Isoprene SOA yields derived from space-based observations of AOD and HCHO

D. J. Jacob, G. G. Abad, K. Chance, J. Jimenez, A. Fried, P. Kim, C. Miller

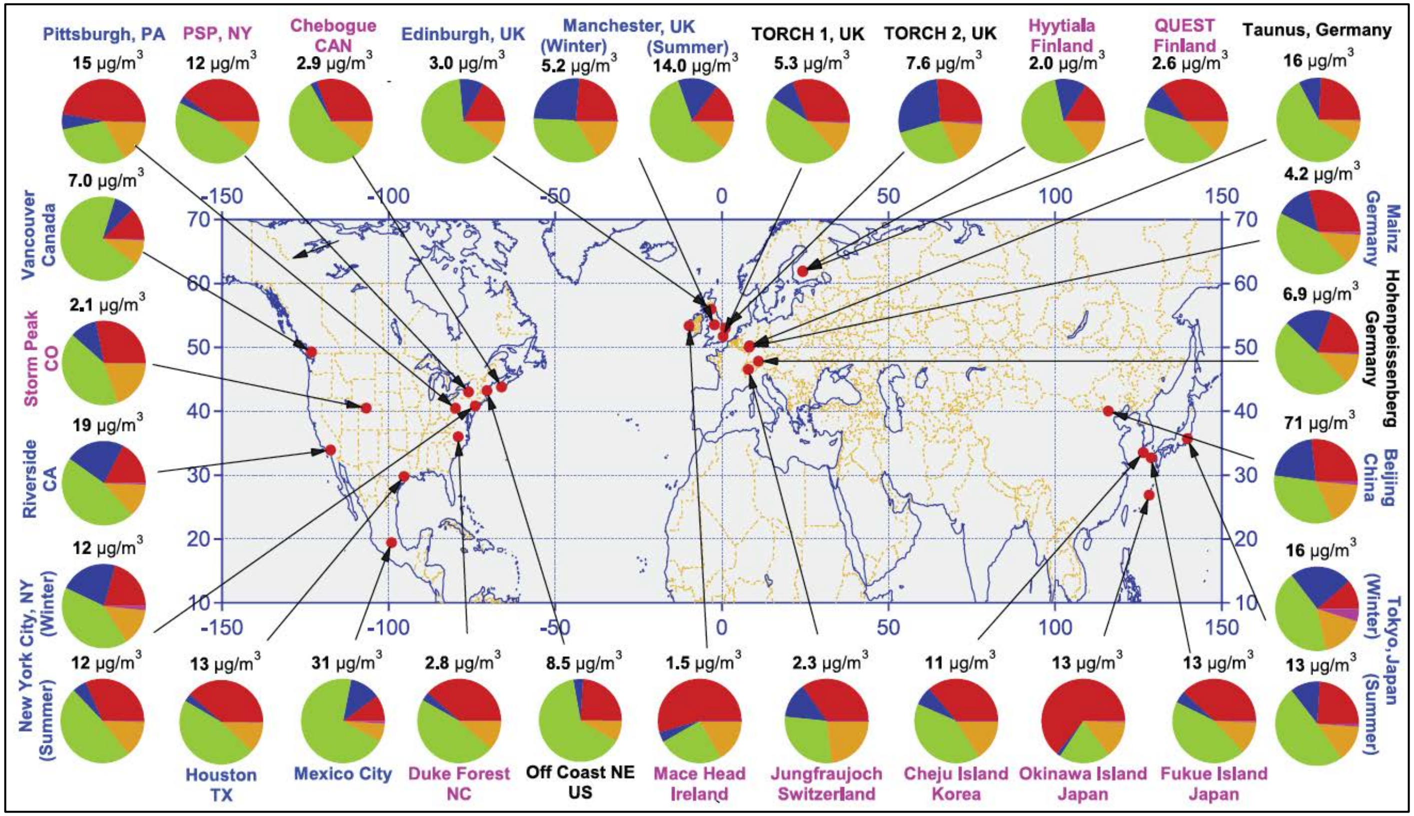
Eloïse Marais

CIRES, CU Boulder

11 February 2015

Organic Aerosol Component Dominates Aerosol Composition

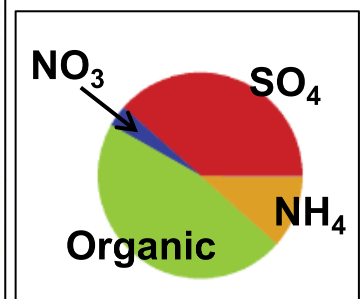
…And is becoming increasingly relevant in the US and Europe



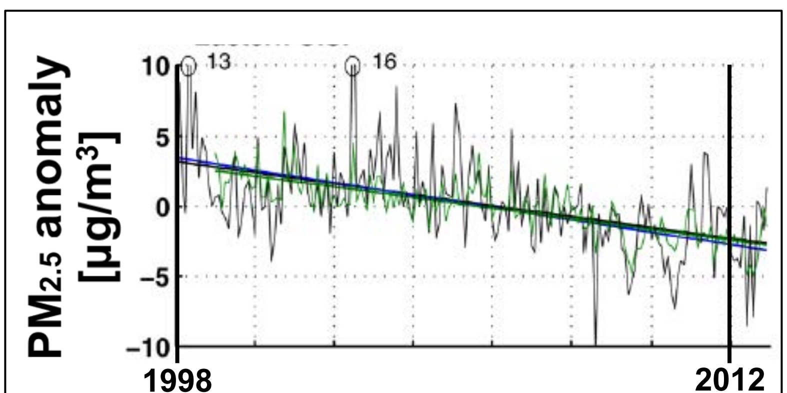
Isoprene SOA of **12-19 %** of organic PM2.5 observed in rural SE US

[Lin et al., 2013]

KEY



Zhang et al. (2007)

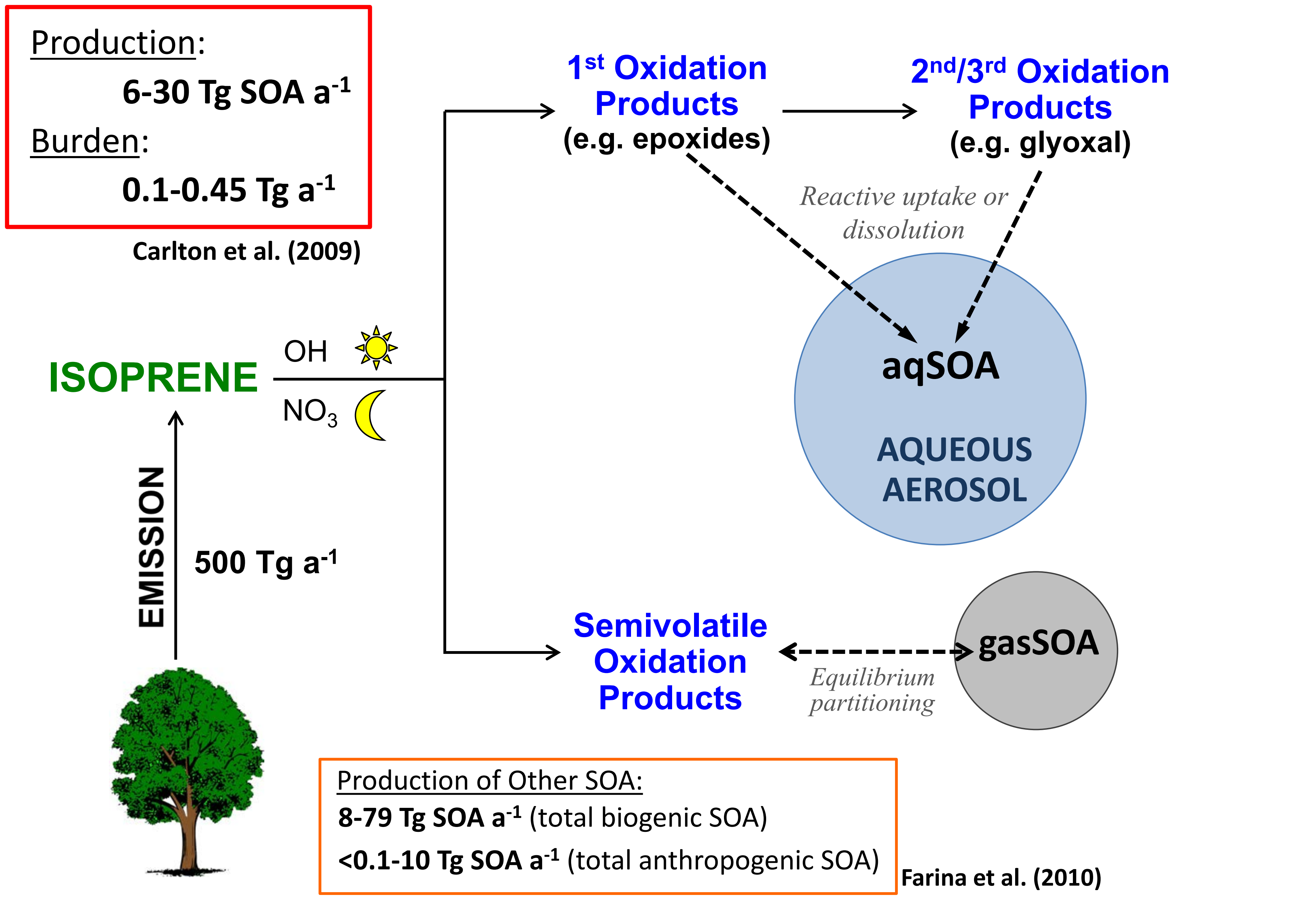


Boys et al. (2014)

Trend in PM2.5 over the eastern US (**-0.4 µg m^{-3} y^{-1}**) obtained with satellite AOD and GEOS-Chem

The trend is attributed to a decline in **inorganic** (NH_4-SO_4-NO_3) **aerosols**.

Formation of Secondary Organic Aerosols from Isoprene



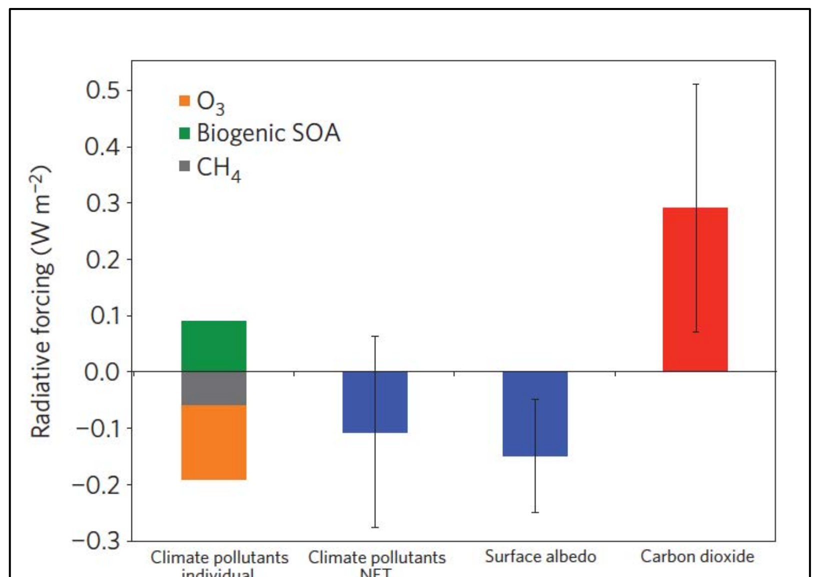
Impact of Isoprene SOA On Climate

Study	DRE [W m^{-2}]*	Evaluation
Hoyle et al. (2009)	**-0.09 or -0.06**	Pre-industrial vs Present SOA
Goldstein et al. (2009)	**-3.0**	Summer vs Winter biogenic SOA
Unpublished**	**-0.02**	Pre-industrial vs Present SOA
O' Donnell et al. (2011)	**-0.31**	With vs Without SOA
Scott et al. (2014)	**-0.08 to -0.78**	With vs without biogenic SOA
Unger (2014)	**+0.09**	1750 vs present landcover

*All results are from global models, except Goldstein et al. (2009), which is obtained with satellite AOD over the SE US

**Value given in Arneth et al. (2010)

Positive forcing if consider effect of landcover change on BVOC emissions:



Unger (2014)

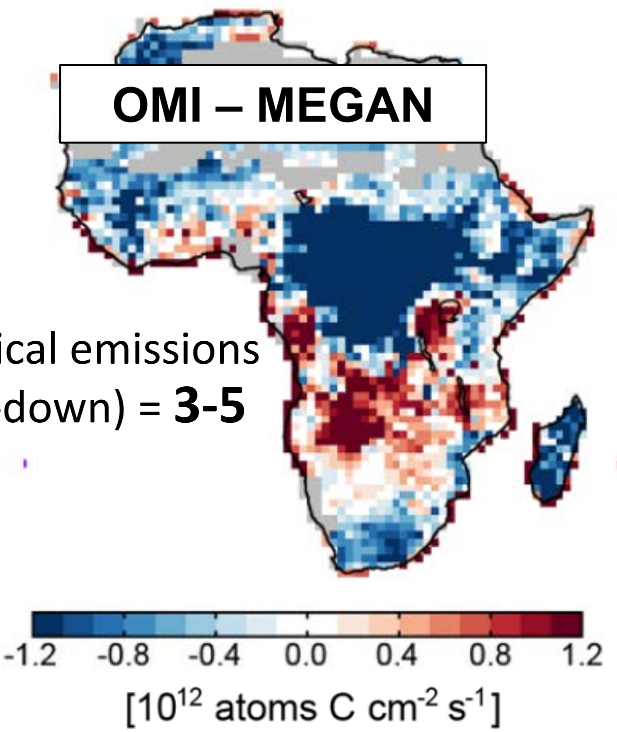
All model studies only include semivolatile SOA precursors

Impact of Isoprene SOA On Climate – Uncertainties

Isoprene Emission Inventory

Uncertain by at least a **factor of 2**

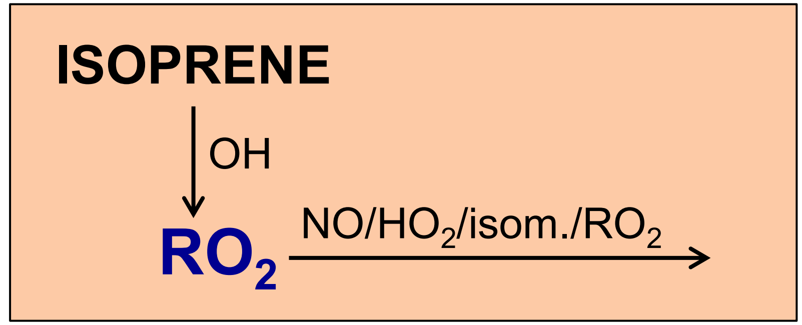
Difference between top-down and MEGAN isoprene emissions



Tropical emissions (top-down) = **3-5**

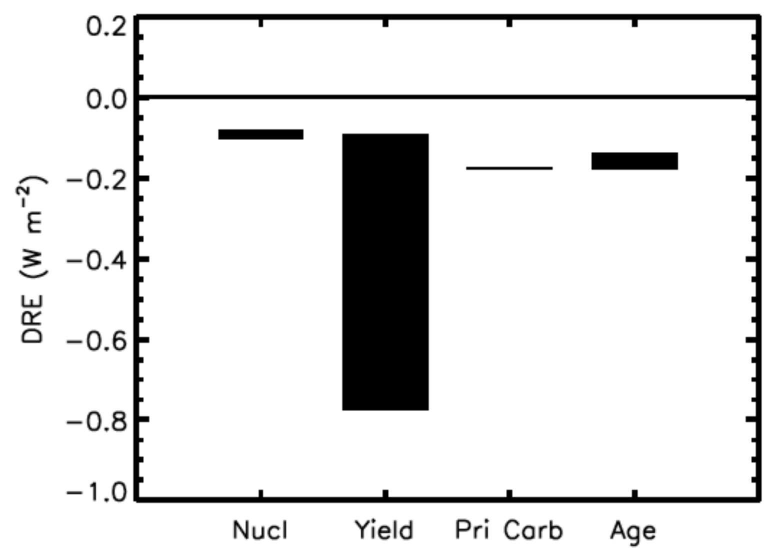
Marais et al. (2012)

Gas-phase Isoprene Oxidation



Yield of SOA from isoprene

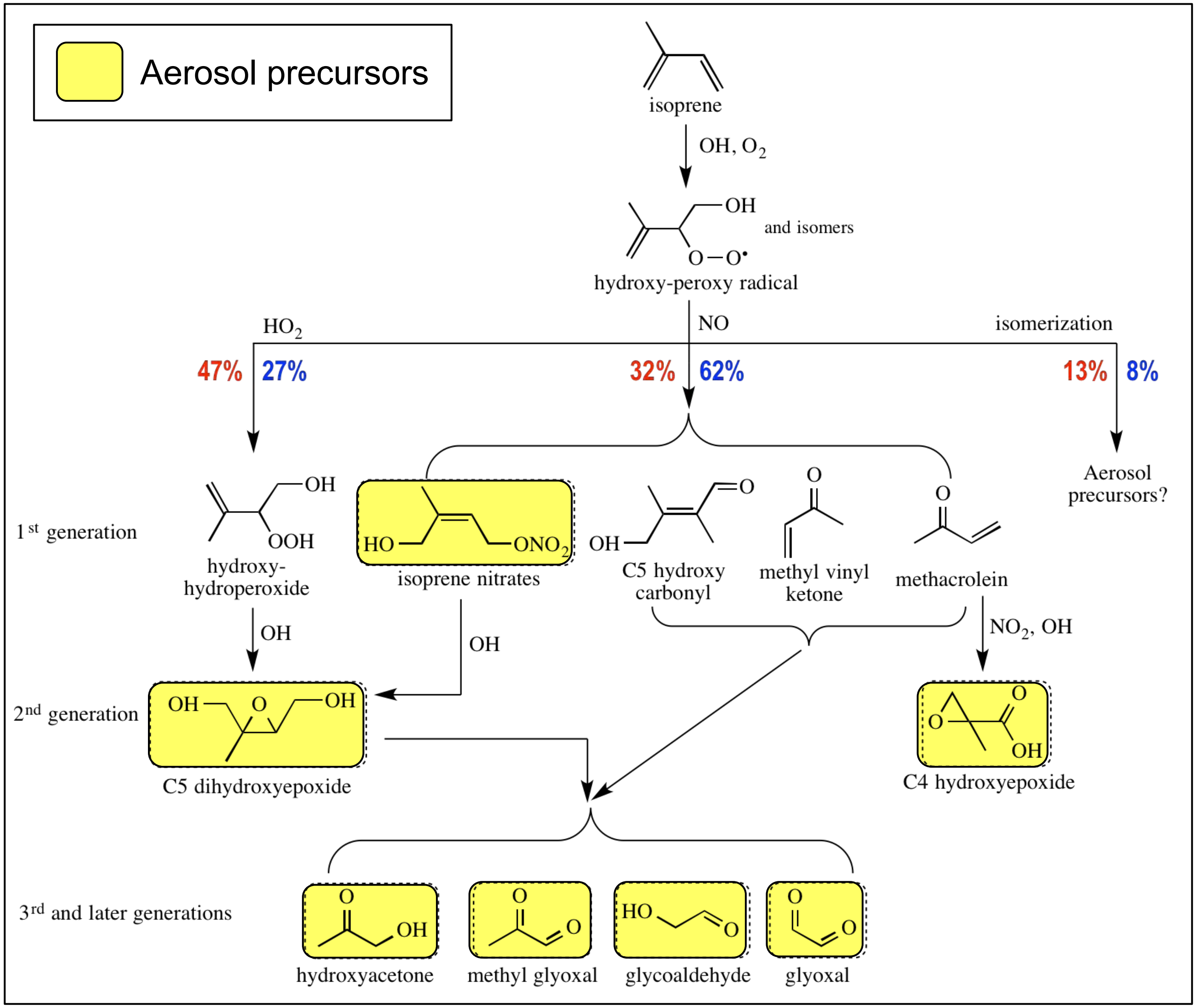
Effect of SOA yield on DRE estimate



Scott et al. (2014)

Aerosol Precursors from Isoprene Oxidation

Aerosol precursors from NO and HO_2 dominant oxidation, but isomerization products are uncertain.



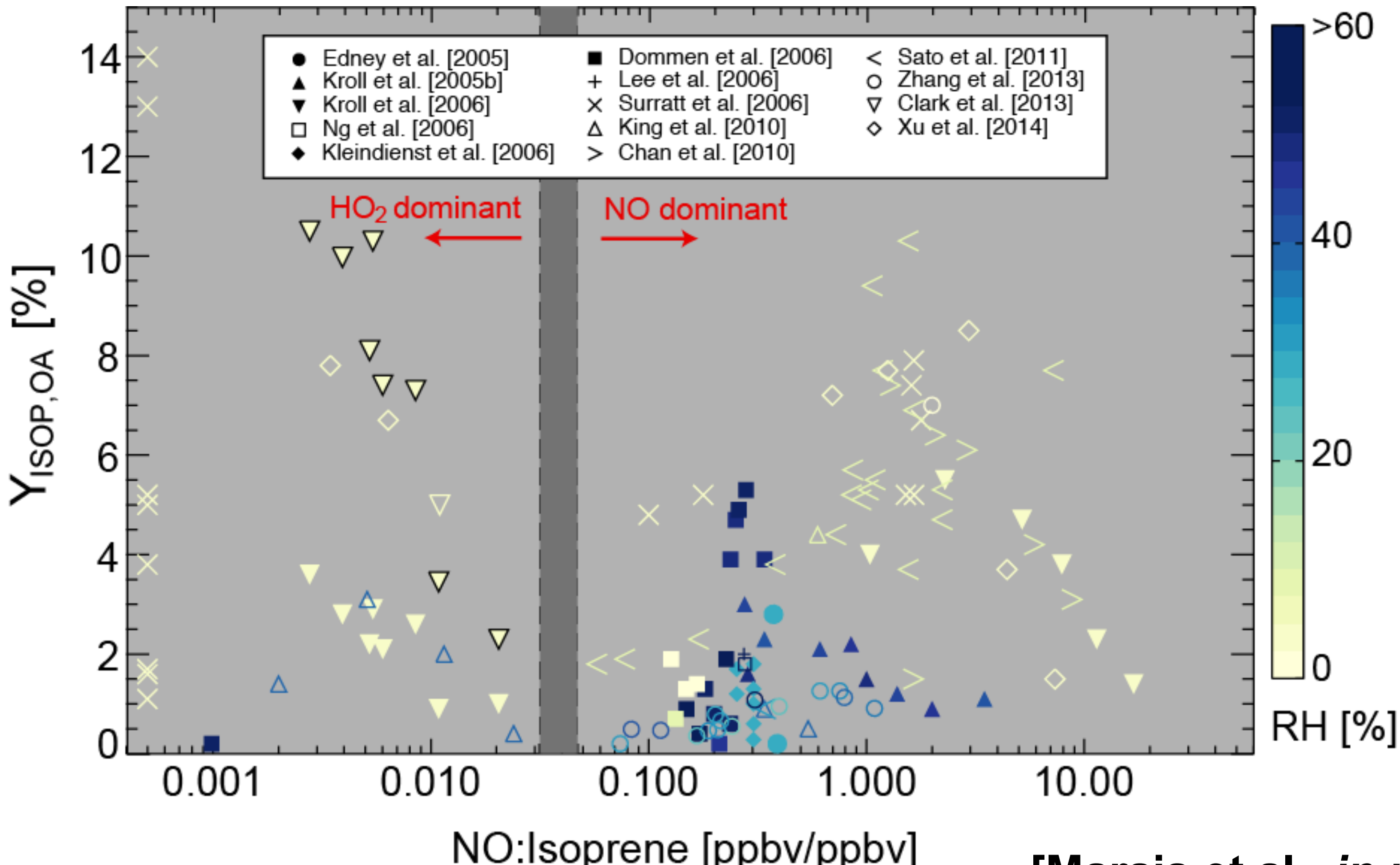
GEOS-Chem branching ratios are for the **southeast US** and **Africa**

[Marais et al, in prep]

Laboratory Estimates of Isoprene SOA Yields

Mass yield of isoprene SOA: $Y_{SOA} = \frac{\Delta SOA}{\Delta isoprene}$

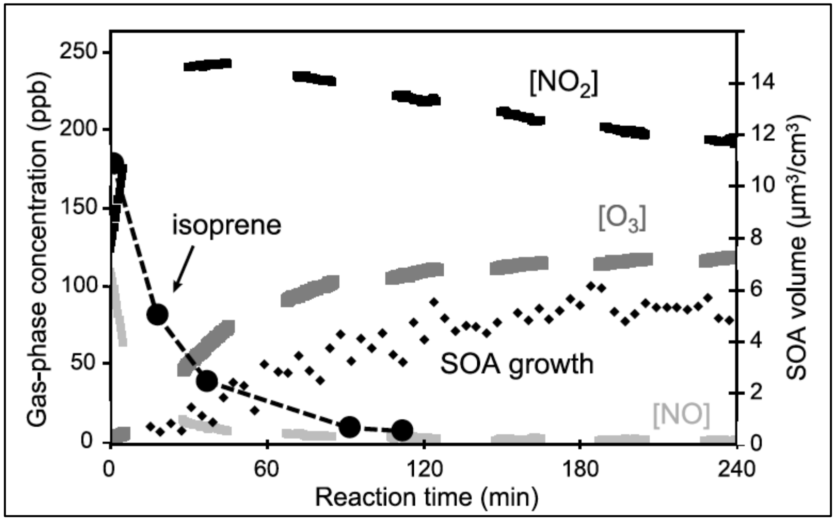
A large range of SOA yields are obtained in chamber studies



[Marais et al., *in prep*, 2015]

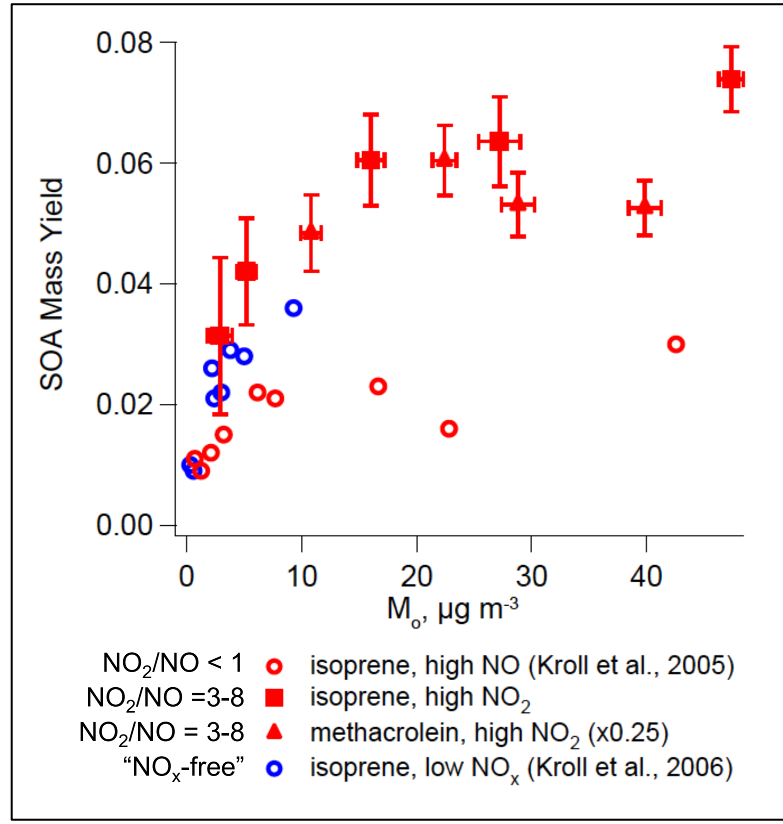
Laboratory Estimates of Isoprene SOA Yields

High initial isoprene (>100 ppbv) and NO_x (> 200 ppbv)



Kroll et al. (2005)

Experimental yields sensitive to chamber conditions (e.g. NO_2/NO)



Chan et al. (2010)

α-pinene yields:

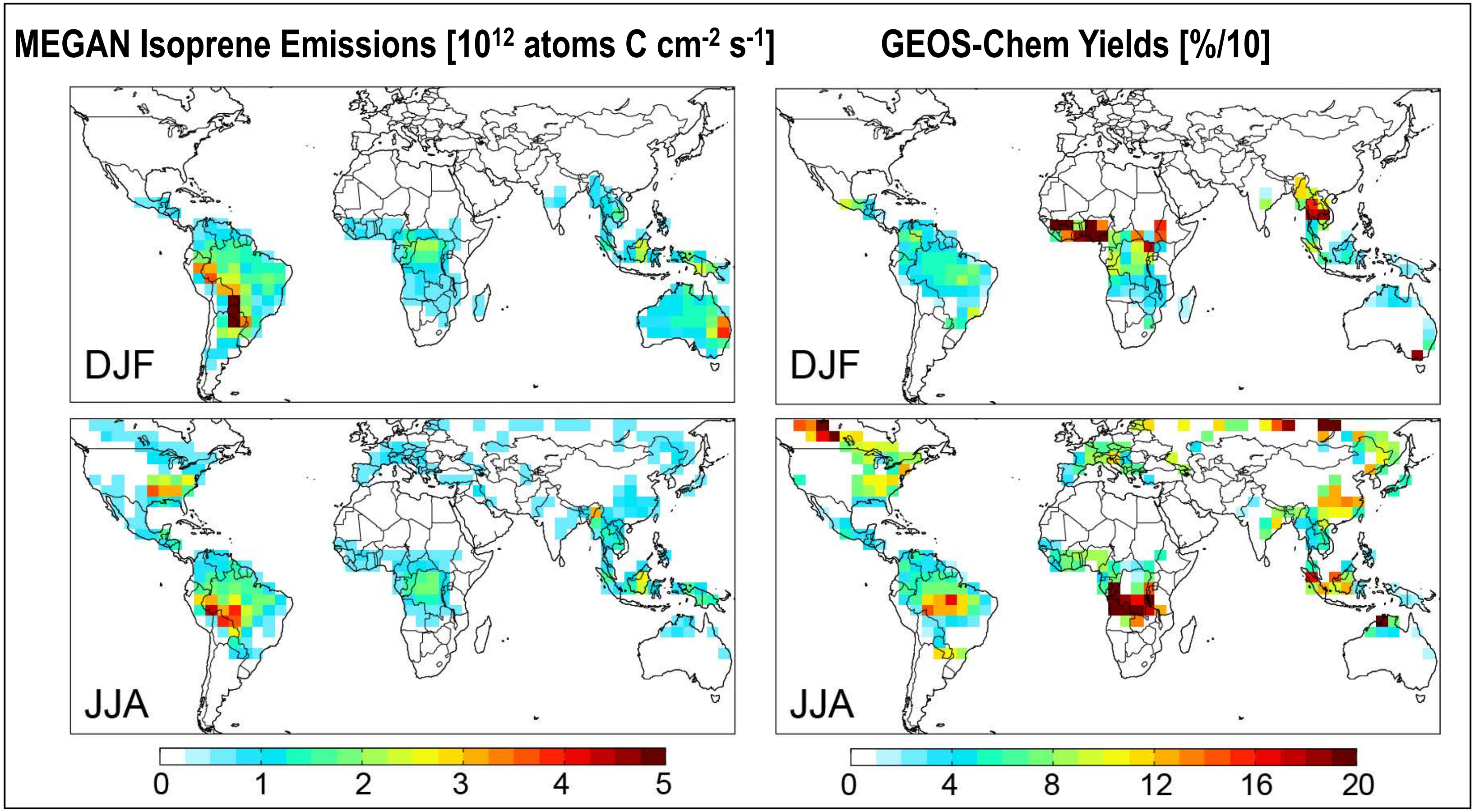
20% from dark ozonolysis and reaction with OH (less variability than isoprene)

[Shilling et al., 2008; Carlton et al., 2009; Eddingsaas et al., 2012]

Isoprene SOA In GEOS-Chem

Yield in GEOS-Chem: $$Y_{SOA} = \frac{\text{net SOA produced}}{\text{isoprene lost}}$$

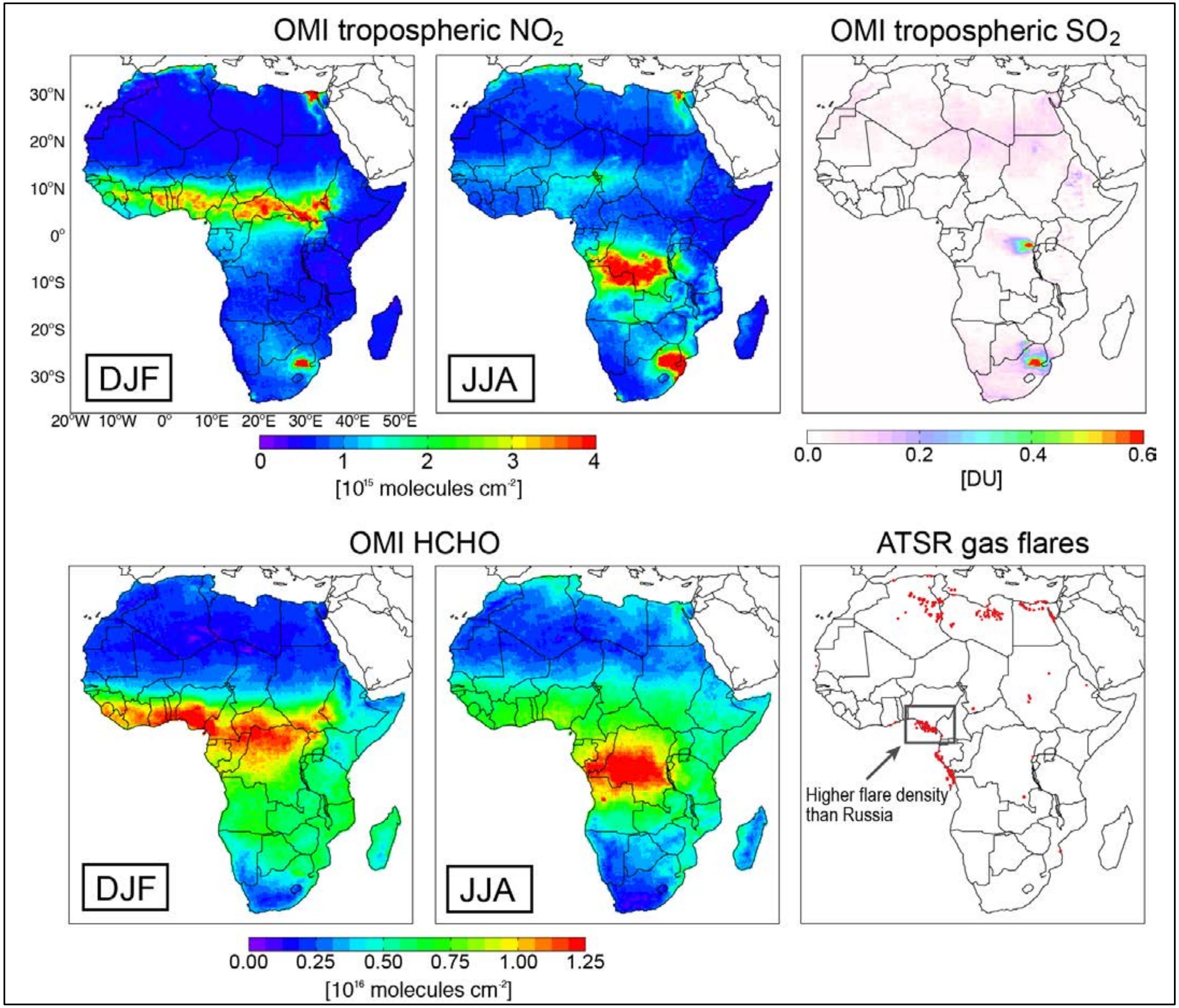
GEOS-Chem Yields not spatially consistent with isoprene emissions:



Depends heavily on **pre-existing aerosols** (biomass burning)

Satellites Enhance Understanding of Atmospheric Composition

…in particular in Africa, where ground observations are sparse



Surface NO_x is from biomass burning, soils, and fossil fuels.

Reactive NMVOCs predominantly from inefficient sources of combustion.

Similar **SO_2 emissions** from non-eruptive volcanoes and fossil fuels.

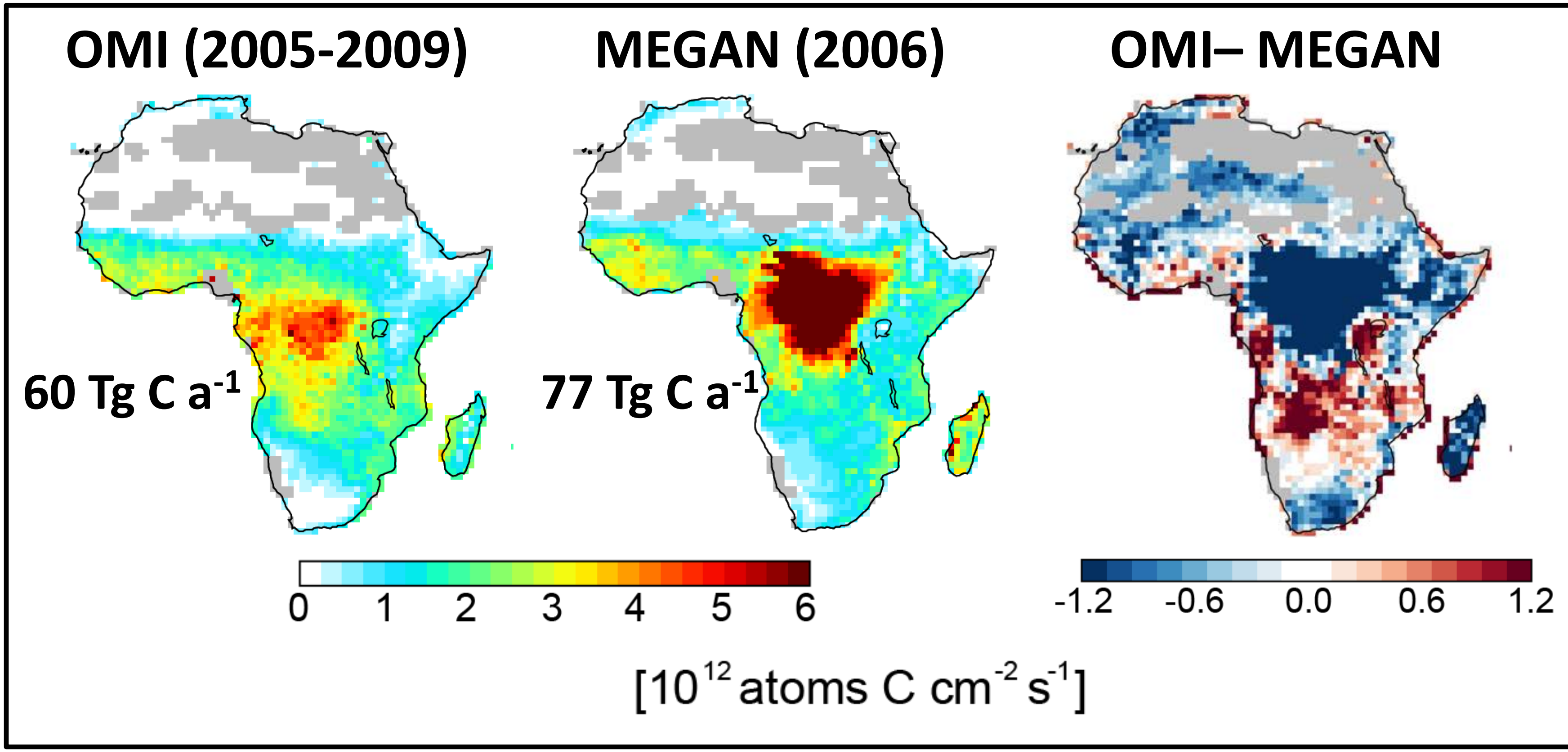
NO_2 → surface NO_x sources

HCHO → reactive NMVOCs

Gas flares → wasted natural gas

Obtain satellite-derived isoprene emissions in Africa

Maps: **Annual Average Isoprene Emissions (12-15 LT)**
Numbers Inset: **Total (24h) isoprene emissions**



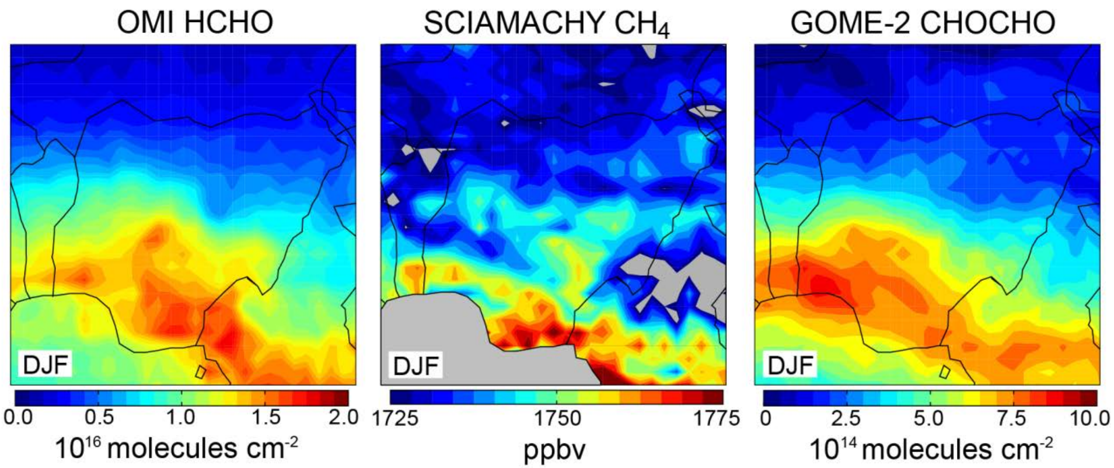
Small difference in total emissions, but large regional discrepancies .

[Marais et al., ACP, 2012]

Constrain air quality in Nigeria

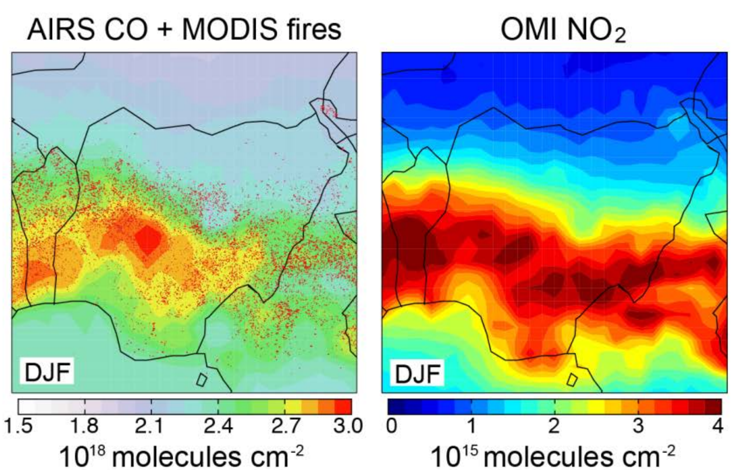
Biomass burning, inefficient combustion in Lagos, and extensive natural gas leakage and flaring visible from space.

Anthropogenic Volatile Organic Compounds

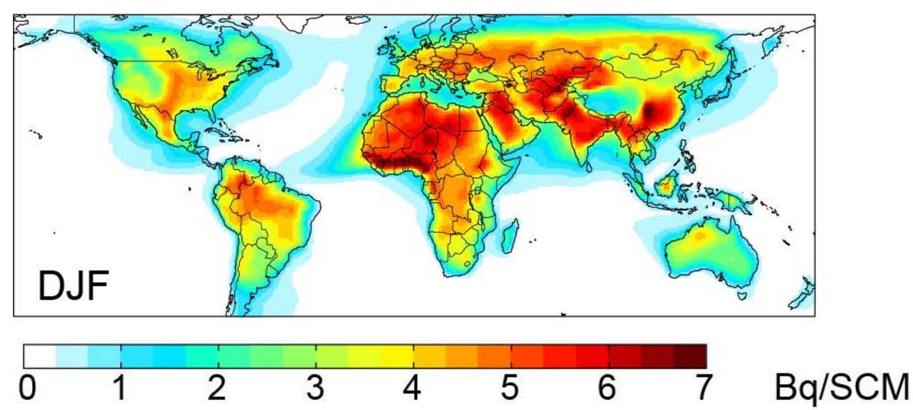


[Marais et al., AE, 2014]

Seasonal open fires

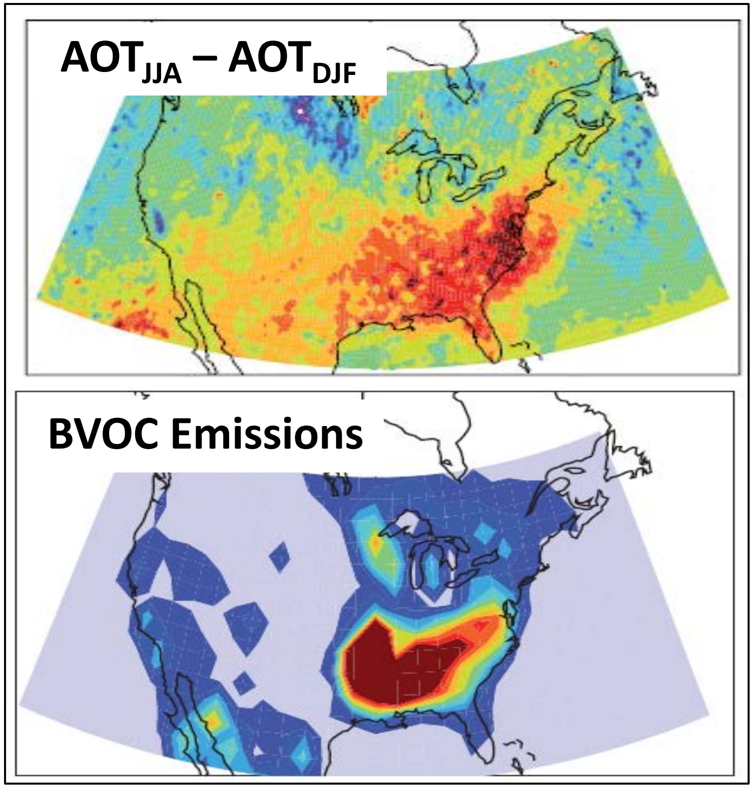


Severely restricted ventilation (GEOS-Chem mean radon-222)

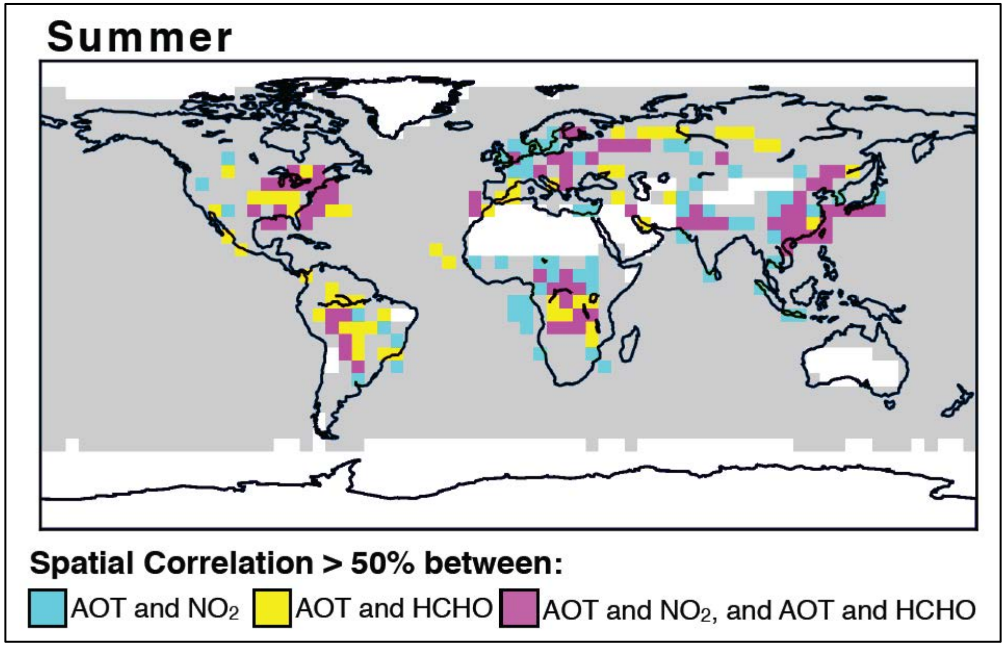


Spatial consistency between **satellite AOD** and **HCHO**

Relation between AOD and biogenic emissions from satellite observations



Goldstein et al., 2009



Veefkind et al. (2011)

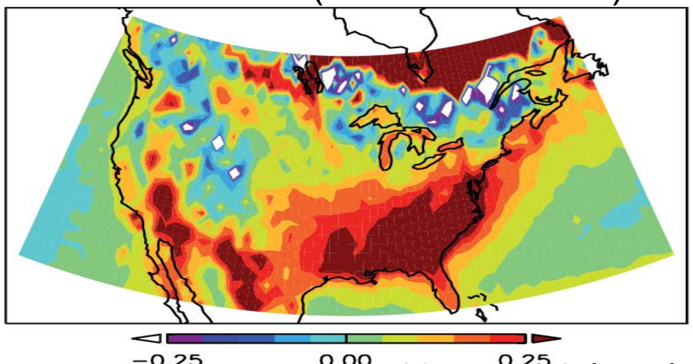
Correlation between **AOD and HCHO only** suggests a large biogenic SOA source from isoprene

Contamination from **biomass burning** limits spatial coverage of this correlation

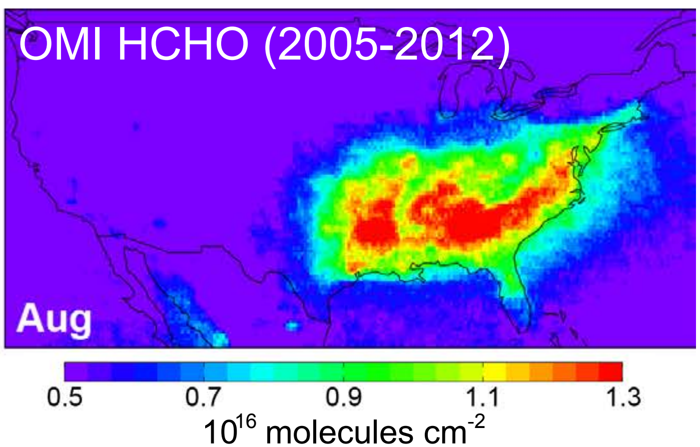
Use **spatial consistency** between **satellite AOD** and **HCHO** to estimate isoprene SOA yields for two environments:

Southeast US (SEUS)

MODIS AOD (summer - winter)



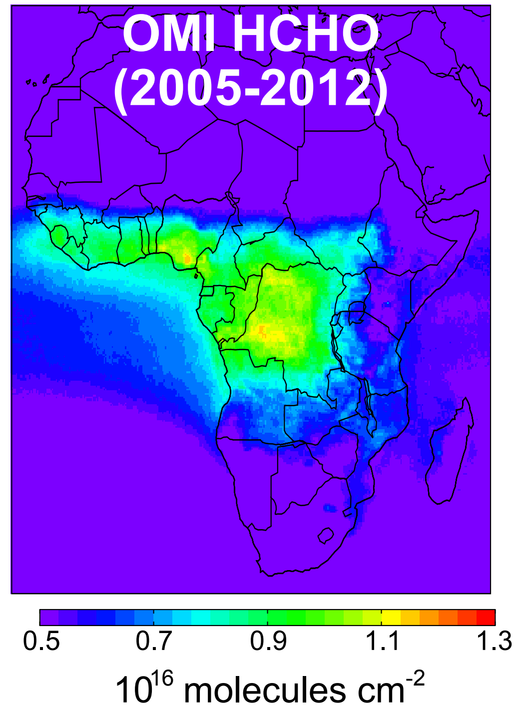
Goldstein et al. (2009)

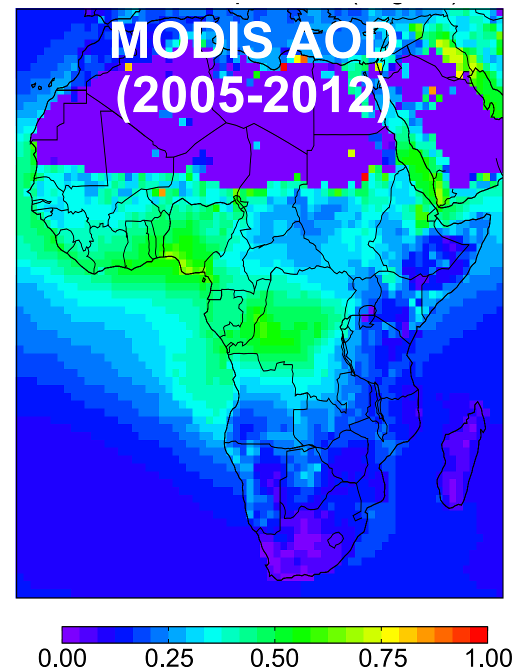


Summertime HCHO: **isoprene**

Summertime AOD: **biogenic SOA**, **SO_4, NO_3, NH_4**

AFRICA

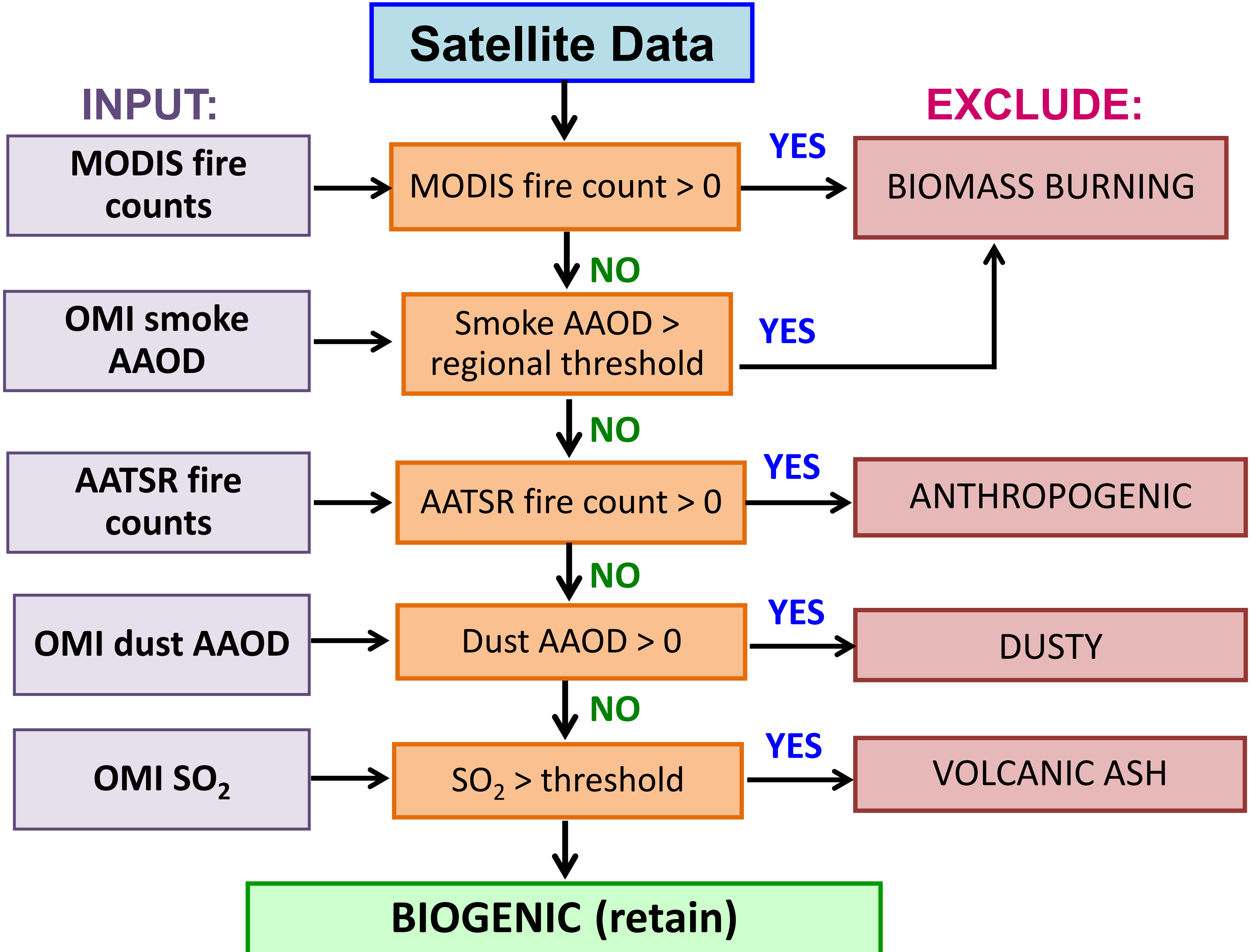




HCHO: **isoprene**, **biomass burning**, **anthropogenic**

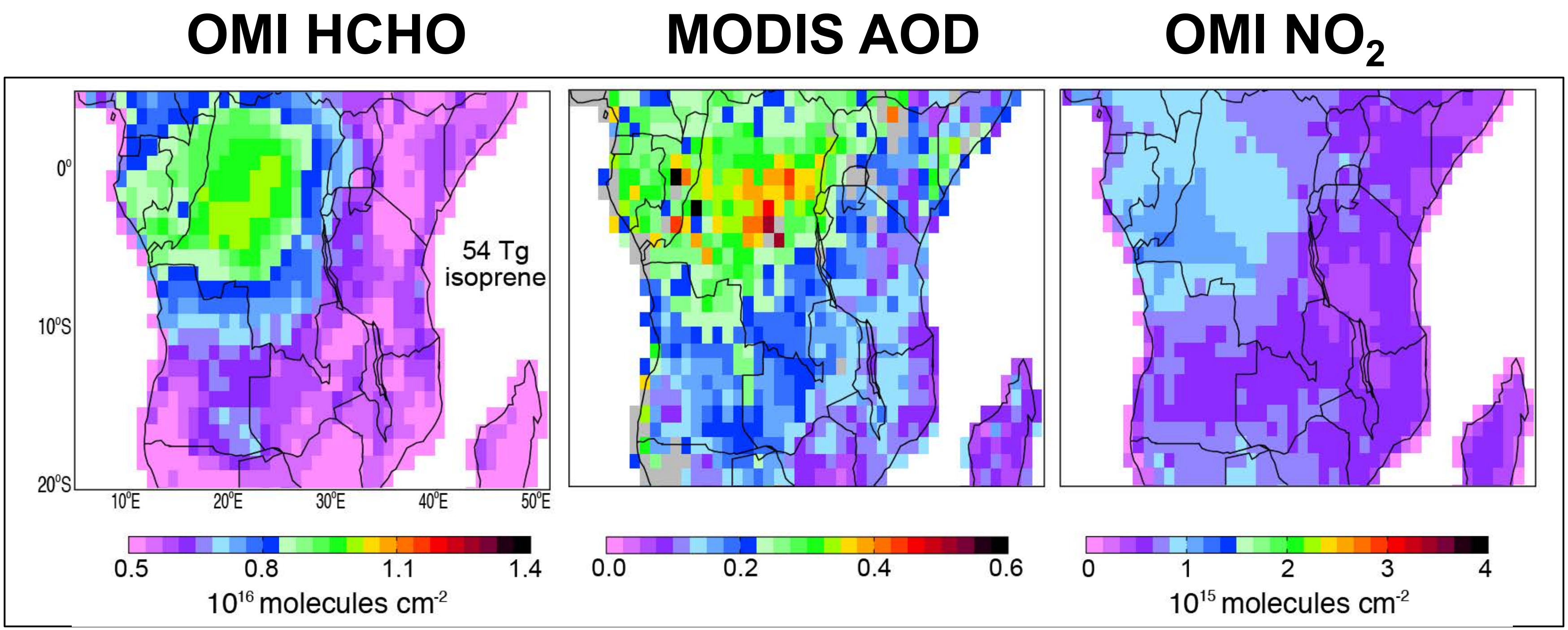
AOD: **biogenic SOA**, **biomass burning**, **dust**, **volcano**

Isolate biogenic signal of HCHO and AOD



Collocation of biogenic satellite HCHO and AOD - Africa

Satellite biogenic AOD and HCHO are spatially consistent in central Africa (annual data for 2005-2012)

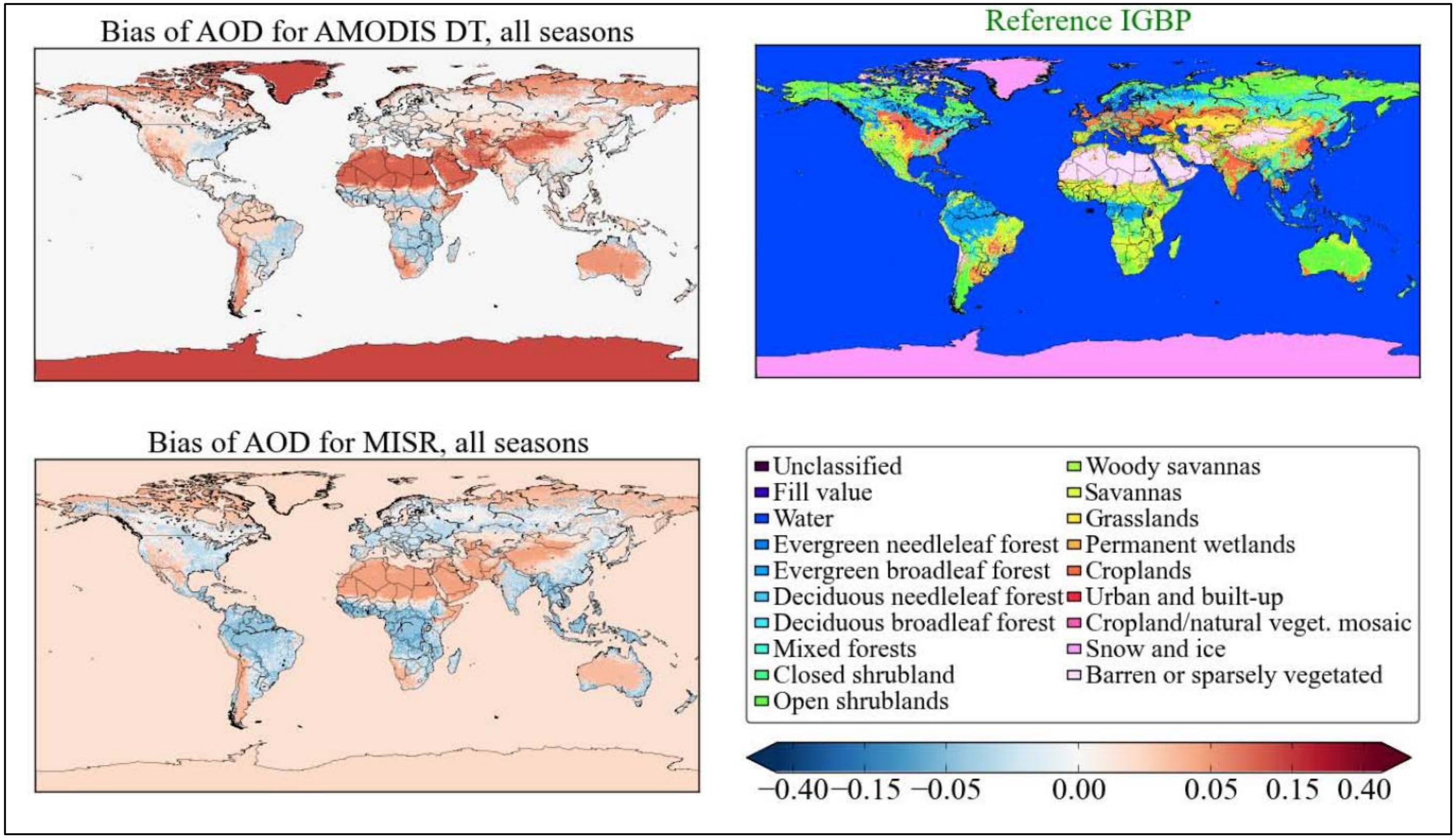


Dust and anthropogenic influence north of 5°N, so only consider central Africa

Effect of land cover type on satellite AOD

Evaluate assumptions about **type and optical properties** of underlying **terrestrial surface**

Land cover type dependent biases for MODIS (Aqua) and MISR

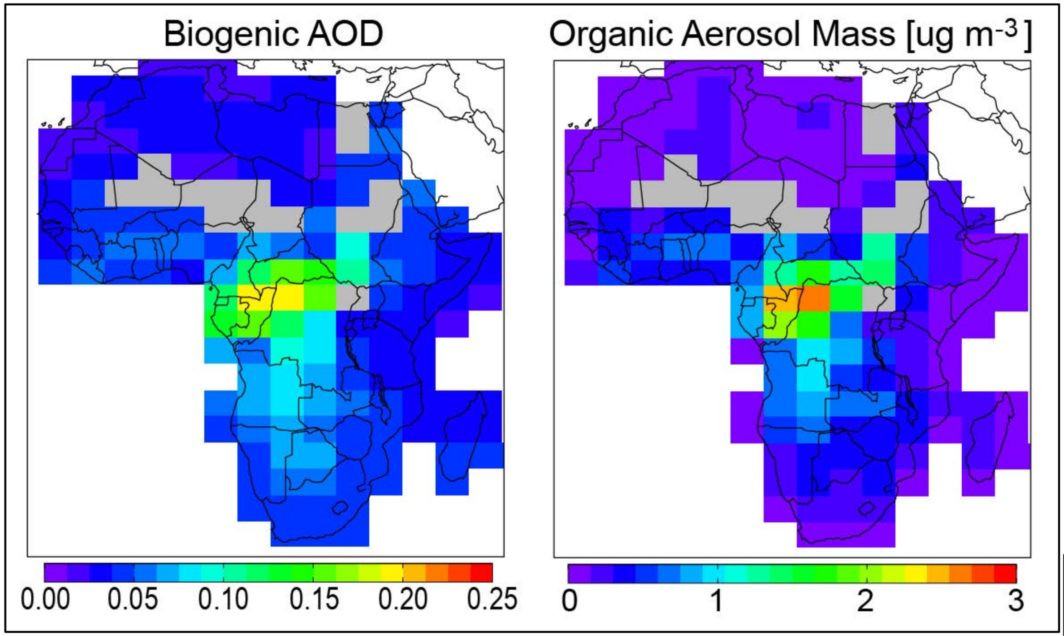


Petrenko & Ichoku (2013)

Strong correlation and low bias (**<0.05**) for AOD over tropical vegetation (**evergreen broadleaf forests**) → ideal, dark surface for satellite AOD.

GEOS-Chem Underestimates Biogenic AOD in Africa

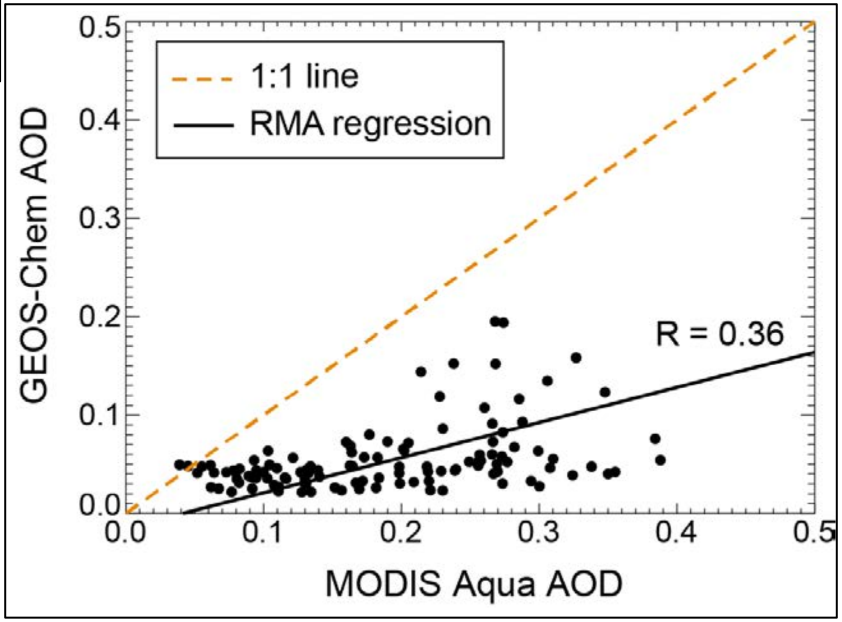
GEOS-Chem (4 × 5°) annual biogenic AOD and biogenic aerosol mass loading



Updates to GEOS-Chem:

- Use **improved MEGAN** isoprene emissions (Africa only)
- Update gas-phase **isoprene oxidation** (minor)

Comparison of GEOS-Chem and MODIS biogenic AOD

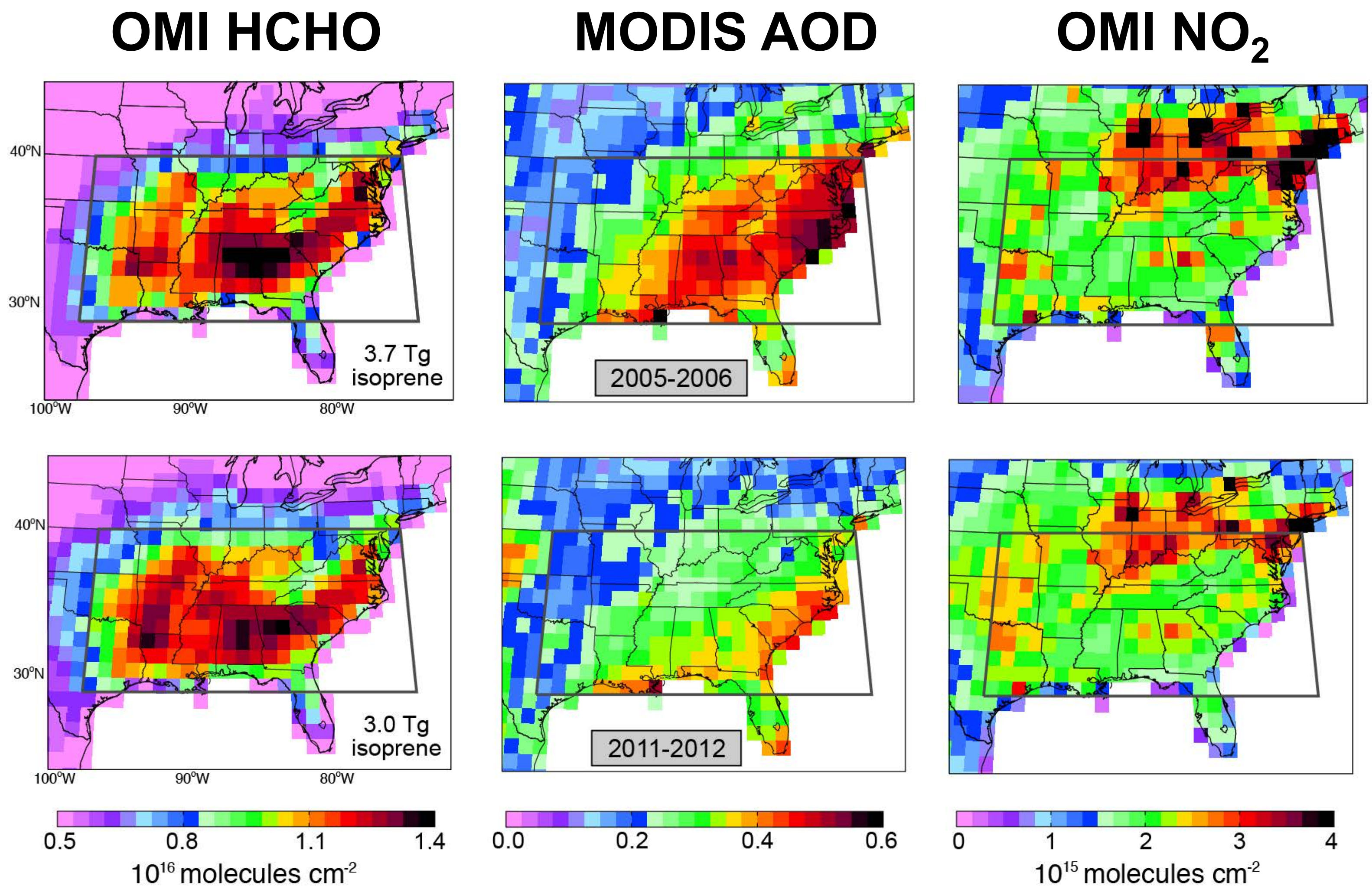


Data have been filtered to remove:

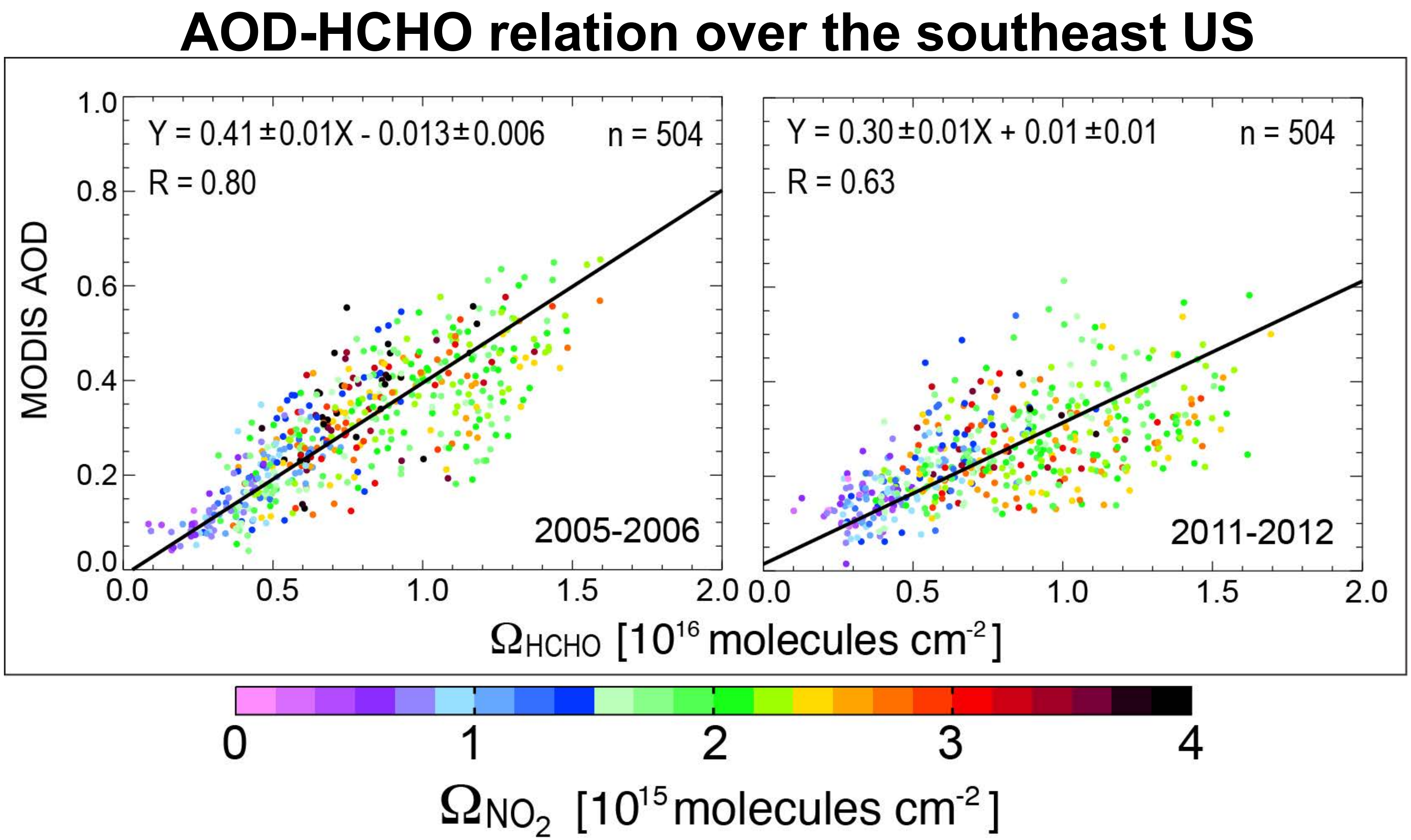
- Biomass burning
- Dusty scenes
- Anthropogenic influences
- Non-eruptive volcanic ash

HCHO and AOD collocation – Southeast US

Satellite biogenic AOD and HCHO are spatially consistent in southeast US (June-August mean)



Satellite HCHO and AOD collocation – Southeast US



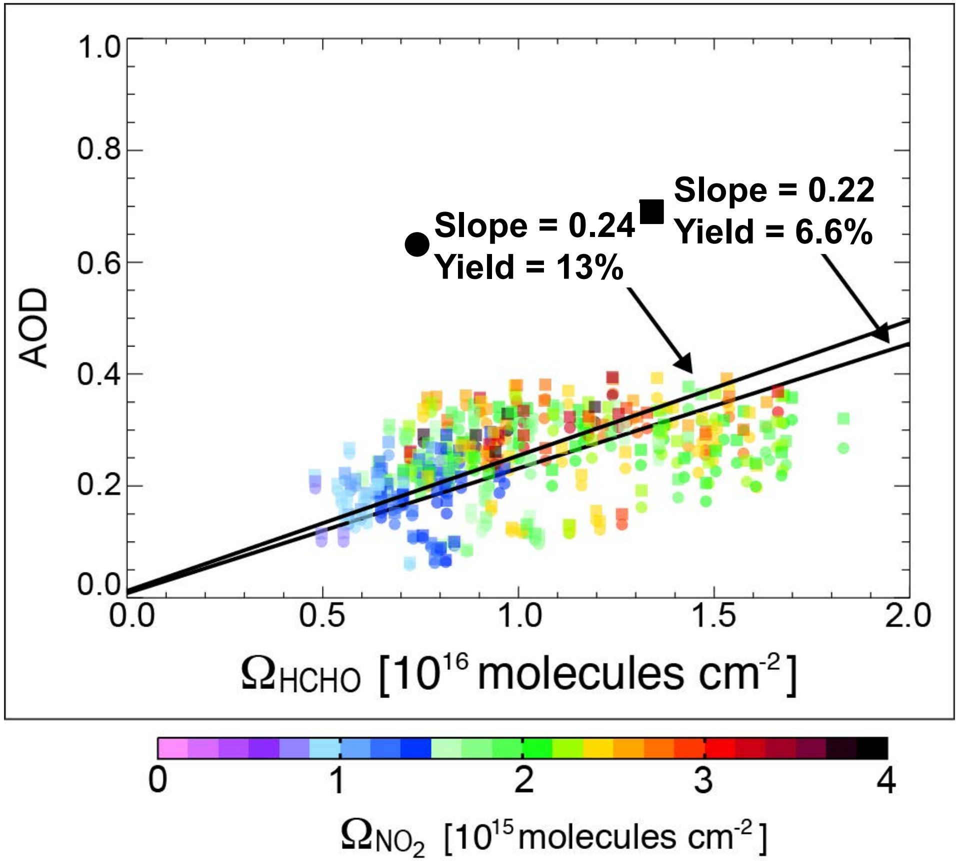
Individual points are monthly means in June-August at 2º × 2.5º grid

Reduced slopes consistent with reduced anthropogenic AOD

Southeast US is at high levels of NO_x (OMI NO_2 > 1 × 10^{15} molecules cm^{-2})

GEOS-Chem AOD-HCHO relationship – Southeast US

Sensitivity of AOD-HCHO to isoprene SOA yields

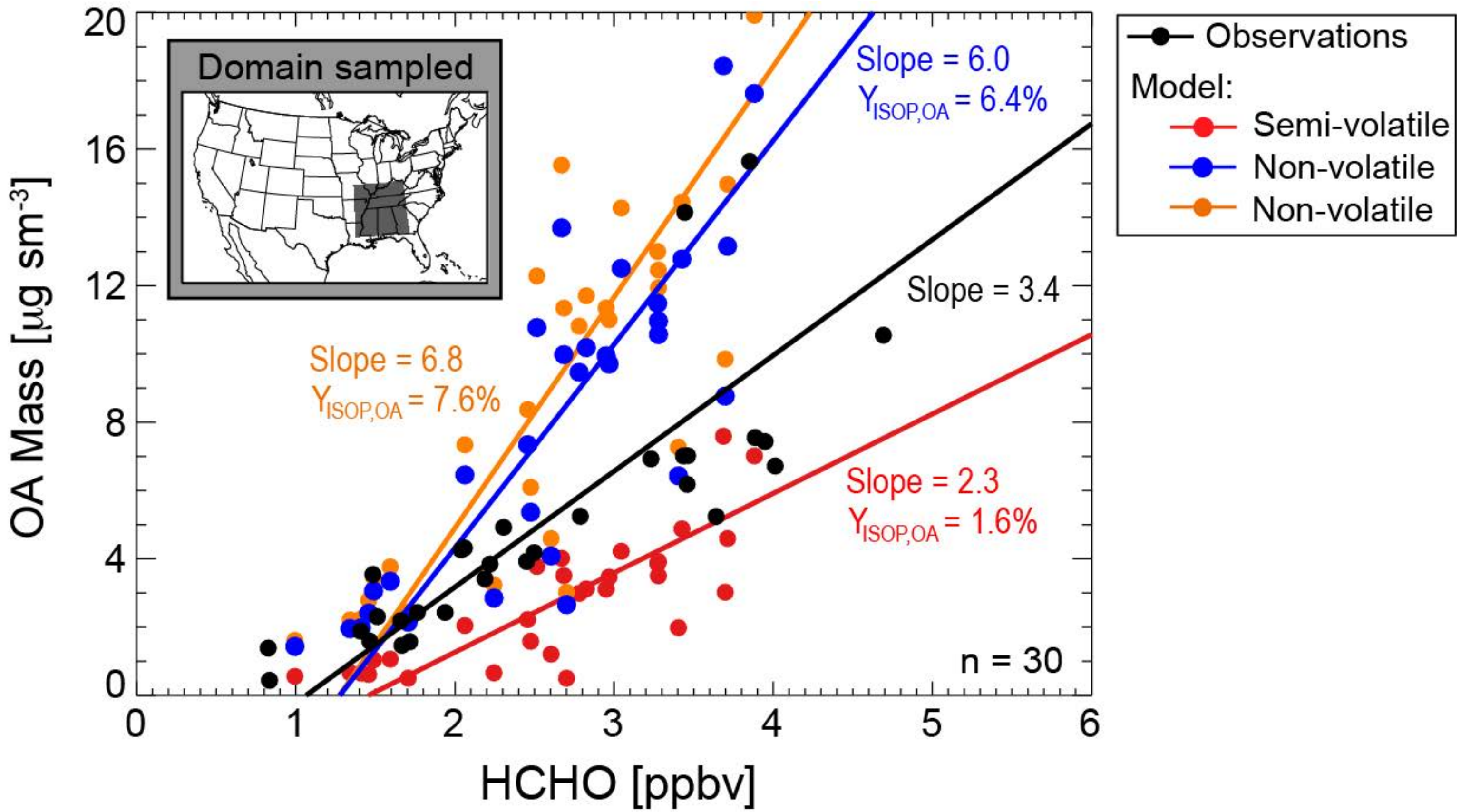


Individual points are model monthly means at the satellite overpass time

Dampened sensitivity of GEOS-Chem AOD to yields of isoprene SOA

AOD-HCHO relationship – SEAC^{4}RS

Boundary-layer (< 2 km) OA mass and HCHO mixing ratios over the SEAC^{4}RS domain



Observations are AMS OA (PI Jimenez) and CAMS HCHO (PI Fried).

Individual points are daily means on the GEOS-Chem 2º × 2.5º grid.

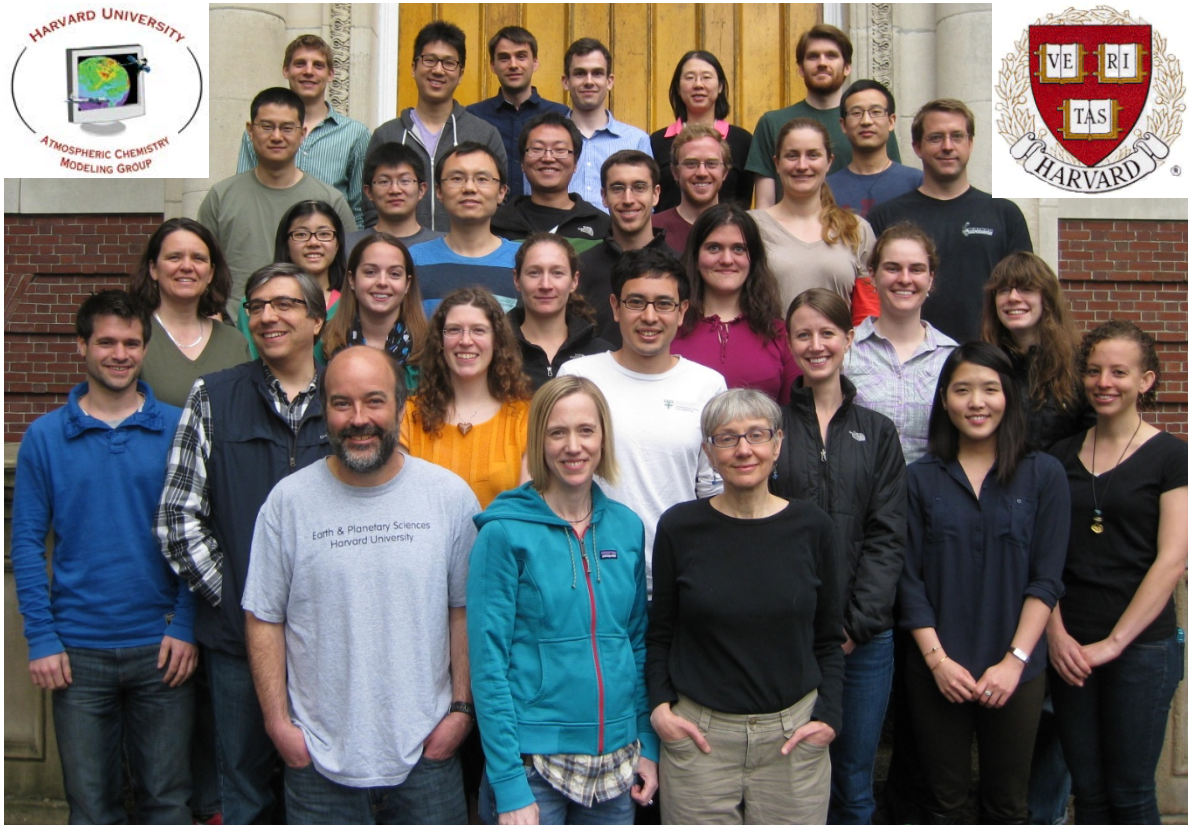
SEAC4RS OA-HCHO slope suggests a yield of ~3%

Concluding Remarks

- Africa is a mixed oxidation regime, while in the southeast isoprene reacts predominantly by NO oxidation.
- Yields in current SOA parameterizations are lower than the chamber yields used in the simulation.
- SOA formation is favored where pre-existing OA mass loading is high
- Current SOA parameterizations in CTMs underestimate OA in Africa and the southeast US.
- There is a strong linear relationship between AOD and HCHO to estimate yields of isoprene SOA, but GEOS-Chem AOD-HCHO is only weakly sensitive to the underlying yields.
- Over the southeast US SEAC4RS sampling domain yields of isoprene OA are ~3.0%.

Acknowledgements

Jacob/Mickley research group



Funders/Fellowship Organizations

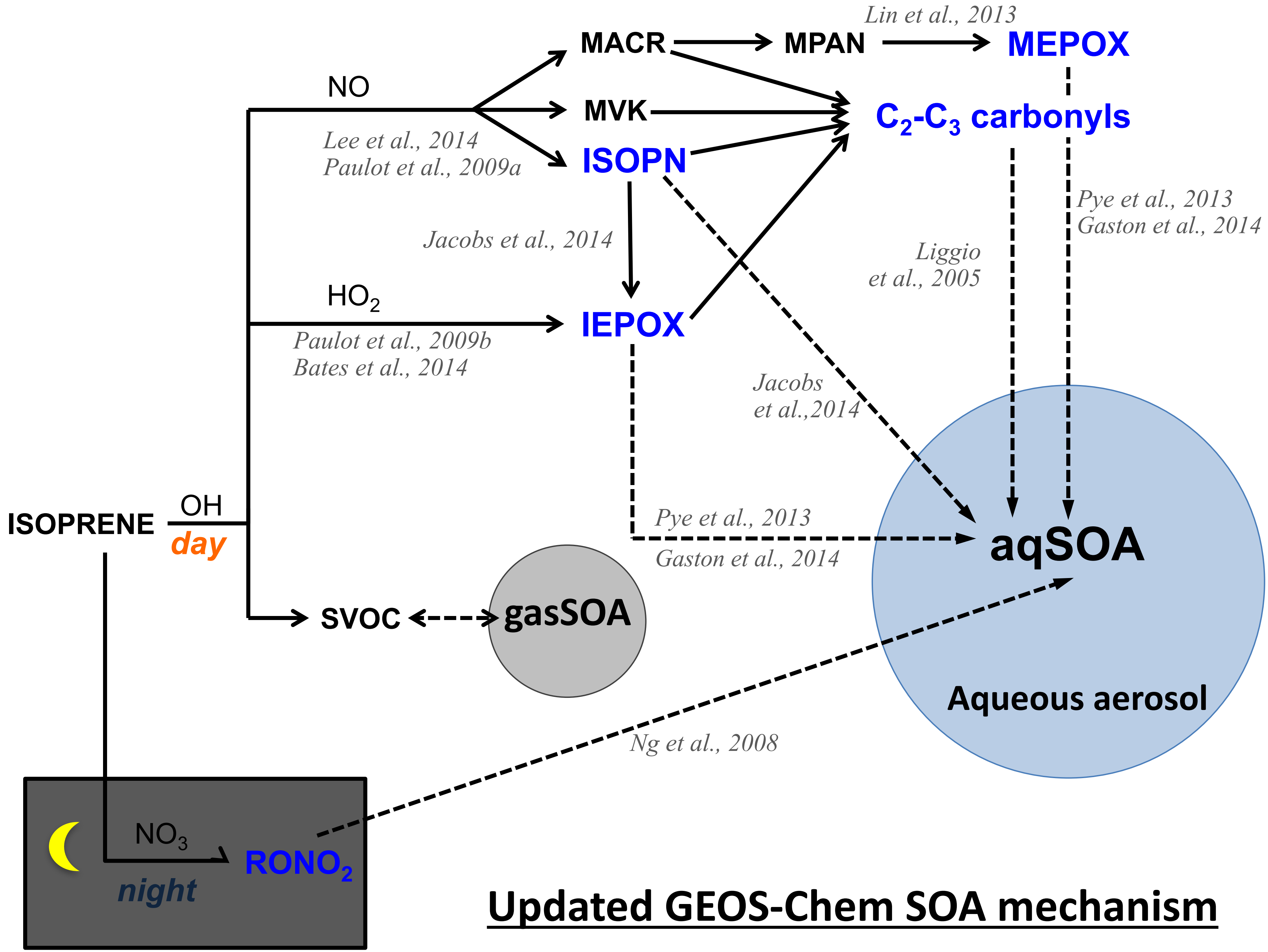
NASA

NRF National Research Foundation

FULBRIGHT

Schlumberger Foundation Faculty for the Future

SUPPLEMENTARY SLIDES



MACR
MPAN
Lin et al., 2013
MEPOX
NO
MVK
C2-C3 carbonyls
Lee et al., 2014
Paulot et al., 2009a
ISOPN
Jacobs et al., 2014
Pye et al., 2013
Gaston et al., 2014
Liggio
et al., 2005
HO2
IEPOX
Paulot et al., 2009b
Bates et al., 2014
Jacobs
et al.,2014
ISOPRENE
OH
day
Pye et al., 2013
Gaston et al., 2014
aqSOA
SVOC
gasSOA
Aqueous aerosol
Ng et al., 2008
NO3
RONO2
night
Updated GEOS-Chem SOA mechanism

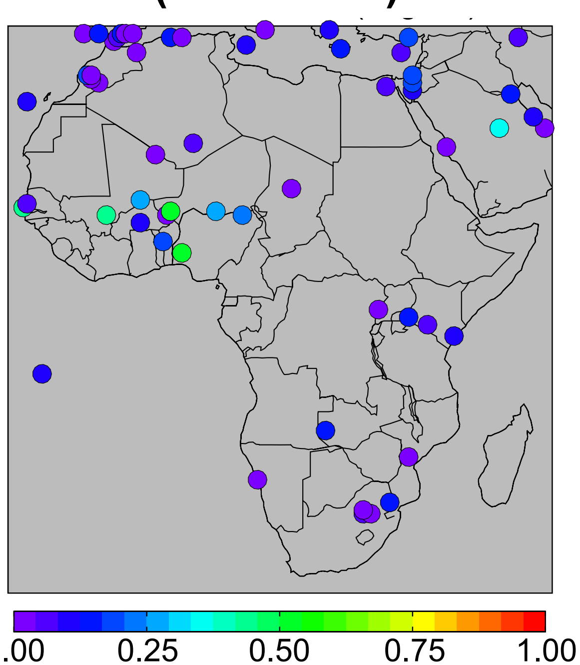
Satellites Products Used in this Work

Species	Instrument	Coverage	Overpass Time	Product	Reference
HCHO	OMI	Daily	13:30 LT	CfA retrieval	Abad et al. (2014)
AOD	MODIS (Aqua)	Daily	13:30 LT	Dark Target C005	Levy et al. (2010)

Evaluate satellite AOD in Africa with AERONET data

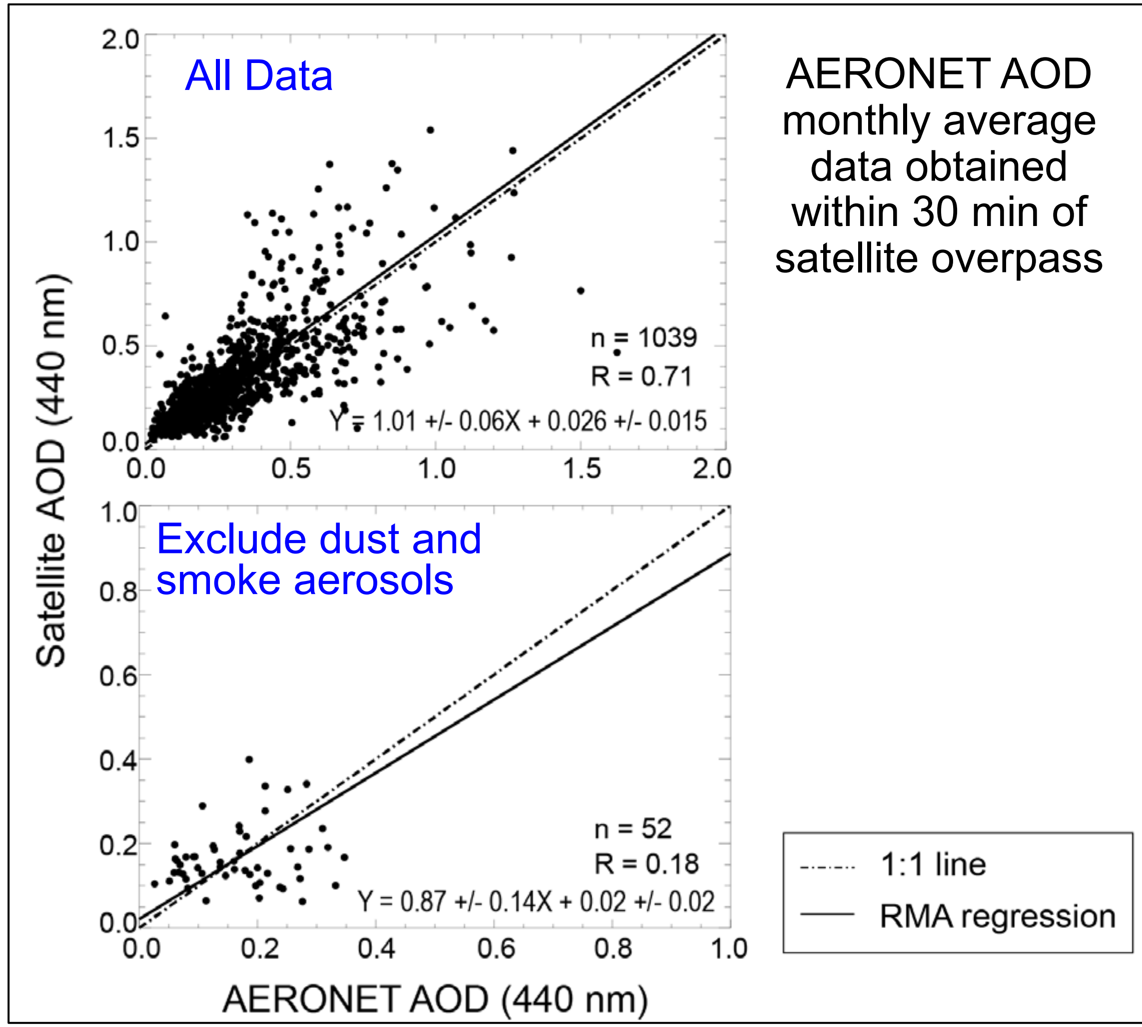
AERONET AOD
represent the ground truth
(uncertainty of 0.01-0.021)
[Holben et al., 1998;
Eck et al., 1999]

AERONET AOD (440 nm) (2004-2013)



Relatively good coverage, but nothing in the tropics

MODIS (Aqua) AOD versus AERONET AOD

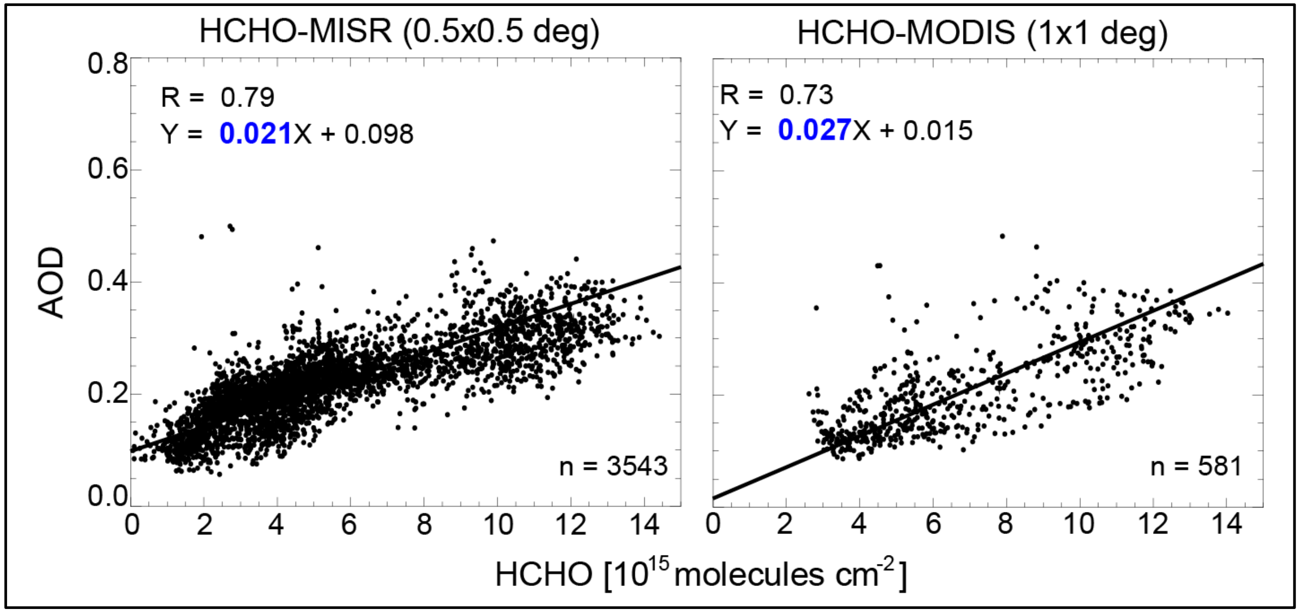


Few observations remain after filtering for dust and smoke for a rigorous evaluation of satellite AOD.

Use SEUS to Develop Method for Inferring Top-down Yields

STEP ONE: Evaluate relationship between biogenic AOD and biogenic HCHO

Satellite AOD and OMI HCHO over the US in summer (JJA)

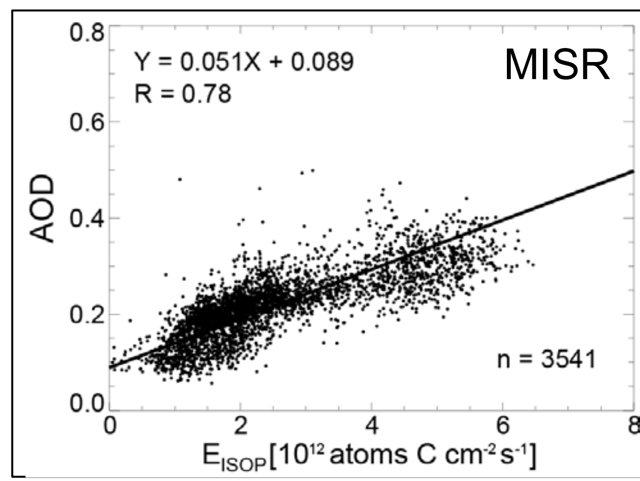


Higher slopes for **Africa** of **0.07** (lower HCHO)

Same as figure above, but with top-down E_{ISOP}

Regression slope is the sensitivity of AOD to biogenic HCHO (or isoprene)

Convert OMI HCHO to isoprene emissions indicates an increase of **0.05 in AOD** with every **1×10^{12} atoms C cm^{-2} s^{-1}** isoprene



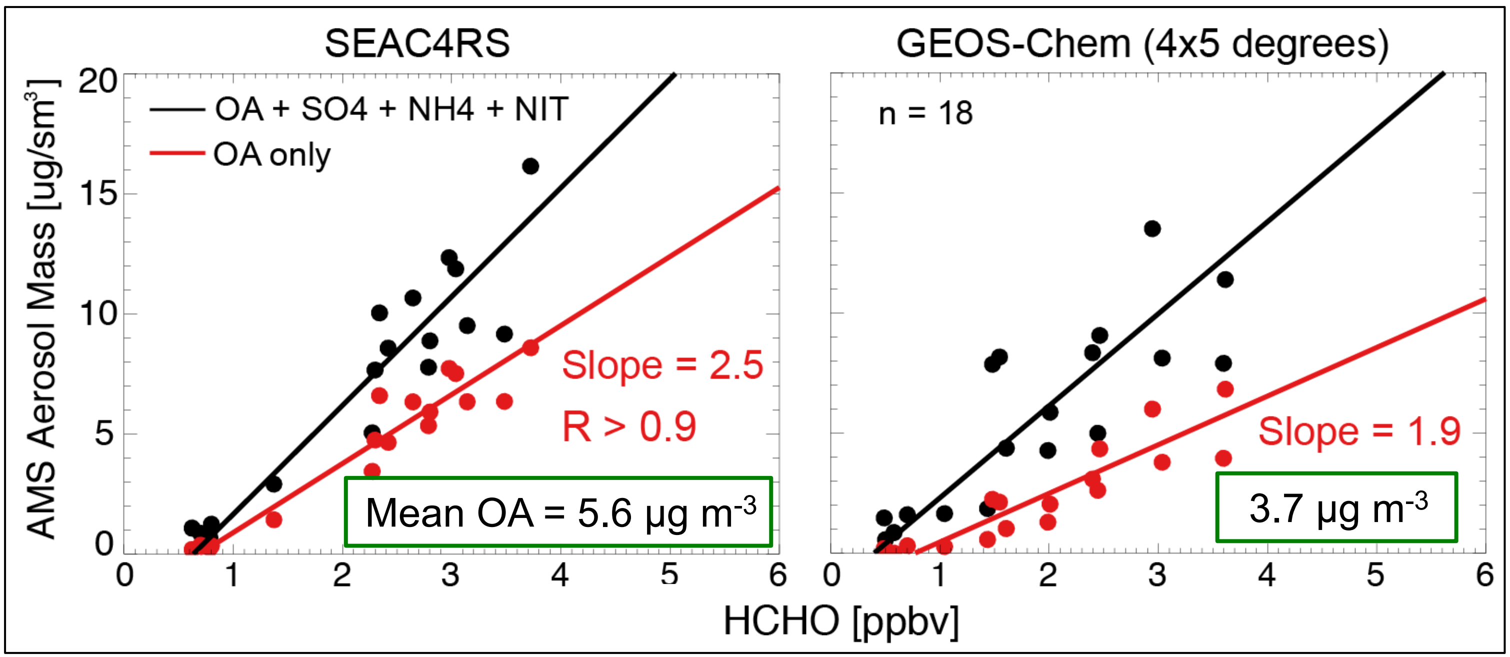
Boundary-layer HCHO and OA Variability in the SEUS

Measurements over the SEUS (Aug-Sep 2013)

HCHO from LIF (PI: Hanisco) and **aerosol mass loading** from AMS (PI: Jimenez)

GEOS-Chem global model at 4 × 5°

Boundary-layer (< 2 km) relation between OA and HCHO



SEAC4RS data are regridded to the GEOS-Chem 4 × 5° grid

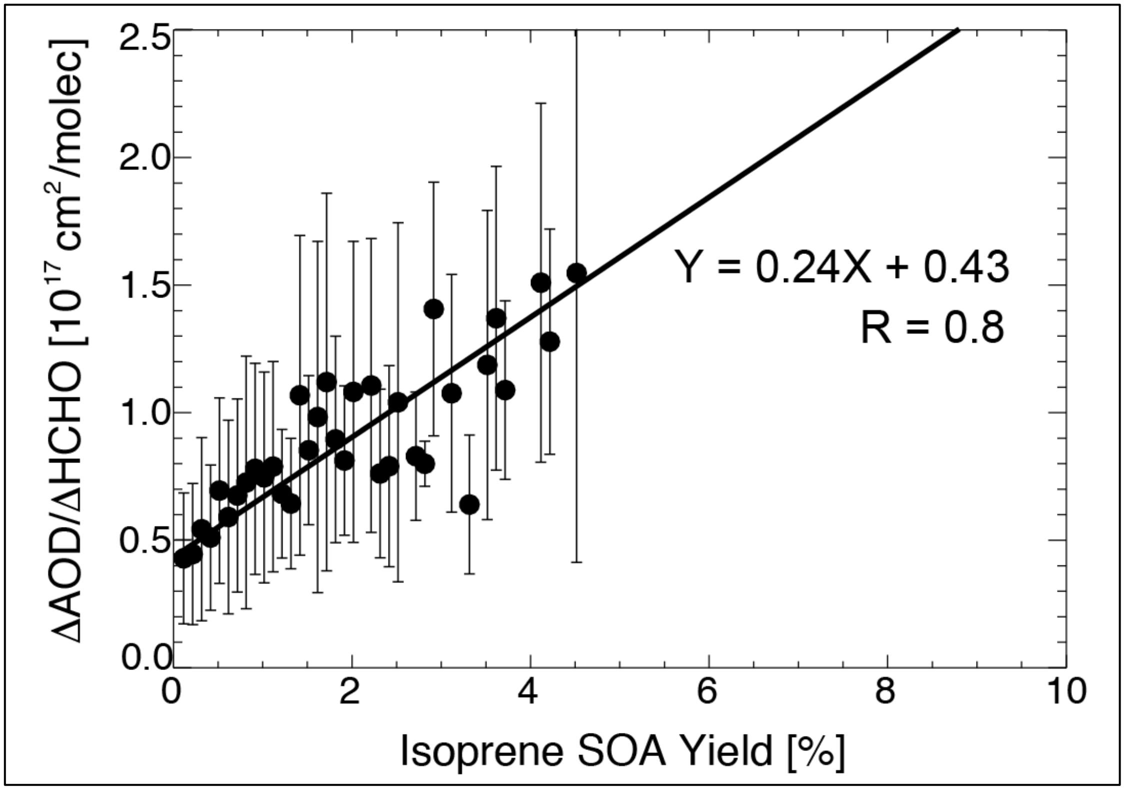
➡ GEOS-Chem underestimates organic aerosol (OA) amount and yield over SEUS.

Convert (ΔAOD/ΔHCHO) to an isoprene SOA Yield

Satellite **ΔAOD/ΔHCHO** = **2.1×10^{-17}** $molec^{-1}\ cm^2$ (MISR)

2.7×10^{-17} $molec^{-1}\ cm^2$ (MODIS Aqua)

GEOS-Chem relation between ΔAOD/ΔHCHO and Y_{SOA}



Points are GEOS-Chem 24-hour average 4 × 5° gridsquares over the US.

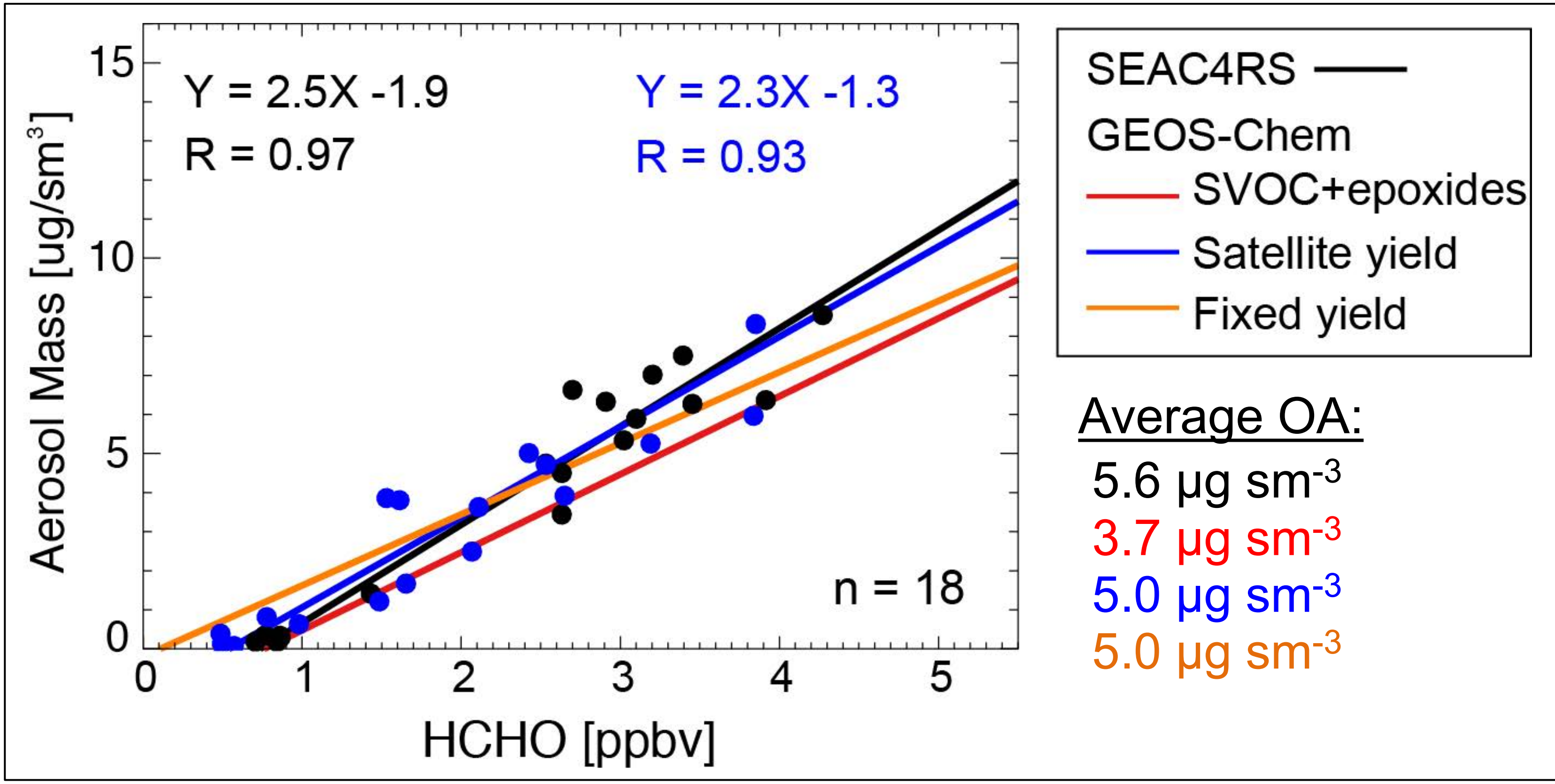
Δ is the difference between a standard GEOS-Chem run and a simulation with **double MEGAN isoprene emissions.**

Satellite **Y_{SOA}** = **7** % (MISR) (*7 kg SOA per 100 kg of isoprene*)

10 % (MODIS Aqua)

Test satellite-derived SOA during SEAC4RS

Boundary-layer (< 2 km) relation between OA and HCHO



Slope obtained with MISR yields (7%) is lower (2.3) and MODIS yields (10%) is higher (2.7) than SEAC4RS (2.5)

Improved sensitivity of OA to isoprene emissions (HCHO) over the SE US