Characterization of Chemical Mechanisms used in Top-Down VOC Emission Estimates

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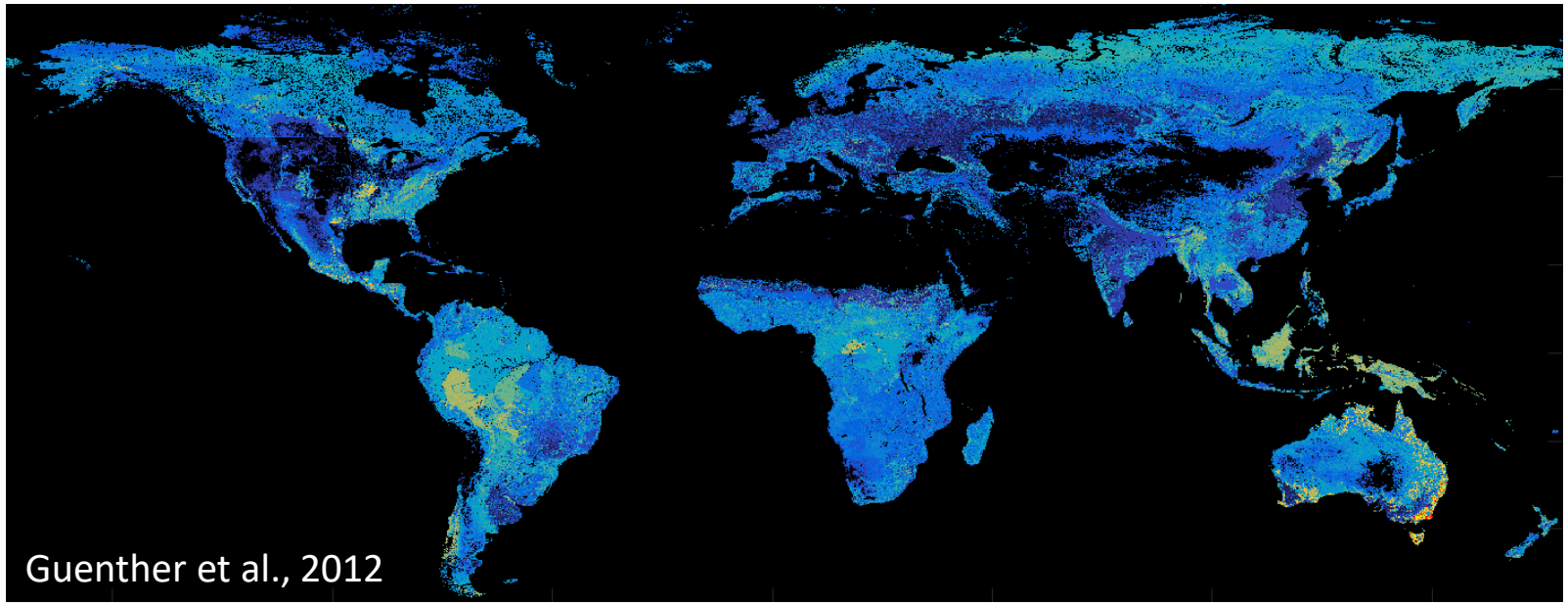
ACM meeting

Dec 5 2018

Getting emissions right is the first step in predictive air quality modeling.

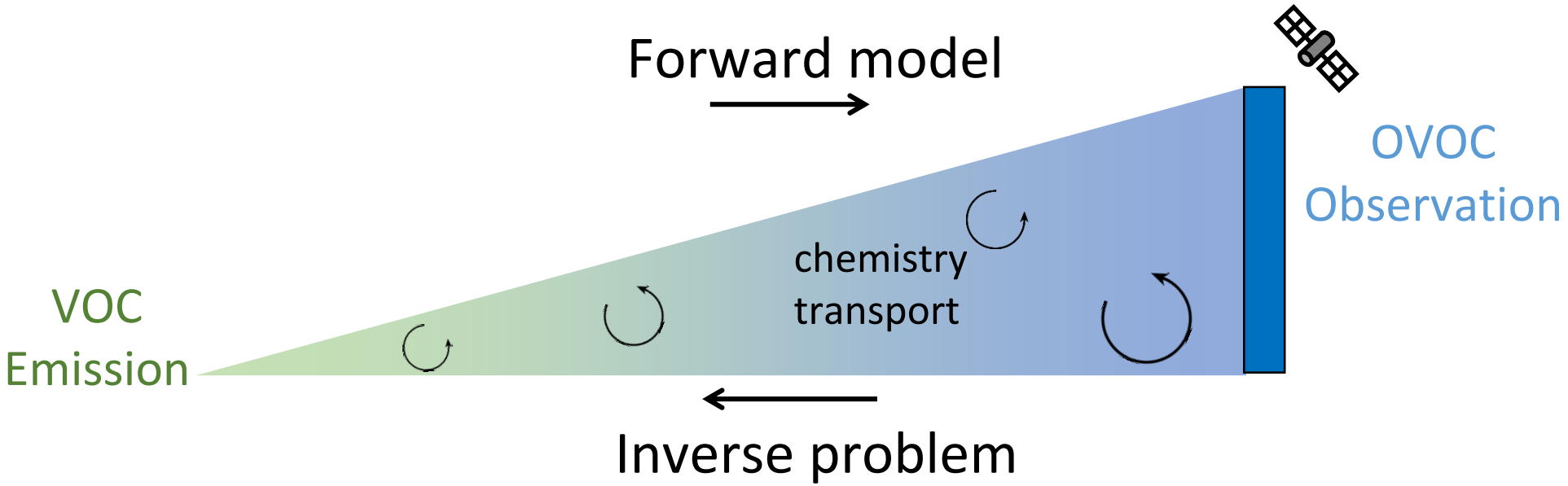
MEGAN v2.1 base emission factors (ε_{isop})



- VOC emissions are especially challenging
- Unlike chemistry, emissions are a moving target
- But if we understand the chemistry, we can back out the emissions.

Applications/Implications

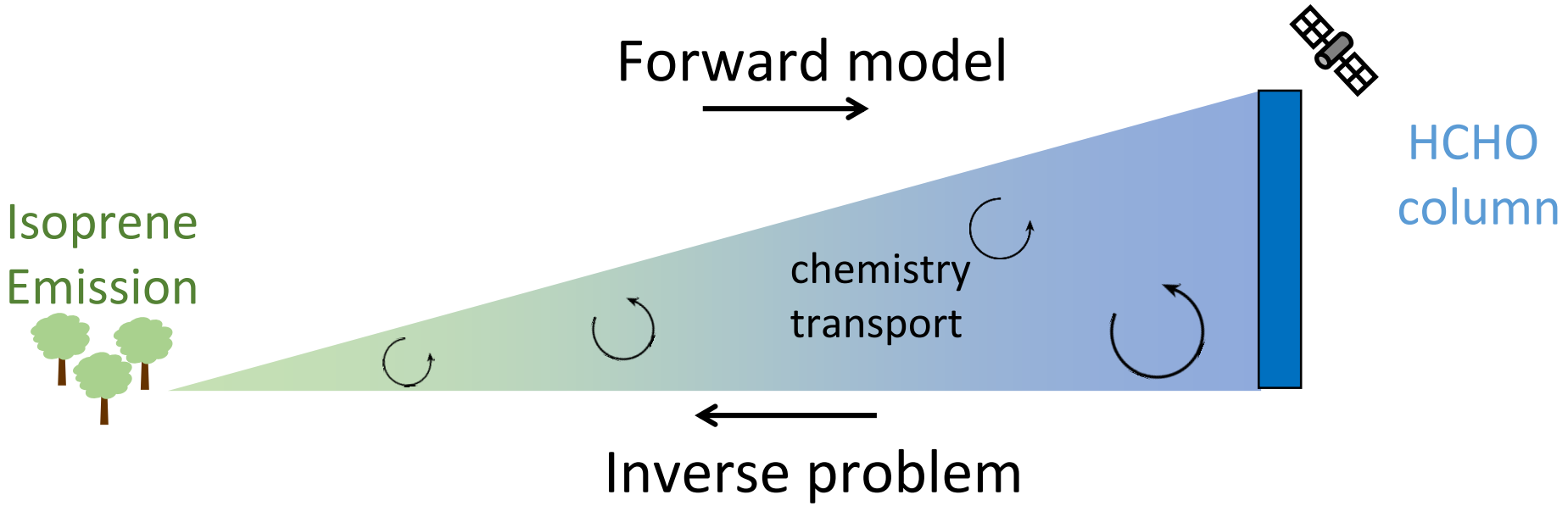
Can current chemical mechanisms be used to improve VOC emission estimates?



Are OVOC yields well constrained?

What are the uncertainties in our observations?

First: a success story

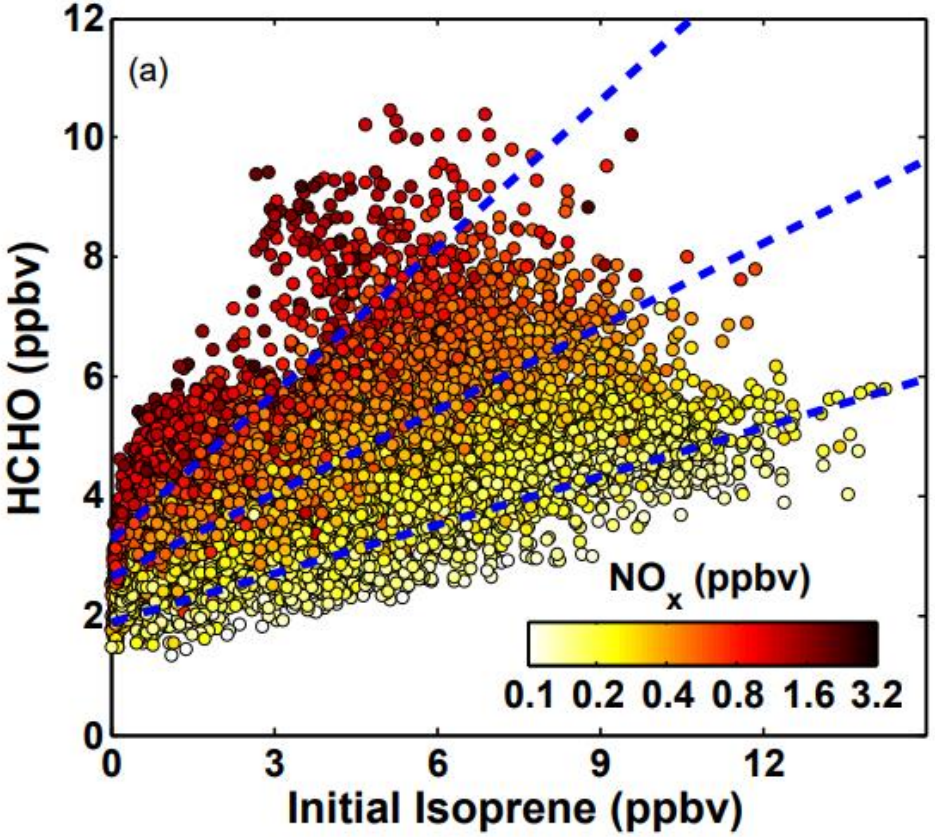


High-resolution inversion of OMI formaldehyde columns to quantify isoprene emission on ecosystem-relevant scales: application to the southeast US.

Kaiser et al., 2018

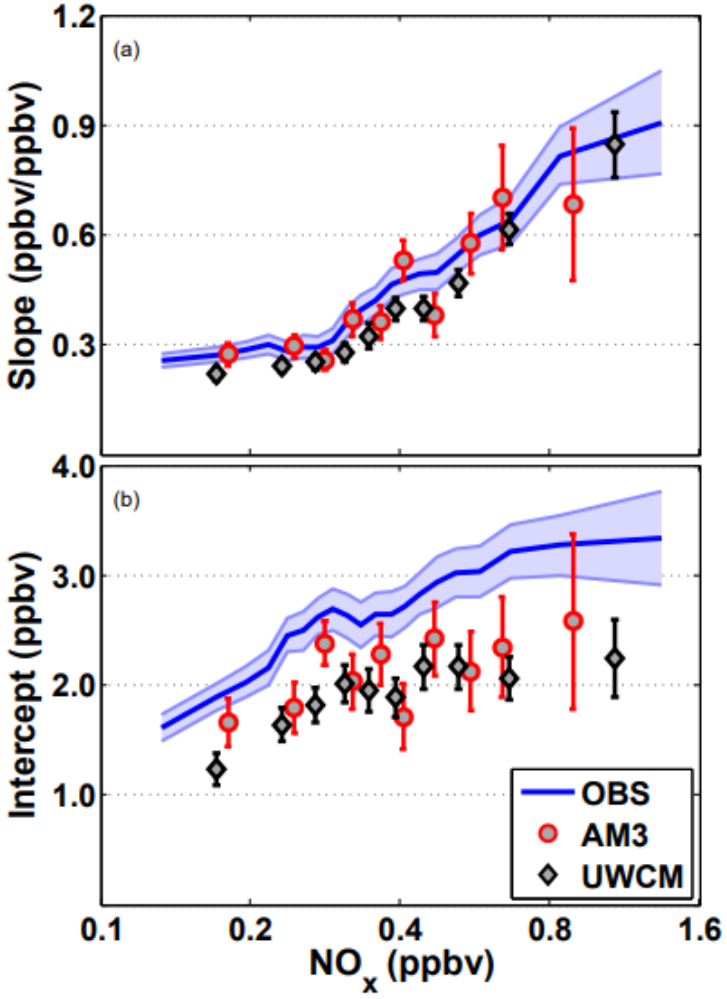
Mechanism must reproduce the observed VOC-NO_X-OVOC relationship.

SENEX 2013 observations:



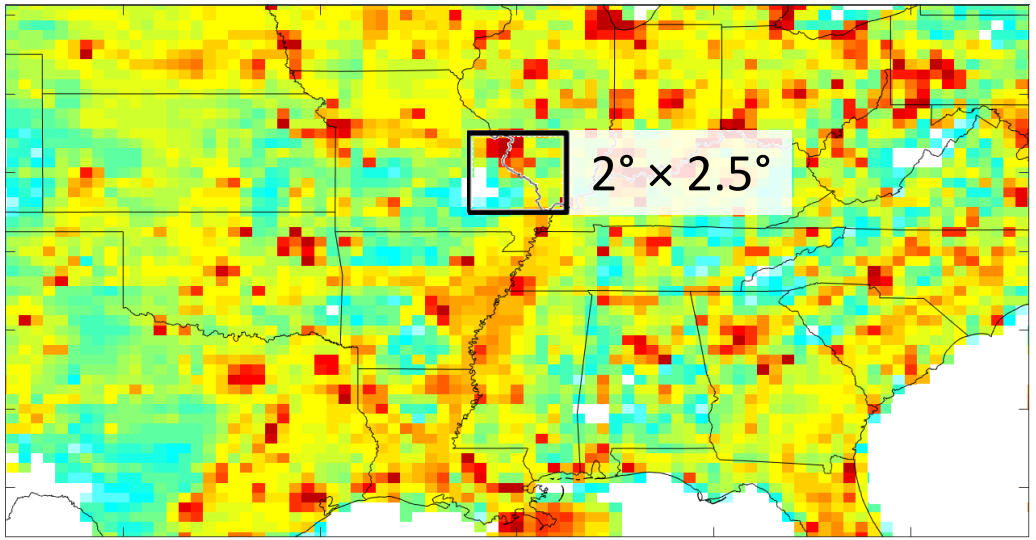
Wolfe et al., 2016

Comparison with box model:

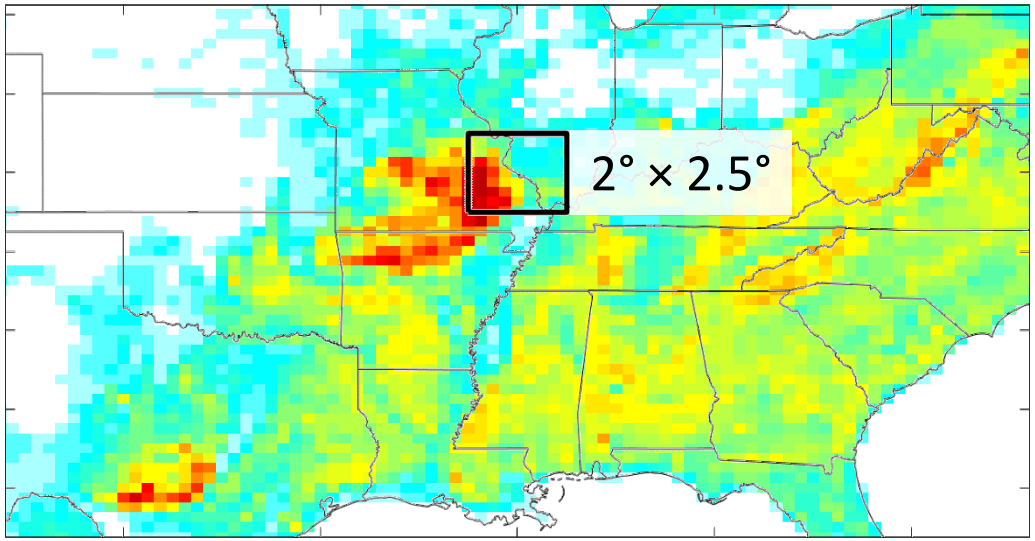


NOx emissions need to be right, and spatially resolved.

NO_x emissions (log scale) 0.25° × 0.3125°



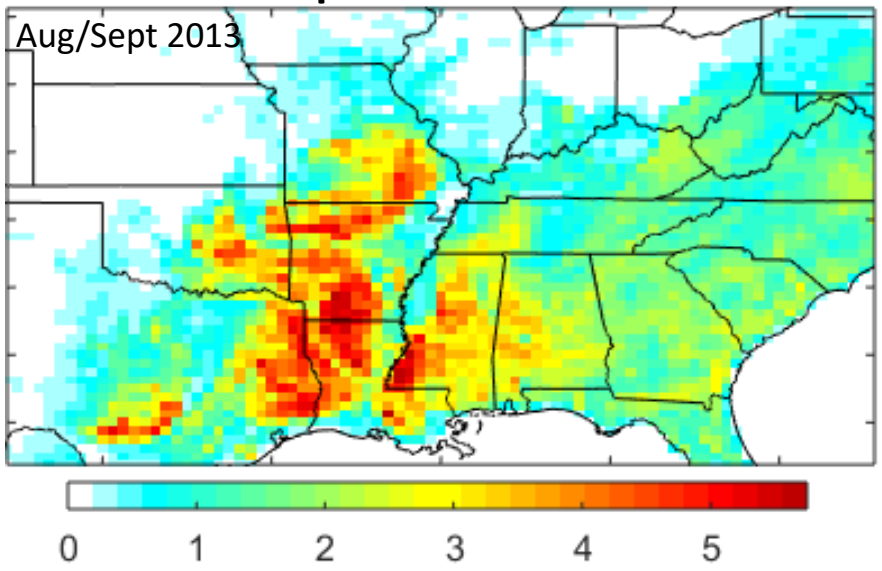
Isoprene emission factor 0.25° × 0.3125°



- HCHO has much higher larger under high NOx conditions!
- Coarse simulations show a high HCHO bias (Yu et al., 2016)
- GEOS-Chem NOx constrained using a suite of observations (Travis et al., 2016)

Isoprene and HCHO during SEAC^{4}RS

Prior isoprene emissions

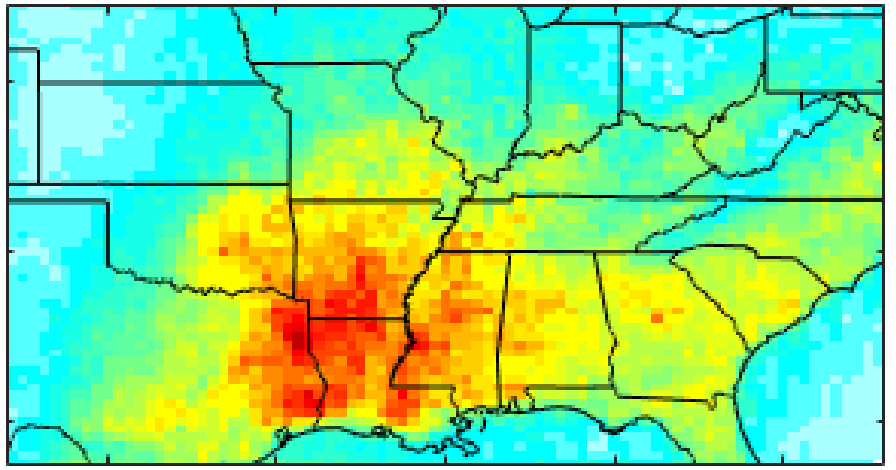


10^{12} atom C cm^{-2} s^{-1}

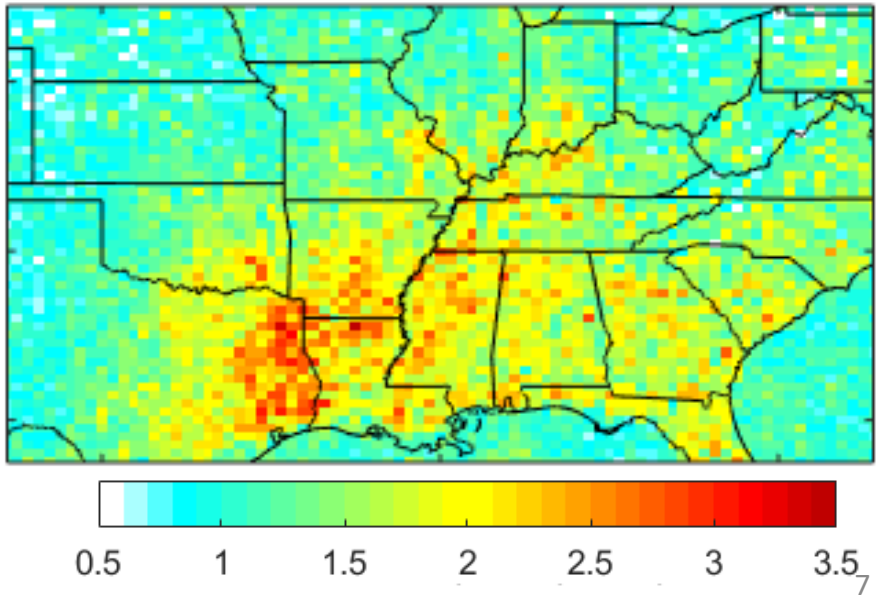
Forward model

Inverse analysis

Modeled HCHO column



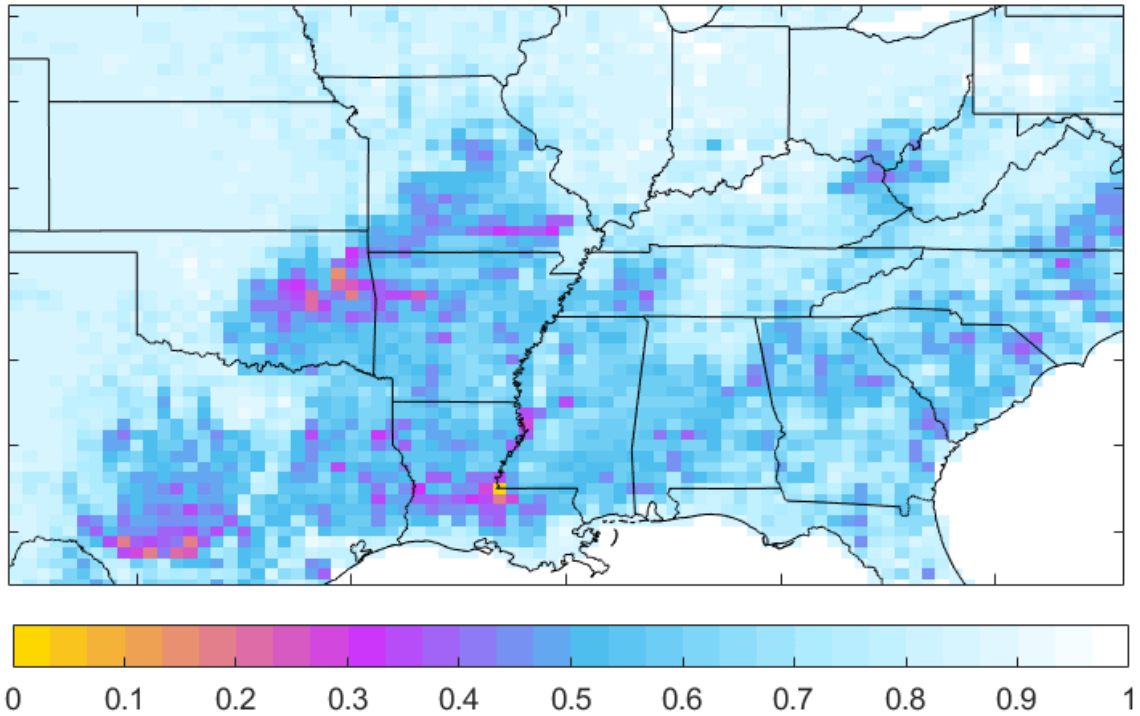
Observed HCHO column



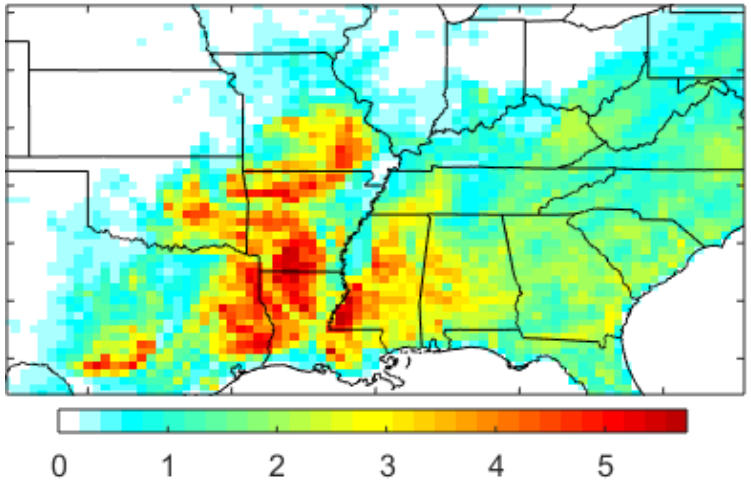
10^{16} molecules cm^{-2}

HCHO observations indicate modeled isoprene emissions are biased high by 40%.

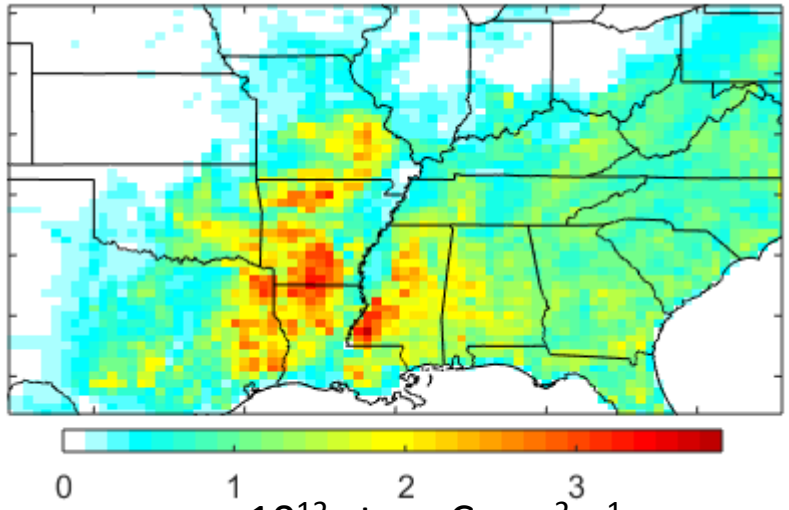
Optimal scaling factors



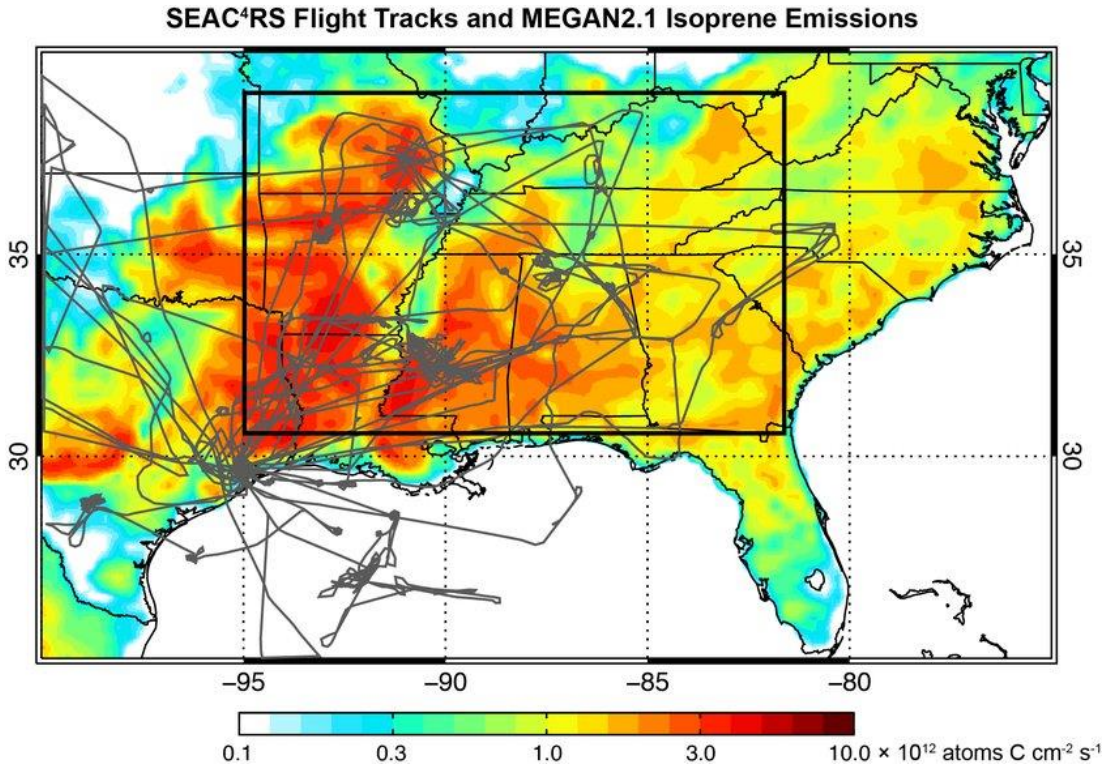
Prior isoprene emissions



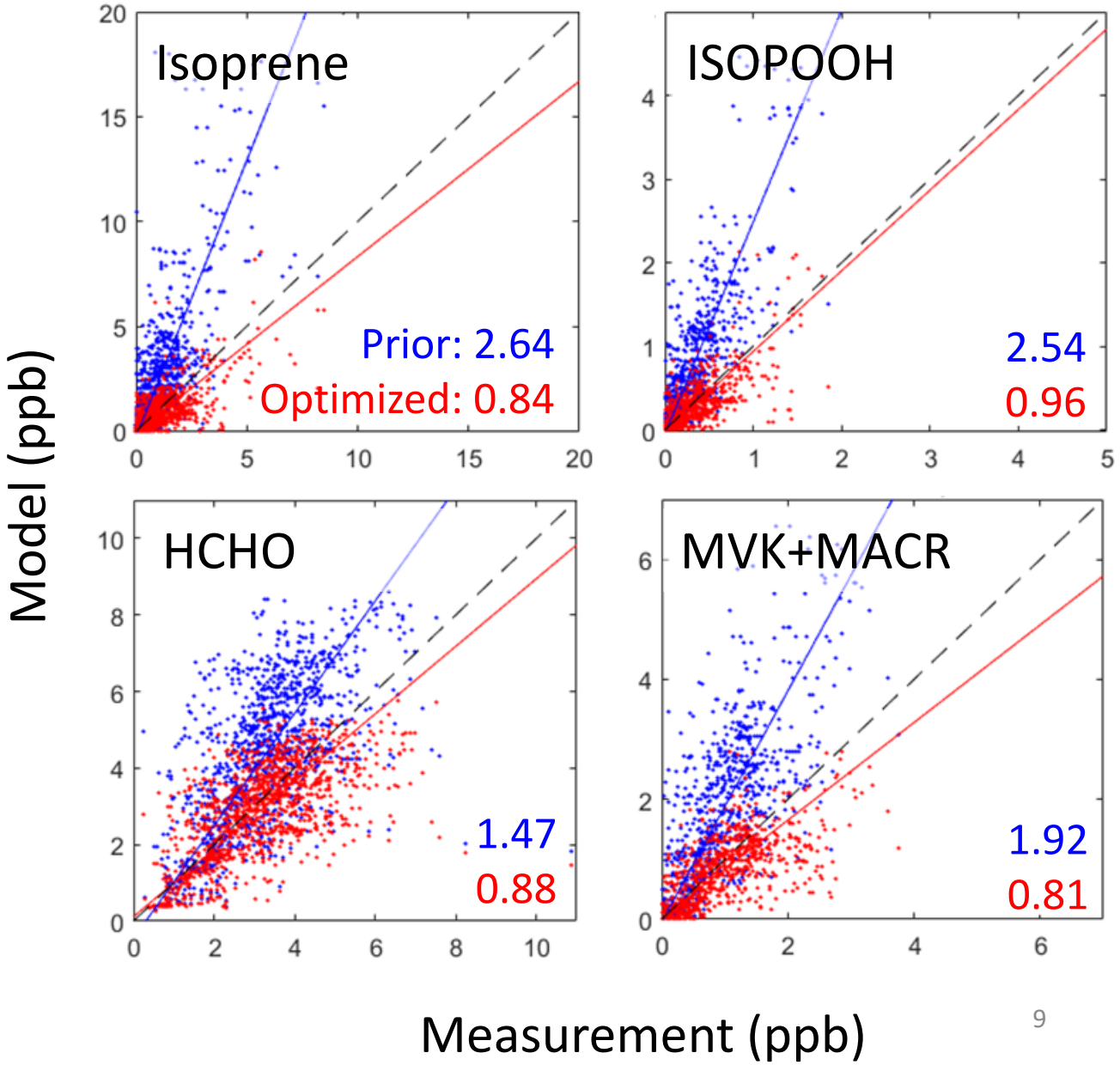
Optimized emissions



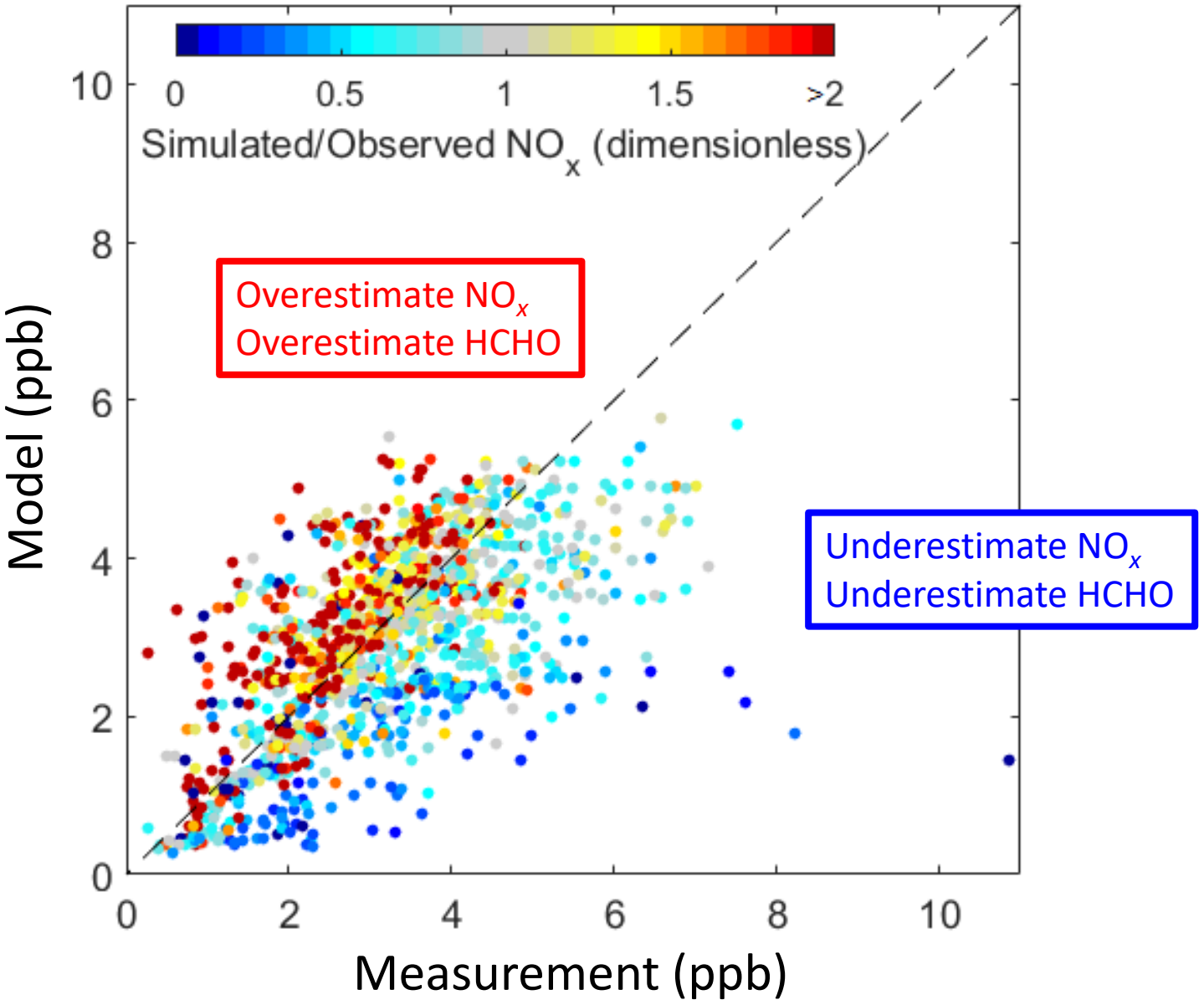
Optimized emissions produce better agreement with SEAC^{4}RS



Kim et al., 2015

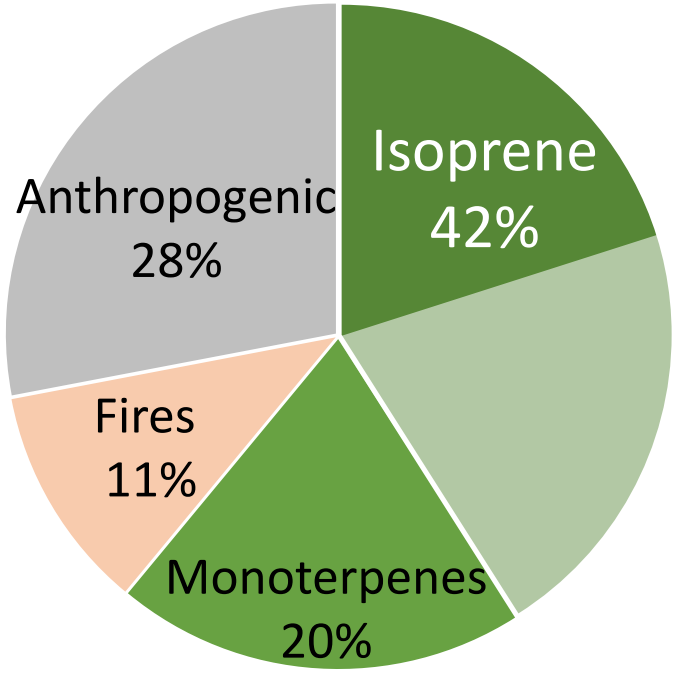


Right NO_x is crucial for accurate inversions



Implications for air quality

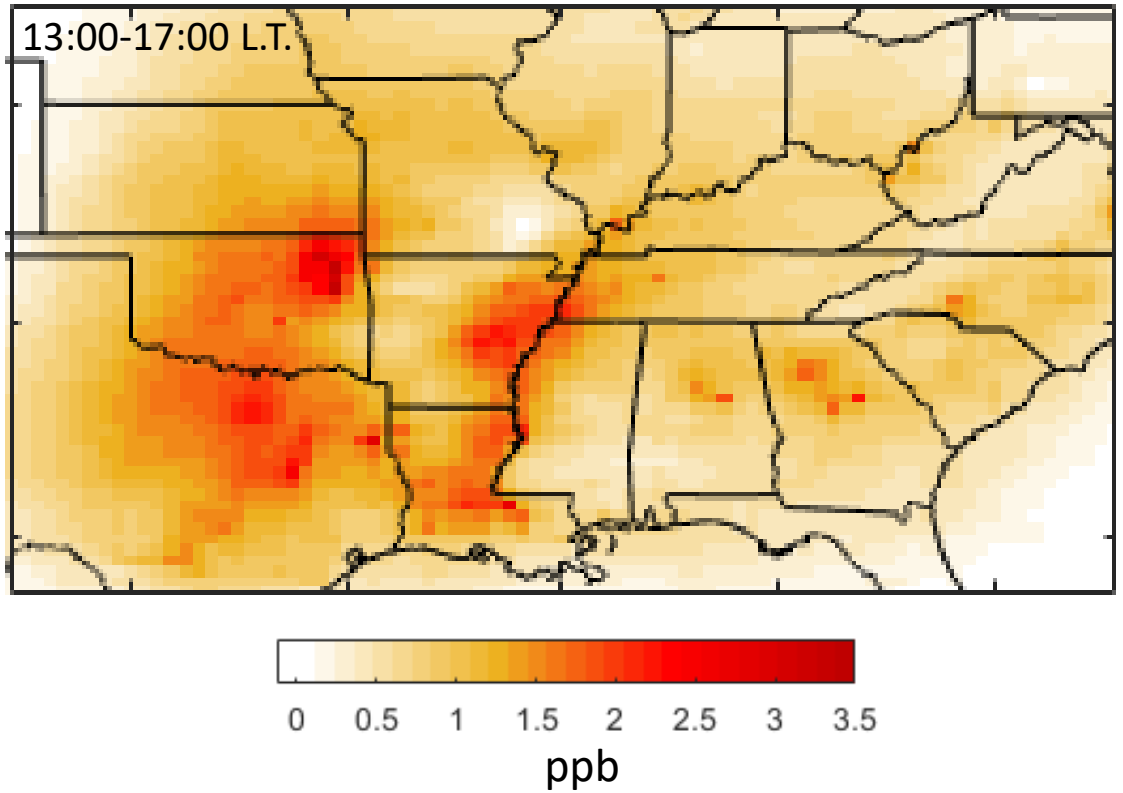
Shifting organic aerosol sources



Halved!

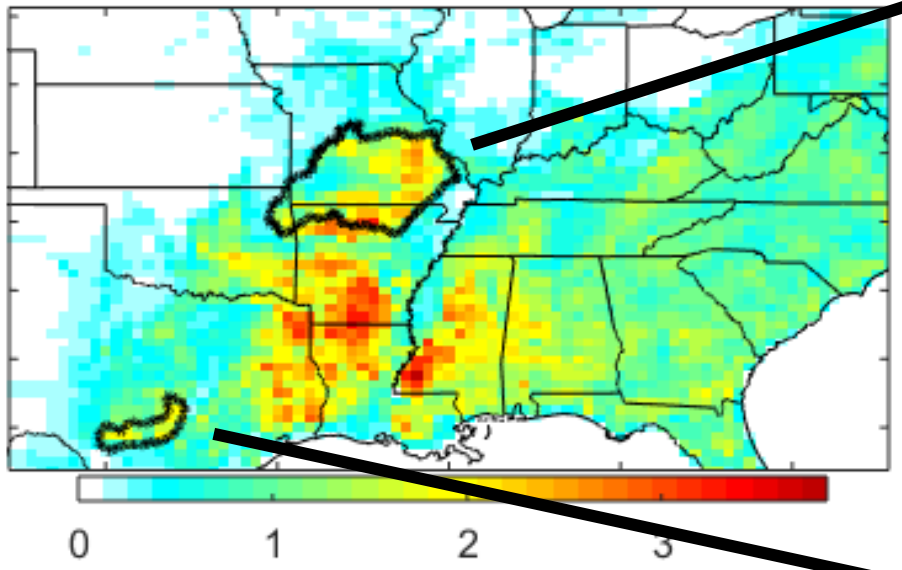
Kim et al., 2015

Reduction in surface O_3



Evaluating updated emission inventories

Optimized emissions



10^{12} atom C cm^{-2} s^{-1}

Ozarks: 46 reduction%

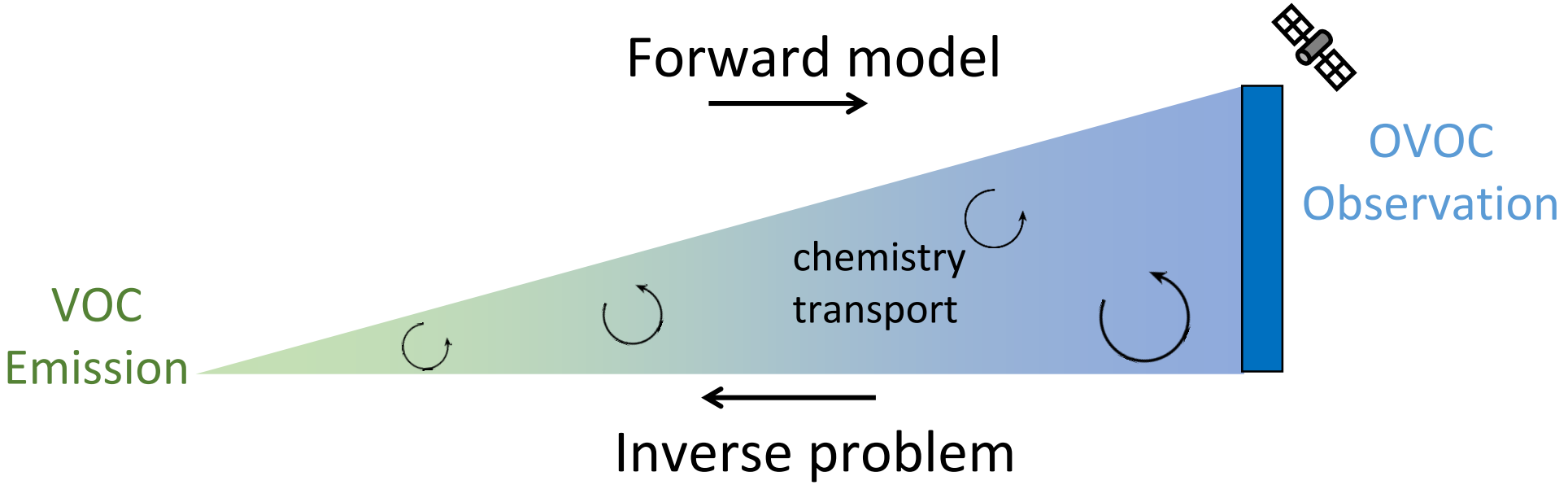
- Dense mixed pine-oak forest
- Vertical heterogeneity in canopy structure?

Yu et al., 2017.

Edwards Plateau: factor of 3 reduction

- Oak/Juniper/Elm
- Fraction of forest cover varies widely in landcover maps

Extending this framework:



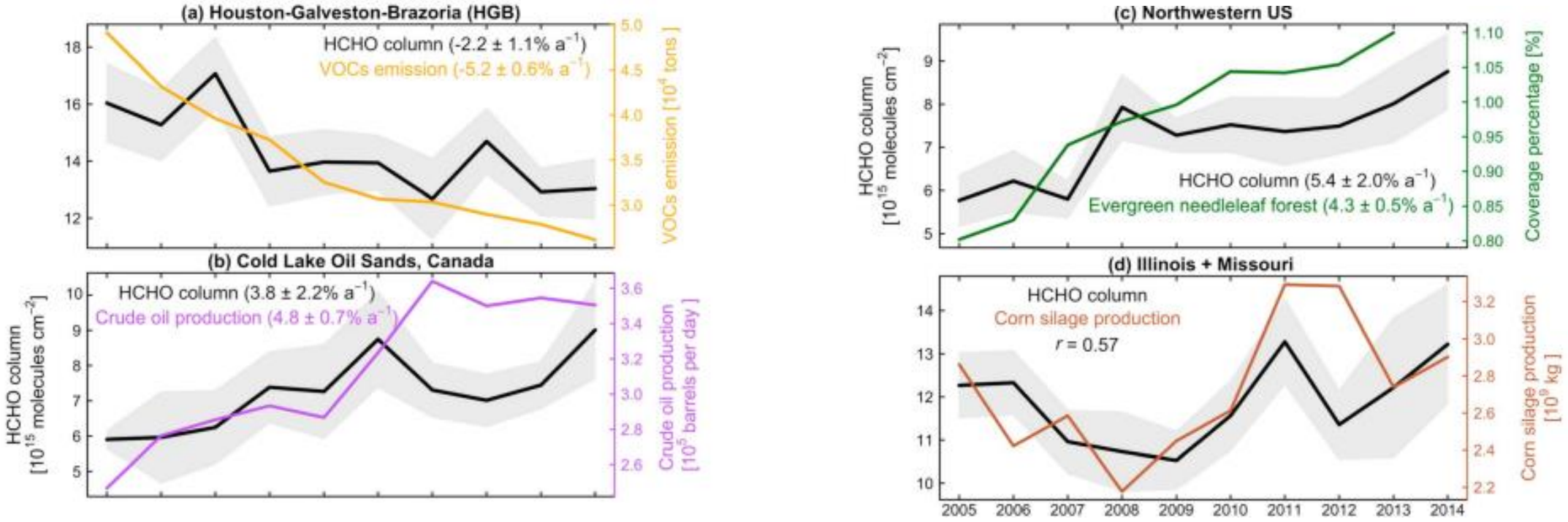
Are OVOC yields well constrained?

- What is the status for HCHO yields from other VOCs?
- What other OVOCs would be useful?

What are the uncertainties in our observations?

- The next generation of satellites

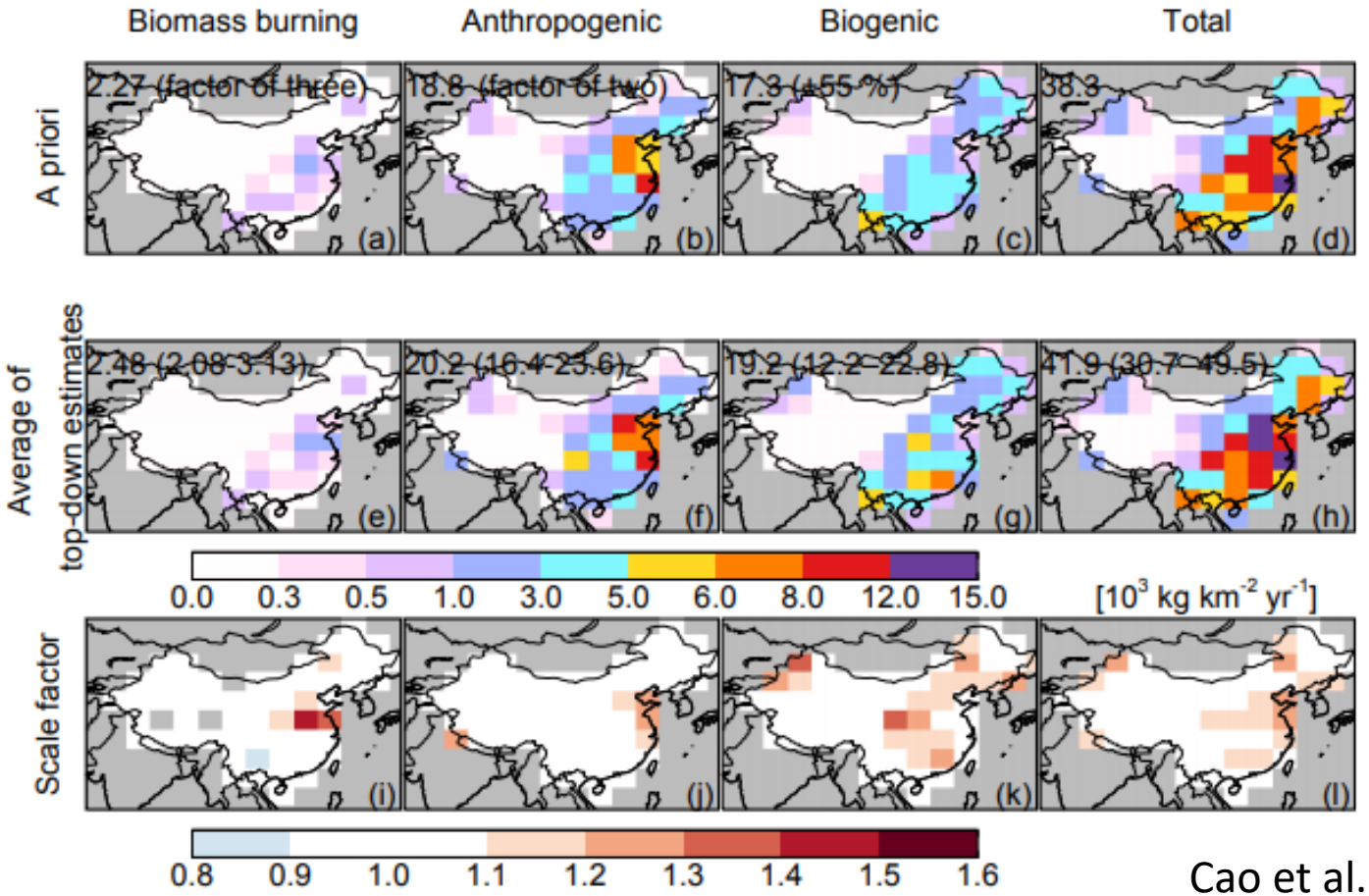
Using HCHO to constrain other VOCs



What *are* the HCHO precursors?
What are the OVOC yields?
Are direct emissions significant?

Zhu et al., 2017

Using glyoxal and formaldehyde together



Cao et al., *ACP,* 2018

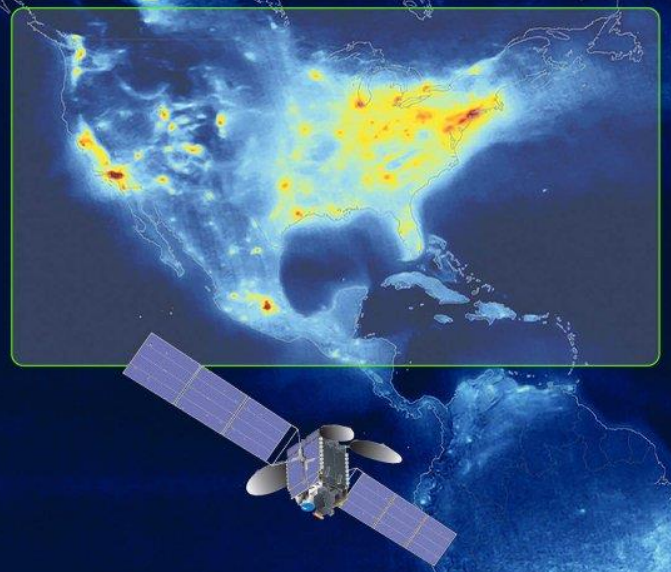
Mechanism based on MCM v 3.1

NOx dependency of glyoxal yield across VOCs has received much less attention.

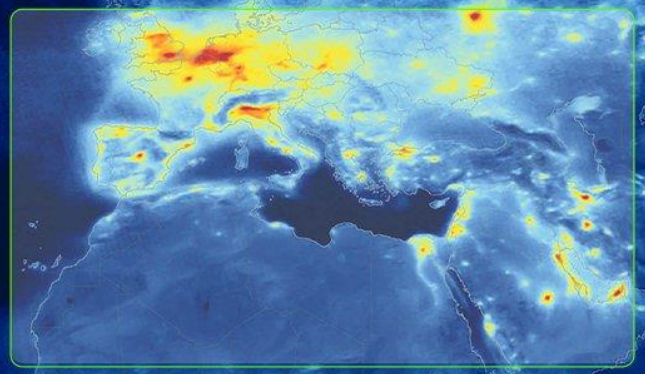
The differing yields provide new information to constrain top-down studies.

The context of this work: a new era of satellites

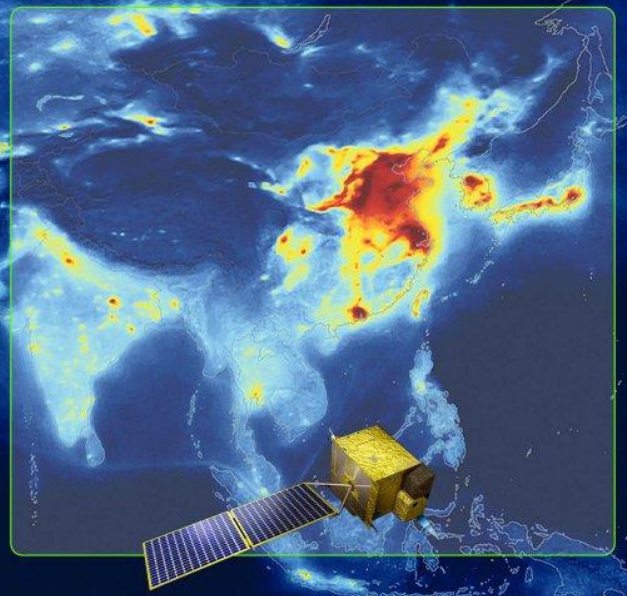
TEMPO *(hourly)*
Tropospheric Emissions:
Monitoring of Pollution



Sentinel-4 *(hourly)*



GEMS *(hourly)*
Geostationary Environmental
Monitoring Spectrometer



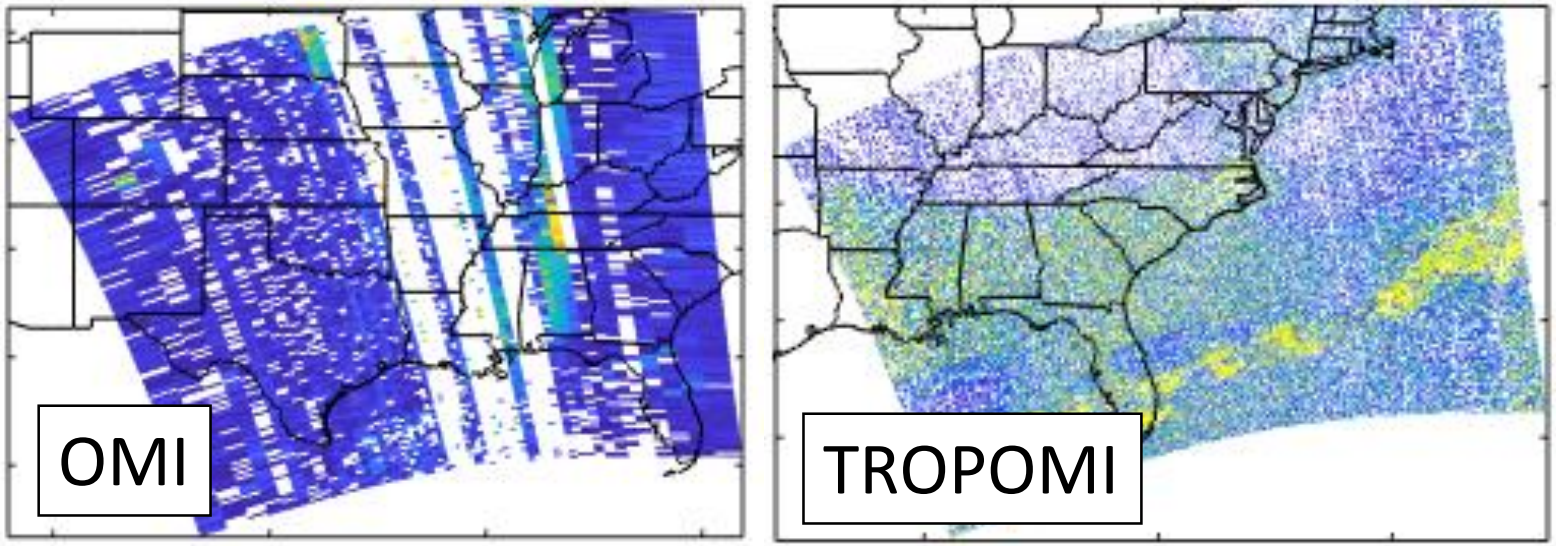
Sentinel-5P *(once per day)*

GaoFen-5 *(once per day)*

TROPOMI is on-line!

https://s5phub.copernicus.eu

Oct 27 HCHO VCDs
(unfiltered for QC)



More pixels… More *successful* pixels… More information!

Acknowledgements

- SENEX team
- SEAC^{4}RS team
- Daniel Jacob
- Harvard Atmospheric Chemistry Modeling Group
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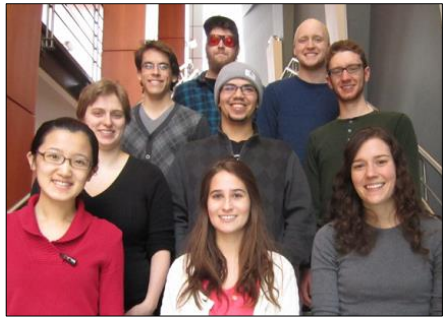

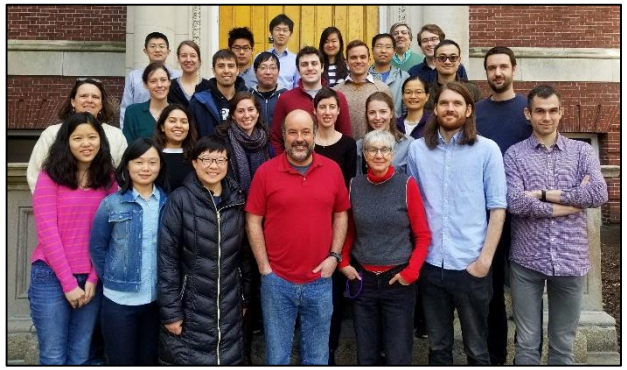

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