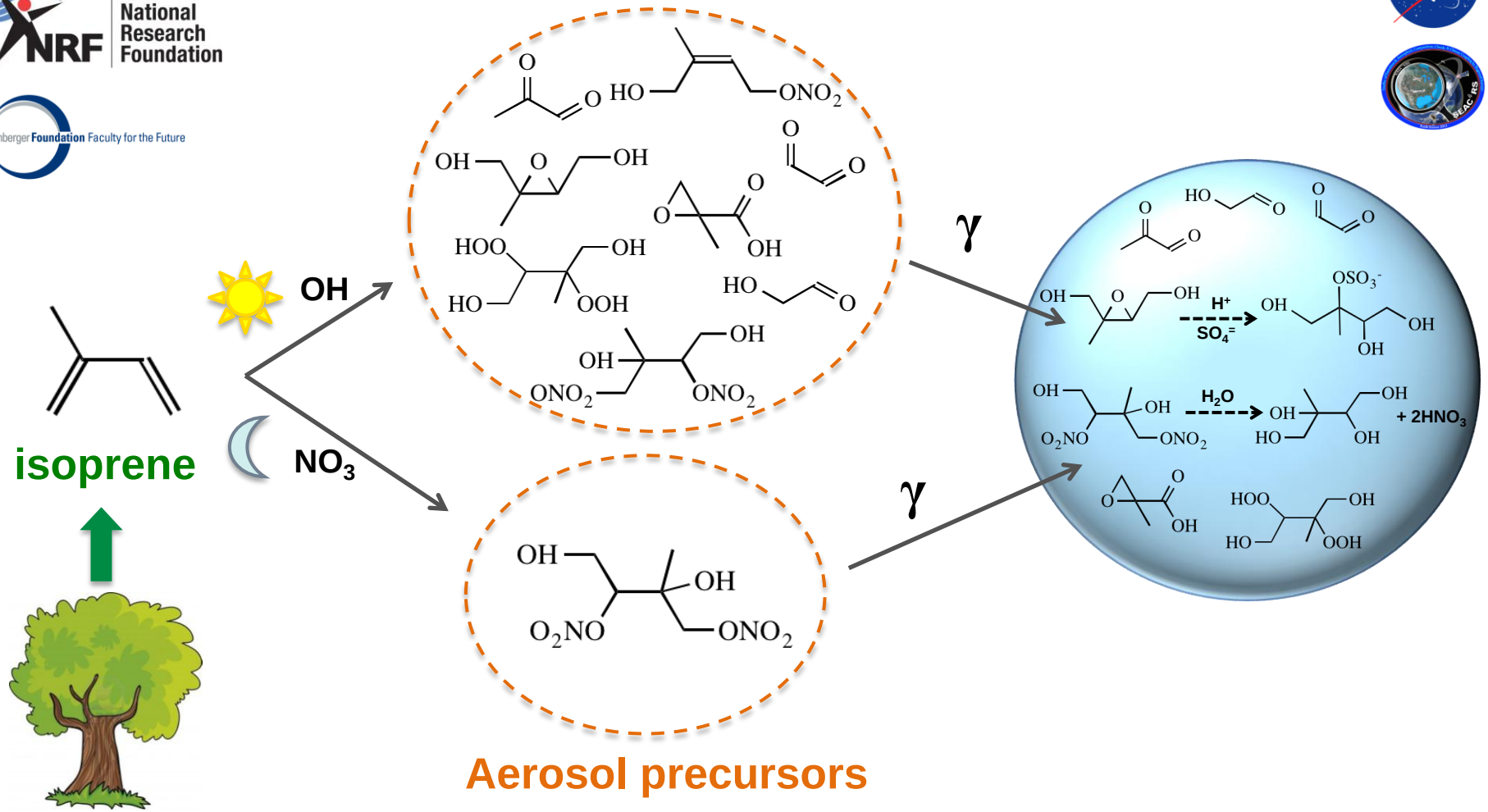
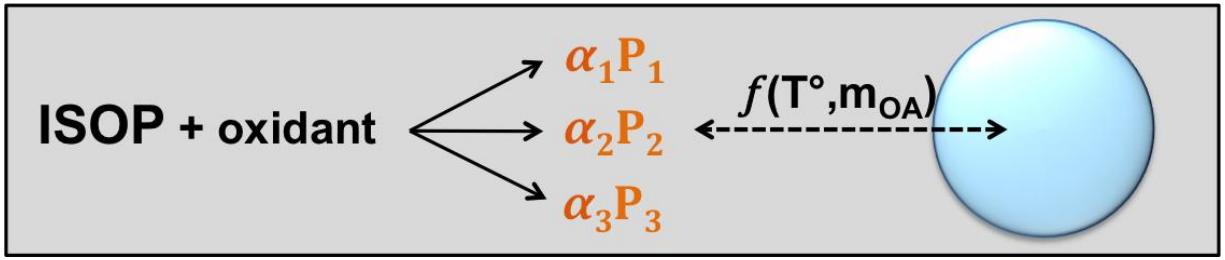
Improved understanding of southeast US isoprene SOA using ground (SOAS), aircraft (SEAC^{4}RS), satellite (MODIS, OMI), and model (GEOS-Chem) data



Eloise A. Marais (*emarais@seas.harvard.edu*)
D. J. Jacob, L. Zhu, P. Kim, K. Chance, G. Abad, J. Jimenez, J. Krechmer, W. W. Hu, P. Campuzano-Jost, A. Fried, J. Kroll, K. Froyd, J. Liao, V.F. McNeill

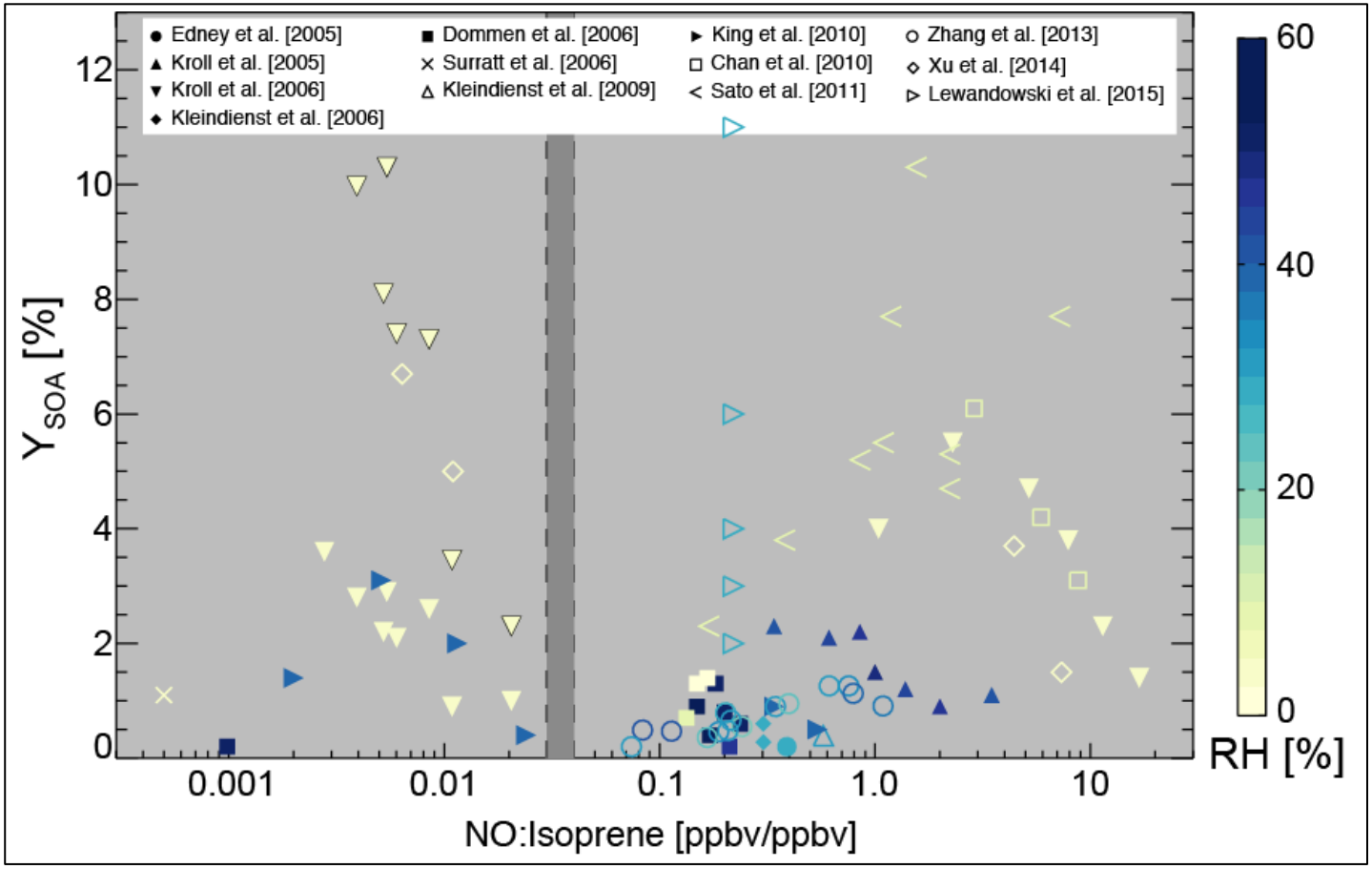
SEAC^{4}RS STM
Pasadena, CA
30 April 2015

Default decoupled GEOS-Chem isoprene SOA scheme



[Pye and Seinfeld, 2010]

Use fixed yields (α_i), despite large variability in **chamber studies**:



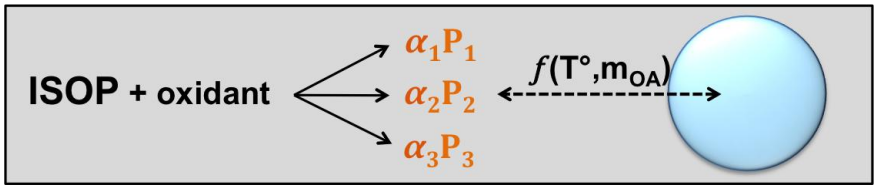
Lab results for **OA < 20 µg m^{-3}**

Yields colored by **relative humidity**

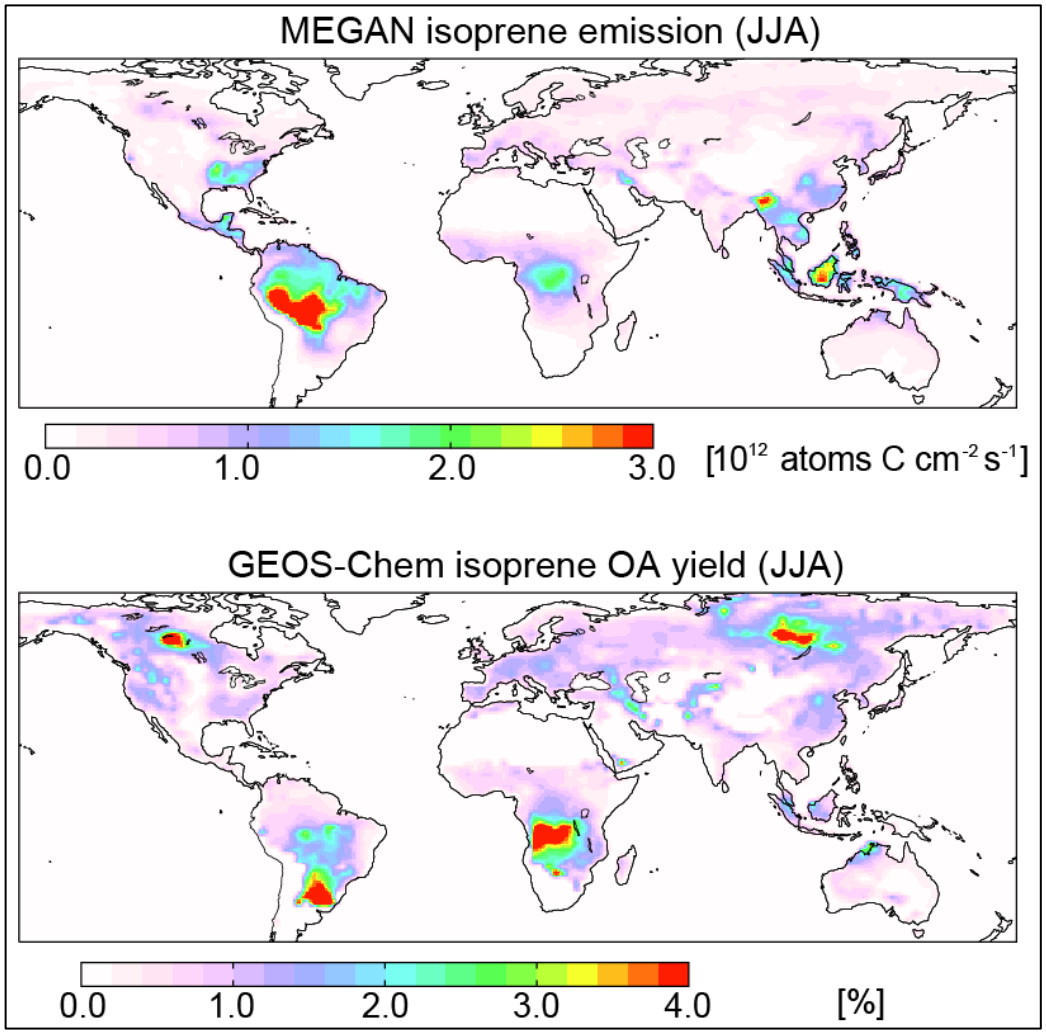
In GEOS-Chem **α_i is ~3% at 10 µg m^{-3}**.

Realized yields are lower

Default GEOS-Chem isoprene SOA scheme



Isoprene Emissions and SOA yields in GEOS-Chem



Isoprene OA mass yield

$$\text{Yield} = \frac{\Delta \text{SOA}_{\text{net}}}{\Delta \text{isoprene}}$$

Very low yield (**~1%**) in the southeast US

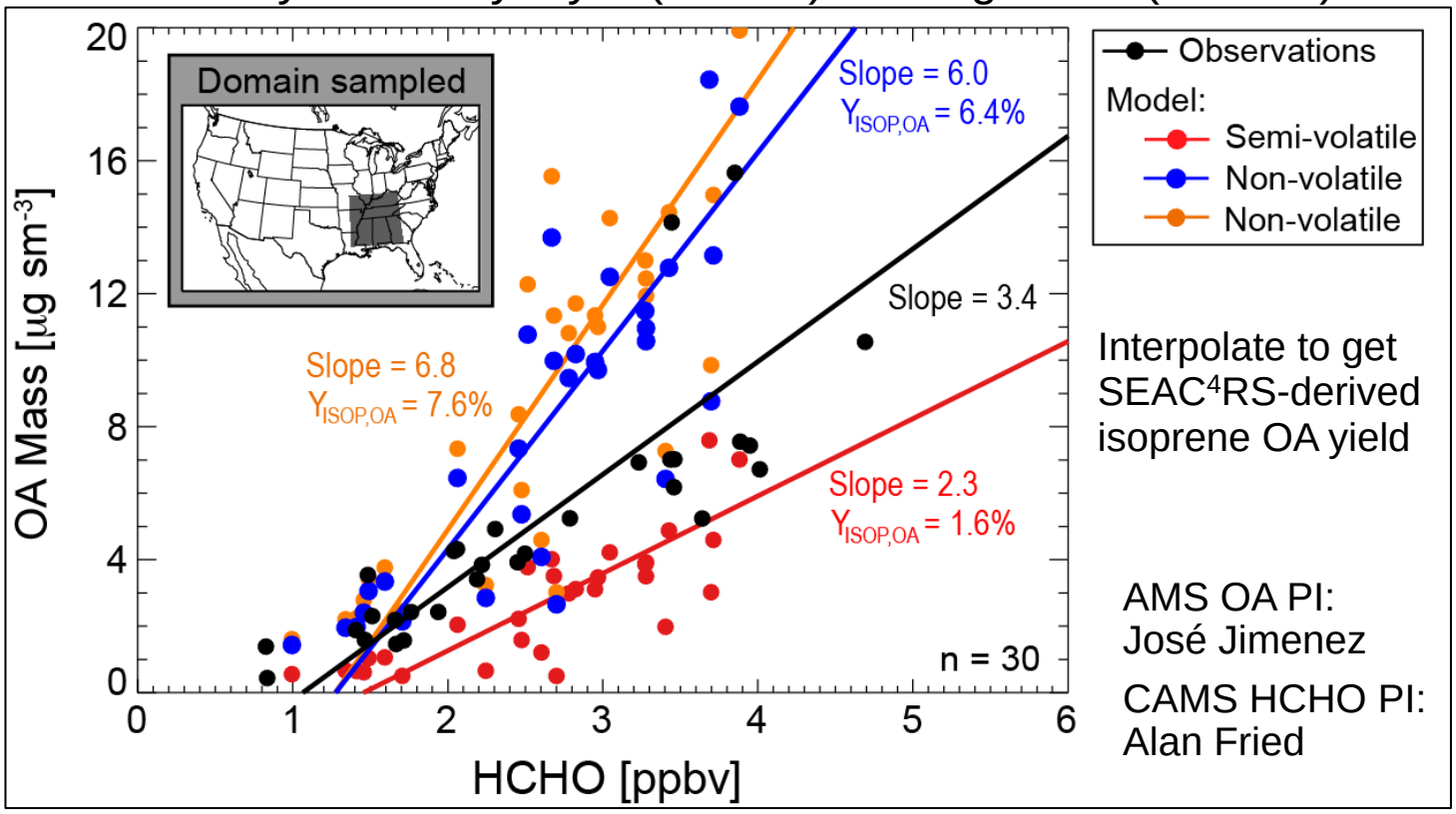
High yields not coincident with isoprene emissions

High yields where there is biomass burning
(high pre-existing aerosol; low temperatures).

SEAC⁴RS-derived isoprene OA yields

OA-HCHO relationship during SEAC⁴RS

Points are daily boundary-layer (< 2 km) mean gridded (2°×2.5°) data



By interpolation, SEAC⁴RS-derived **isoprene OA yield is 3 %**

SEAC⁴RS OA -HCHO relationship provides total isoprene SOA yields.

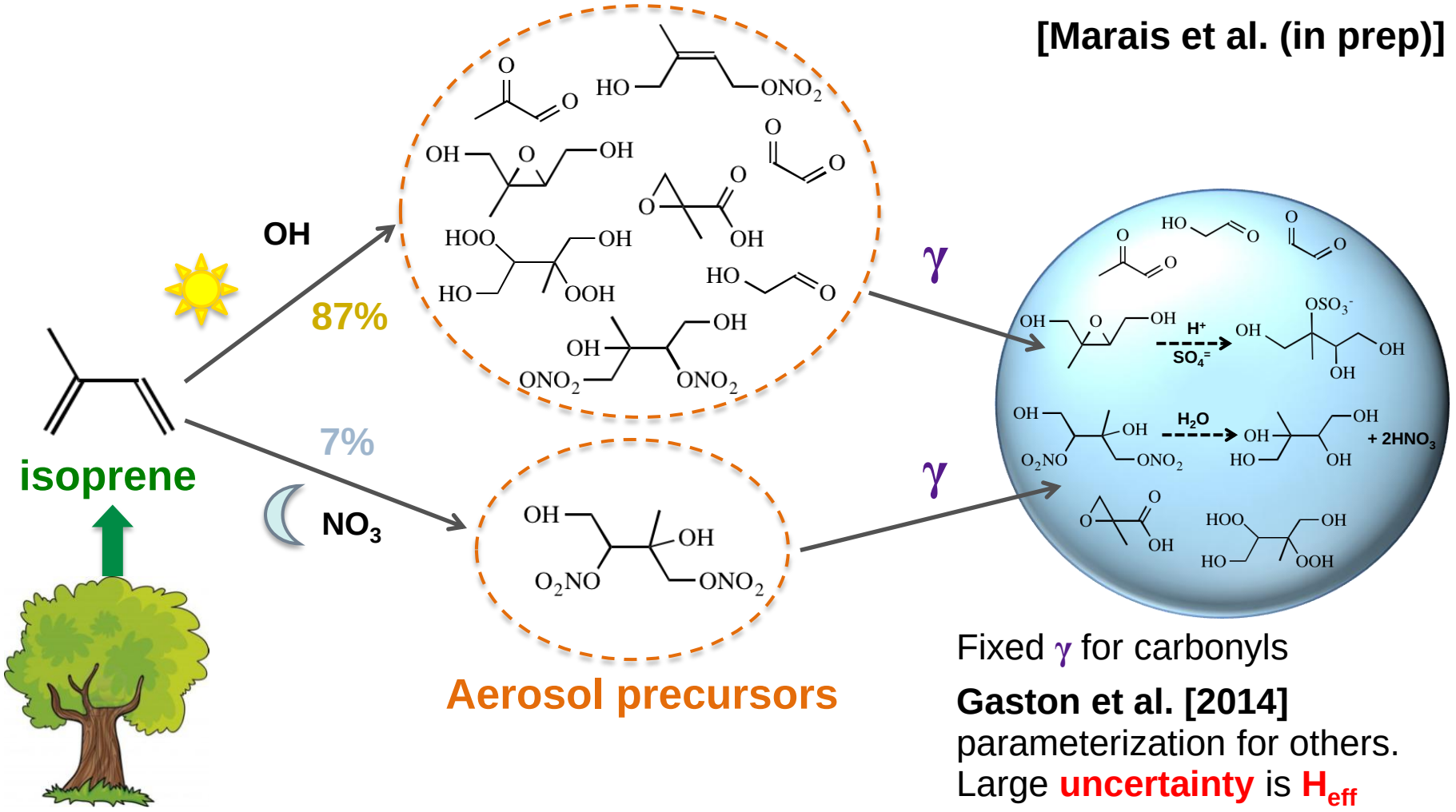
Obtain additional information from AMS-derived isoprene SOA components

New GEOS-Chem isoprene SOA parameterization

Develop a mechanism that couples gas and aerosol phases

Better understand the processed that lead to isoprene SOA formation

[Marais et al. (in prep)]



Fixed γ for carbonyls

Gaston et al. [2014] parameterization for others.

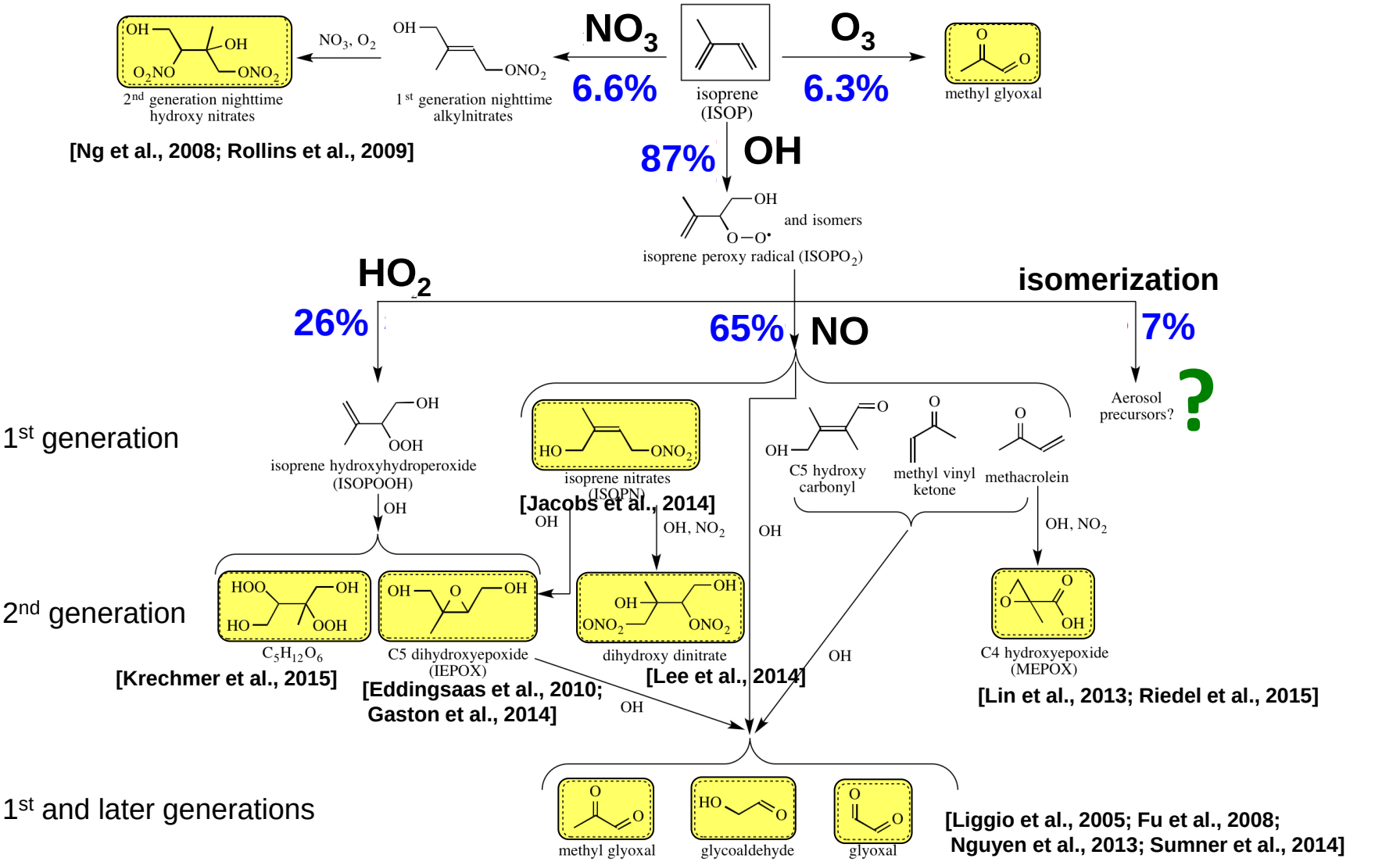
Large **uncertainty** is H_{eff}

Reversible partitioning of semivolatile precursors **retained for ISOP+OH** and **removed for ISOP+NO_3**

Isoprene OA precursors in GEOS-Chem

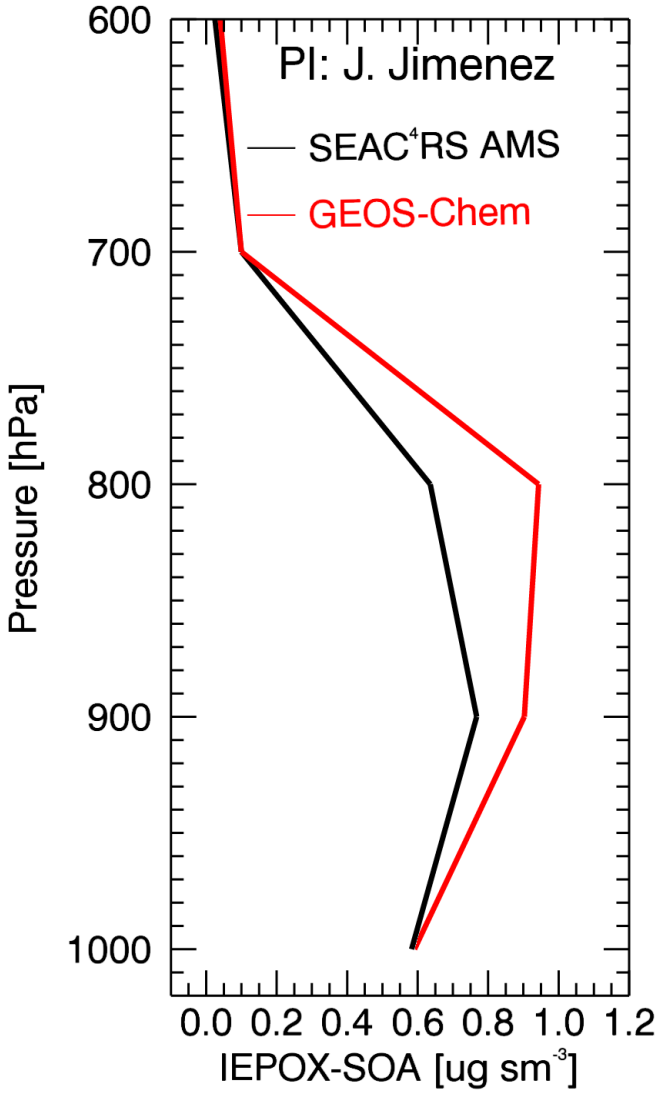
GEOS-Chem branching ratios:
Southeast US (Jun-Aug)

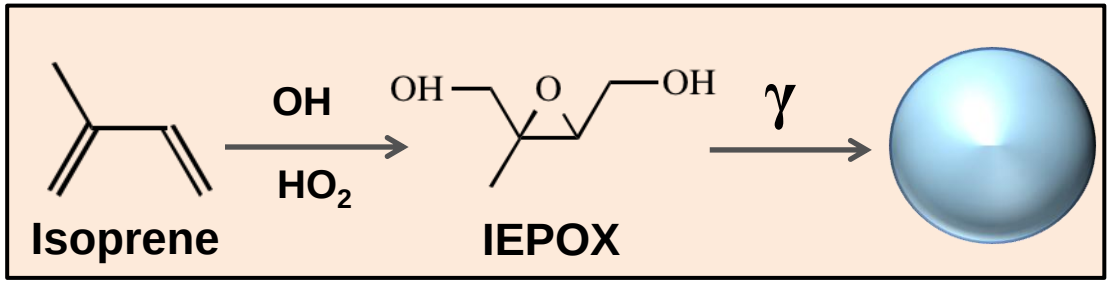
Aerosol precursors



GEOS-Chem isoprene OA during SEAC^{4}RS

Vertical profile of AMS-derived IEPOX-SOA and GEOS-Chem IEPOX-SOA





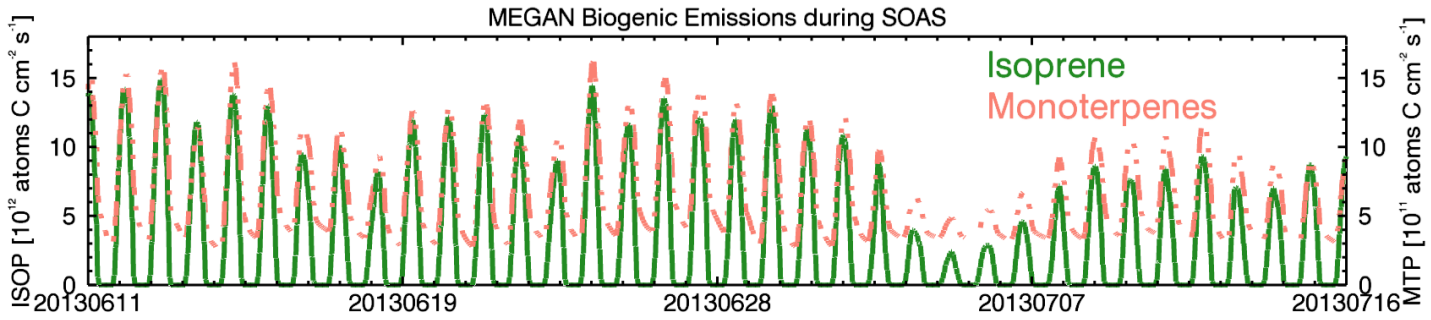
Relatively uniform distribution throughout the boundary layer

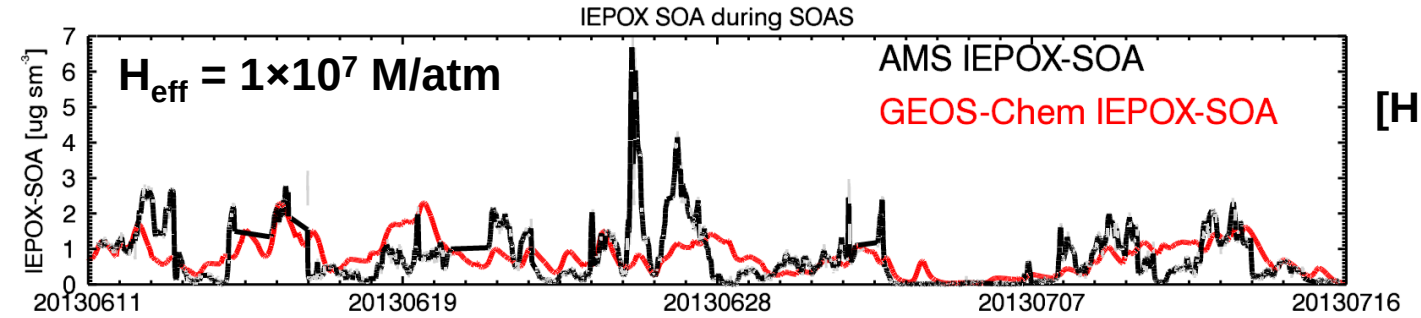
Slight model overestimate likely caused by gas-phase overestimate in IEPOX, but also likely due to uncertain uptake dynamics.

IEPOX-SOA data provided by P. Campuzano-Jost

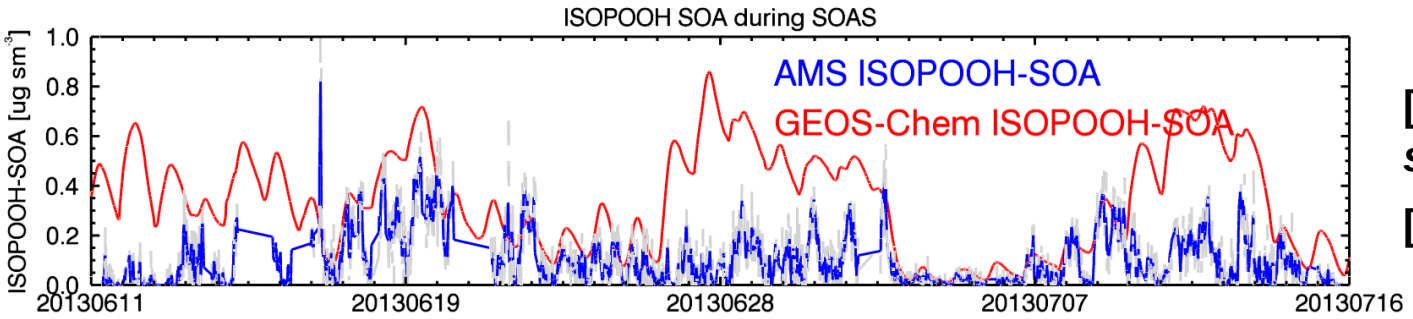
SOAS AMS and GEOS-Chem isoprene OA

Timeseries of MEGAN biogenic emissions, and GEOS-Chem and AMS IEPOX-SOA and ISOPOOH-SOA (Centreville, AL)





[Hu et al., ACPD, 2015]



[Krechmer et al., *submitted*, 2015]

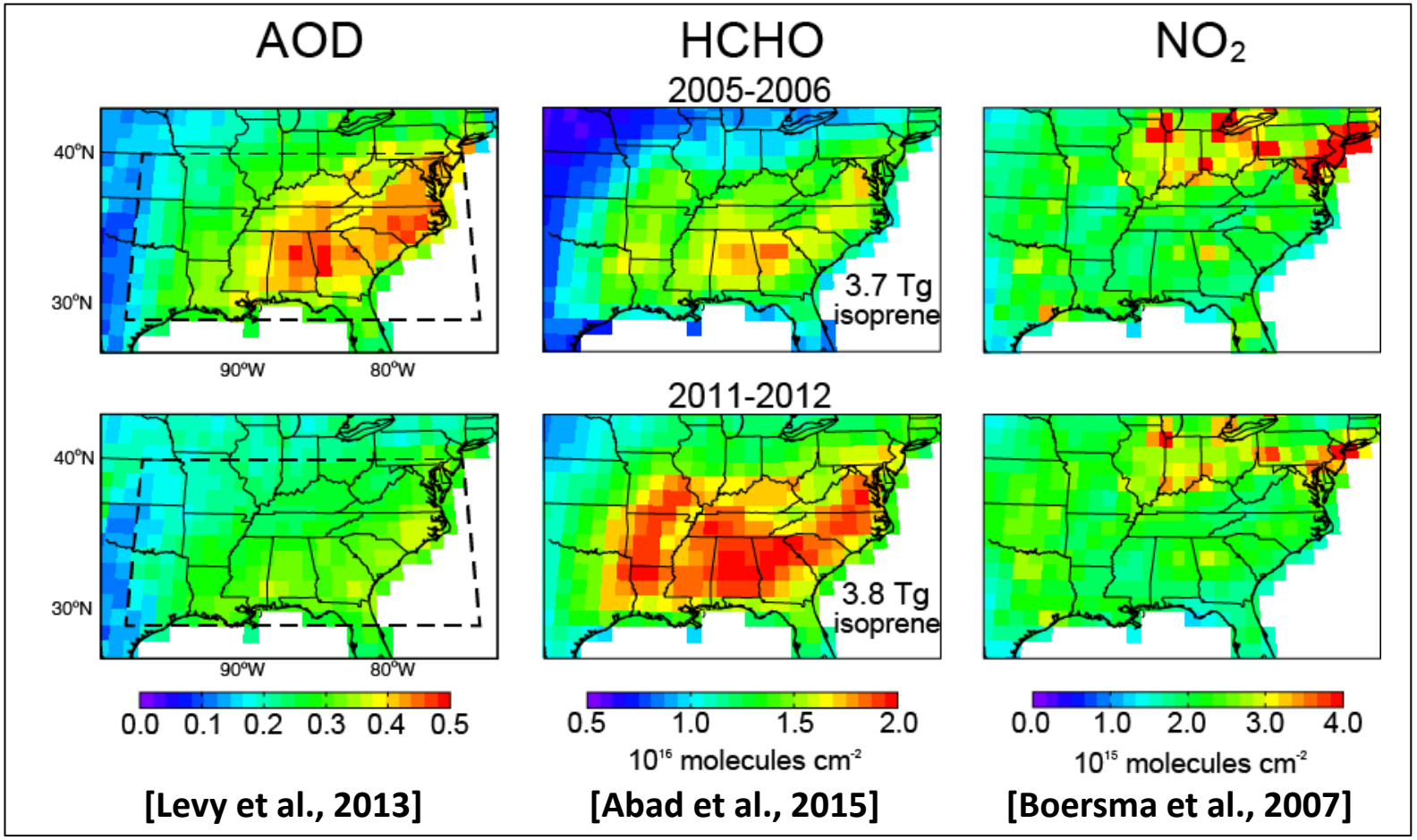
[Liu et al (in prep)]

IEPOX-SOA is ~2 µg m^{-3} during SOAS. ISOPOOH-SOA is an order of magnitude lower.

GEOS-Chem captures temporal variability during SOAS. GEOS-Chem IEPOX-SOA sensitive to effective Henry's constant (H_{eff})

Satellite-derived isoprene OA yields

Satellite Observations over the southeast US in June-August



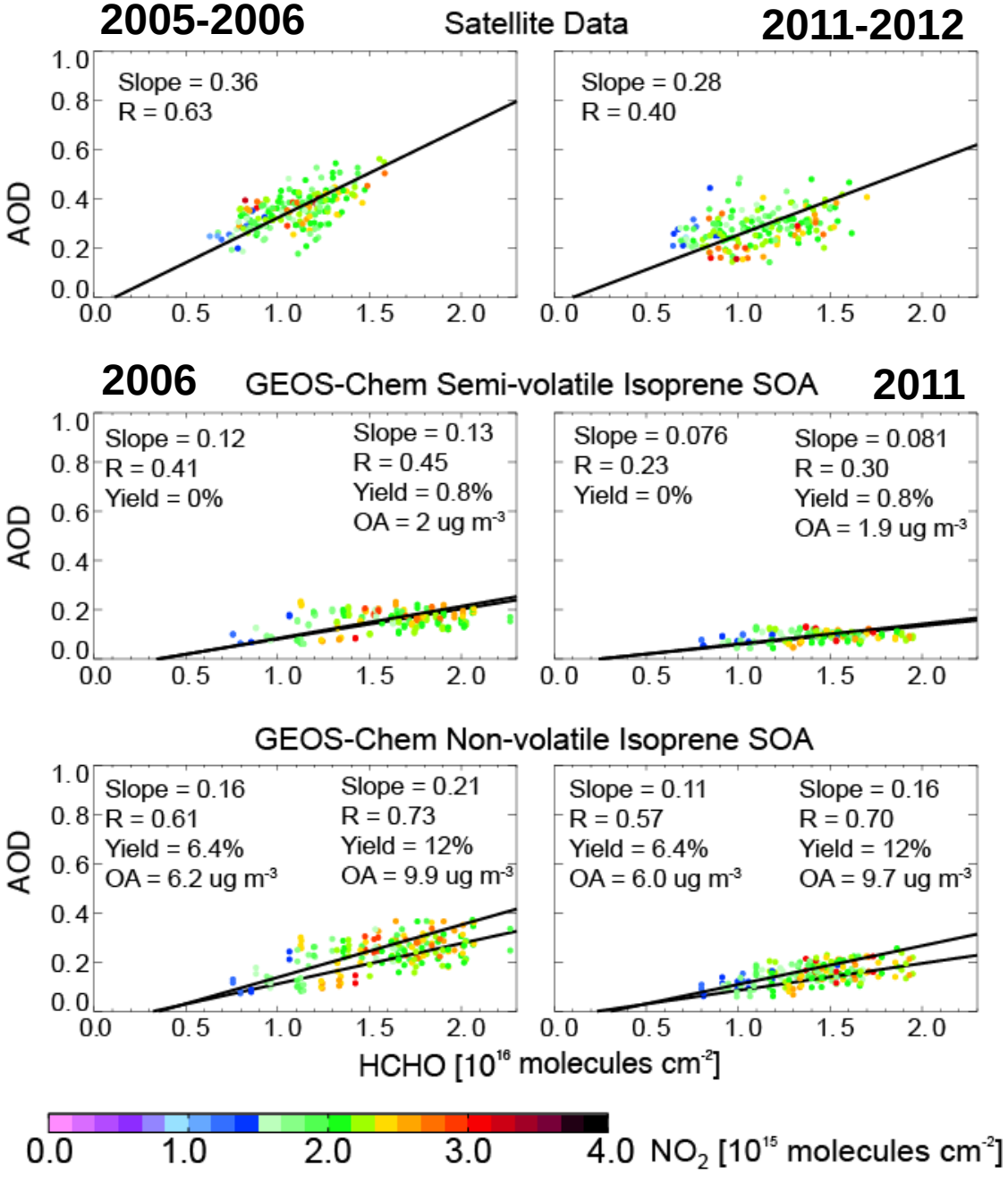
Large decline in anthropogenic emissions, so consider 2 time periods.

Coincident enhancements in **AOD and HCHO** **[Veefkind et al., 2011]**, but AOD includes **ubiquitous inorganic aerosols**.

Interpret **relationship between satellite AOD and formaldehyde** with the GEOS-Chem CTM.

Test effect of **sulfate mass loading** on isoprene OA yields using top-down approach.

Satellite-derived isoprene OA yields



Points are monthly mean 2x2.5 degree data colored by tropospheric column NO_2.

Top panel: Satellite AOD-HCHO

Middle: GEOS-Chem semivolatile partitioning with and without isoprene SOA

Bottom: GEOS-Chem with irreversible uptake of isoprene SOA and 2 different yields

GEOS-Chem AOD-HCHO relationship has little sensitivity to changes in the underlying isoprene SOA

Concluding Remarks and Future Work

- Isoprene OA yields are ~3% from fitting relationship between observed OA and HCHO over the SEAC4RS domain.
- Resolved isoprene OA components during SEAC4RS and SOAS campaigns provide constraints on isoprene SOA formation
- Test model simulation of aerosol-phase IEPOX organosulfate using PALMS IEPOX-organosulfate measurement
- Satellite-derived yields limited by low AOD sensitivity to isoprene SOA yields
- Replace monoterpene SOA volatility basis set scheme with monoterpene SOA formation that couples the gas and aerosol phases.
- Estimate the contribution of individual isoprene SOA components to the isoprene SOA budget using GEOS-Chem