Adjoint-based inverse modeling of OMI HCHO to constrain isoprene emissions

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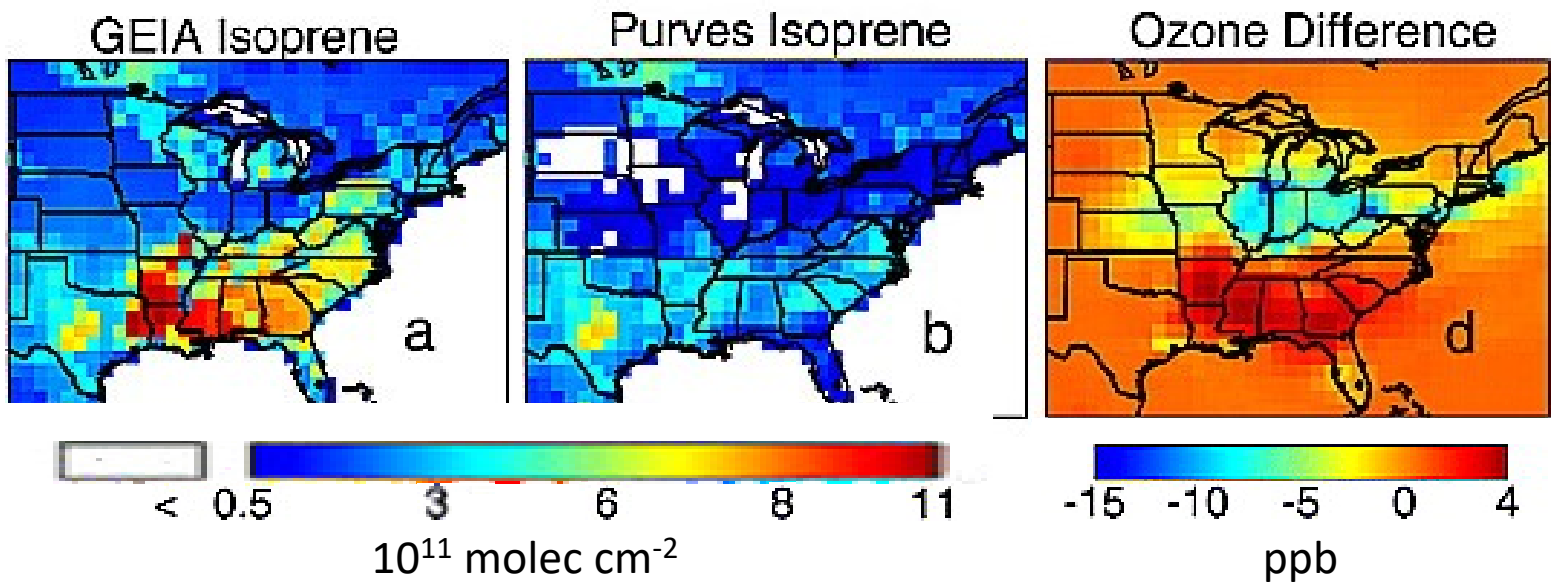
TEMPO science meeting

1 June 2017

Why isoprene?

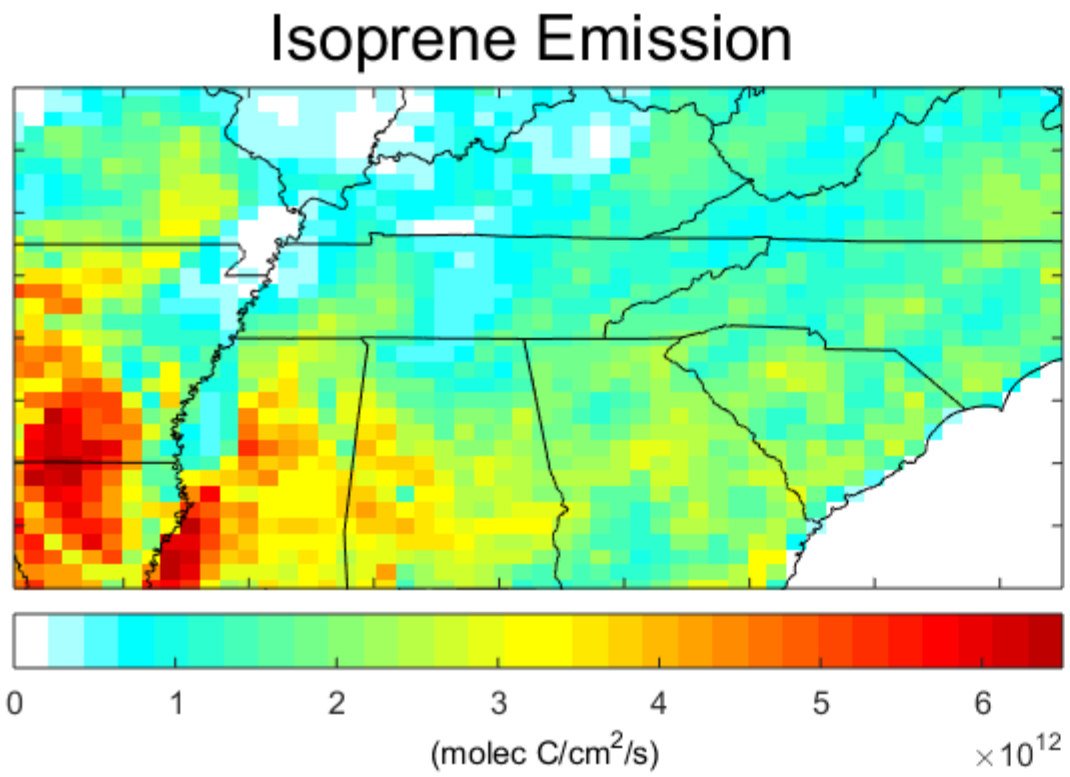


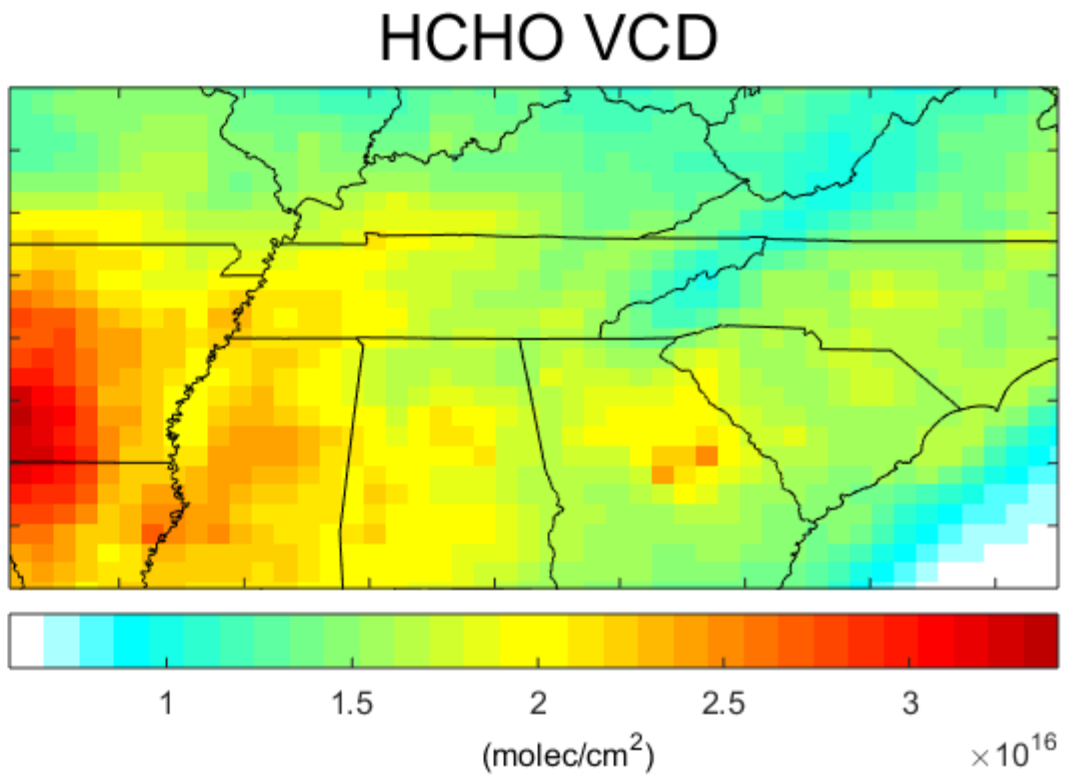
Fiore et al. (2005)

Isoprene is a major precursor to secondary pollutants.
Uncertainty in bottom-up inventories is at least a factor of 2.

HCHO-E_{ISOP} Link

GEOS-Chem 1-14 August 2013:

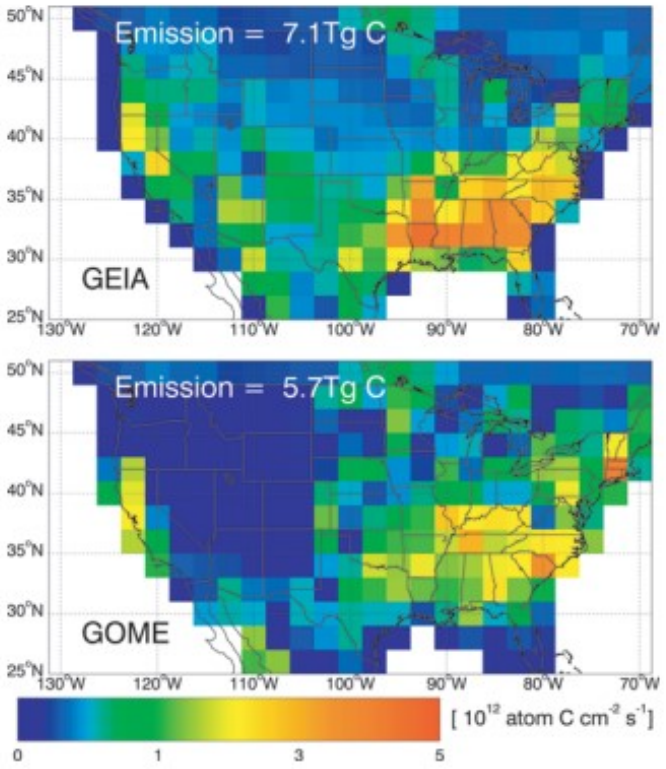




E_{ISOP} variability drives HCHO variability--
HCHO provides a top-down constraint on isoprene emissions.

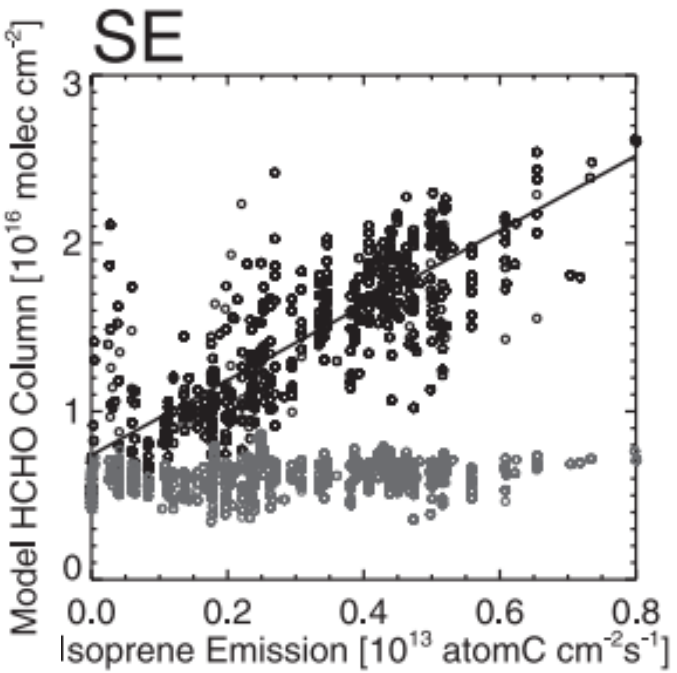
Linear approximations

$$\Omega_{HCHO} = SE_{isoprene} + B$$



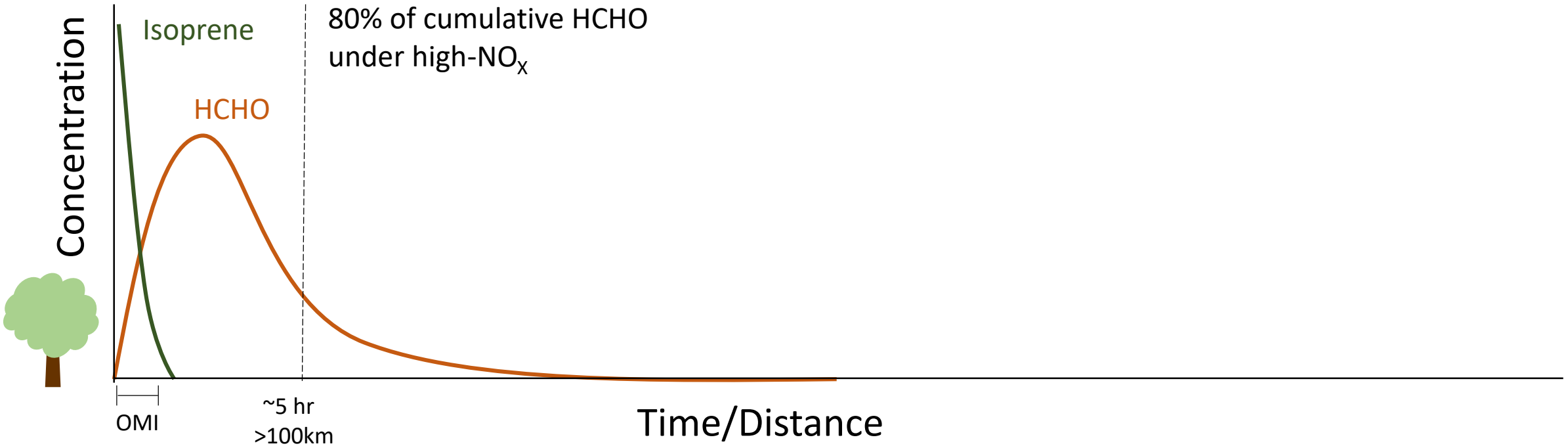
se GEOS-Chem
derive slope

Apply slope to
observed Ω_{HCHO}

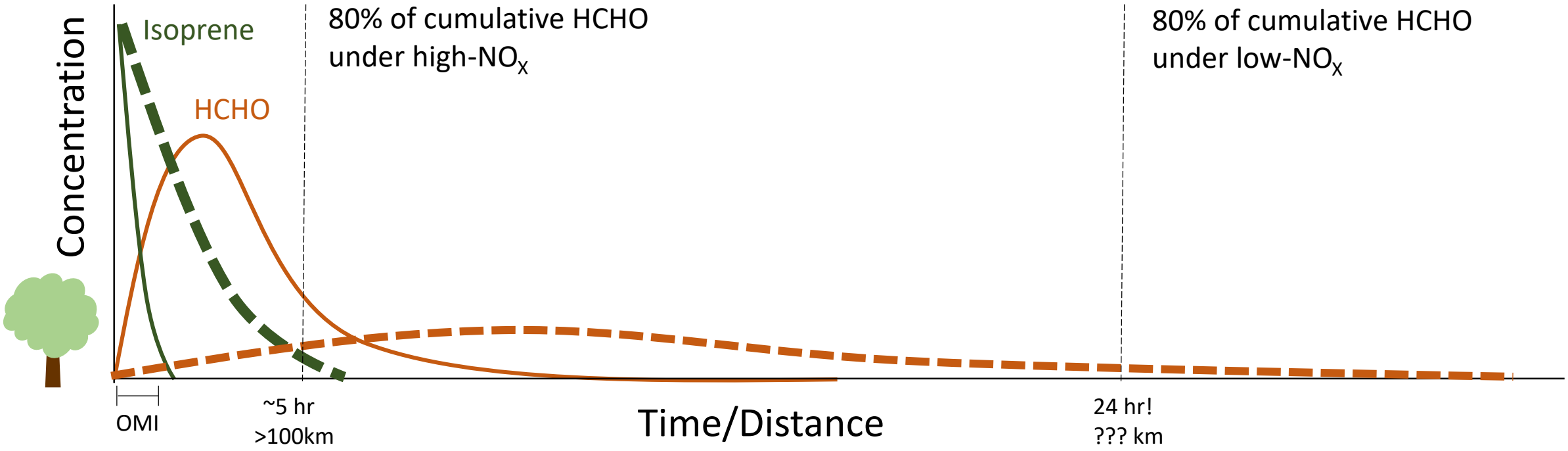


Palmer et al. (2003)

The smearing problem



The smearing problem



Fine resolution analysis not possible with linear approach!

Adjoint-based approach

Minimize cost function J(x):

A priori term

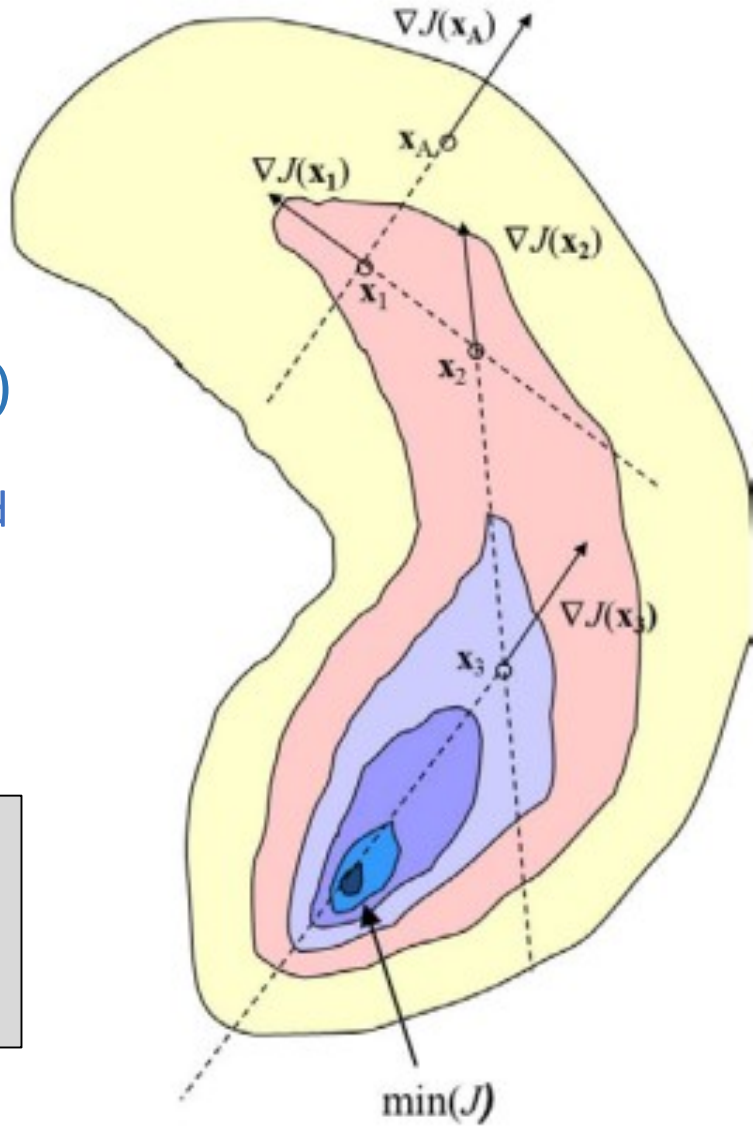
Observational term

$$J(x) = (x - x_A)^T S_A^{-1} (x - x_A) + (F(x) - y)^T S_O^{-1} (F(x) - y)$$

How different are modeled and optimized isoprene emissions?

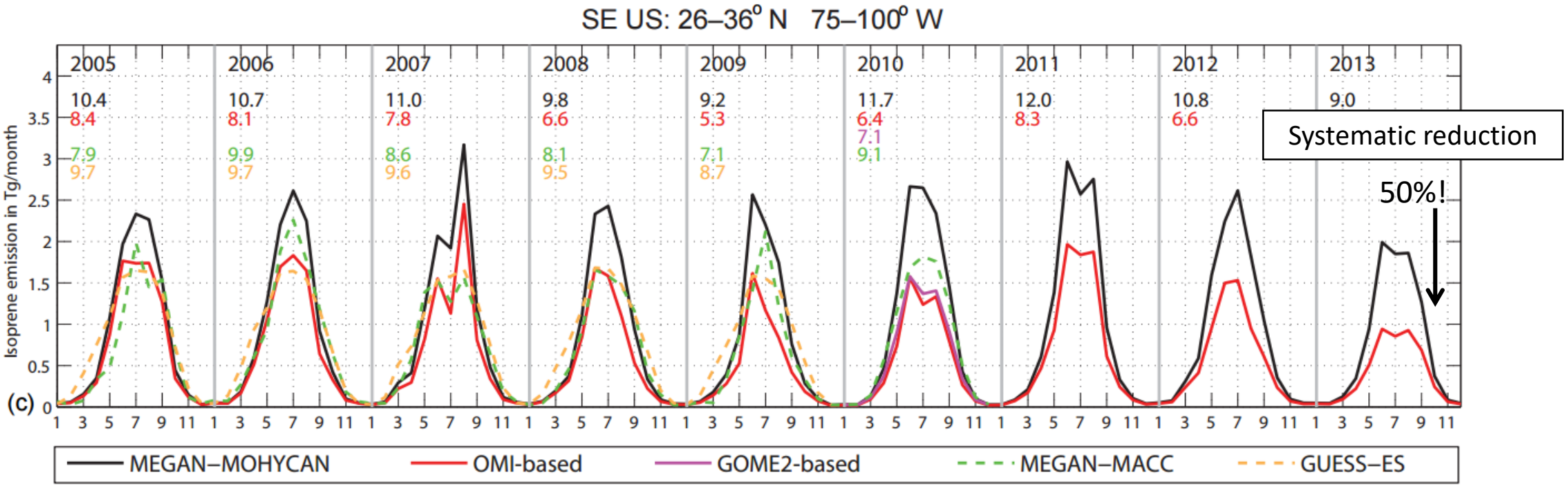
How different are modeled and measured HCHO columns?

(1) Retain information from bottom-up estimates
(2) Error is taken into account
(3) Spatial displacement of E_{ISOP} and HCHO fully represented



Brasseur and Jacob (2017)

Adjoint-based approach



Bauwens et al. (2016)

Let's test this as higher resolution using best knowledge from SEAC^{4}RS!

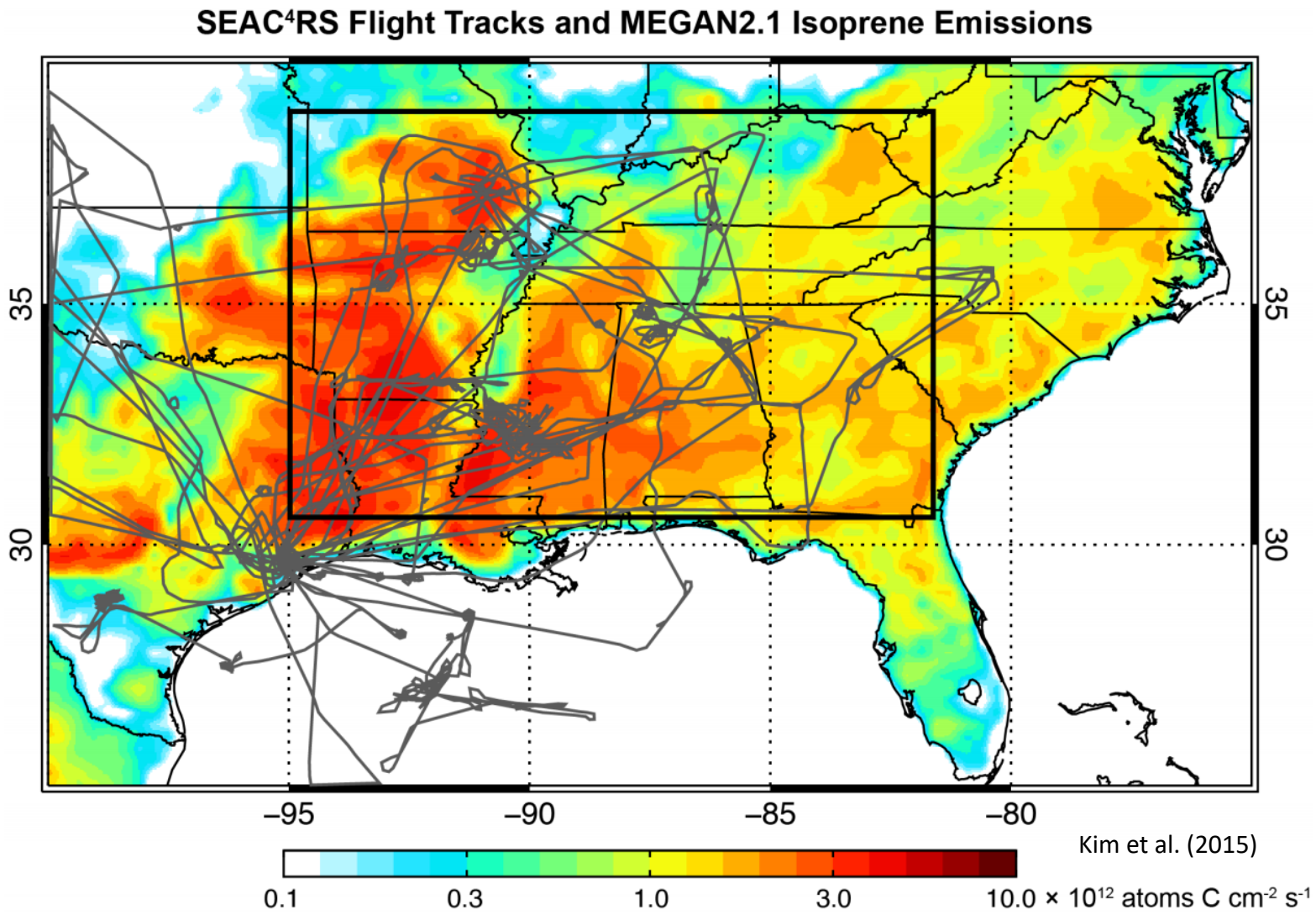
Lessons from SEAC^{4}RS

(1) Impact of resolution

(2) Confidence in our isoprene oxidation mechanism

(3) Constraints on other influential factors

(4) Correcting biased satellite retrievals



Kim et al. (2015)

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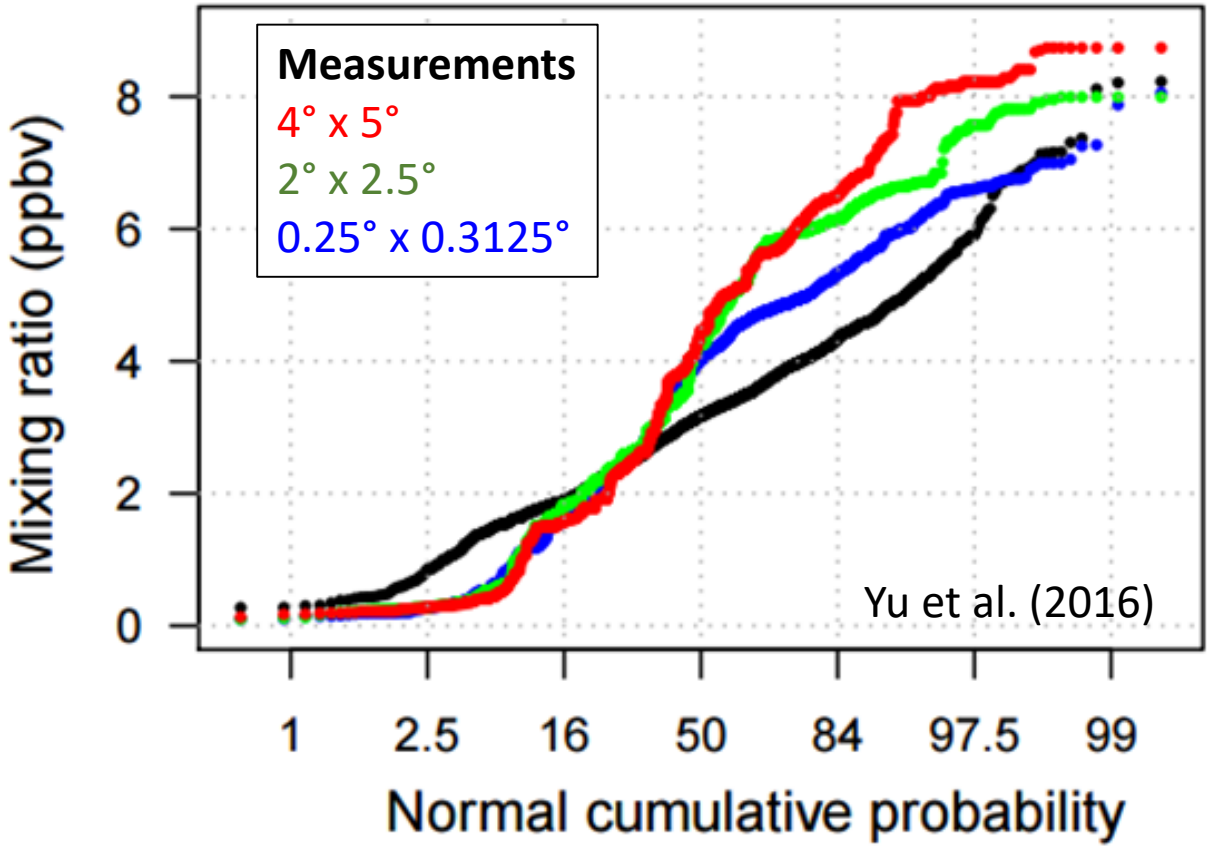

Fine resolution captures isoprene/NOx segregation, and is needed to explore heterogeneity of E_{ISOP}

Lessons from SEAC^{4}RS

(1) Impact of resolution

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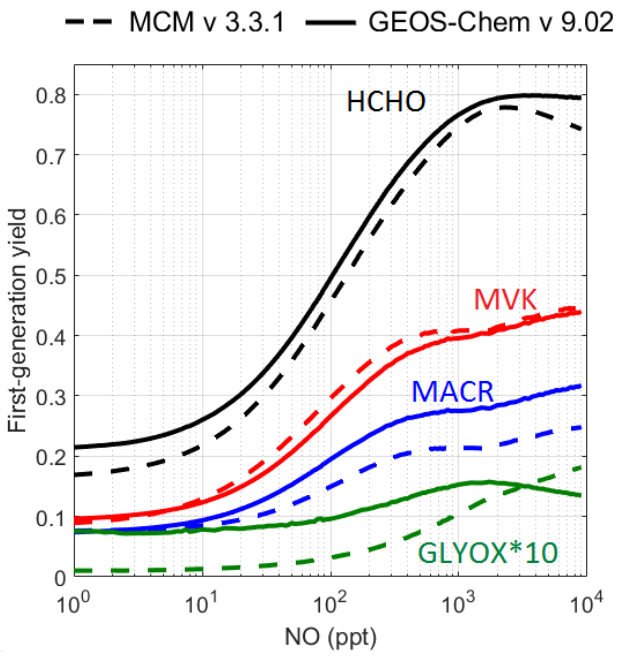
GEOS-Chem is up-to-date and agrees well with observations and fully explicit mechanisms



- Increase of $ISOPO_2 + HO_2$ → ISOPOOH yield
- Update IEPOX + OH reaction rates
- Revise ISOPN + OH reaction products
- Inclusion of HPALD formation, oxidation and photolysis
- Revision of ISOPO2 self-reaction rate and productions
- …

Fisher et al. (2016); Kim et al. (2015); Marais et al., (2016); Travis et al (2016).

Lessons from SEAC^{4}RS

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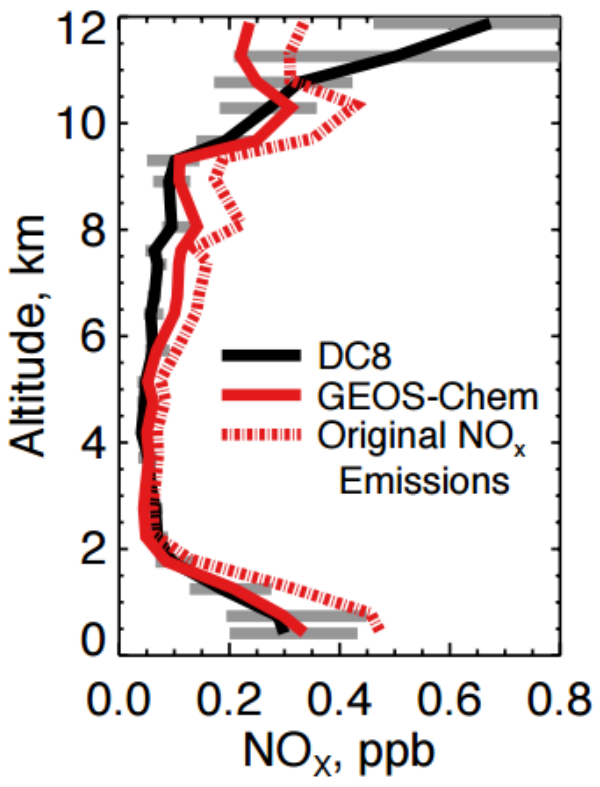
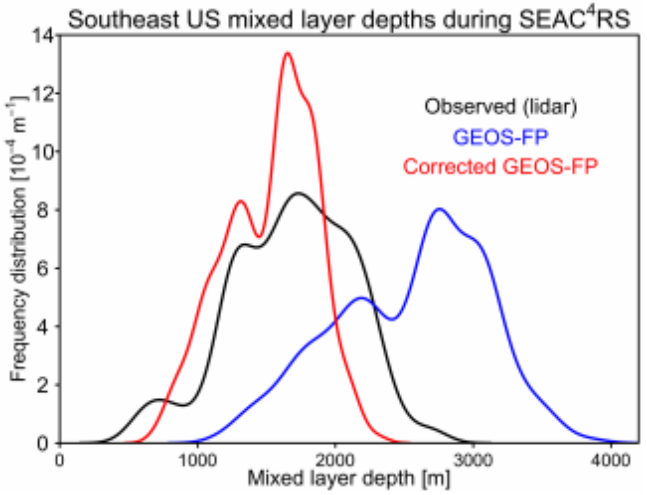
NO_X emissions



Travis et al. (2016)

PBL heights



Zhu et al. (2016)

Exhaustive analysis of the forward model allows us to isolate the role of E_{ISOP}

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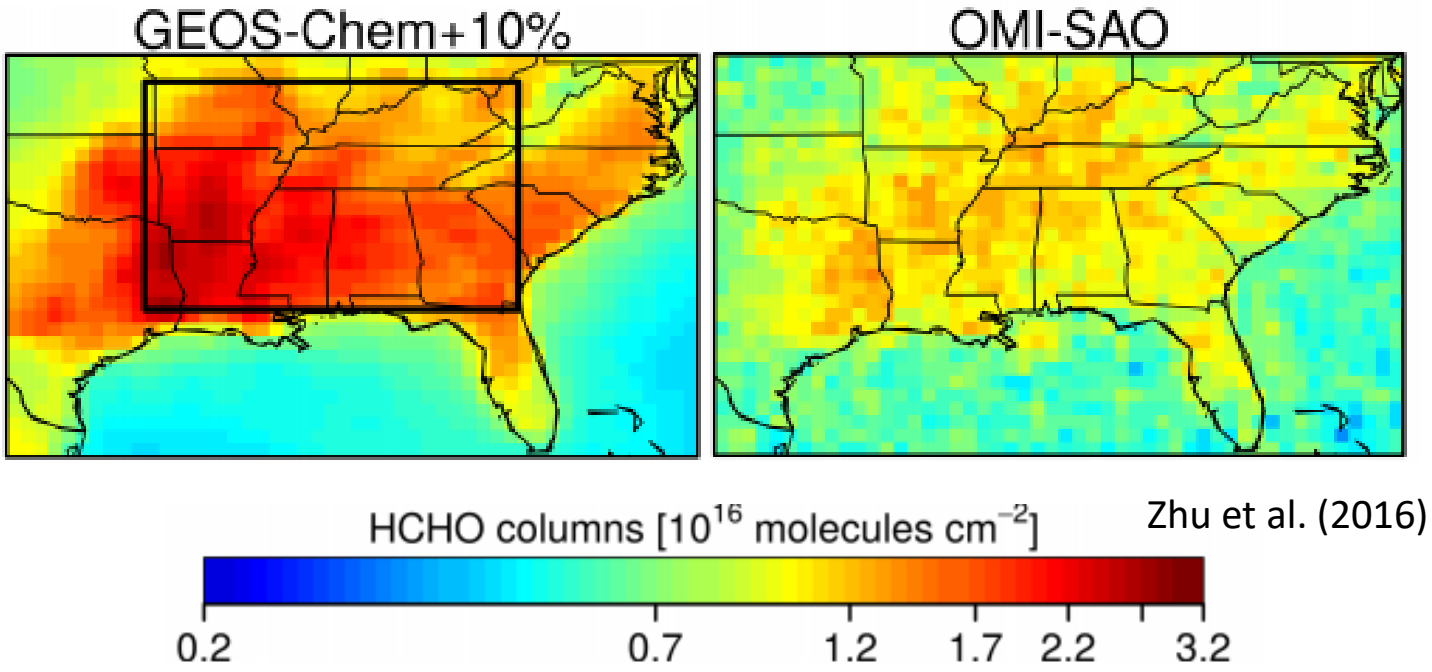


Zhu et al. (2016)

Uniform -37% bias related to spectral fitting and scattering weights

Final notes on the inversion setup

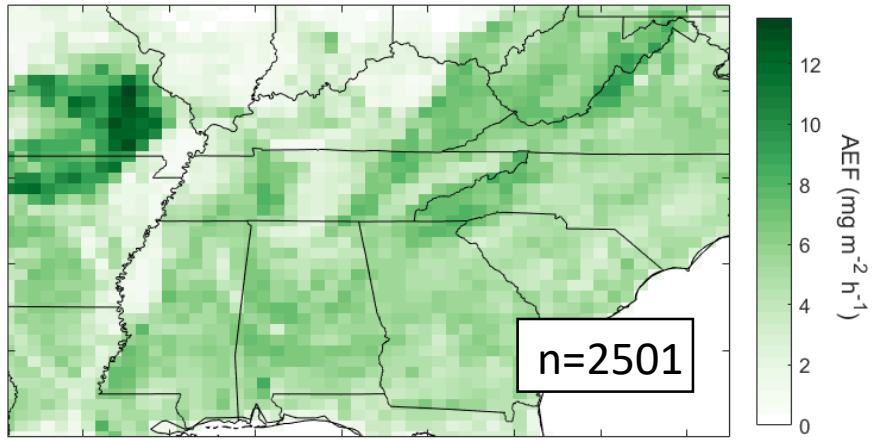
$$J(x) = (x - x_A)^T S_A^{-1} (x - x_A) \quad + \quad (F(x) - y)^T S_O^{-1} (F(x) - y)$$

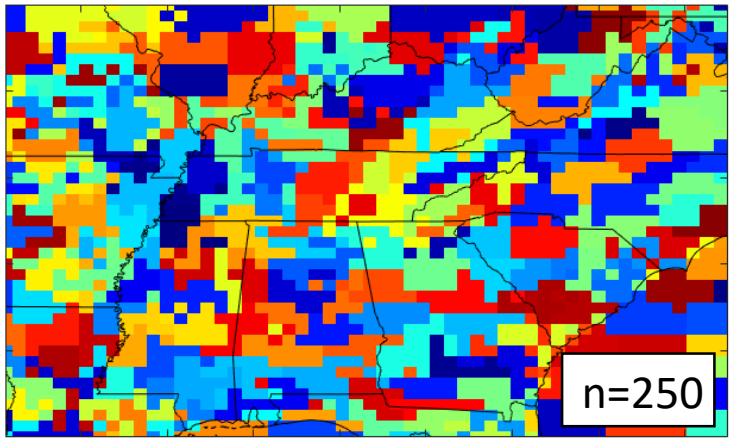
MEGAN v2.1 100% ~70% (RRE method)

Slant column densities

GC SCD = GC VCD x OMI-based AMF

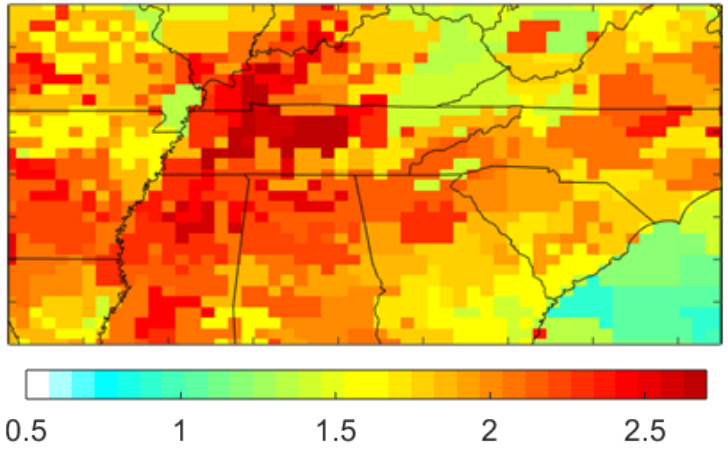
We produce a more relevant coarse grid for calculation of ∇J:





Inversion results

OMI SAO Ω_{HCHO}



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

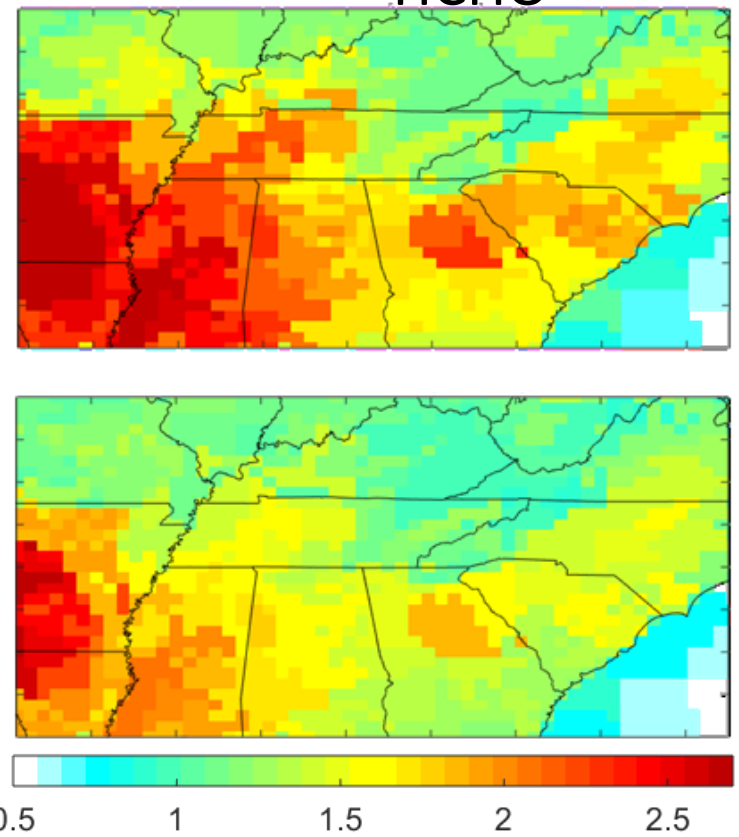
1-14 Aug 2013

GC Ω_{HCHO}

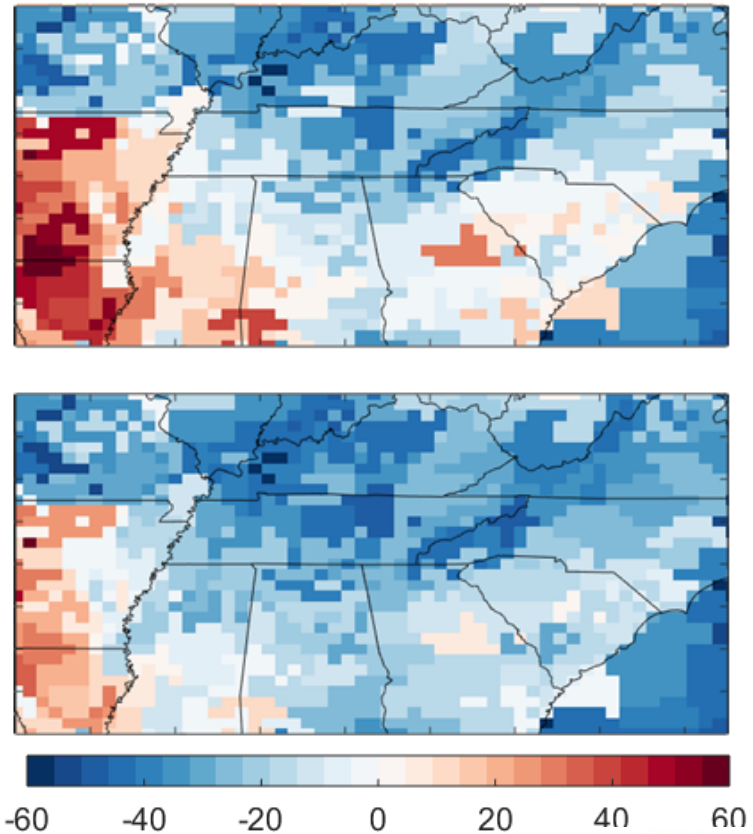
Bias

Prior

Posterior



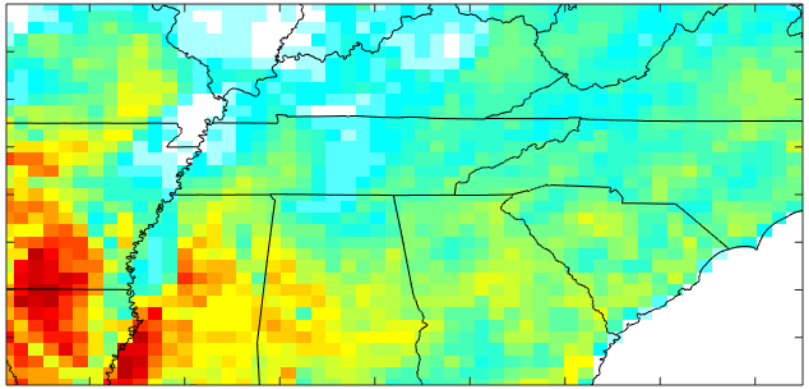
10^{16} molec cm^{-2}



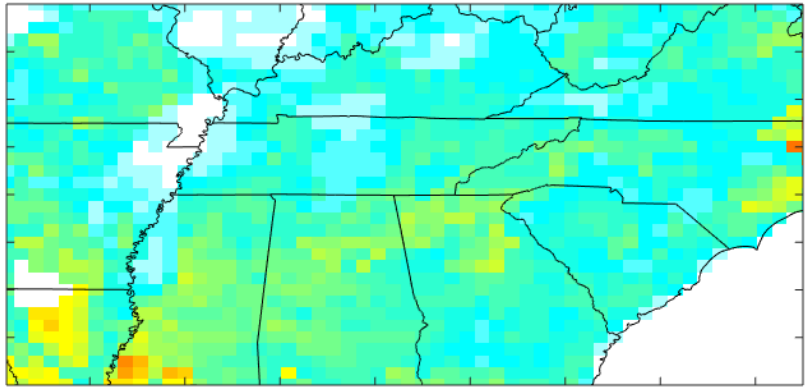
(%)

Inversion results

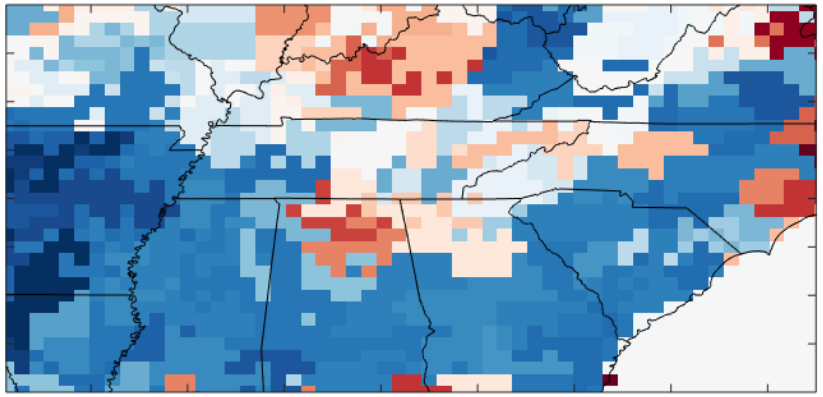
Prior

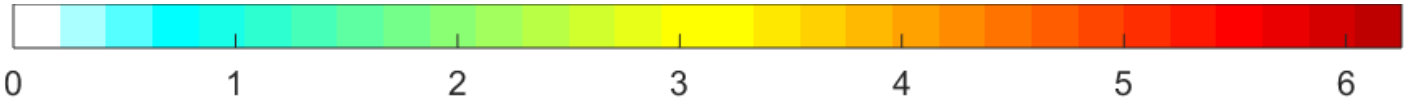


Posterior

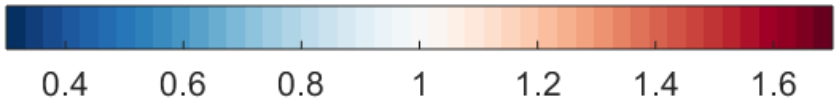


Correction

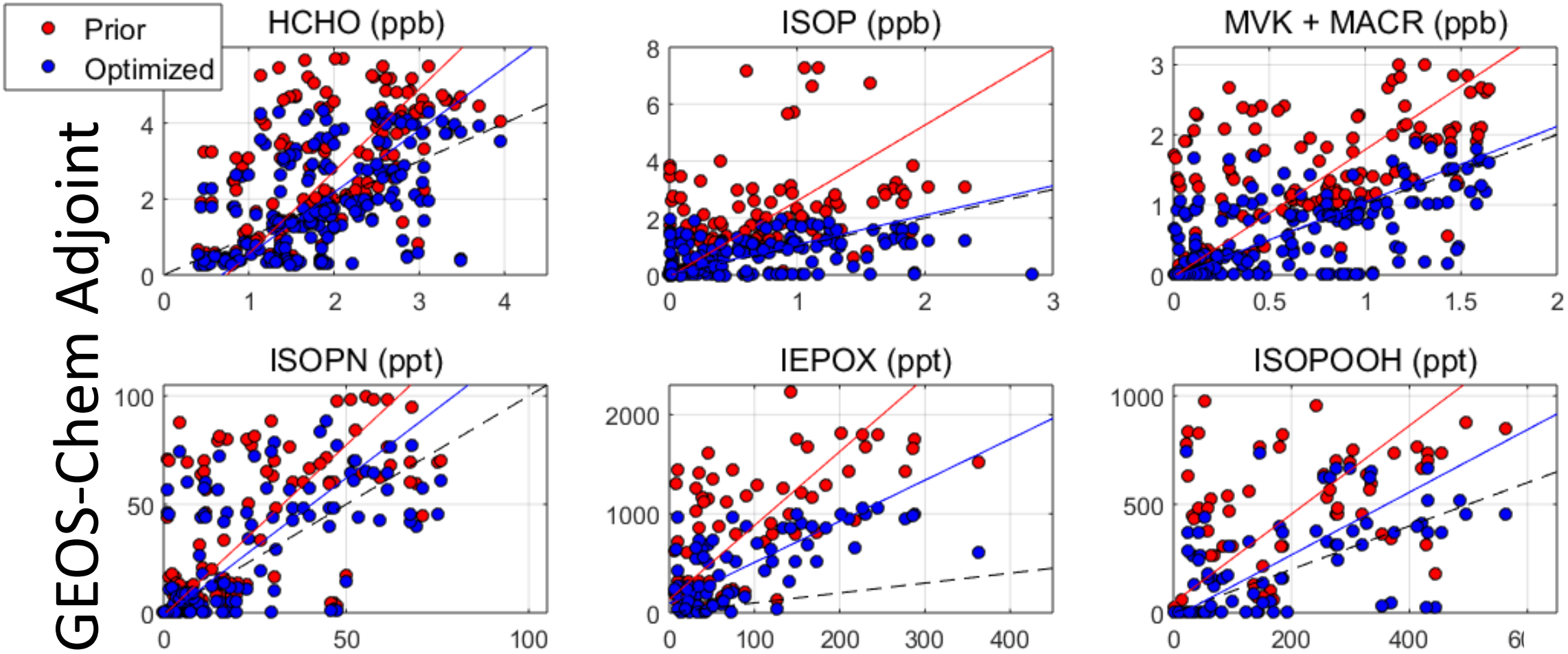




Isoprene emissions (10^{12} atms C cm^{-2} s^{-1})

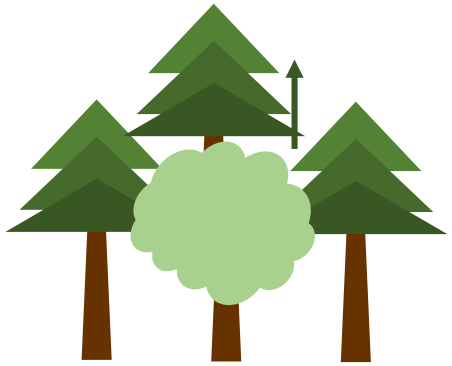


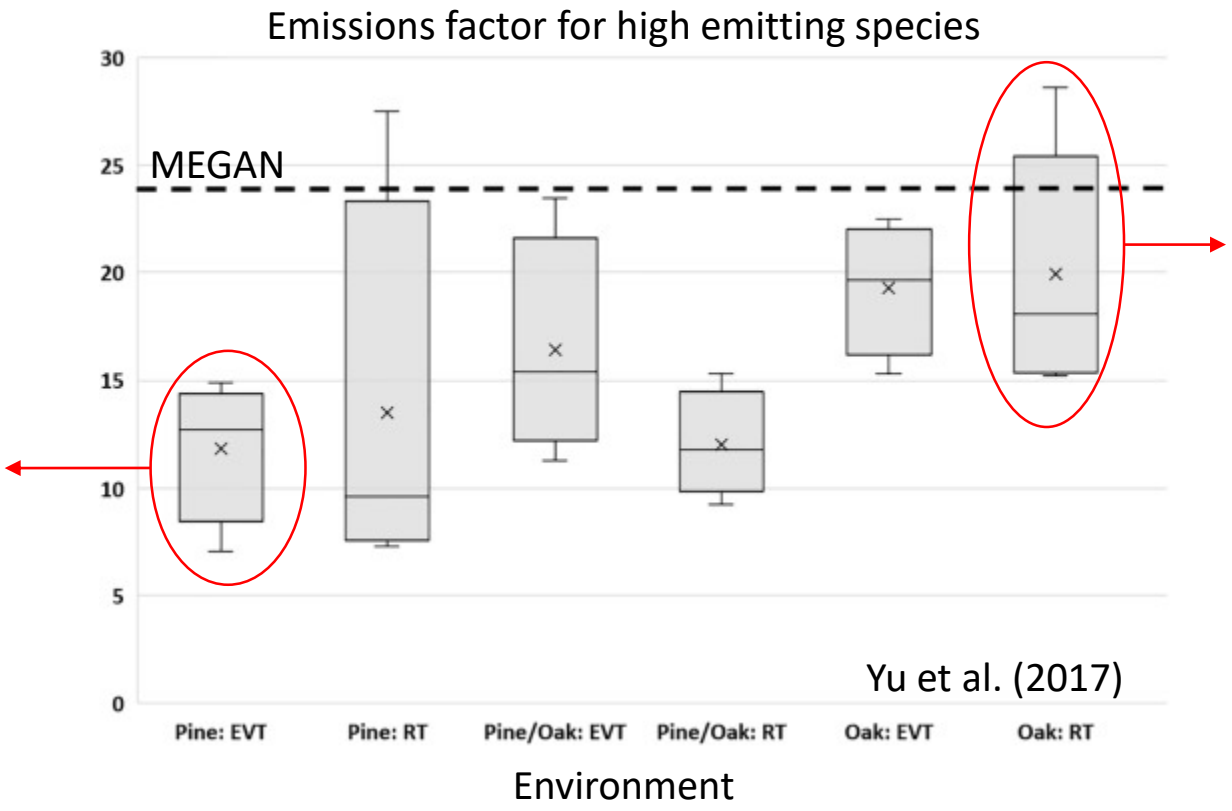
Comparison with SEAC4RS measurements

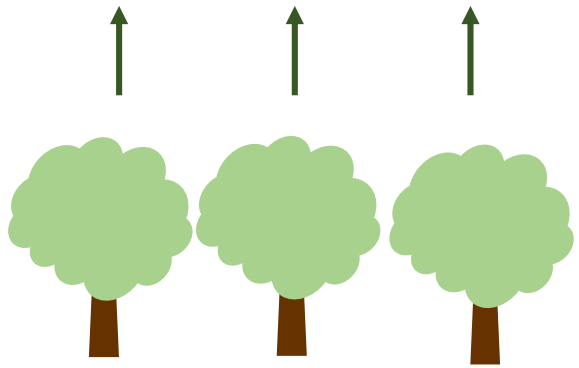


News from the bottom-up world

Emissions ~2x lower that expected

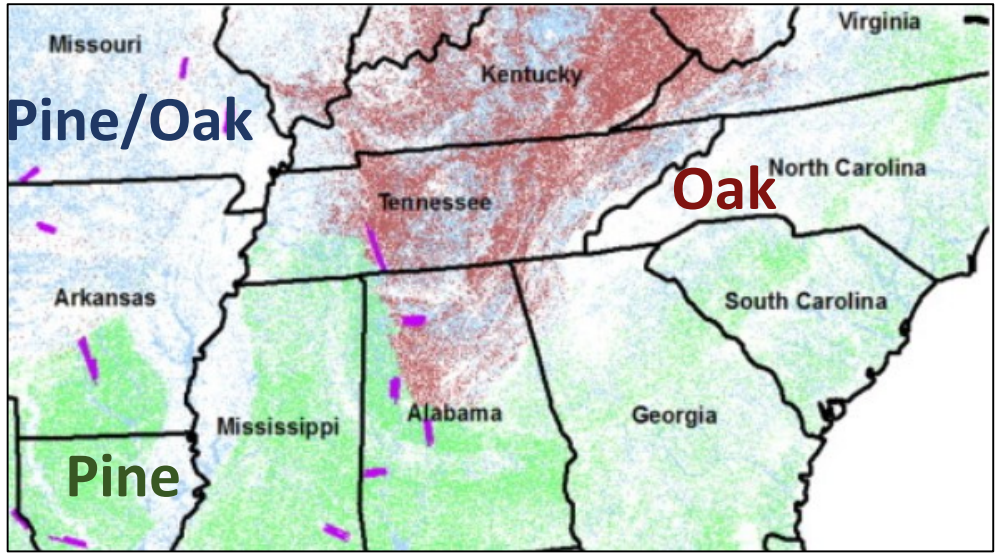




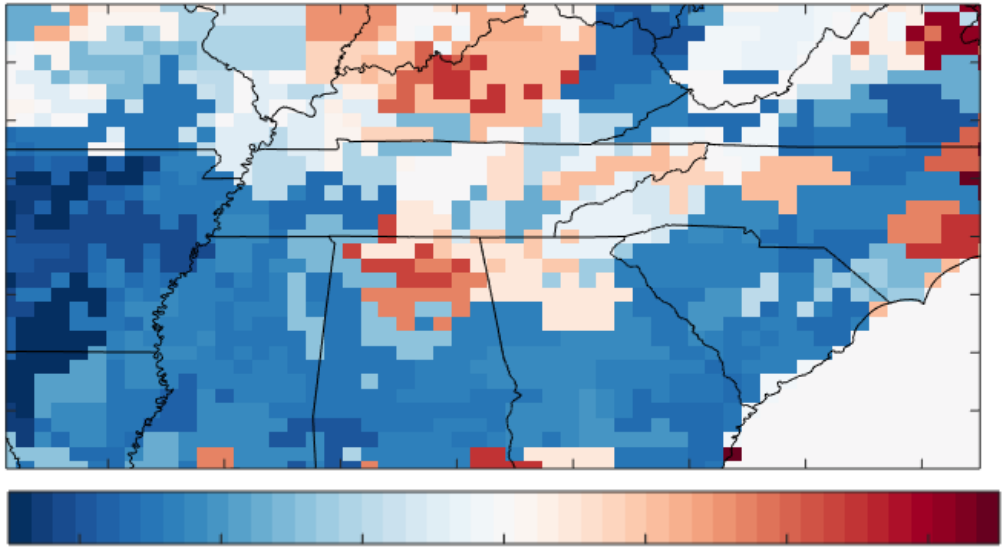


Emissions well captured

News from the bottom-up world



Yu et al. (2017)

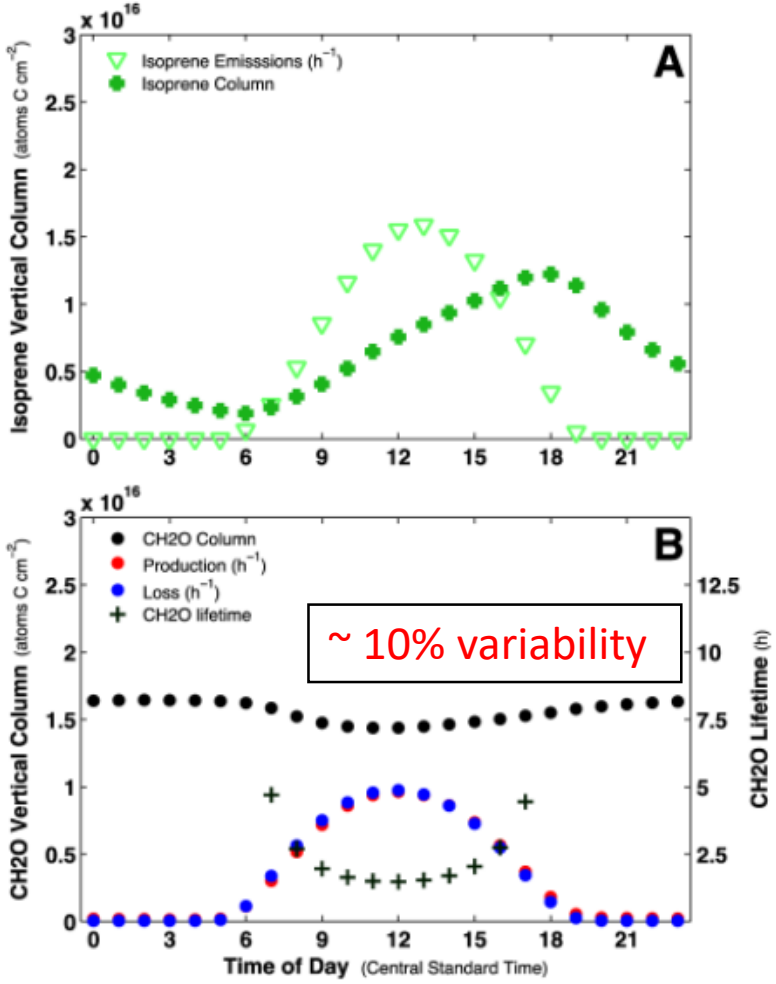


Scaling Factor

Similar conclusions from different methods!

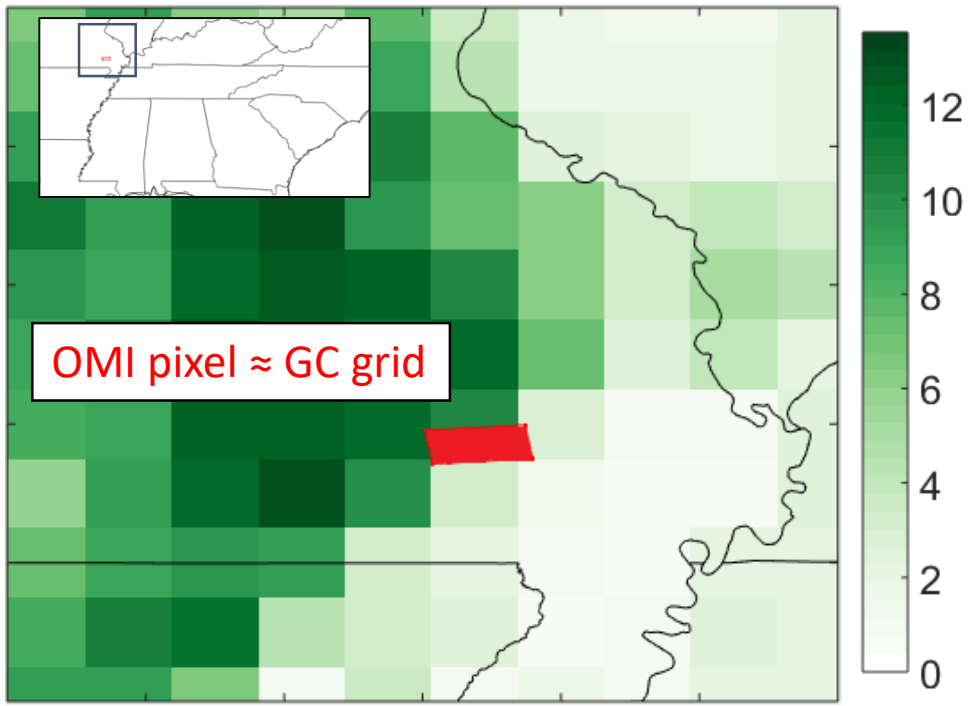
Improvements offered by TEMPO

Diurnal variability



Valin et al. (2016)

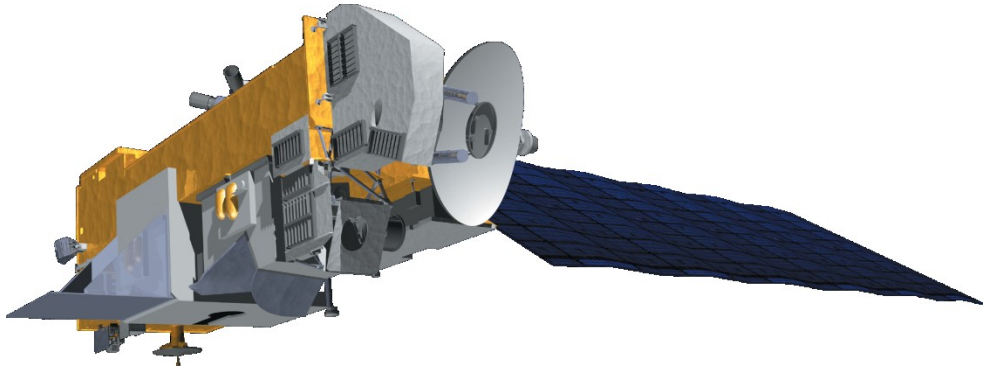
Increased spatial resolution



TEMPO ≈ 75 x more information than OMI

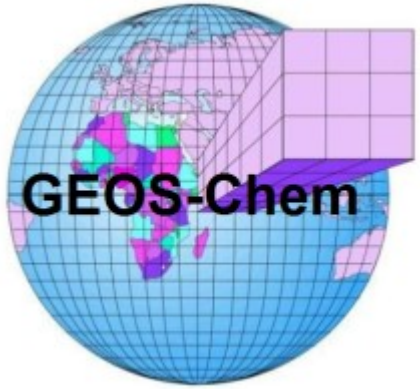
Summary

Recipe for a more accurate and useful top-down isoprene emission estimate:



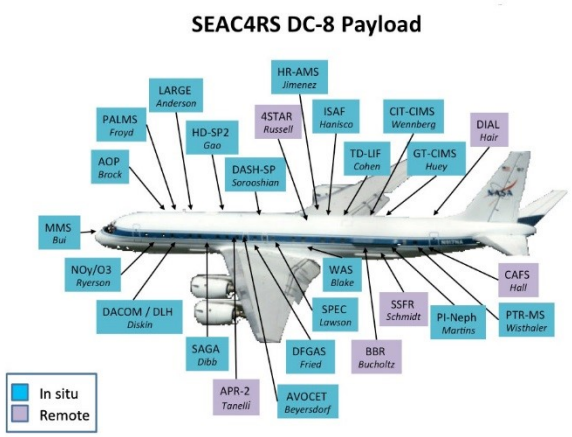
Bias-corrected satellite retrievals

+



Validated fine-resolution model and it's adjoint

+



Plentiful field data

TEMPO retrievals will provide far better constraints for this sort of analysis!

Bottom-up calculations

$$F = \gamma \varepsilon_{isop}$$

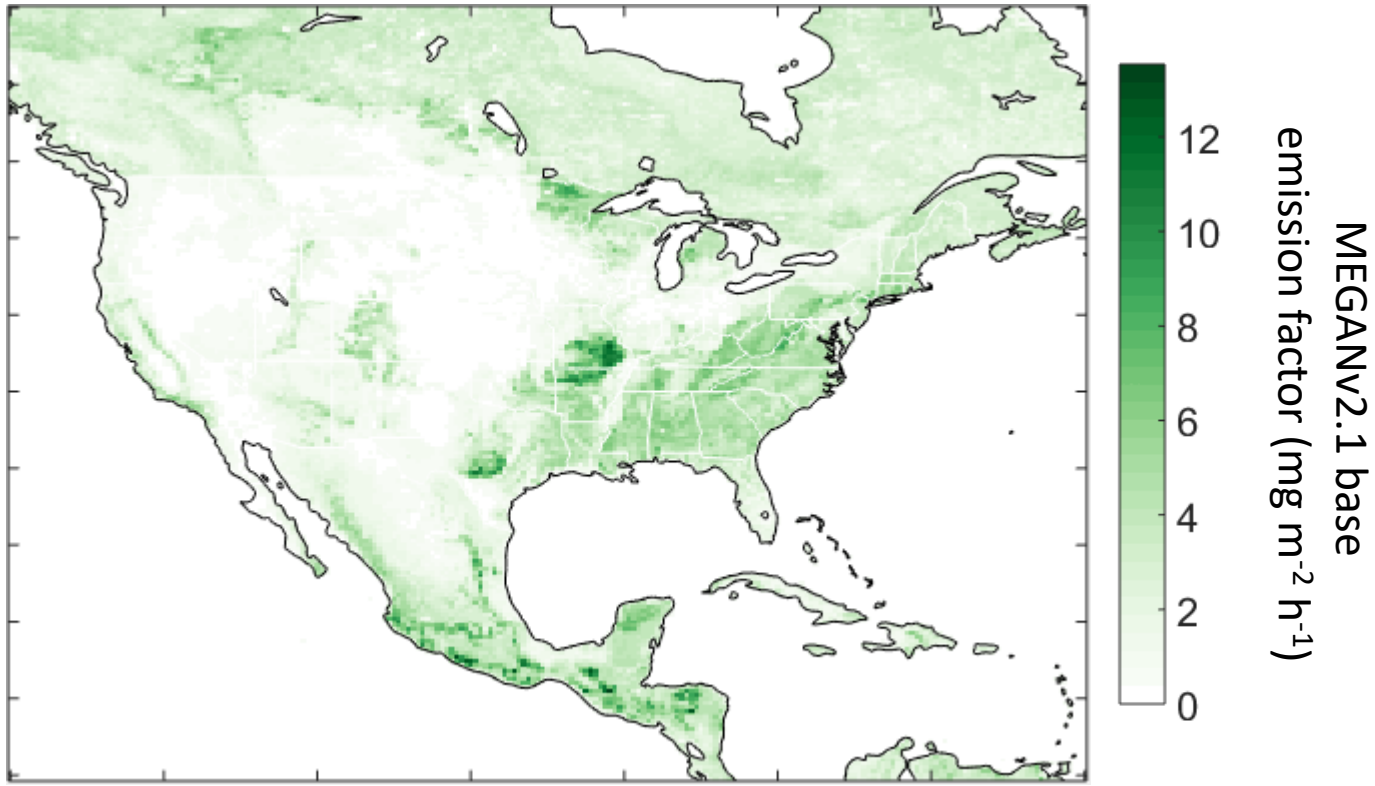
$$\gamma = \gamma_{LAI} \cdot \gamma_{Age} \cdot \gamma_{T} \cdot \gamma_{PAR} \cdot \gamma_{Soil\ Moisture}$$

Uncertainty from:

(1) Limited vegetation data

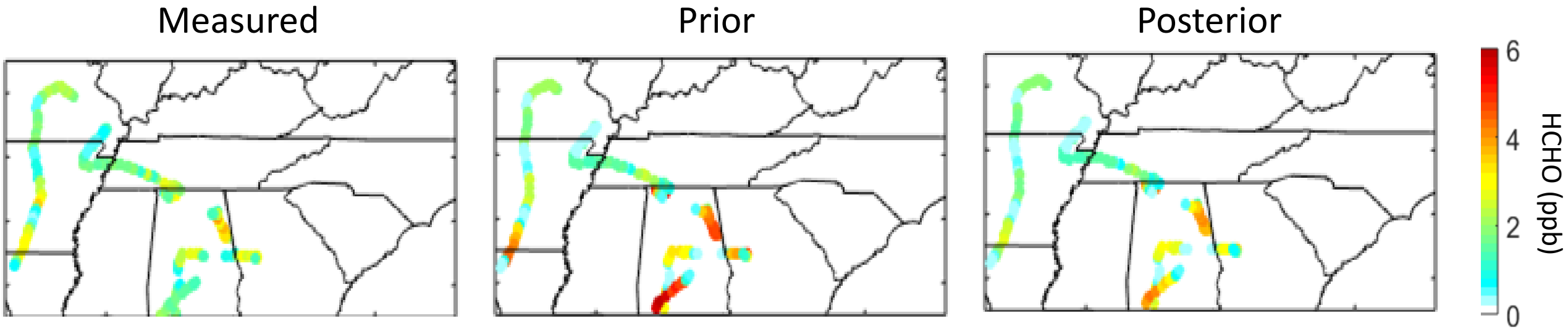
(2) Choice of landcover

(3) Response functions and environmental parameters



Guenther et al. (2012)

Comparison with SEAC4RS measurements



Improvements offered by a ground-based network

DNPH+HPLC

- O_3, NO_2 interferences
- 24h averages reported
- Some long term record at EPA sites

Mid IR Absorption

- Online, direct detection
- Hourly averages easily achievable
- Testing underway in Keutsch lab

Monitoring network on pace to keep up with satellite developments!