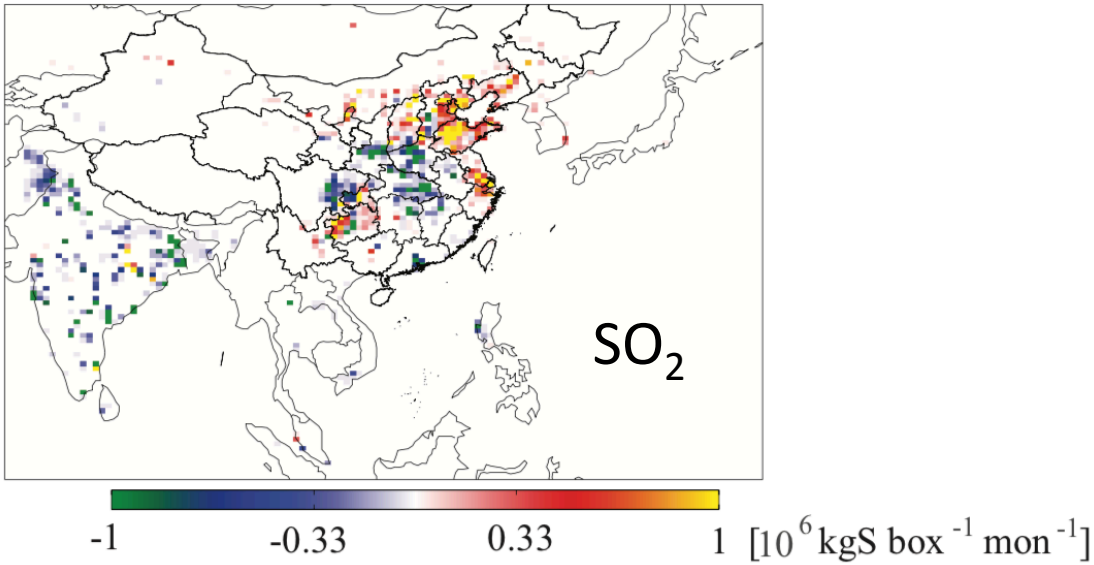
OMI NO_2, NASA standard L2 product (V3)
OMI SO_2, BIRA L2 product

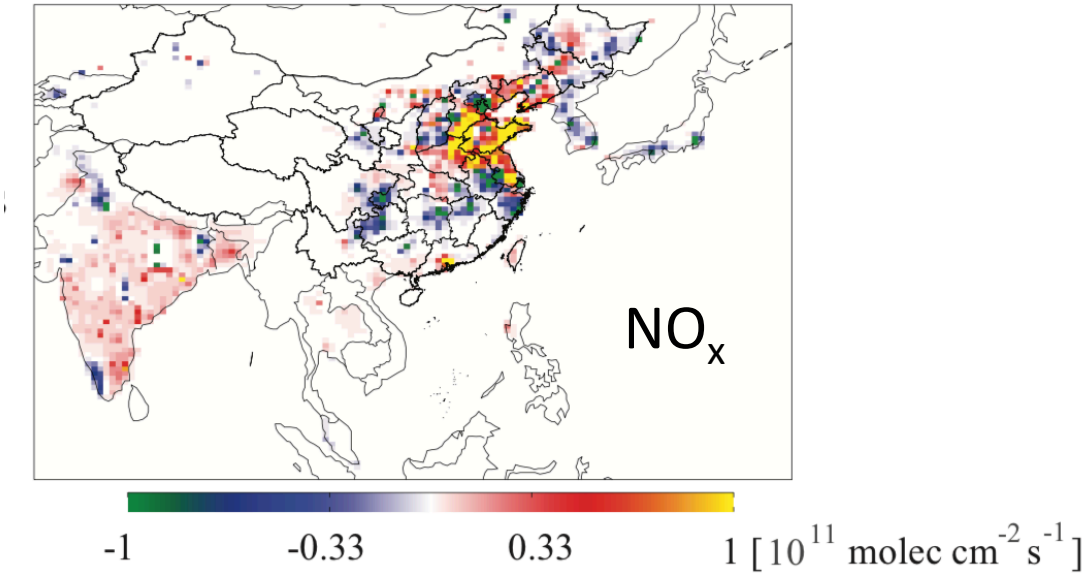
NO_2 column density



Joint v.s. single species posterior emissions better match OMI SO_2 observations (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., in revision)

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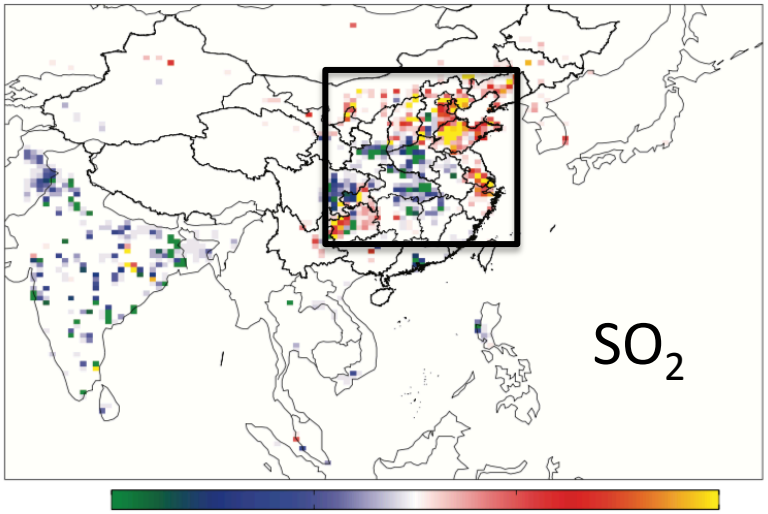
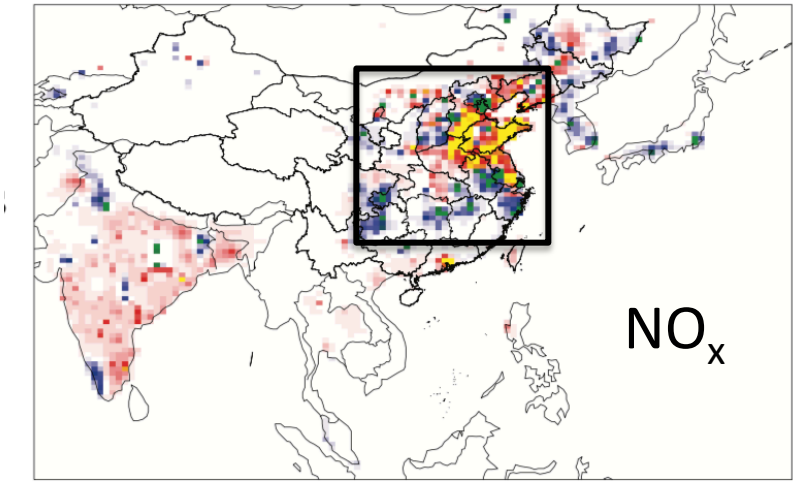
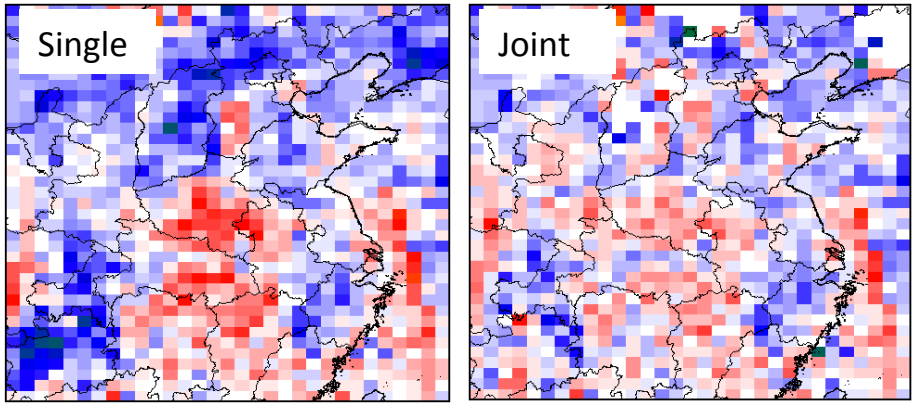


-1 -0.33 0.33 1 [10^6 kgS box^{-1} mon^{-1}]



-1 -0.33 0.33 1 [10^{11} molec cm^{-2} s^{-1}]

SO_2 columns (GEOS-Chem − OMI)



-1 -0.33 0.33 1 [DU]

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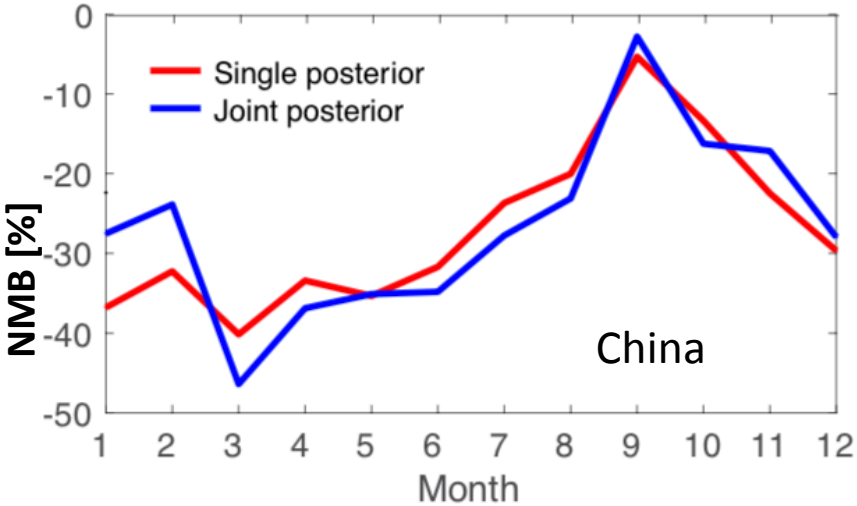
Improved surface concentration using joint posterior emissions when observation uncertainties are large

Surface SO_2 GEOS-Chem vs measurements



- 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.

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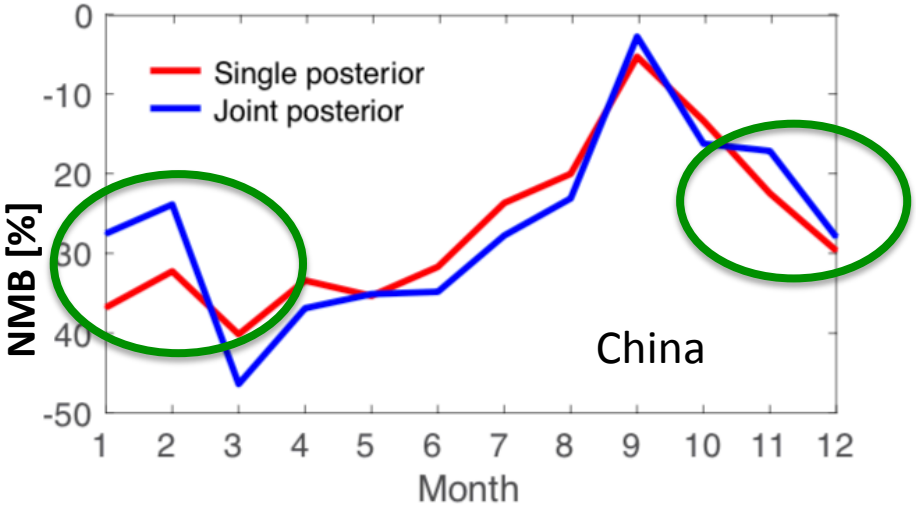

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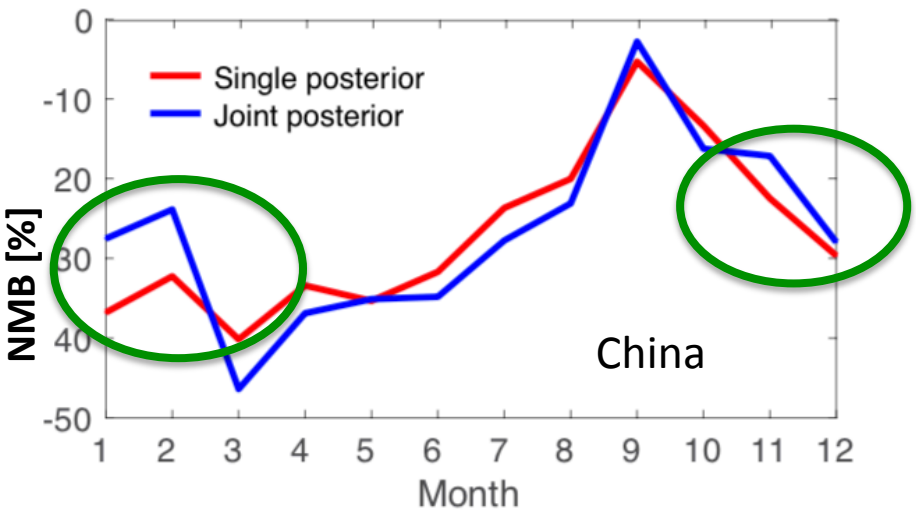
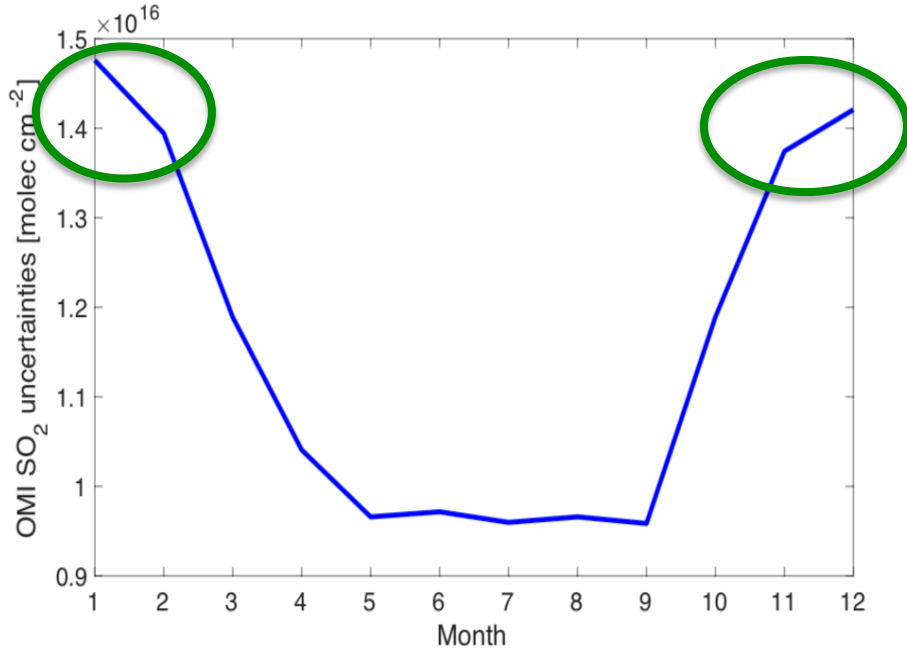


Uncertainties in OMI SO_2 observations (2010)



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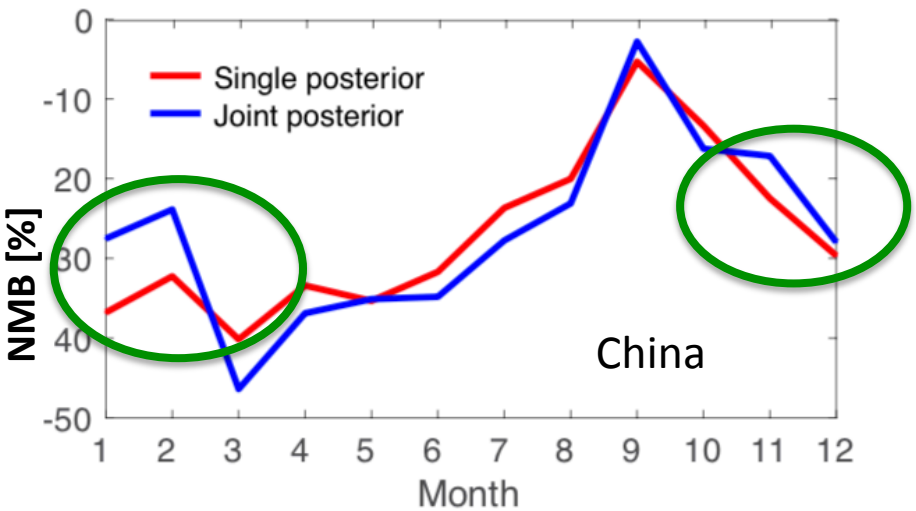

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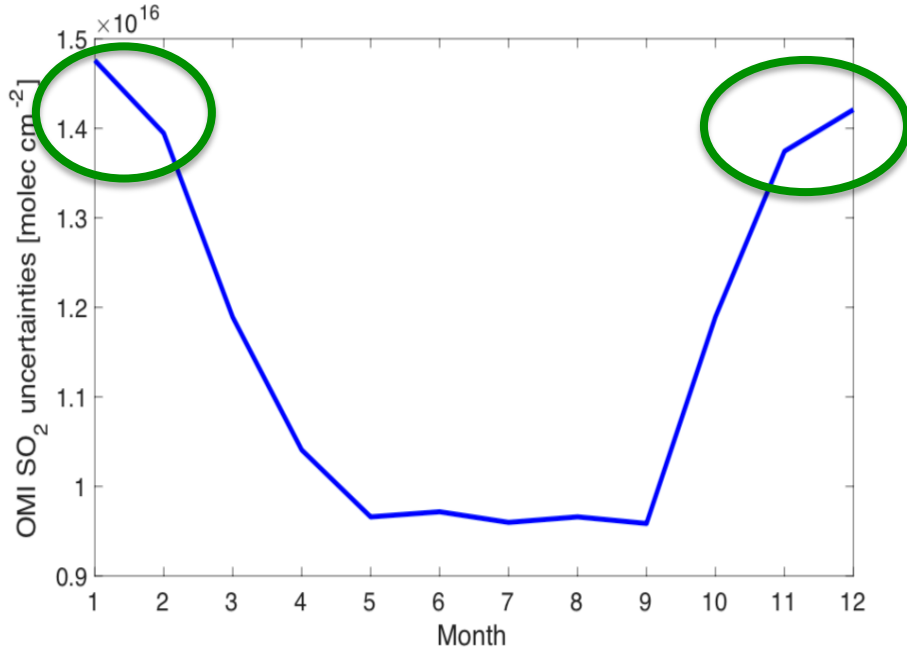
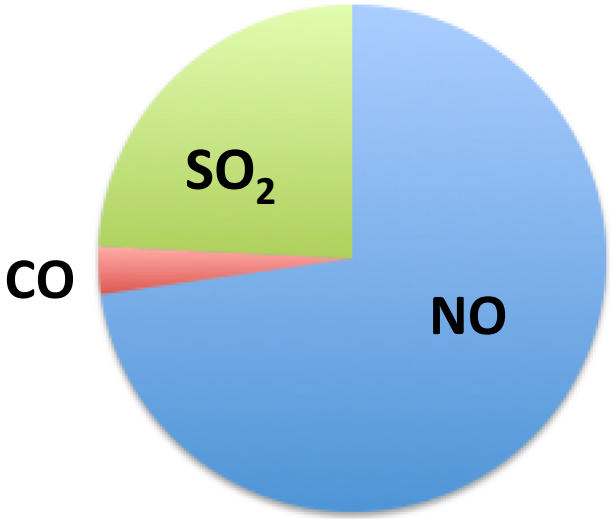


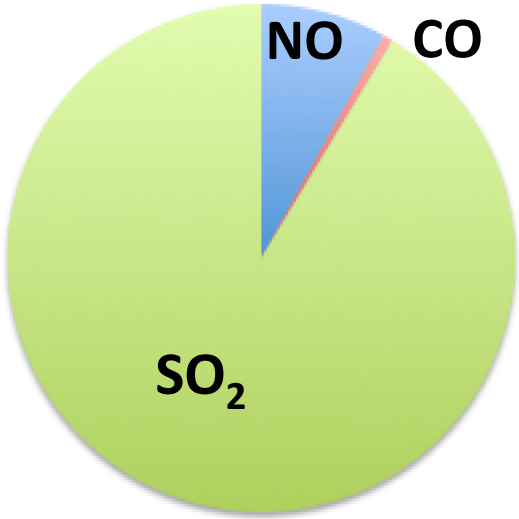
- NMB and NMSE of SO_2 joint posterior reduce by up to 26% and 18% compared to single species posterior in most months between November and February.
- Similar behavior for NO_2 and over India.

Co-emitted pollutants and their uncertainties are correlated

Transportation



Energy



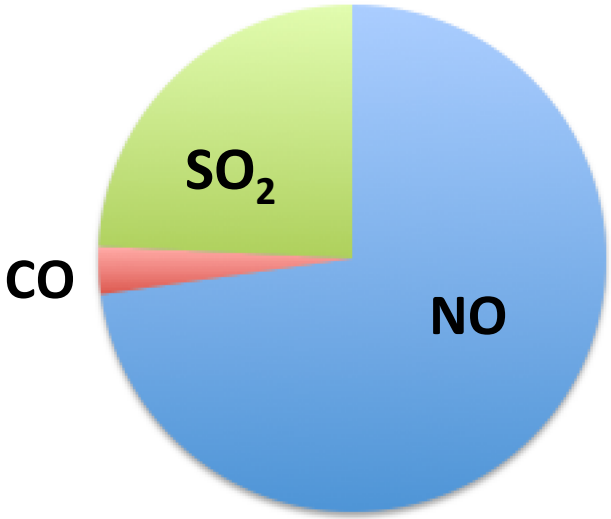
Similar ratio of NO_x, SO_2 and CO emissions in the same sector, yet very different across sectors.

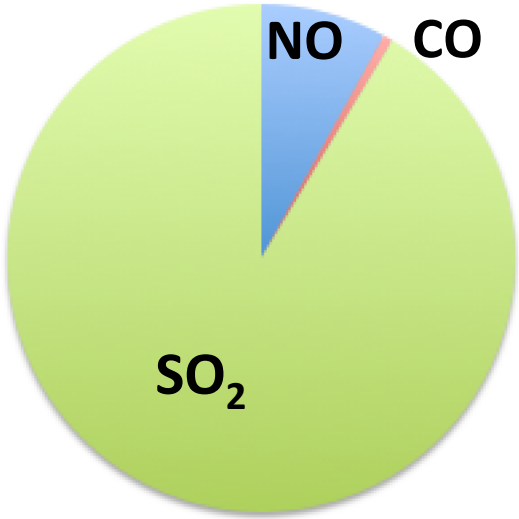
Co-emitted pollutants and their uncertainties are correlated

Transportation

Energy

Sector-based emission scaling factor





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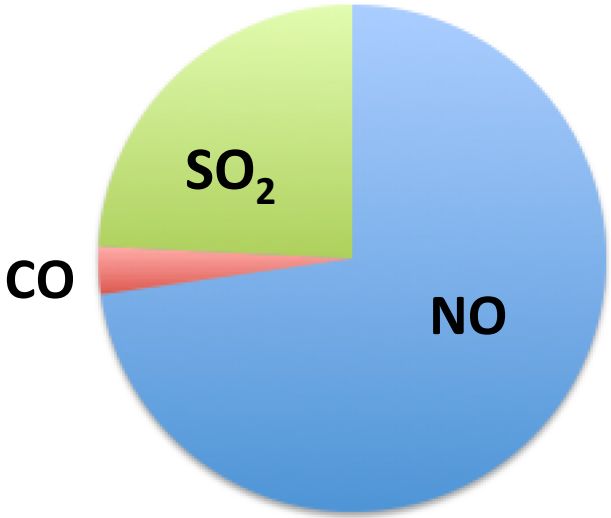
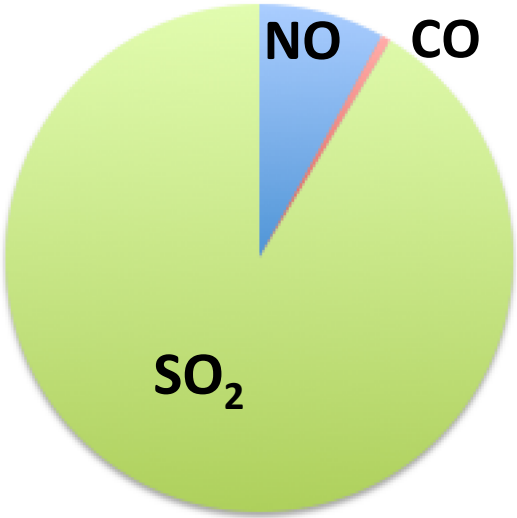
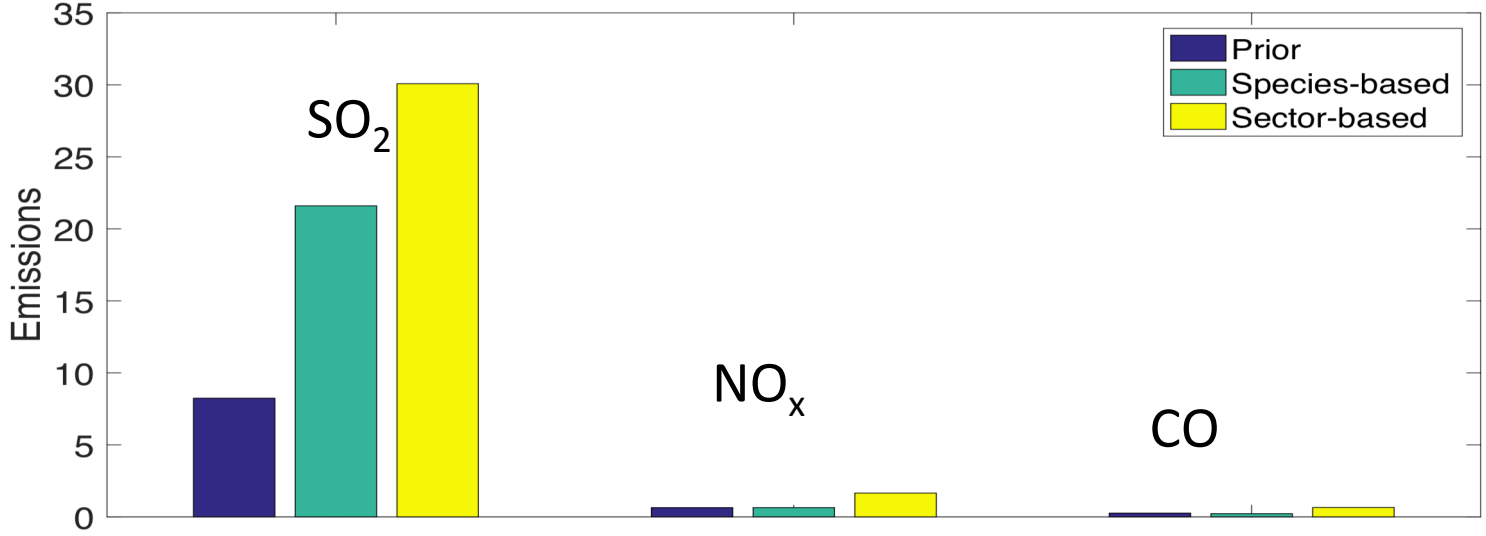




Assimilate:
MOPITT CO
OMI NO_2
OMI SO_2

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

Different posterior emissions using sector- and species-based inversions



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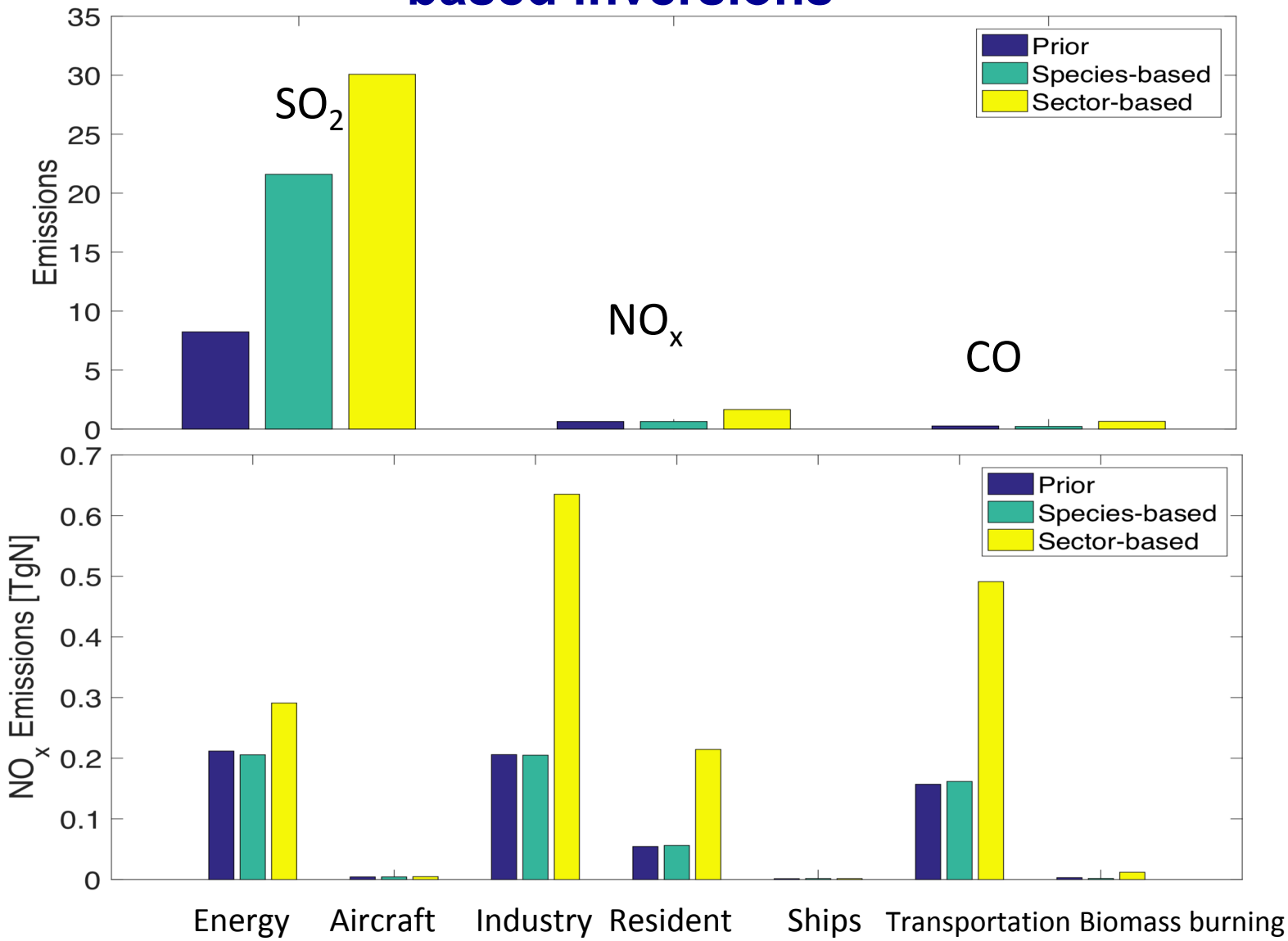

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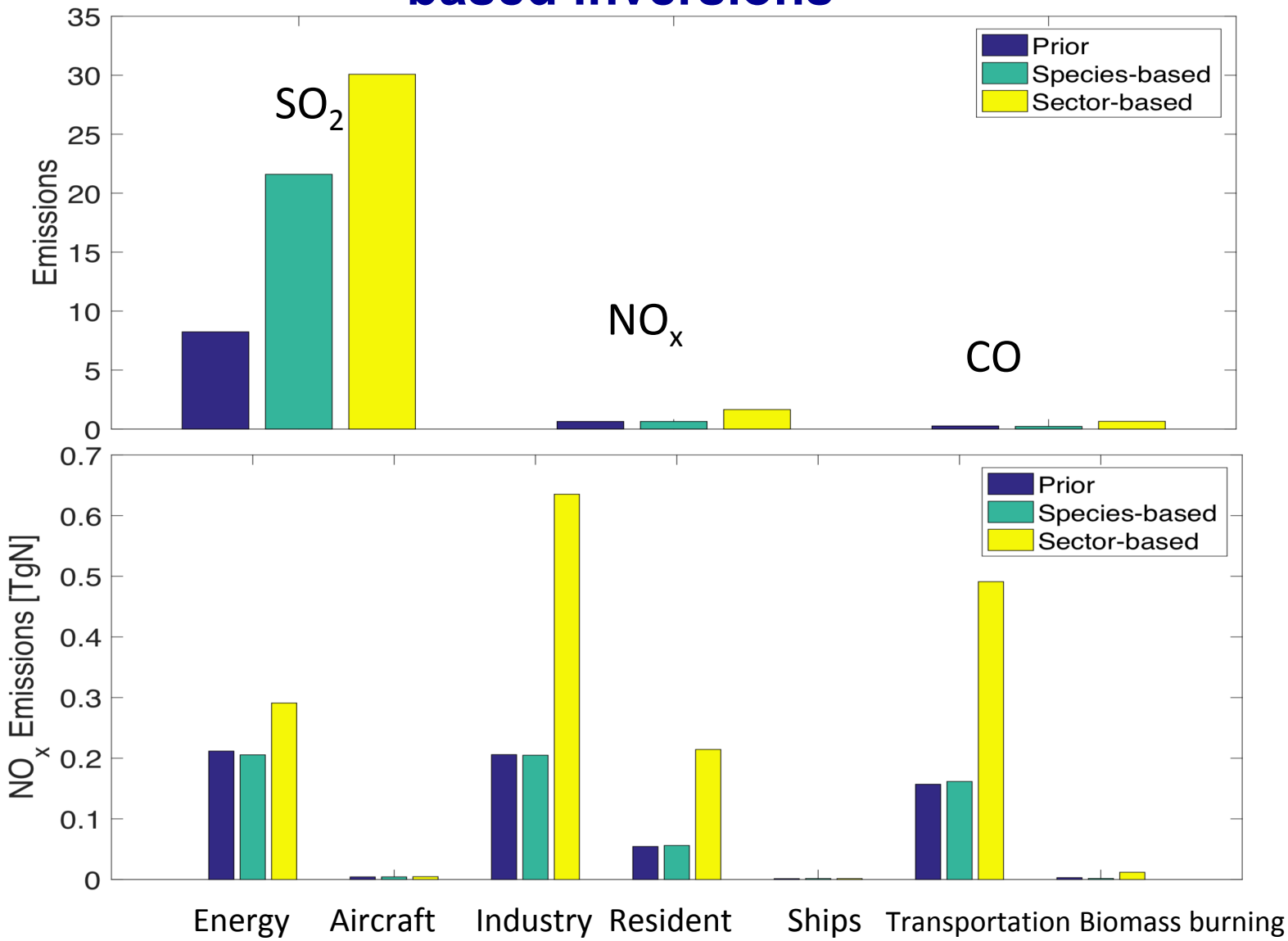
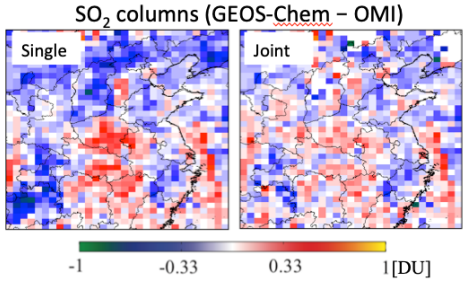


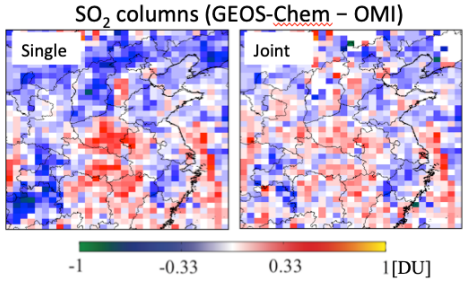
Total budgets increase with both methods. Yet sector-based emissions change in the correct direction when species-based do not, in comparison to surface observations.

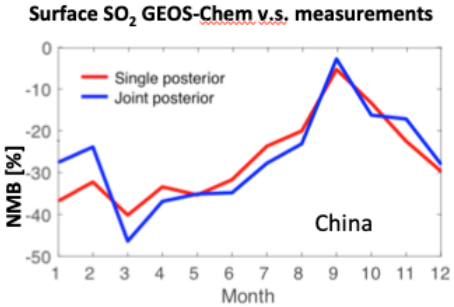
Summary



- Joint inversion reduces error in NO_x and SO_2 top-down emissions through synergistic change of OH concentration.

Summary





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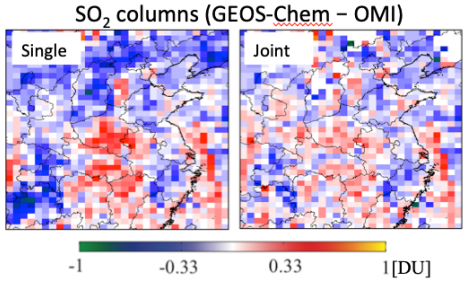
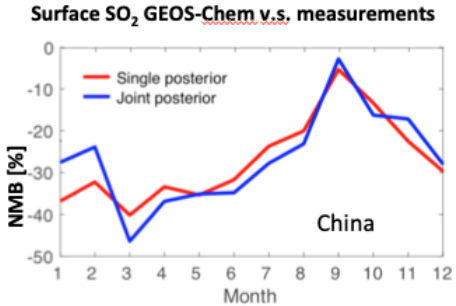
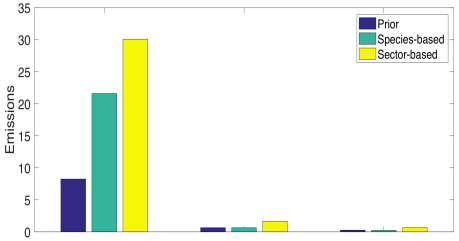
Qu et al., Hybrid mass balance / 4D-Var joint inversion of NO_x and SO_2 emissions in East Asia, *in revision*.

Summary







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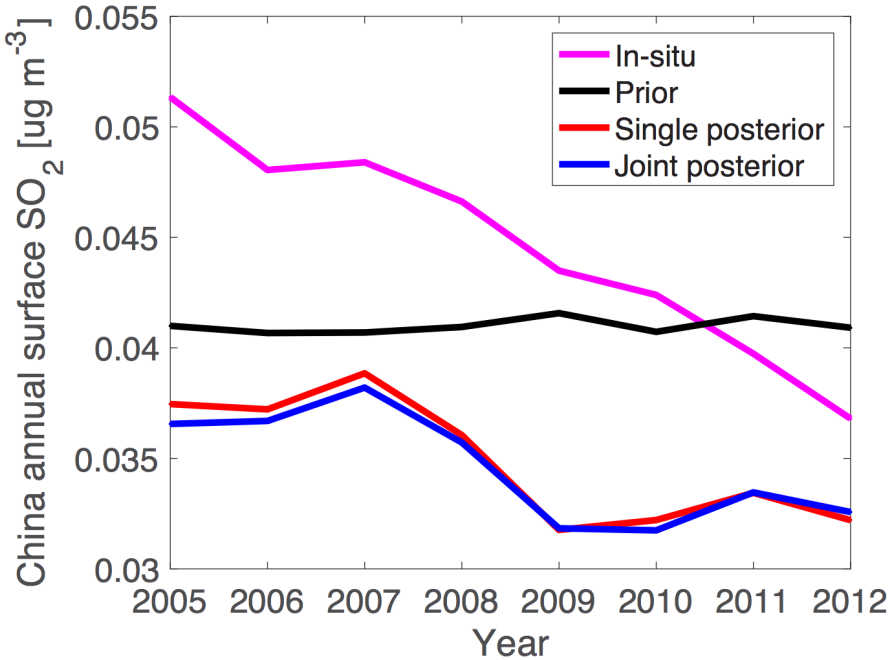
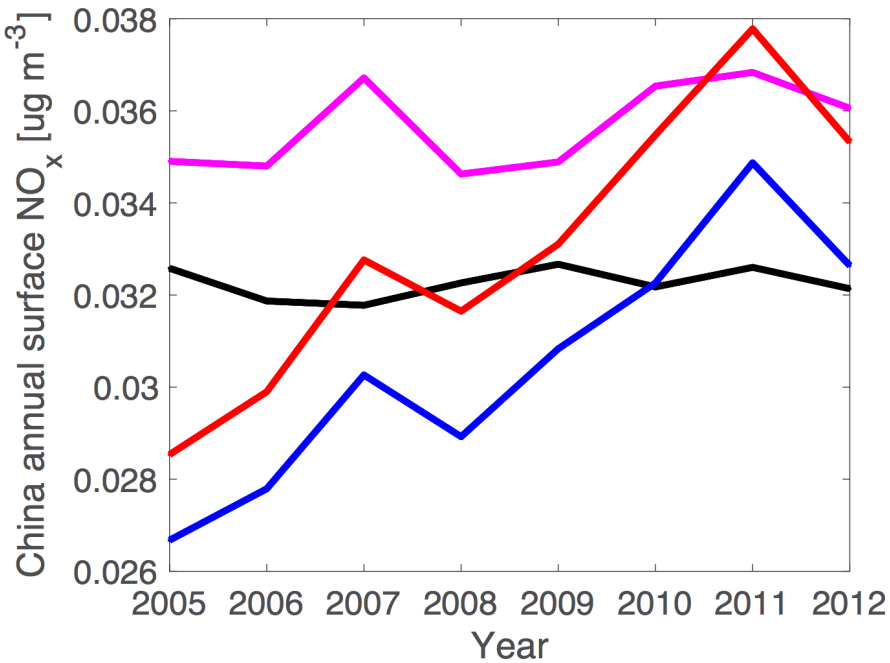
- Sector-based posterior emissions are larger than species-based posterior emissions, and change in the correct direction when species-based inversion does not.

Qu et al., Hybrid mass balance / 4D-Var joint inversion of NO_x and SO_2 emissions in East Asia, *in revision*.

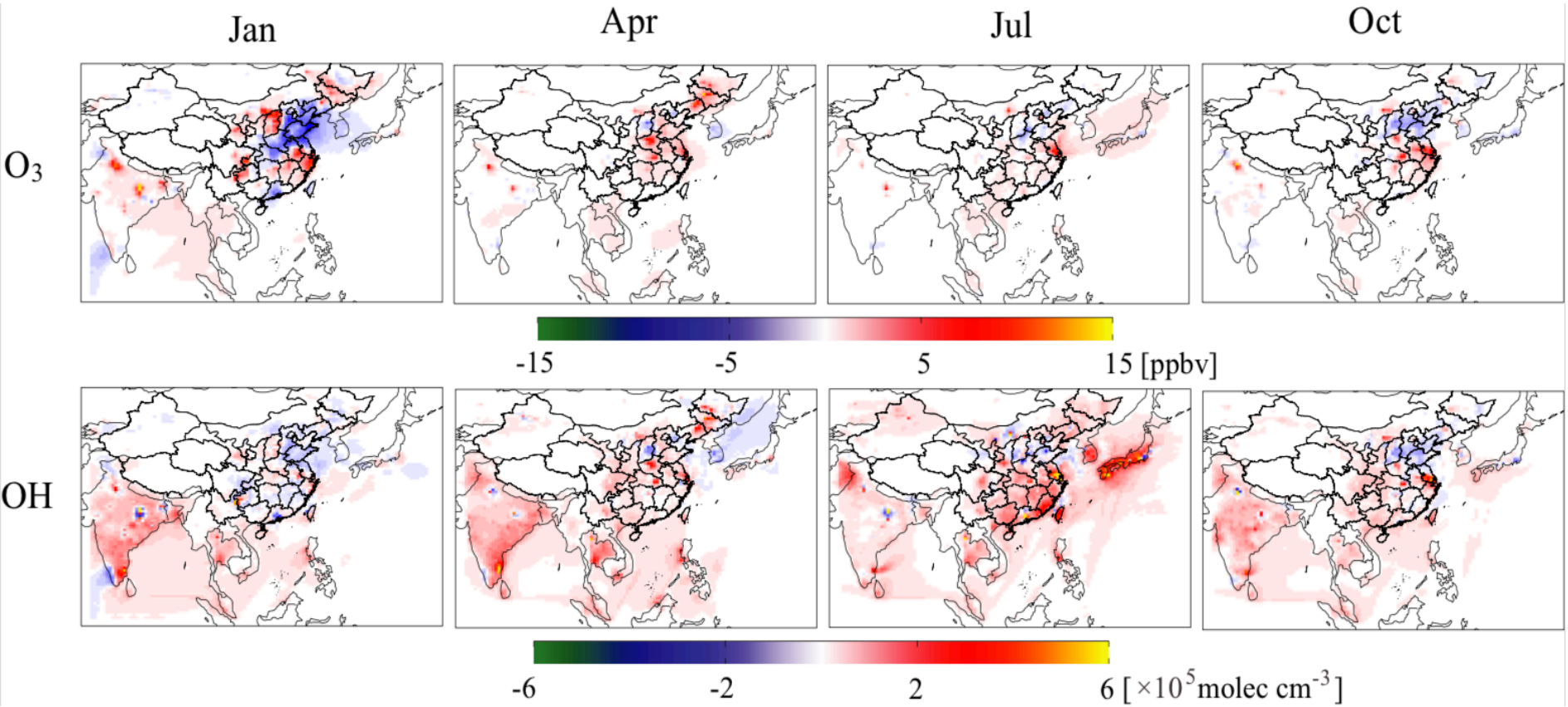
Use OMI data: trend of surface SO_2 concentration



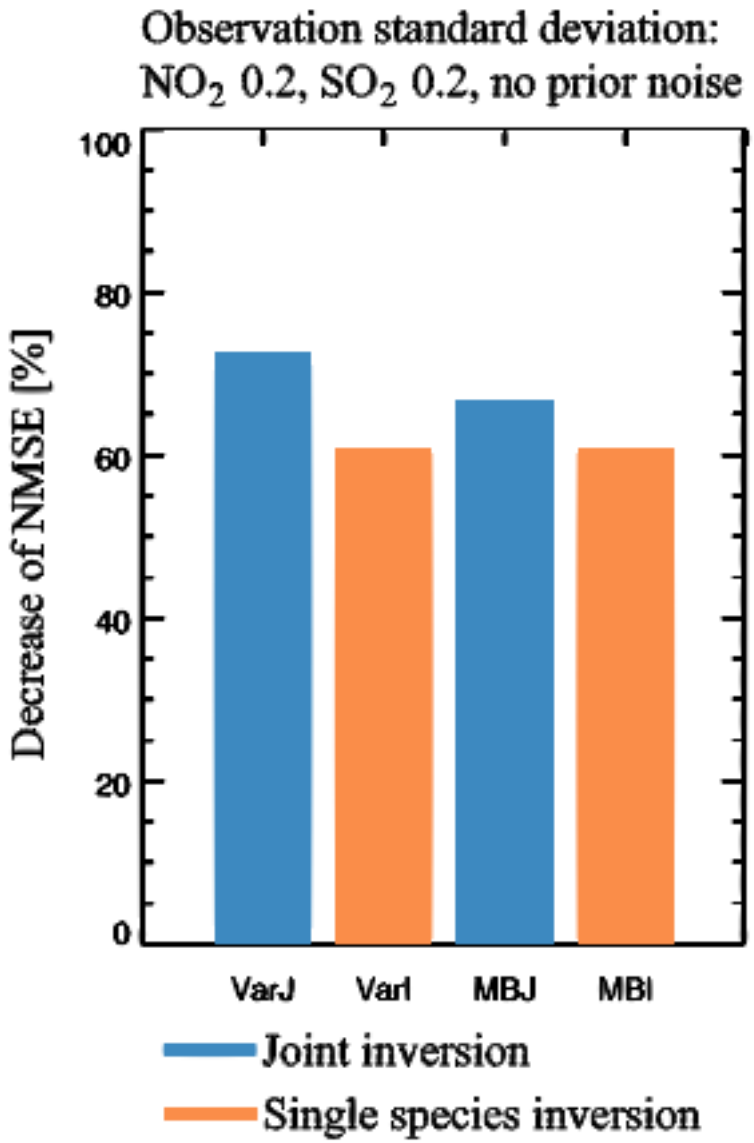


- At domain-wide and annual average, difference between joint and single species inversions are small, as increase and decrease at different regions cancel out

Monthly changes of surface O_3 and OH concentrations go up to 140% and 100% (2010)



Multi-species mass balance method decreases NMSE in pseudo observation test



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