Global isoprene measurements from CrIS constrain emissions and atmospheric oxidation

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Wells et al., *Nature*, 2020

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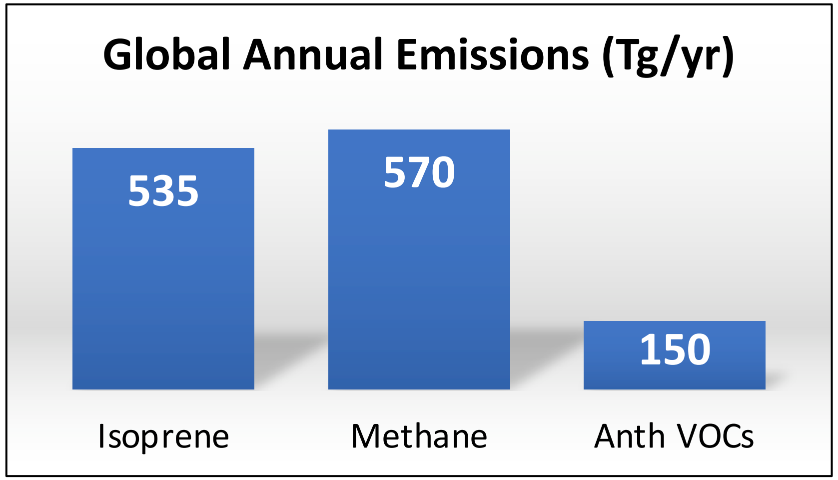
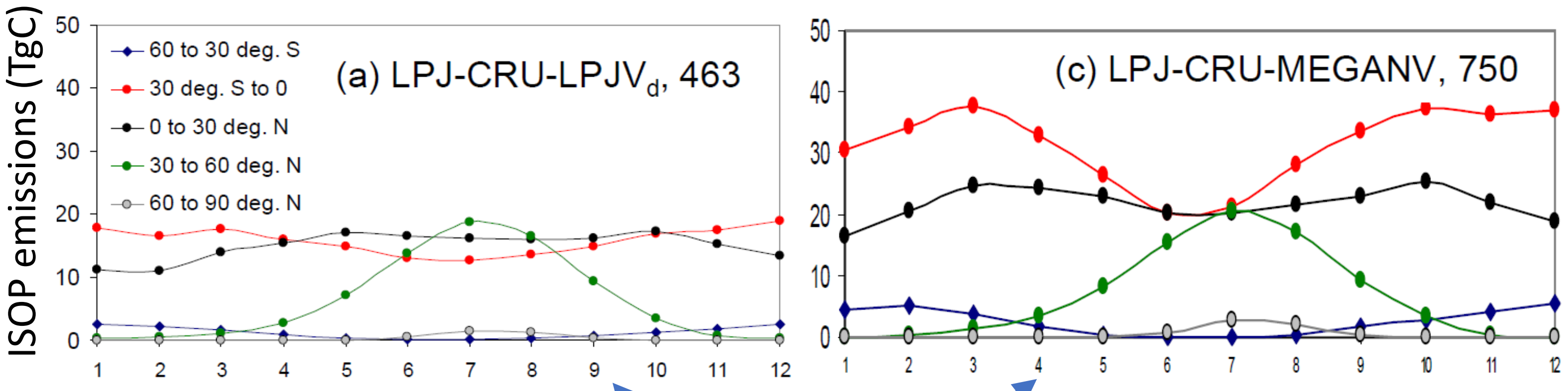
Isoprene = the dominant biogenic VOC emitted to atmosphere; uncertainties in bottom-up emissions and chemistry persist



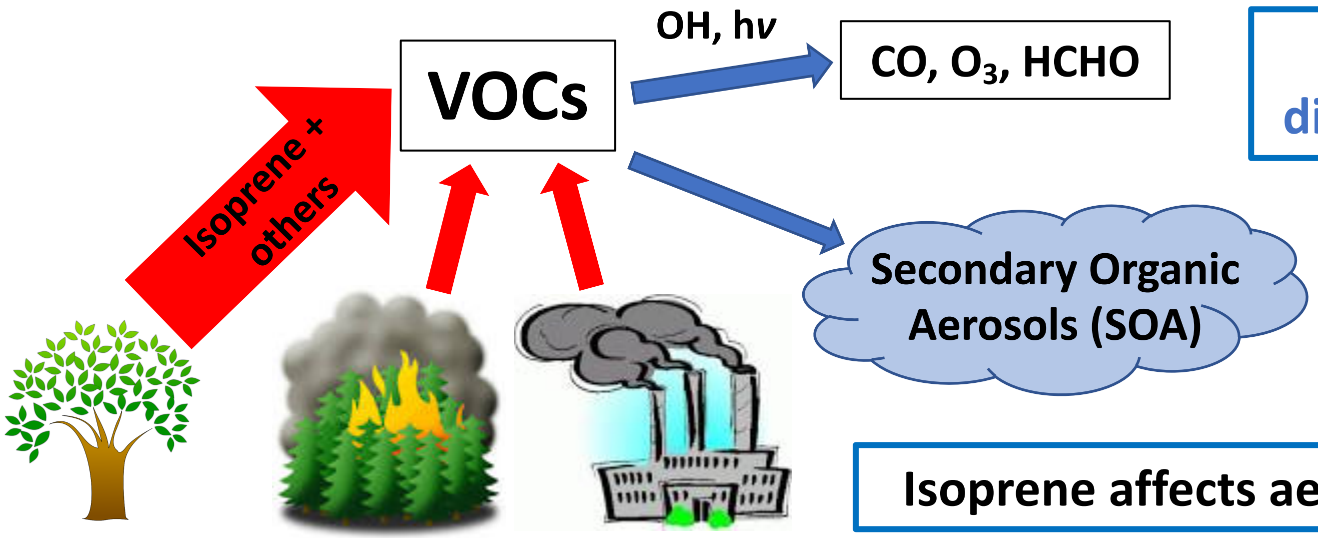
Guenther et al., 2012

Bottom-up emissions sensitive to land cover, meteorology, canopy parametrization, etc.



Arneth et al., 2011

Same emission model & met data, different vegetation = 463 v 750 TgC/y



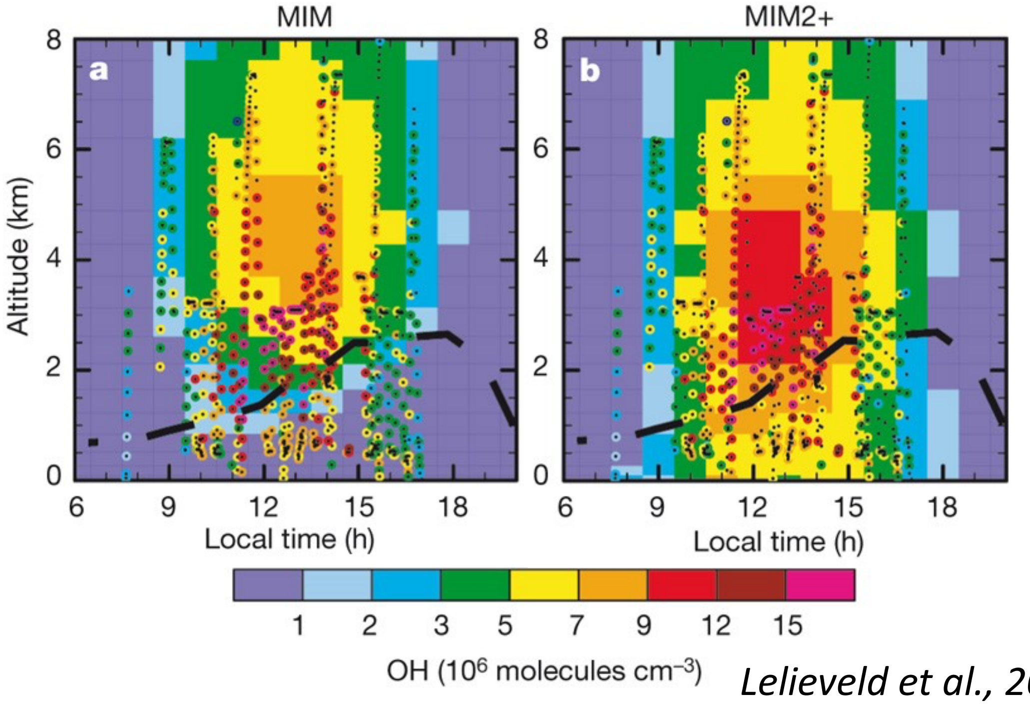
Isoprene affects aerosols, O_3, atmospheric oxidation, N-cycling

Isoprene = the dominant biogenic VOC emitted to atmosphere; uncertainties in bottom-up emissions and chemistry persist

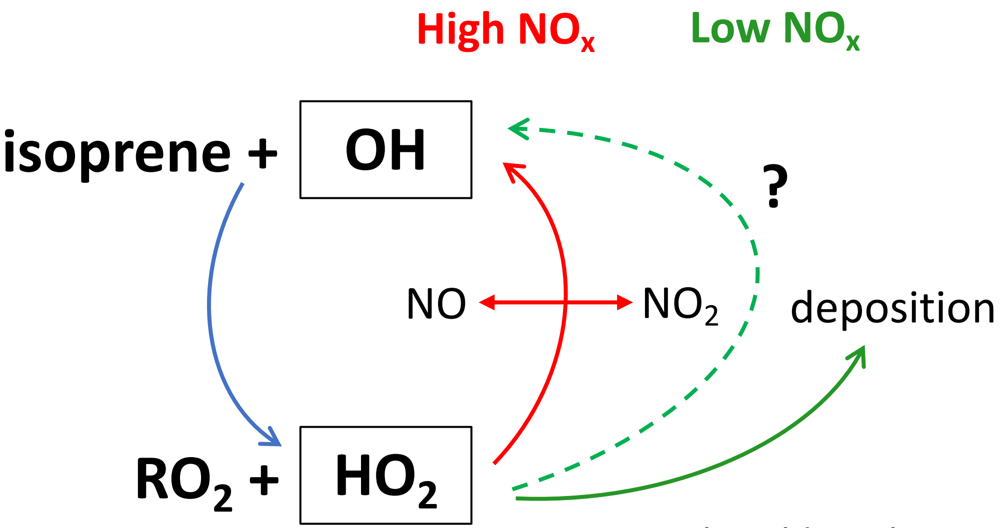
OH = primary atmospheric oxidant
Model OH is generally too low over low-NO_x isoprene source regions

How much OH recycling occurs in high isoprene, low-NO_x conditions?

Model OH with standard mechanism | **Model OH with enhanced recycling**



Lelieveld et al., 2008



e.g., Lelieveld et al., 2008; Fuchs et al., 2013

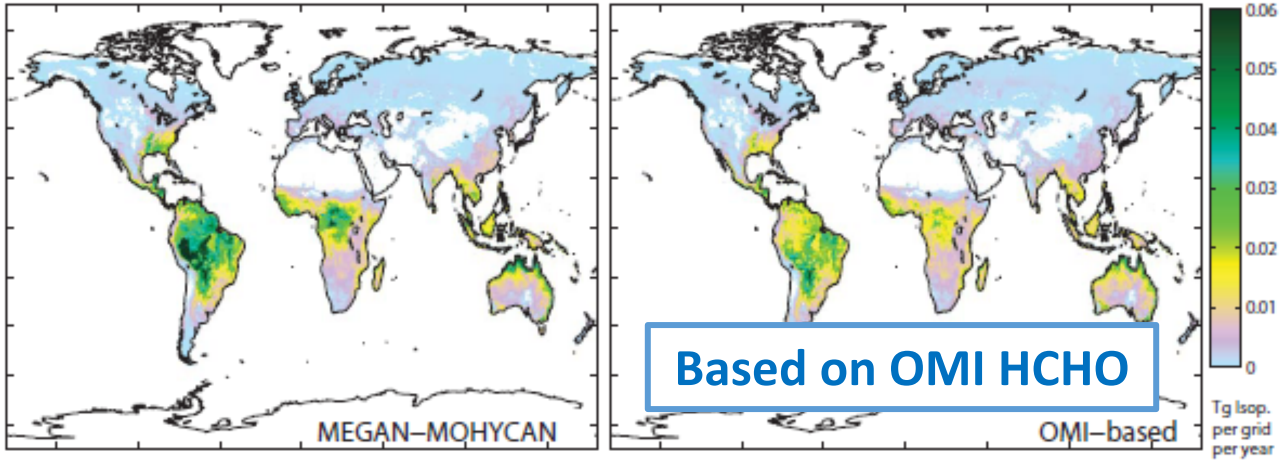
$HO_x = HO_2 + OH$
$NO_x = NO_2 + NO$

Space-based HCHO provides top-down constraints on emissions; uncertain due to chemical complexities and non-isoprene sources

HCHO = formaldehyde, a high-yield isoprene oxidation product

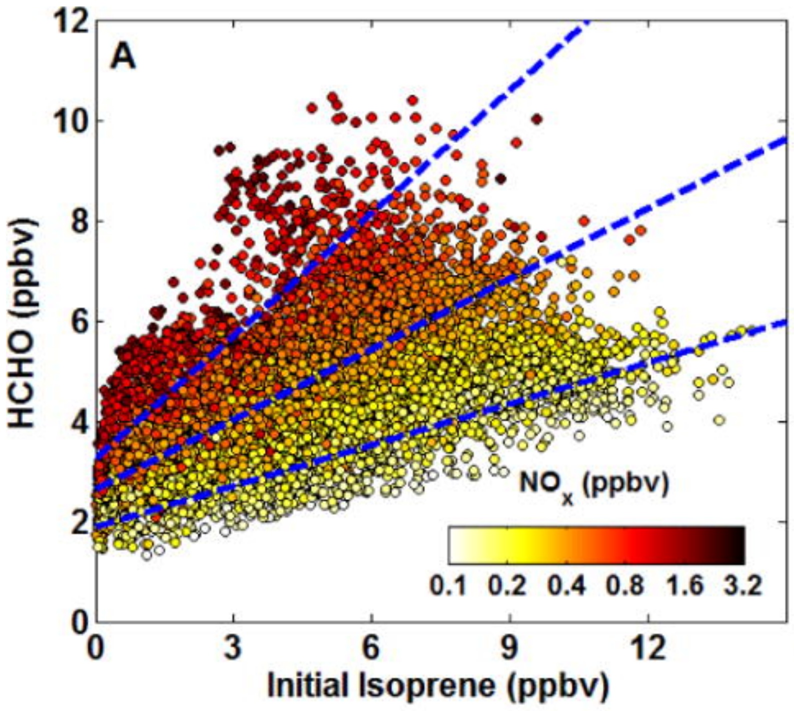
Short lifetime, detectable from space in near-UV

Bottom-up emissions **Top-down emissions**



Based on OMI HCHO

Bauwens et al., 2016



Wolfe et al., 2016

- HCHO yield from isoprene is a non-linear functions of NO_x
- HCHO also produced from fires, other VOCs

We must rely on models to accurately capture these effects

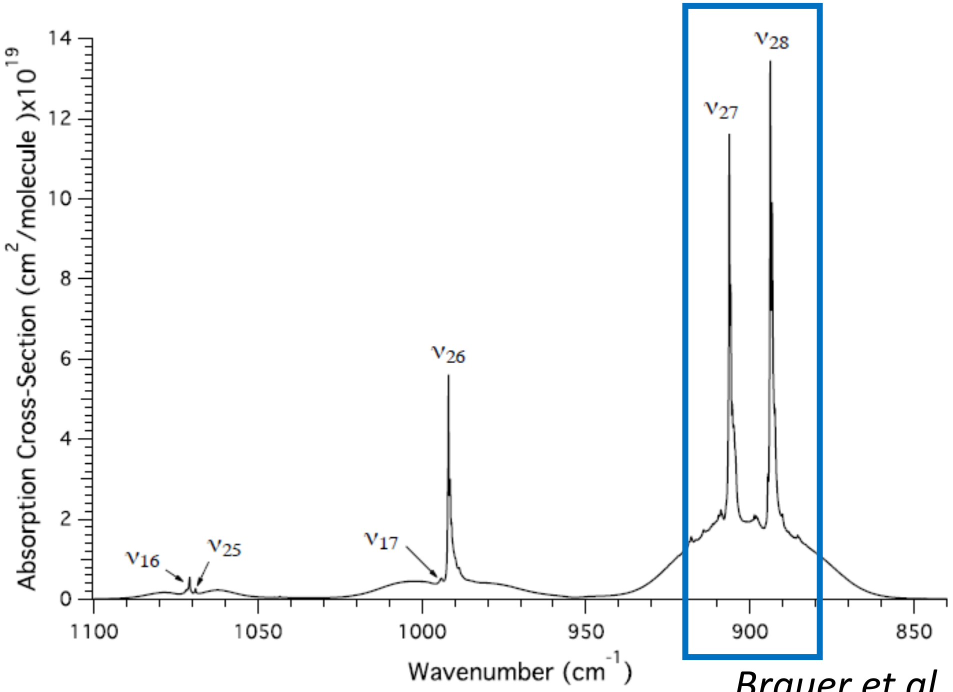
Measurements of TIR absorption cross section of isoprene enable direct measurements of isoprene from space

Cross-track Infrared Sounder (CrIS):

- 10/2011 (Suomi-NPP), 11/2017 (NOAA-20), expected 2022
- Near global coverage twice daily
- Afternoon overpass
- Low noise

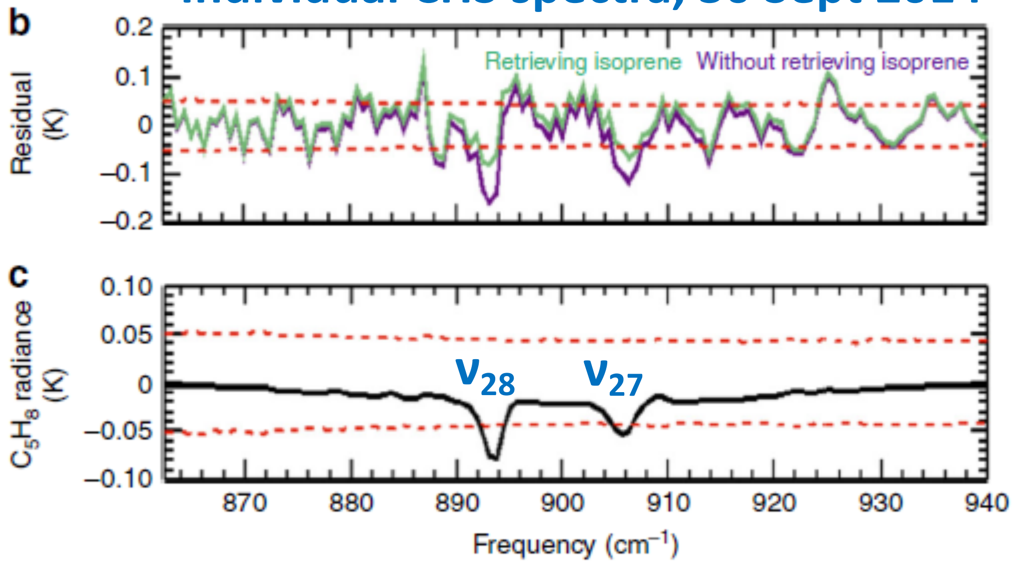
} **Key for isoprene!**

OE retrievals over Amazonia (Fu et al., 2019)

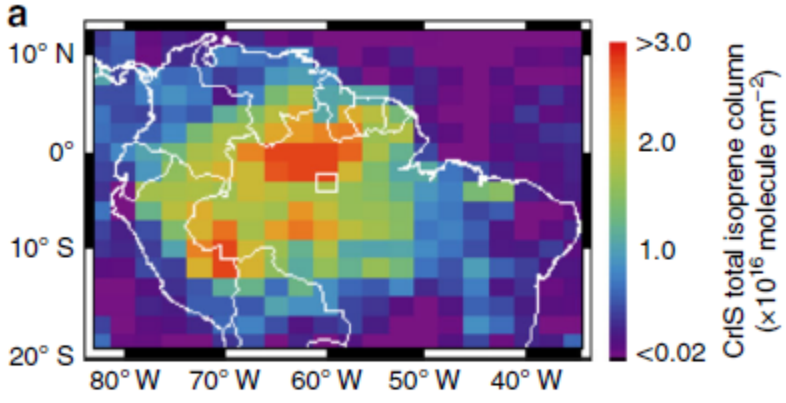


Brauer et al., 2014

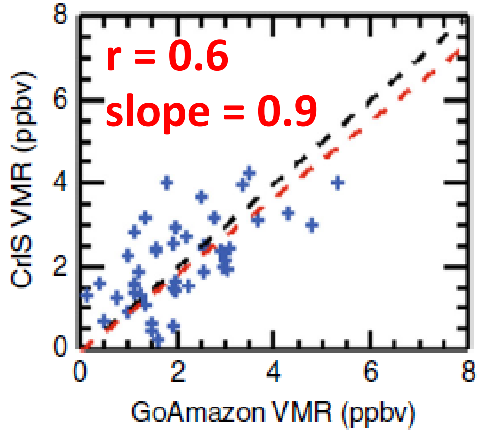
Individual CrIS spectra, 30 Sept 2014



CrIS isoprene column



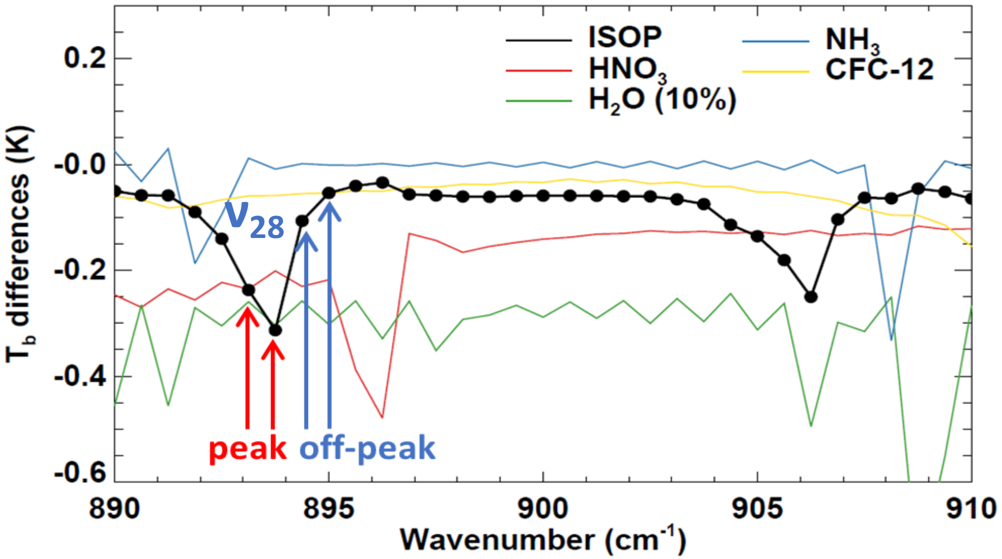
CrIS v aircraft

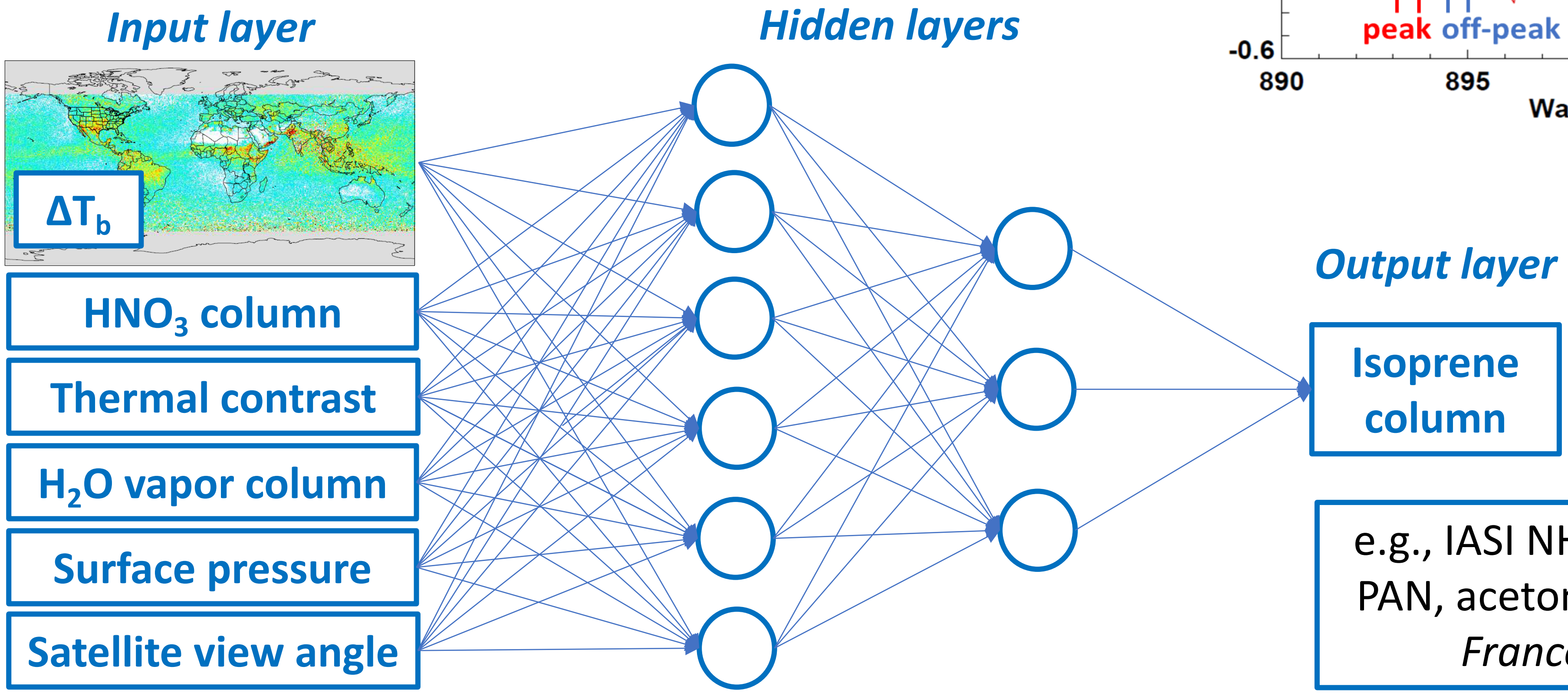


We employ an artificial neural network to derive the global isoprene distribution from the CrIS T_b difference

Monthly-mean T_b difference: $\Delta T_b = T_{b,off\text{-}peak} - T_{b,peak}$

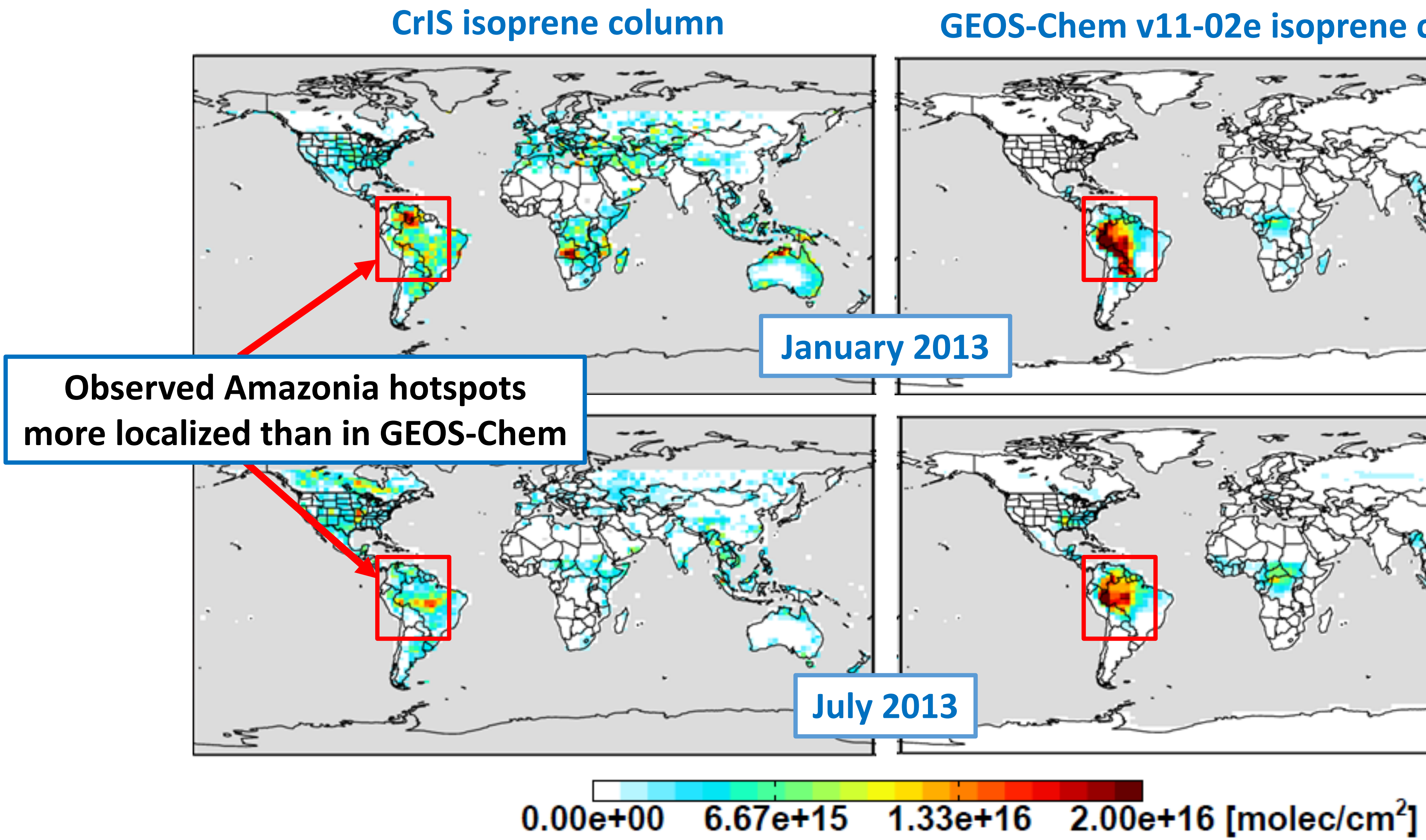
Single-footprint, cloud-screened Level 1B data;
Pro: fast way to process a large dataset (~9e6 spectra/day)
Con: Can be subject to interferences





e.g., IASI NH_3, methanol, formic acid, PAN, acetone (*Whitburn et al., 2016; Franco et al., 2018; 2019*)

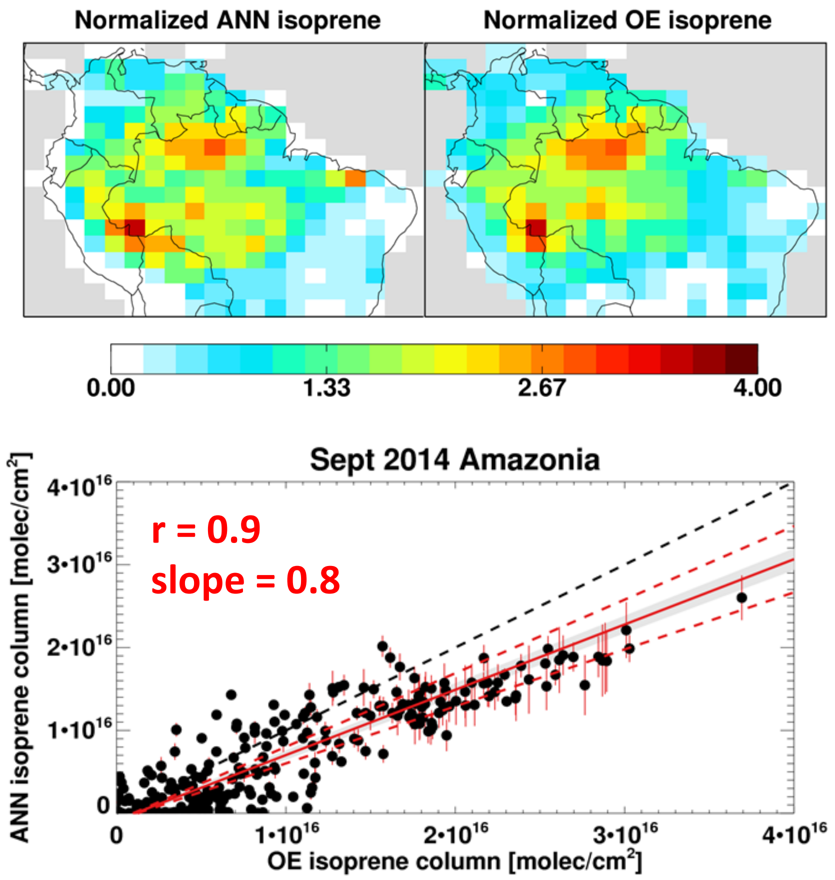
The first global space-based isoprene distribution

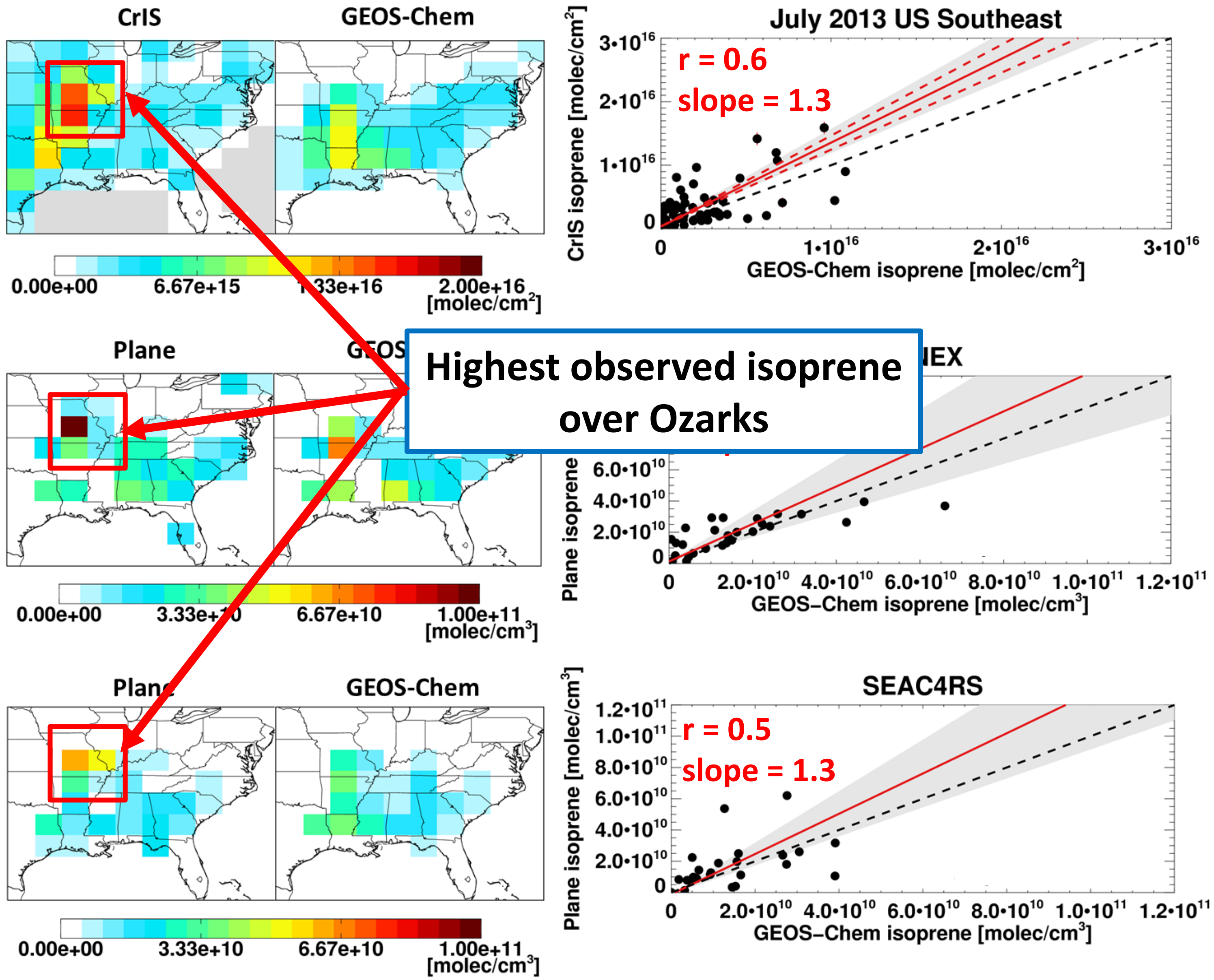


CrIS isoprene is consistent with OE retrievals and aircraft measurements

July 2013, using GEOS-Chem as a transfer standard

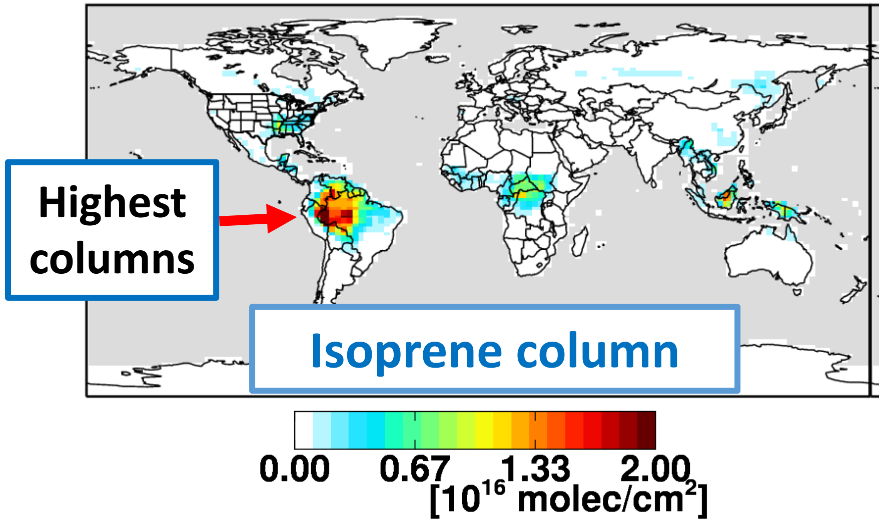
Amazonia OE retrievals (Fu et al., 2019)

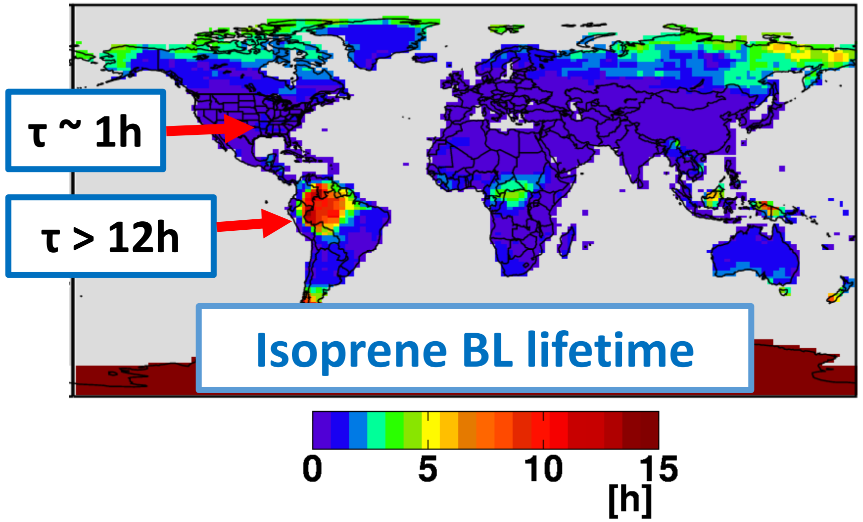


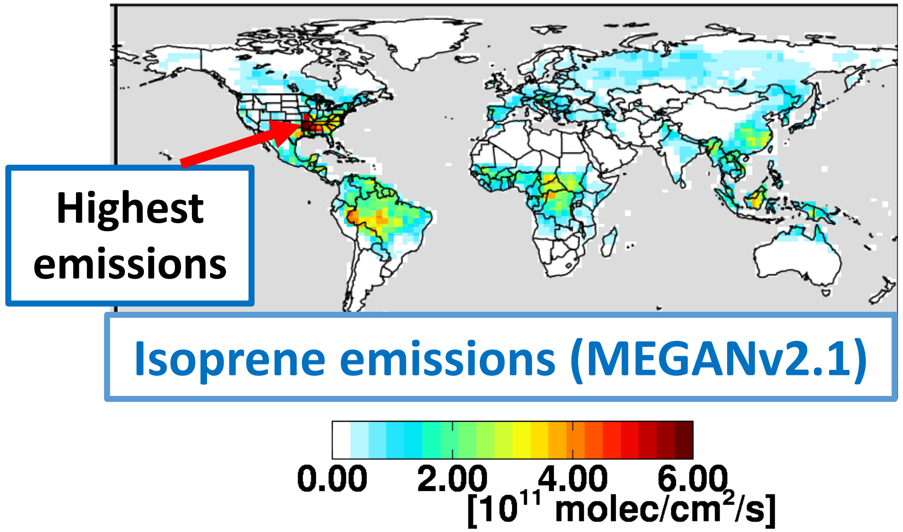


Space-based isoprene and HCHO provide combined constraints on emissions <u>and</u> chemistry in isoprene source regions

GEOS-Chem, July 2013





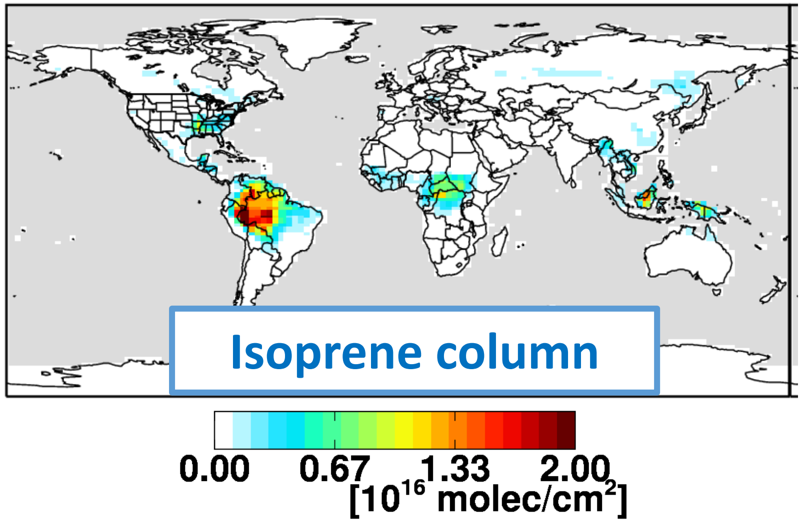


Isoprene column distribution distinct from that of emissions

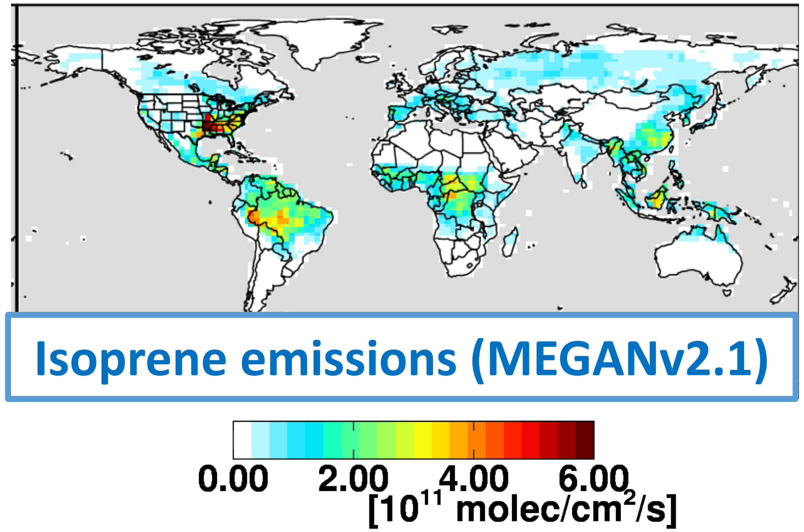
Space-based isoprene and HCHO provide combined constraints on emissions <u>and</u> chemistry in isoprene source regions

GEOS-Chem, July 2013

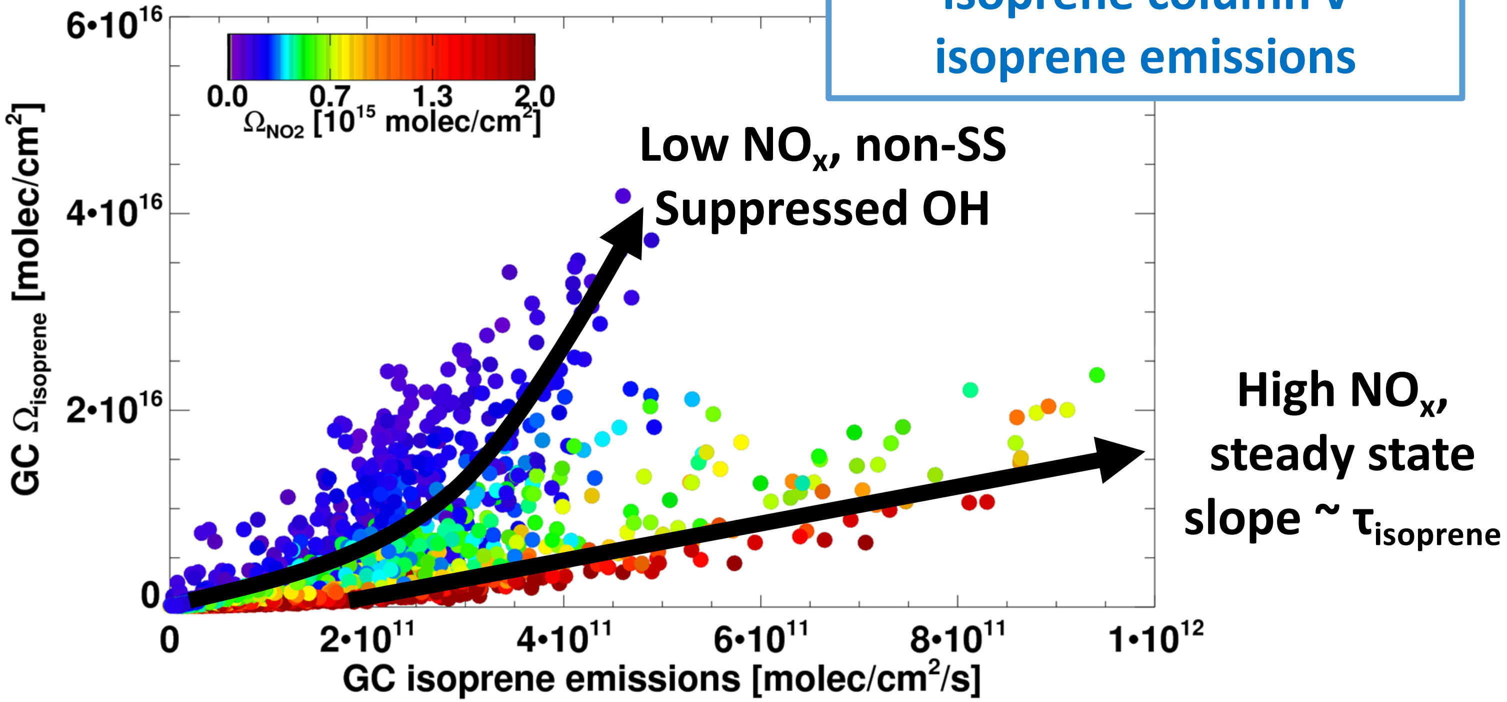
Global ensemble of isoprene column v isoprene emissions



Isoprene column



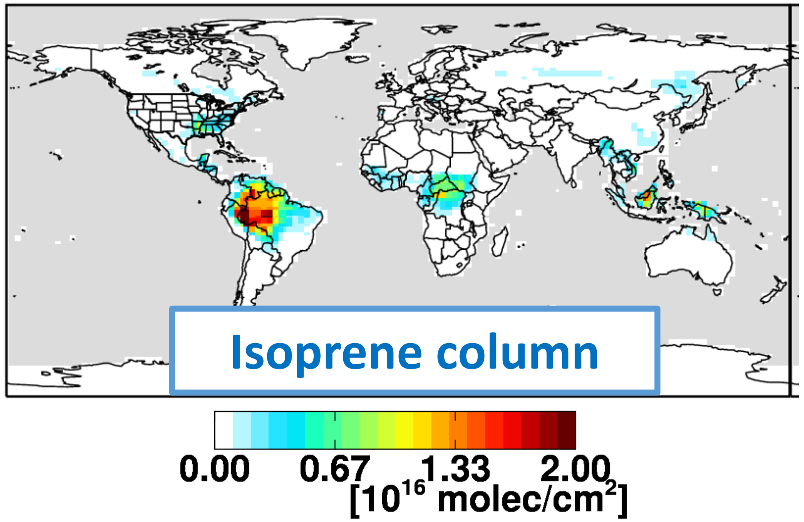
Isoprene emissions (MEGANv2.1)

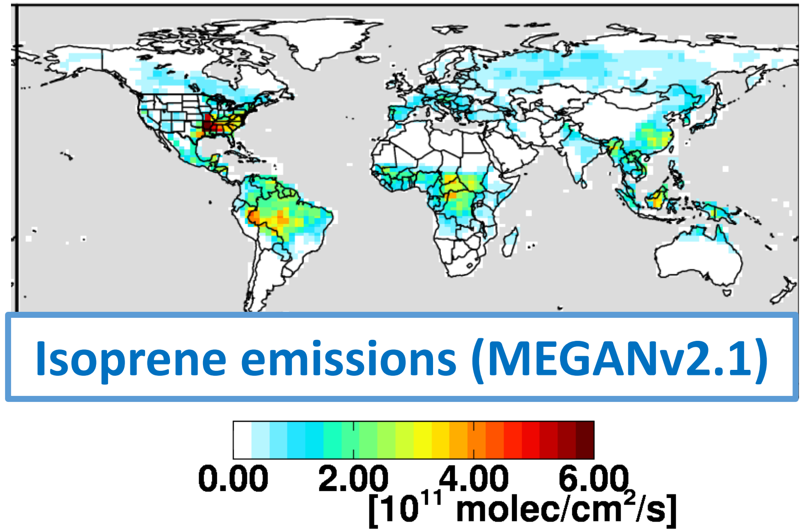


At low NO_x, isoprene increases superlinearly as it begins affecting its own sink

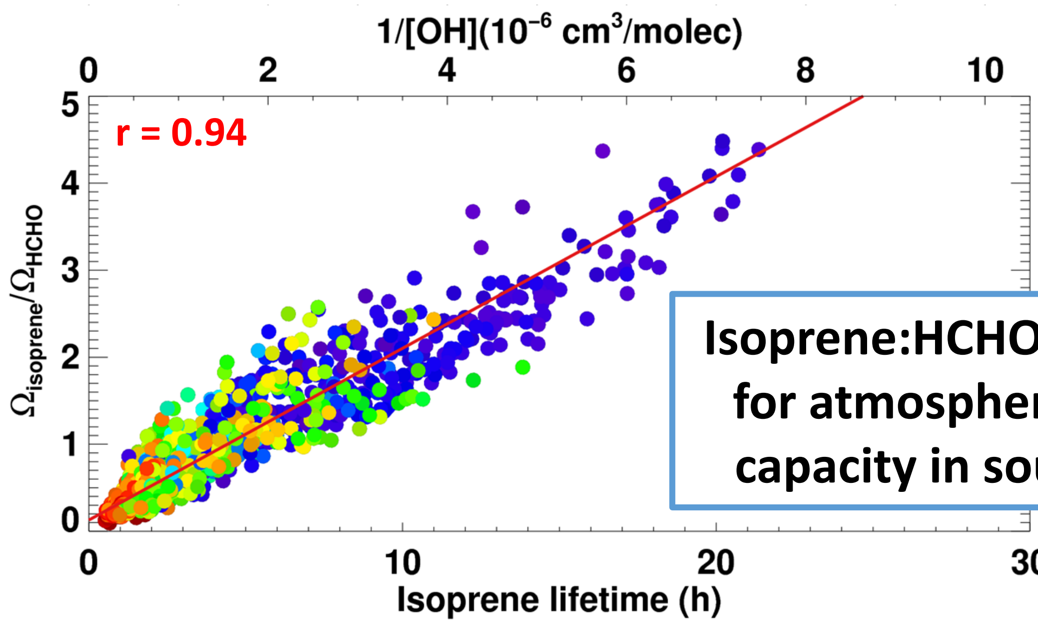
Space-based isoprene and HCHO provide combined constraints on emissions and chemistry in isoprene source regions

GEOS-Chem, July 2013





Global ensemble of isoprene:HCHO column ratio v isoprene lifetime



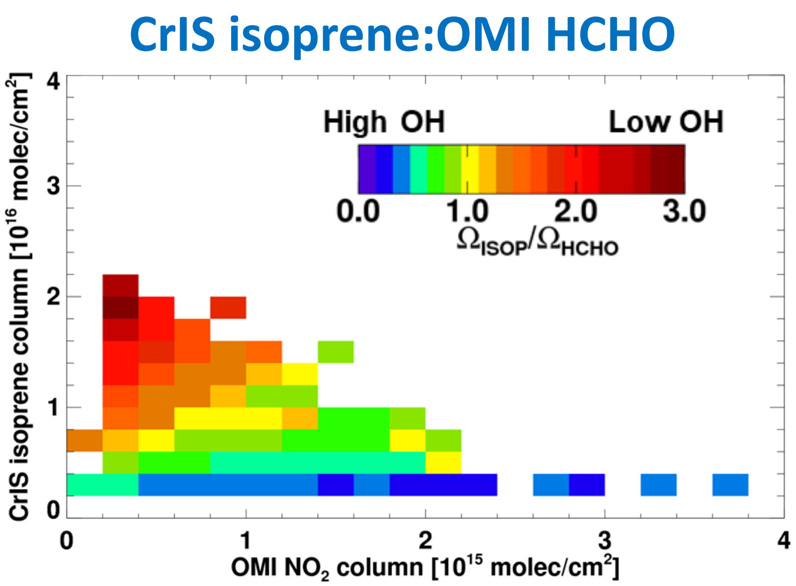
Isoprene:HCHO ratio = proxy for atmospheric oxidation capacity in source regions

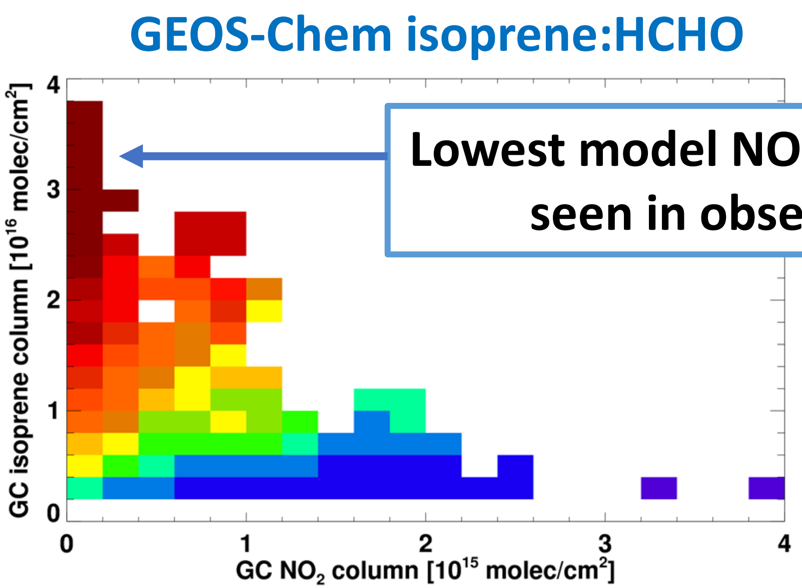
HCHO is more buffered to OH variability:

- Loss to photolysis still occurs at low OH
- Loss proportional to [isoprene]×[OH]

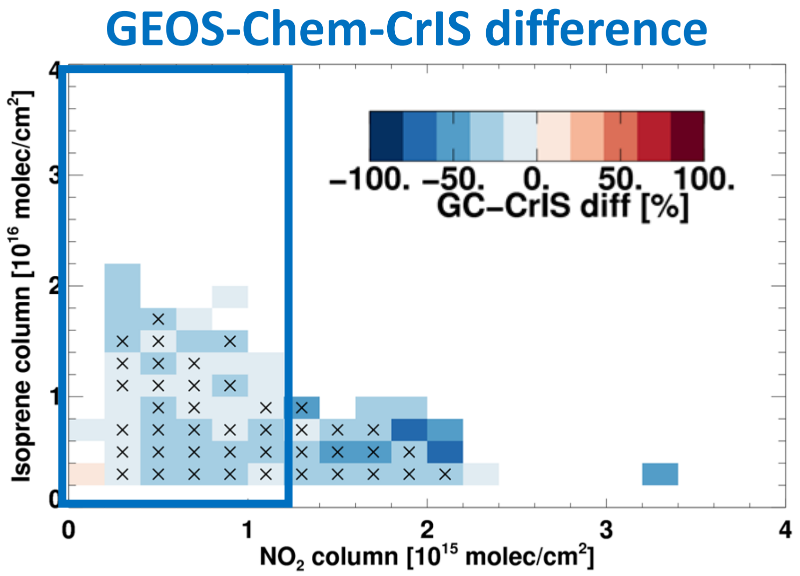
Space-based isoprene:HCHO ratio supports current model treatment of OH chemistry in isoprene source regions

OMI QA4ECV HCHO and NO_2



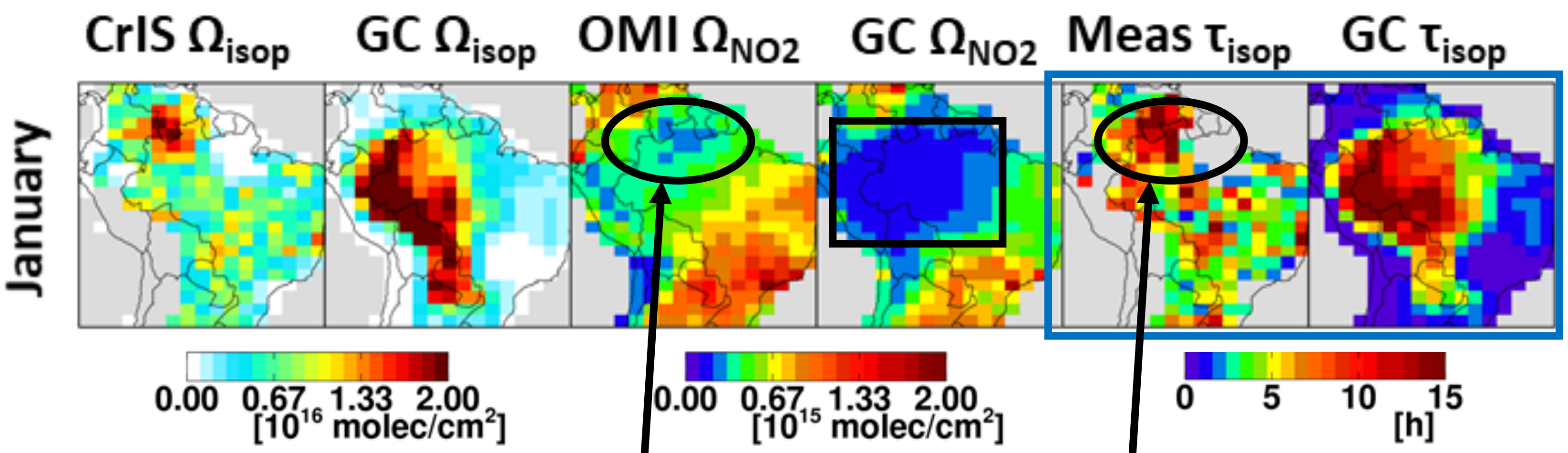


Lowest model NO_2 columns not seen in observations



Agreement within 10-40% at low-to-moderate NO_x argues <u>against</u> substantial missing OH recycling

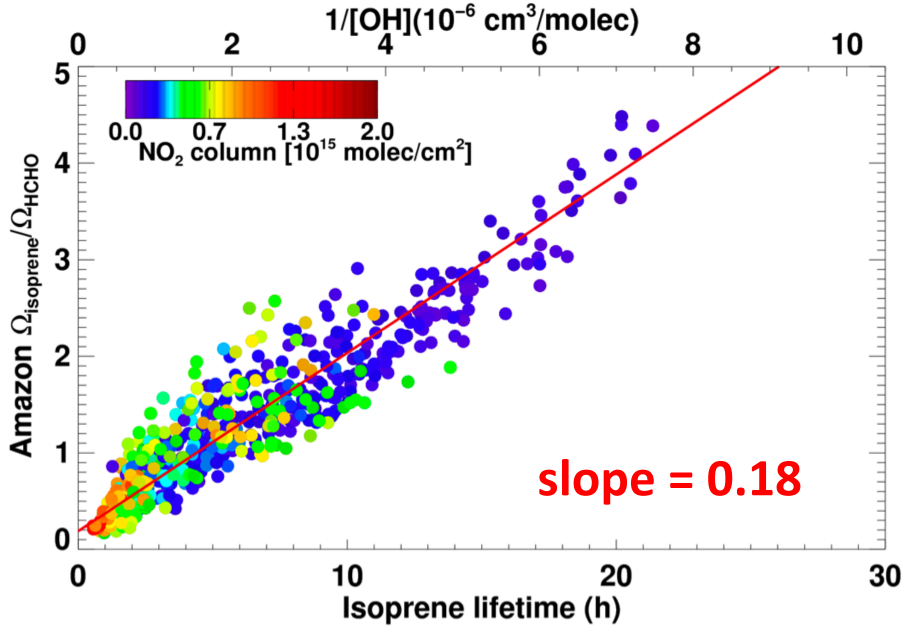
Observed isoprene lifetime consistent with observed NO_2 over Amazonia, large scale NO_x bias evident in model



$\tau_{isop} = \Omega_{isop}:\Omega_{HCHO}/0.18$

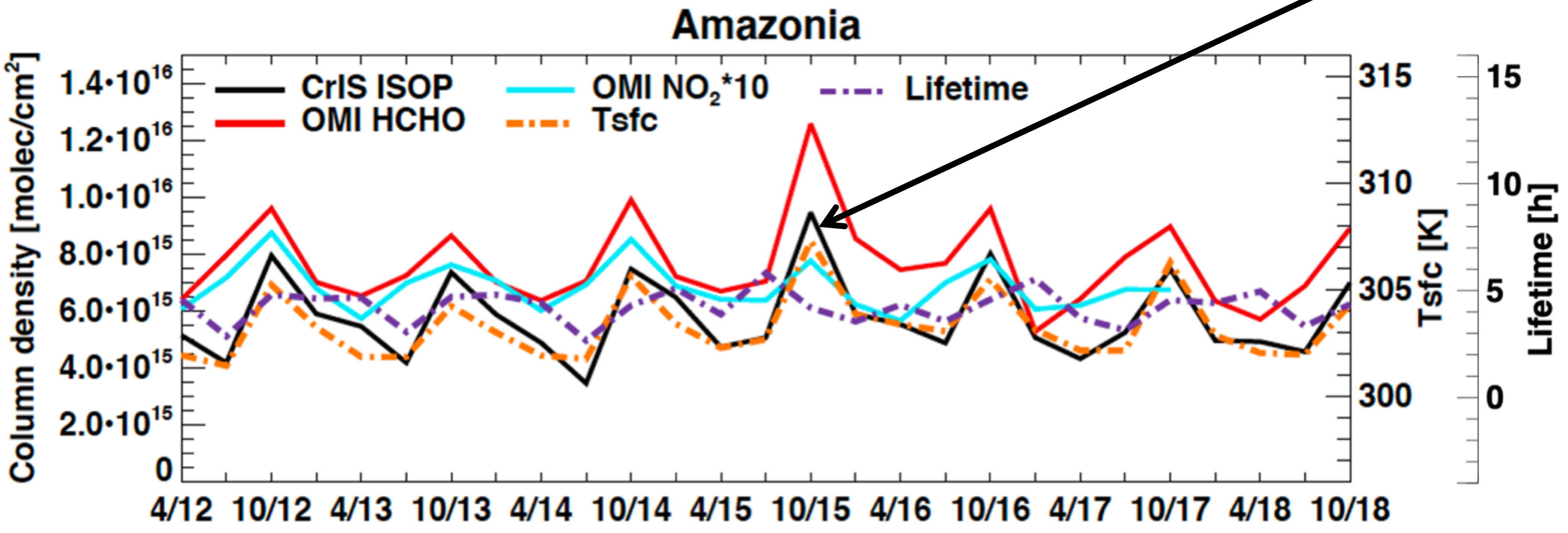
Measured lifetime agrees with chemical expectations = additional confirmation of our approach

Large scale NO_x bias likely due to underestimated soil NO_x emissions (Liu et al., 2016)



A long-term record of isoprene from CrIS will give us new insights into interannual variability and chemistry-climate couplings

Onset of 2015/2016 El Niño = warmer temperatures, drought stress?



April 2012-October 2018

Next steps: next generation CrIS isoprene retrieval based on Hyperspectral Range Index (HRI)

CrIS HRI looks more "isoprene-like" than ΔT_b

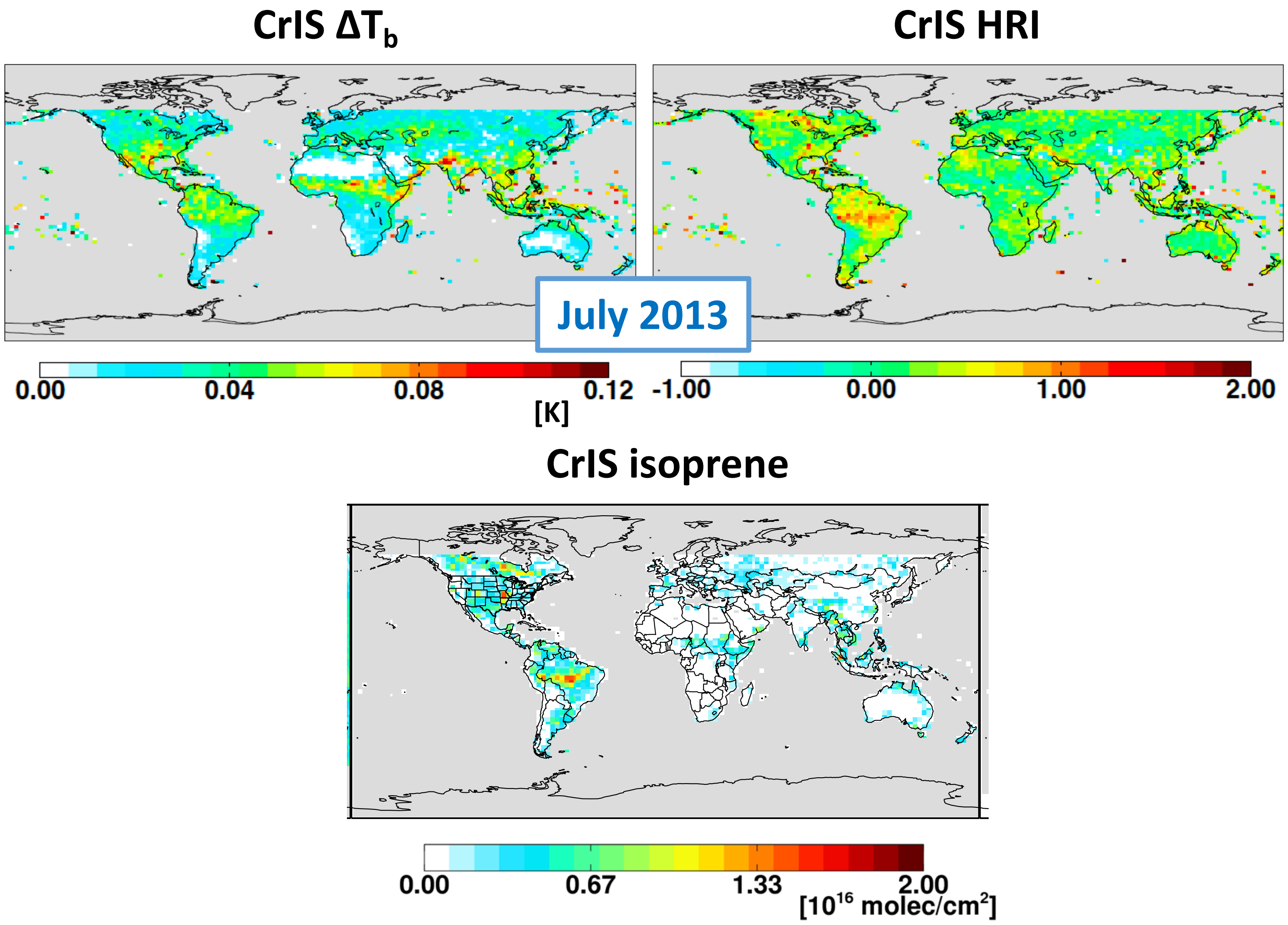
Measured spectrum

Mean background spectrum

$$HRI = \frac{K^T S_y^{-1}(y - \bar{y})}{\sqrt{K^T S_y^{-1} K}}$$

Spectral Jacobian

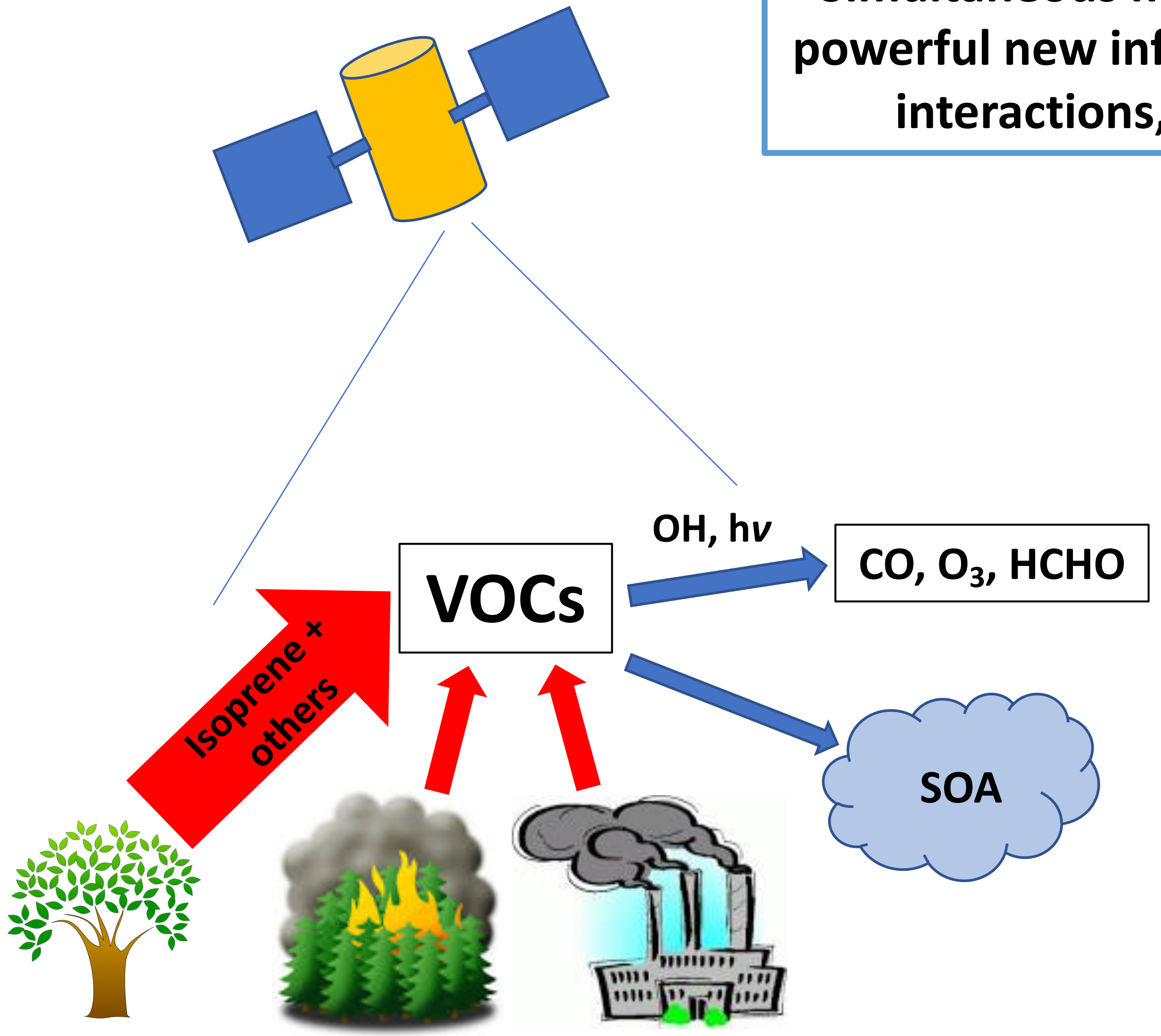
Background spectral covariance



HRI uses full active spectral range for isoprene = enhanced sensitivity, less subject to interferences

Next steps: apply HRI-based retrieval to look at other species, advance understanding of VOC sources and chemistry-climate-ecosystem interactions

Simultaneous measurements of multiple VOCs from CrIS will provide powerful new information to better understand biosphere-atmosphere interactions, biomass burning, and pollution across the globe!



Species	Primary Sources
Methanol	Biosphere, biomass burning
Ethene	Biosphere, biomass burning, vehicles
Ethyne	Combustion
Acetone	Biosphere, biomass burning
PAN	Urban emissions, biomass burning
HCN	Biomass burning
Acetic acid	Biosphere, biomass burning
Ethane	Natural gas, biofuel, biomass burning
Benzene	Combustion

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- Dejian Fu, Kevin Bowman, Evan Manning, Ruth Monarrez, Irina Strickland (JPL)
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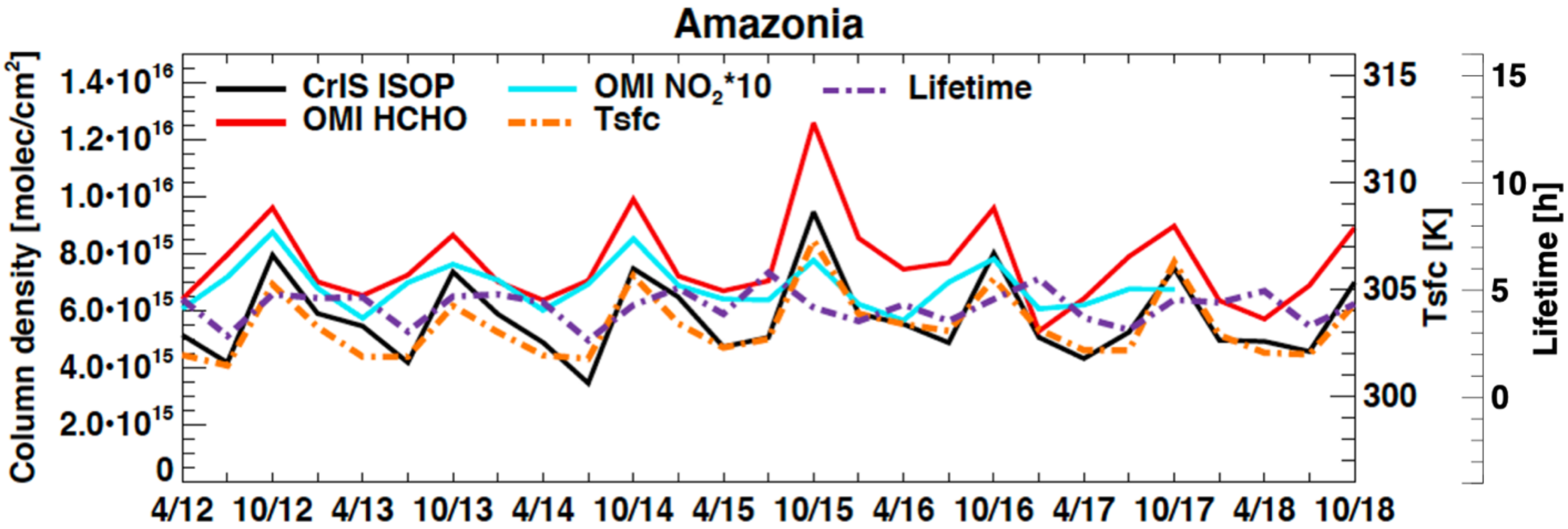
- Eric Edgerton (SEARCH)
- Stephen Springston (BNL)

- NASA ACMAP for funding

Science questions to be answered by long-term TIR sounder records

We can do A LOT of science with even a weak signal! More species would be key to answering the below questions:



1) What does interannual variability tell us about the links between climate and VOC emission drivers?
2) How are VOC emissions (and OH!) changing over time?
3) As anthropogenic emissions decrease in the US and elsewhere, how is atmospheric composition changing?

What should be the highest priorities for new trace gas products?

Species	Primary Sources
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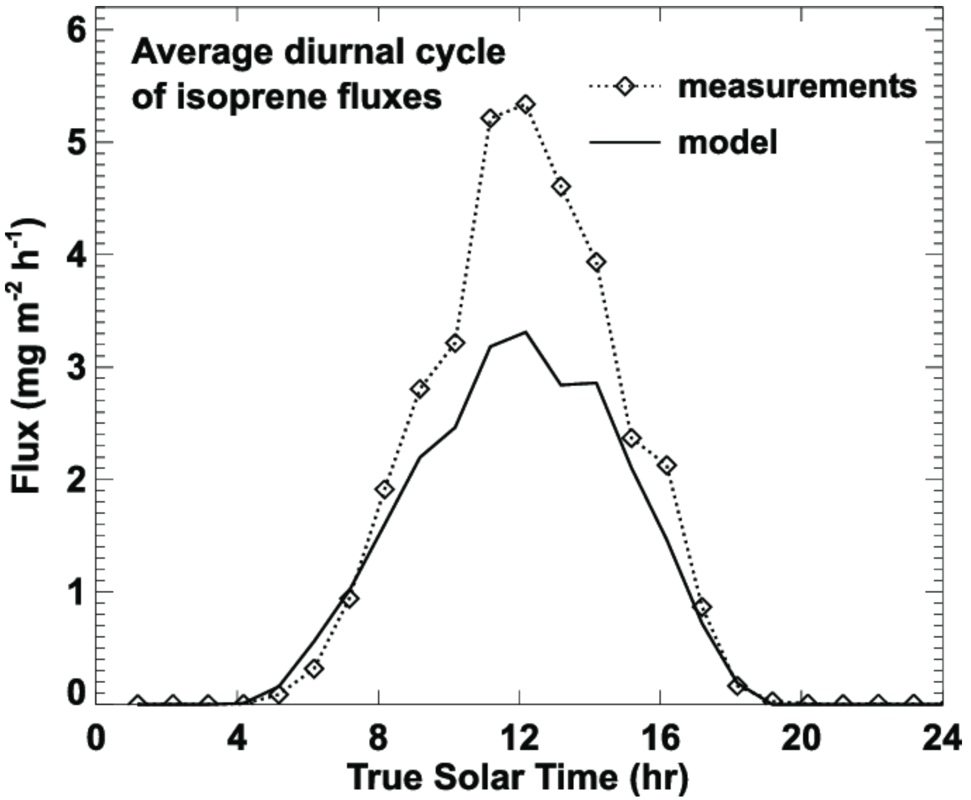
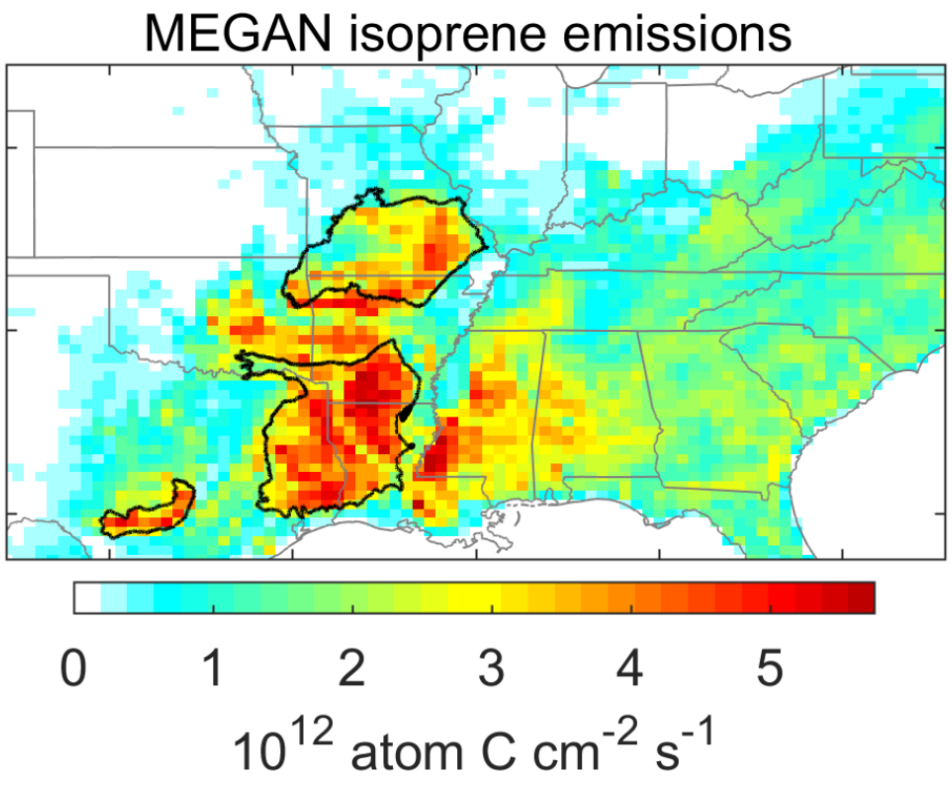
Benzene	Combustion

1) **A greater suite of species active in TIR (not detectable with other sensors!) for doing detailed source apportionment globally over long timescales**
2) **Evaluation of sensitivity (AK) over different source types (biogenic versus biomass burning, etc)**
3) **Near-real-time quick look-type product for specific events (e.g., large wildfires)**

What are the key observational gaps?



Müller et al., 2007



Kaiser et al., 2018

1) Diurnal variability: net biogenic VOC emissions are high during the day and low (or negative!) at night; can we quantify emission processes from space versus just net emission strength?
2) Smaller footprints to look at fire impacts and urban plumes; more information about BVOC emissions as a function of plant type