Comparison of Organic Aerosol (OA) treatments in CESM: Difference in source contribution and lifetimes

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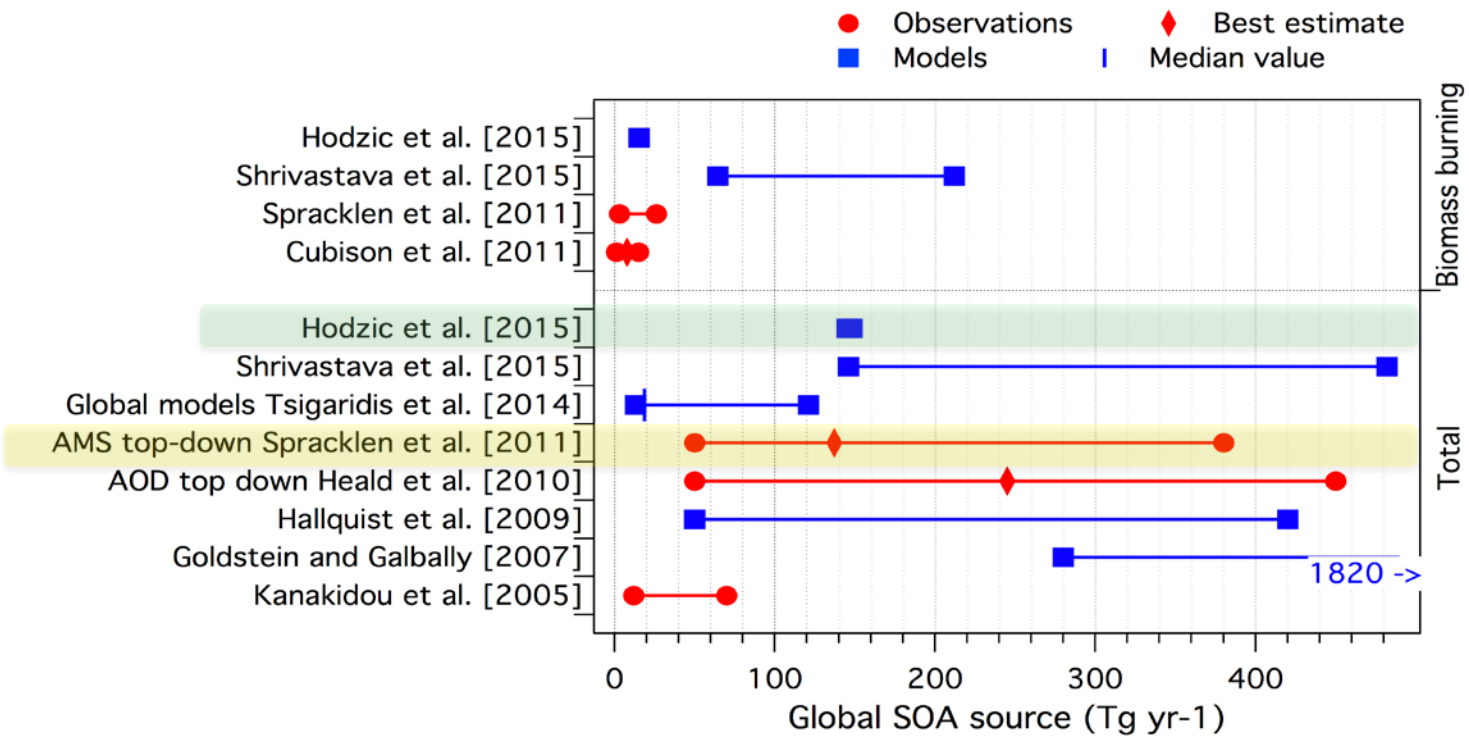
GOALS:

- *Describe the new SOA treatment in CAM5*
- *Discuss the way forward for climate research and for new CMIP runs*

CESM meeting, Boulder, Feb. 2016

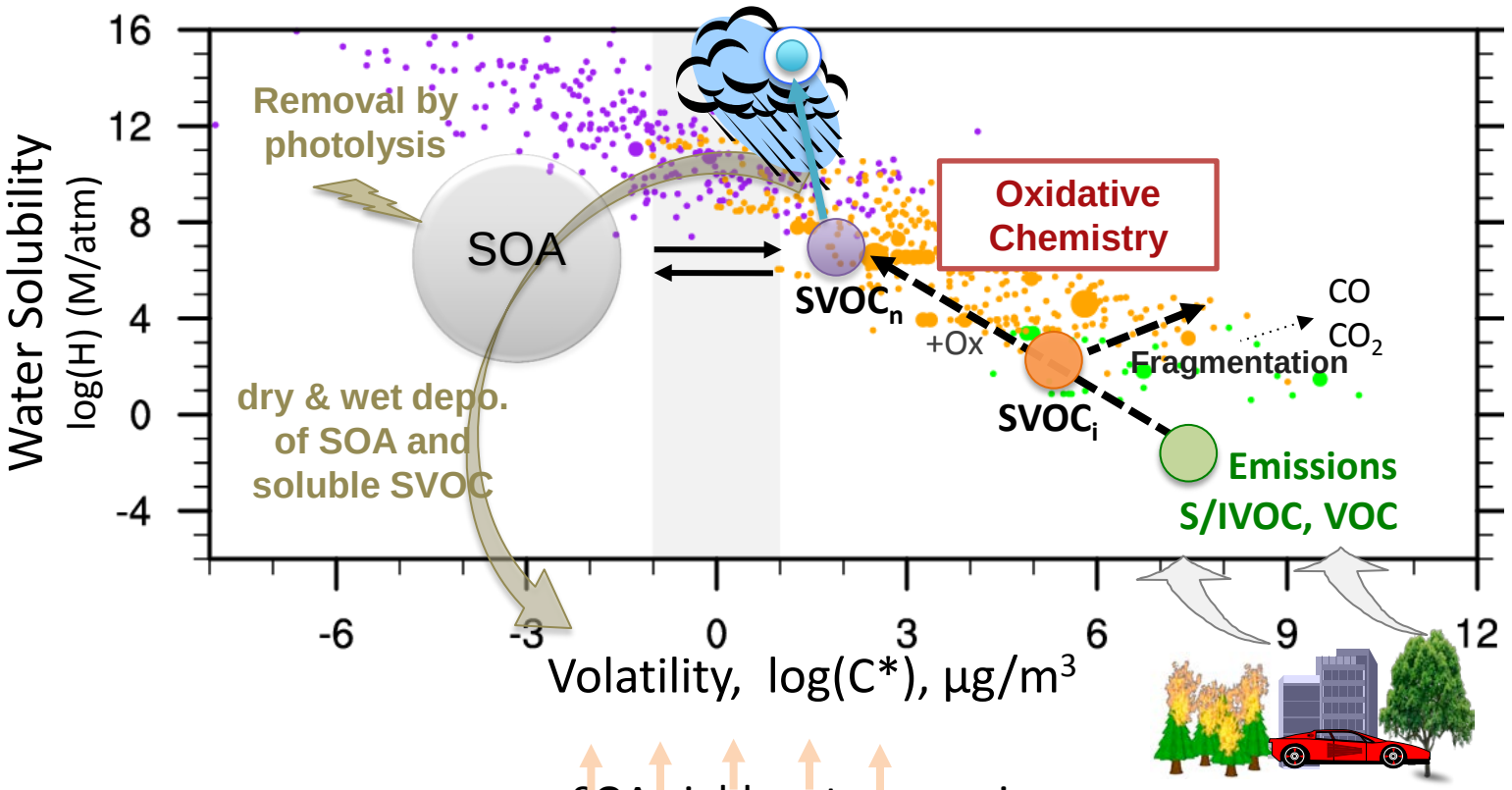
Motivation: Global SOA source is highly uncertain

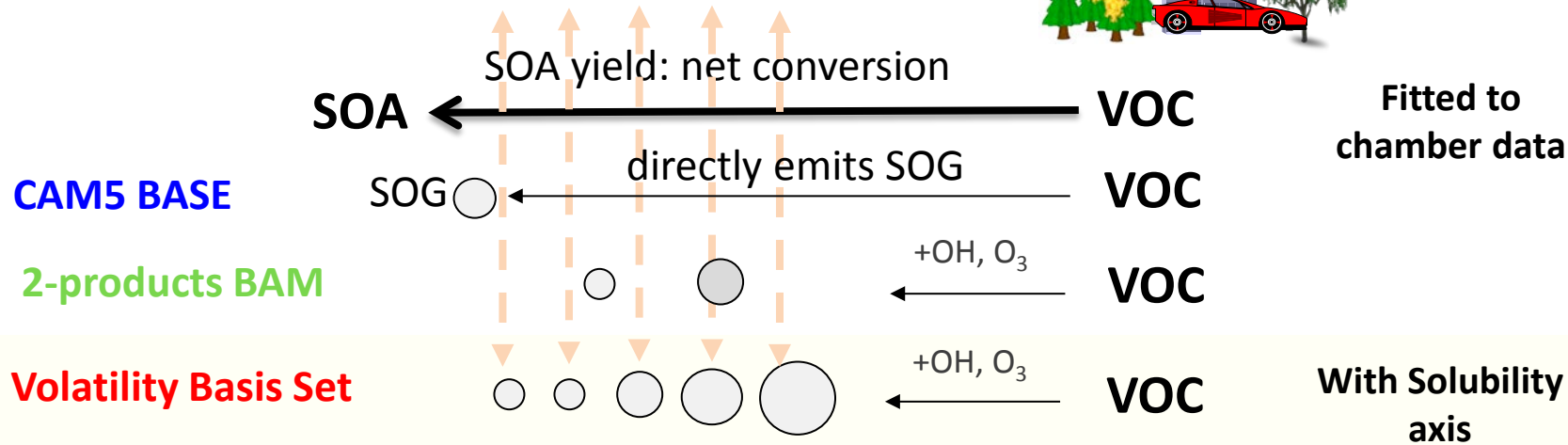


(Hodzic et al. 2015, ACPD)

- Studies constrained by observations point to a source of ~150 Tg/yr
- Current parameterizations mostly fail in predicting those values
- Revised production and removals

Simplistic ways of treating the complex SOA lifecycle

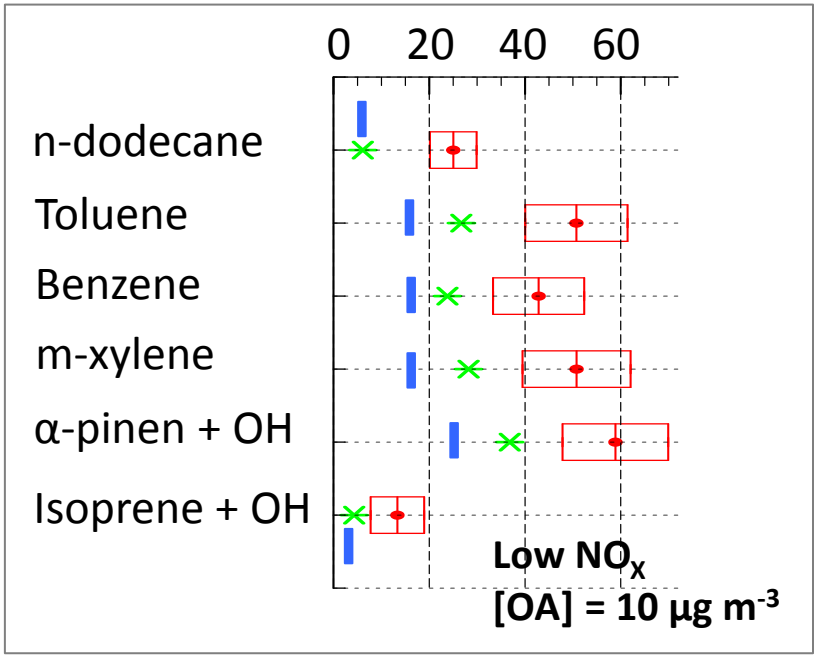




Shortcomings of the *CAM5 BASE* SOA (Liu et al. 2012)

On the production side:

- Does not use oxidants to produce SOG => inaccurate diurnal cycle of SOA, Nb.
- Does not reflect updates in SOA production yields: wall corrected yields indicate a much stronger production



CAM5 BASE: Empirical SOA Yields, sometimes increased by 50%

2PROD BAM: Historic SOA mass Yields with wall loses (Heald et al. 2008)

CAM5 VBS: Wall corrected SOA Yields (Zhang et al. 2014)

- Does not account for S/IVOCs emissions ($C_{>9}$) which are missing in inventories
- SOA production in the aqueous phase is not included

Shortcomings of the *CAM5 BASE* SOA (Liu et al. 2012)

Evidence for extra removal besides dry and wet deposition of SOA:

- SOA particles can undergo *heterogeneous reactions* with OH, O3
- SOA particles are sensitive to *UV light* during atmospheric aging:
  - 50% decrease of the condensed phase α-pinene ozonolysis SOA in 1 week of aging (Epstein et al. 2014)
  - Lifetime of ~10h of lab. α-pinene ozonolysis SOA under UV lamps (Henry and Donahue, 2012; Wong et al. 2014)
  - Heald et al. 2012: when SOA production is increased to match PBL concentrations, SOA levels in the upper troposphere are largely overpredicted

- Effective SOA photolysis rate (Hodzic et al. 2015, ACP)
  - Ambient aerosols with MAC=0.1 m^2/g and QY=1% (1Carbon) JSOA = 0.04% JNO2
  - 40–60% decrease in SOA yields over 10 days of aging at mid-latitudes in summer

2 new model configurations for SOA production and removal

- **VBS-NY CAM5 run (Hodzic et al. 2015, ACPD):**
  updated SOA algorithm uses volatility basis set (VBS)
  - VBS based on the Statistical Oxidation Model fitting of the wall-corrected SOA yields (Zhang et al. 2014).
  - Addition of semi-volatile and intermediate volatility precursors (S/IVOC) from anth. and biomass burning sources (Jathar et al. 2014): $E_{SVOC} = 0.6\ E_{POA}$ and $E_{IVOC} = 0.2\ E_{NMVOC}$
  - S/IVOC yields from explicit chemistry (GECKO-A)

- **VBS CAM5 run:**
  - Volatility dependent Henry's law coeffs
  - Photolytic SOA loss: $J_{[SOA]} = 0.04\%\ J_{[NO2]}$ (Hodzic et al. 2015, ACP)

30 particle species: 5 volatility bins, 2 modes (Aitken and accumulation) and 3 classes including fossil fuel, biogenic and biomass burning. We also have 15 gas species.

Model configuration

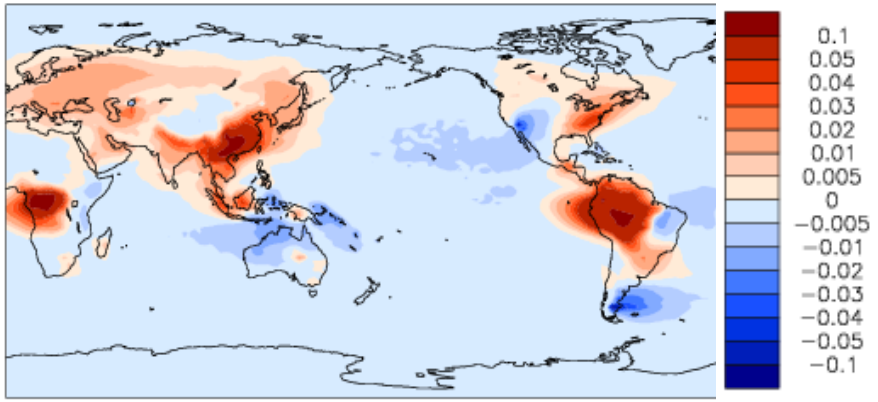
- CAM5.4 (1.9 x 2.5) specified dynamics version, using GEOS5 meteorological analysis (2006-2009).
- MOZART gas-phase chemistry extended to include a better speciation of biogenic and anthropogenic SOA precursors
- SOA precursors emissions:
  - Biogenic MEGAN (Isop, apin, bpin, limon, myrc, sesq)
  - Anthropogenic RCP8.5 (benzene, toluene, xylene and alkanes)
  - Fire from FINN (benzene, toluene, xylene and alkanes)
  - Additional semi-volatile and intermediate volatility organics (S/IVOC) from anthropogenic and biomass burning emissions
- SOA species interact with clouds and the optical calculations

Results: Annual average SOA column burden

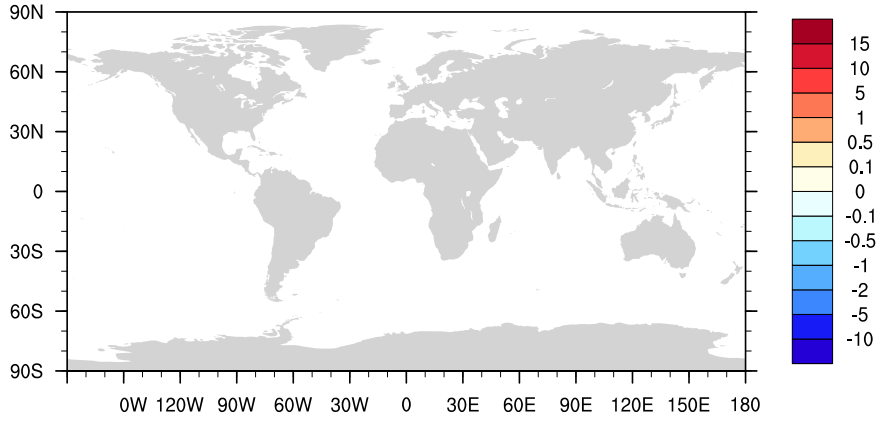
BASE SOA burden=0.66 Tg

VBS SOA burden=0.78 Tg

AOD 550nm: VBS - BASE



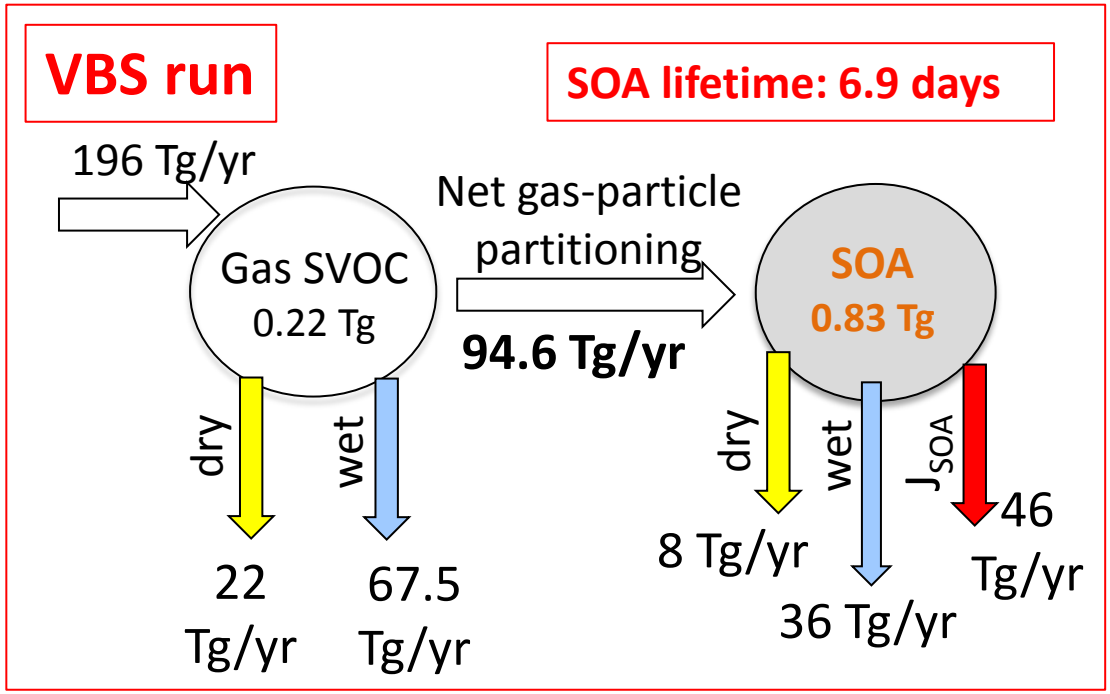
Difference: VBS - BASE

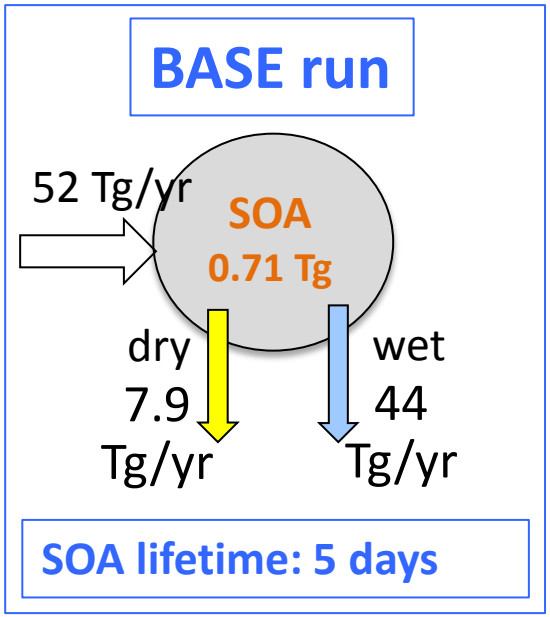


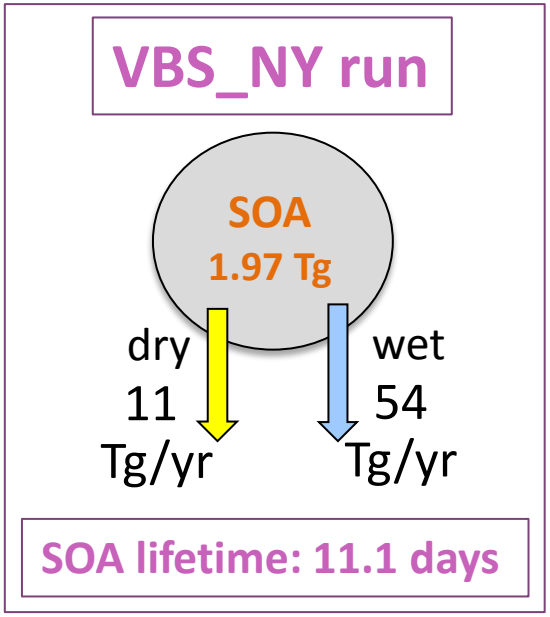
VBS vs. BASE:

- Increased SOA burden in source regions (China, SE U.S., Europe, fire regions)
- Decreased SOA burden in the outflow regions

Results: Annual SOA budgets





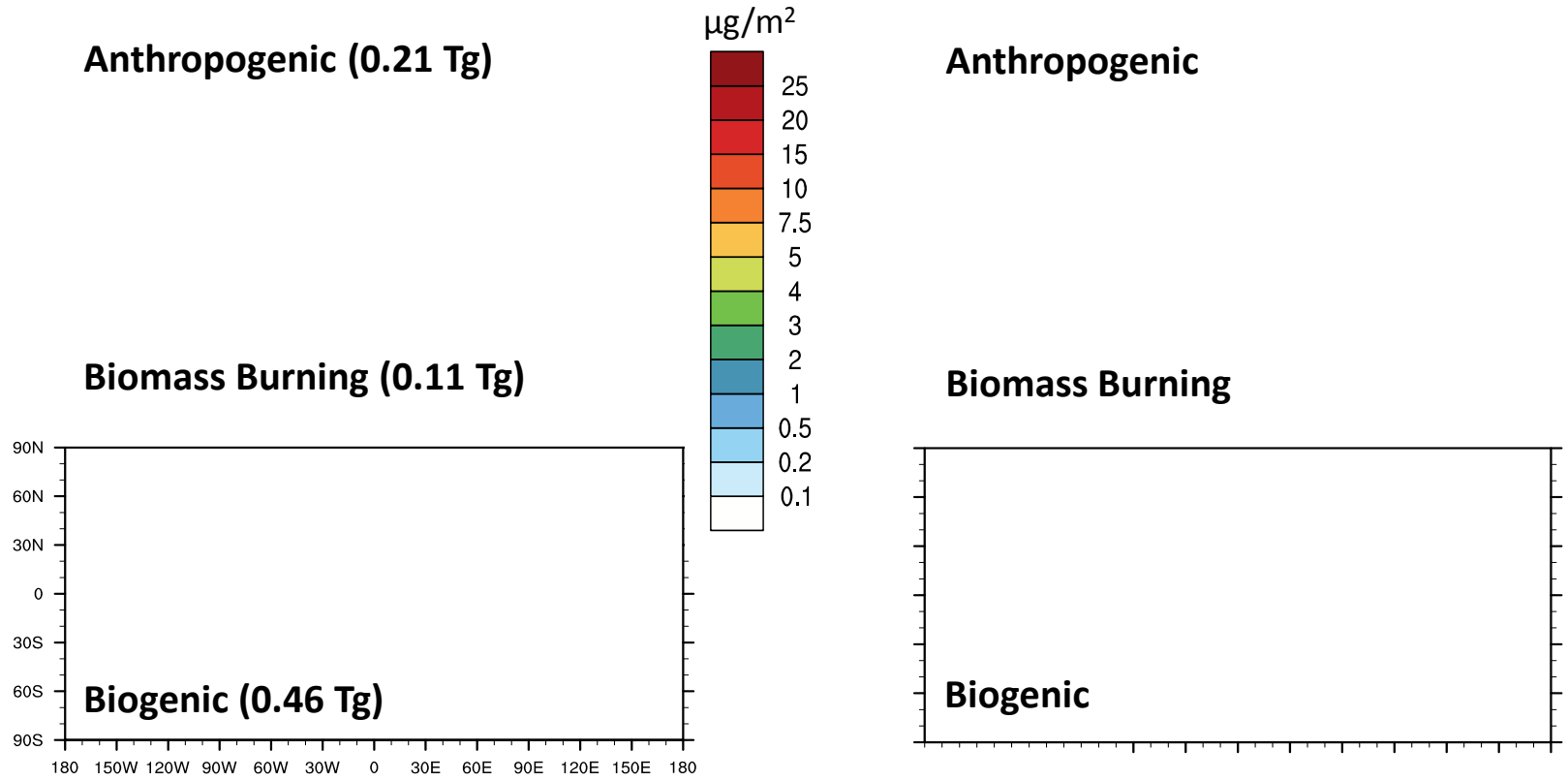


VBS vs. BASE:

- Higher production and higher removals – a more dynamic SOA system
- Updates in the yields alone leads to very high burden

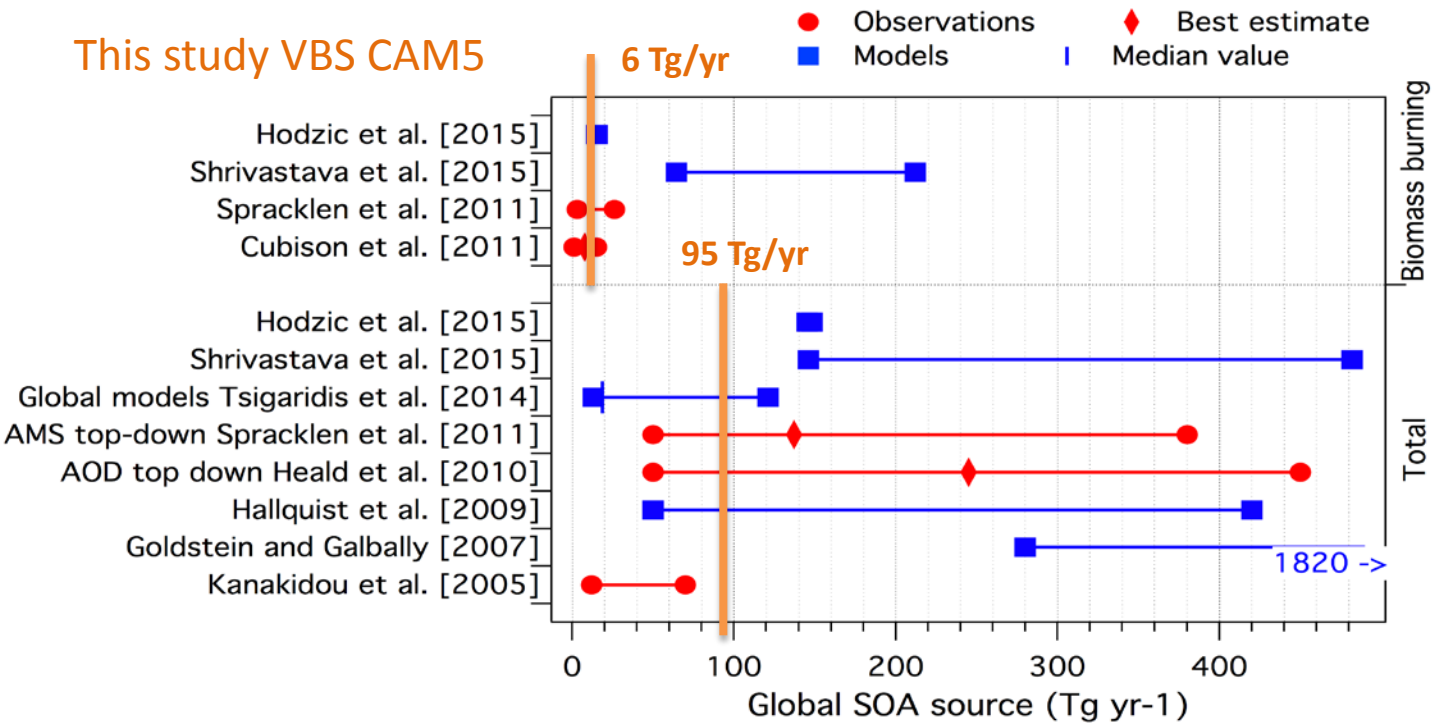
VBS run: source contribution to SOA

% contribution



- Biogenic emissions are the largest source of SOA globally (although treatment of isoprene is work in progress)
- Anthropogenic sources dominate over China and India

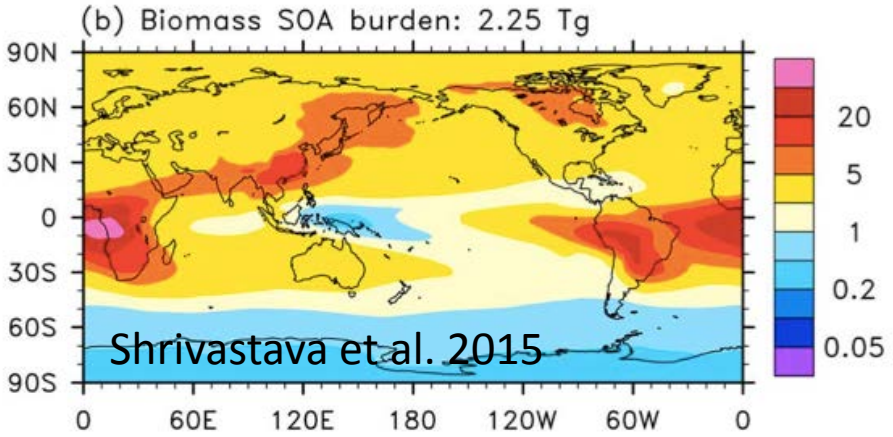
Results: Comparison to previous studies



- Large difference in biomass burning

(a) Biomass SOA burden: 0.11 Tg

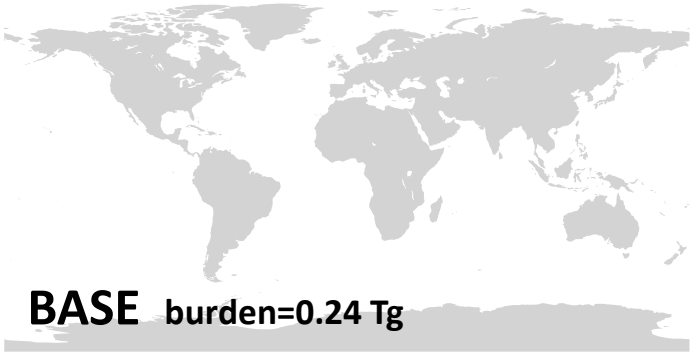
This study VBS CAM5

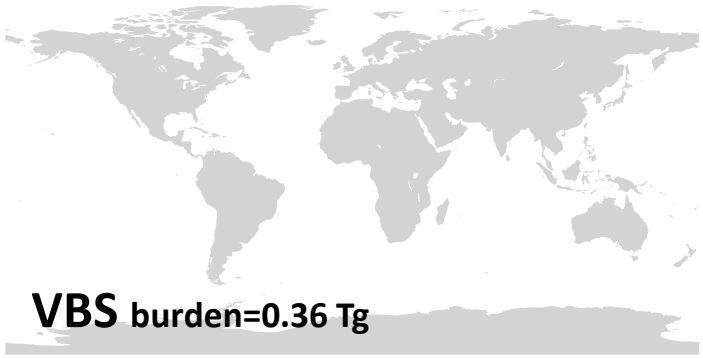


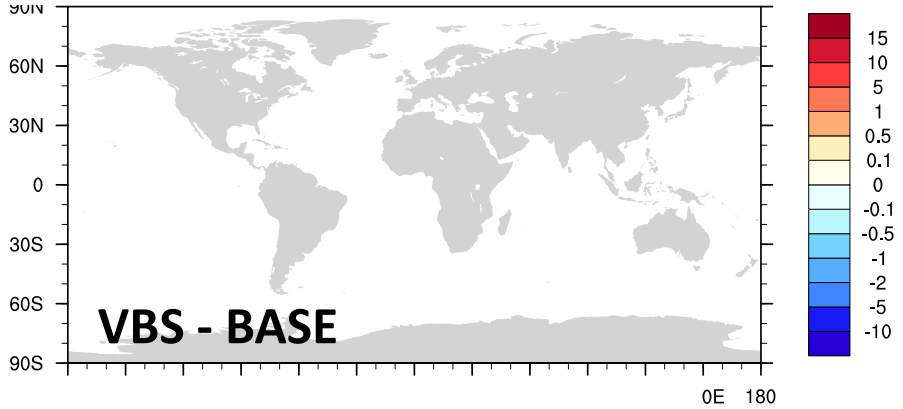
Results: SOA near the surface

- Annual SOA burden (up to 1.5km)

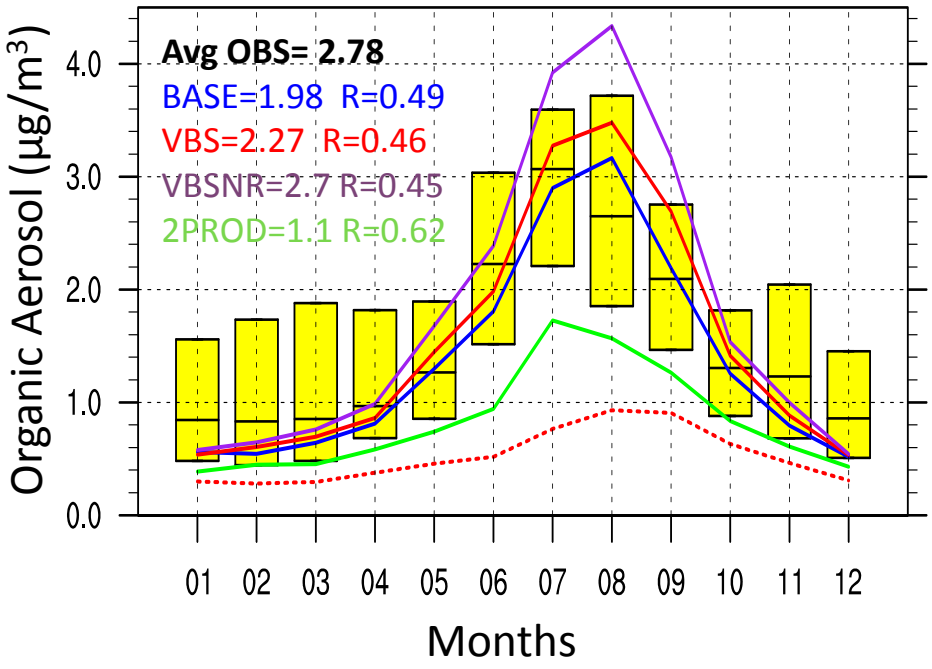
µg/m²







- Comparison with the U.S. IMPROVE data (2005-2008)



Results: Zonal average SOA (µg/kg)

µg/kg

BASE

VBS_NY: updated production only

VBS

2PROD - BASE

VBS NY - BASE

VBS - BASE

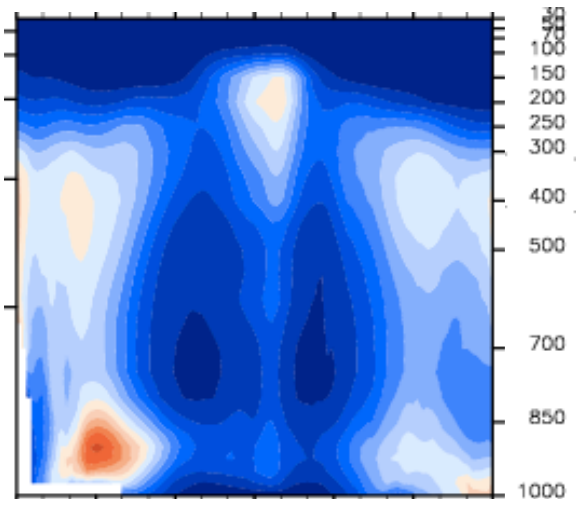
µg/kg

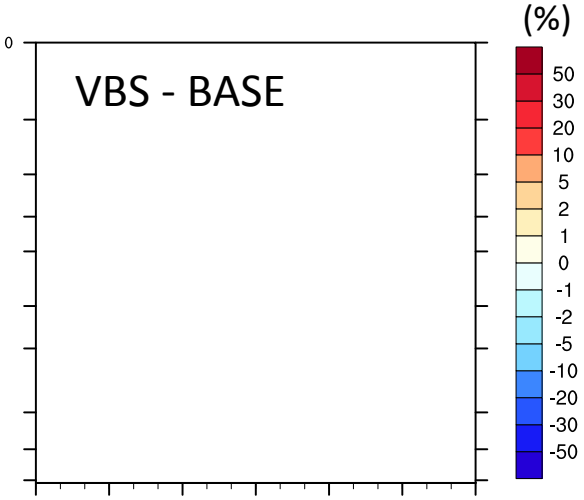
Results: Zonal average Number Concentrations (#/cm^3)

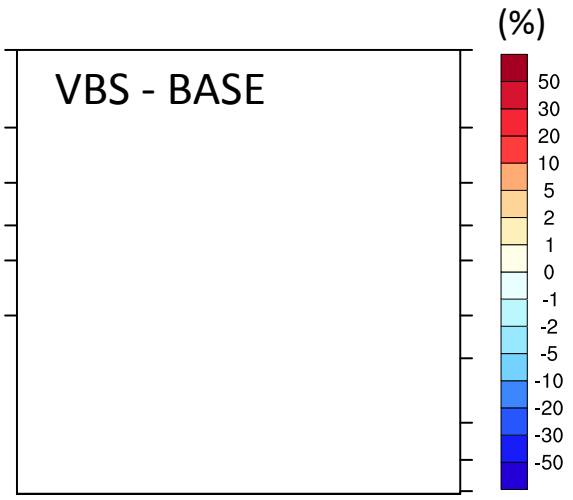
VBS: Accumulation #/cm^3

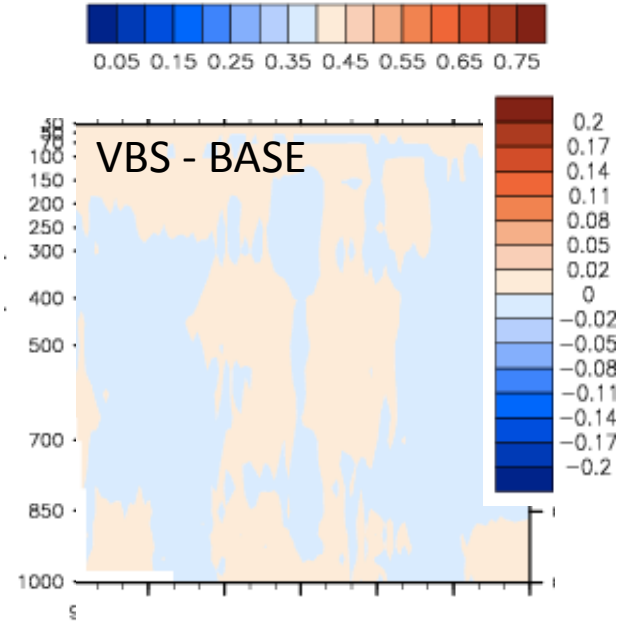
VBS: Aitken Number #/cm^3

VBS: Cloud fraction



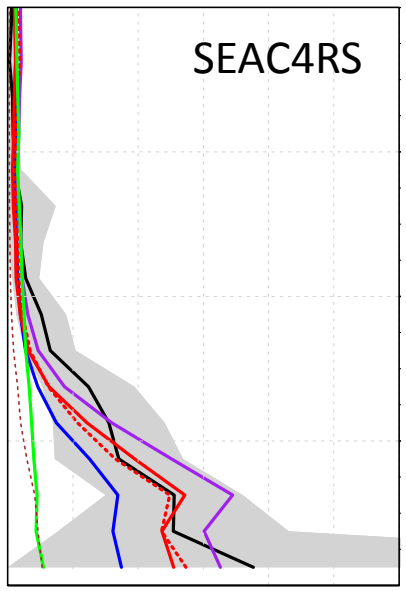


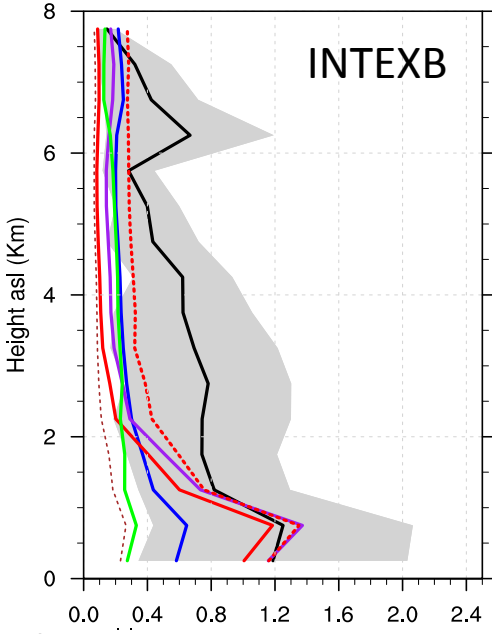


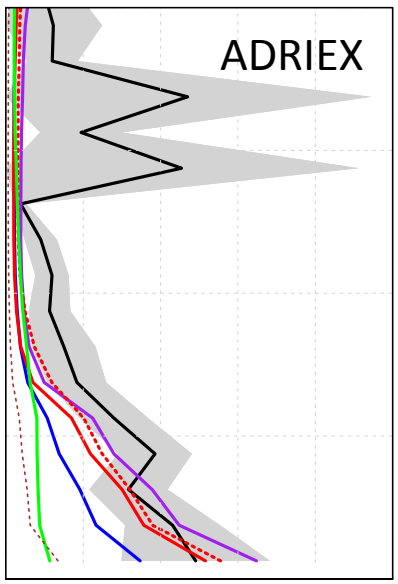


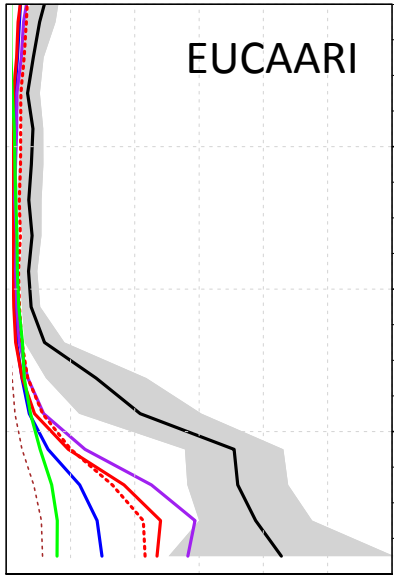
Not sure this is meaningful bc we are using specified dynamics

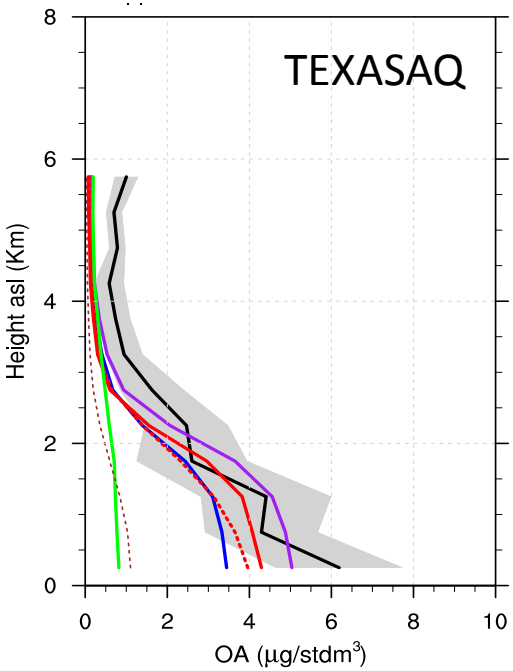
Comparisons with aircraft OA measurements

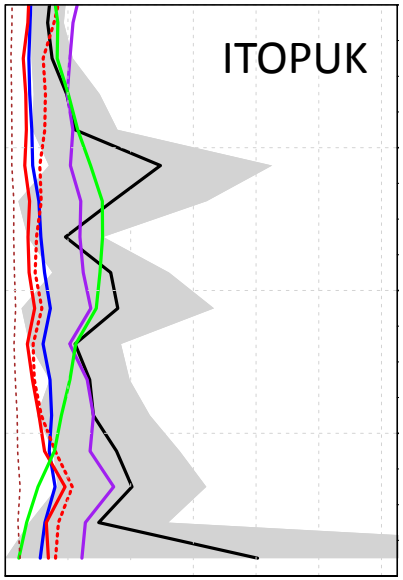












OBS

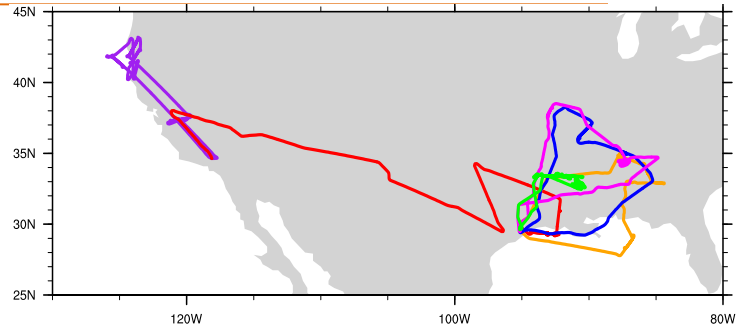
CESM 2x2

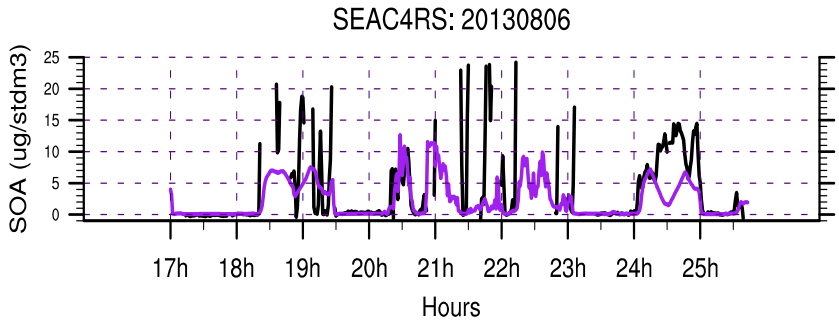
- **BASE**
- **VBS**
- **VBSNY**
- **2PROD**
- **POA**

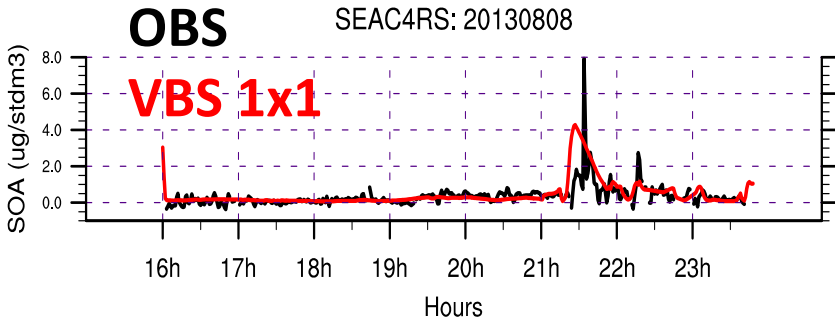
- **VBS 1x1**

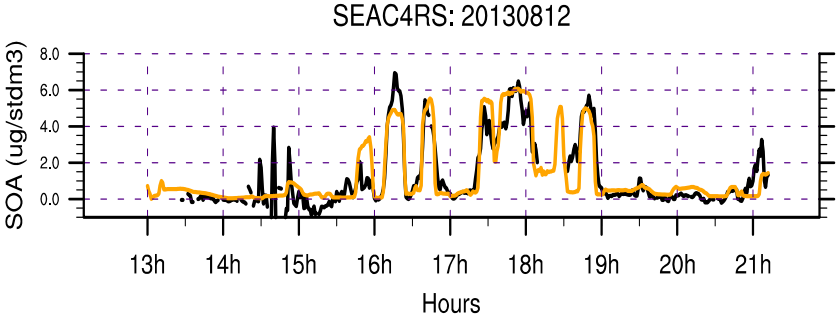
CAM5 VBS 1x1 : Comparisons with SEAC4RS SOA measurements

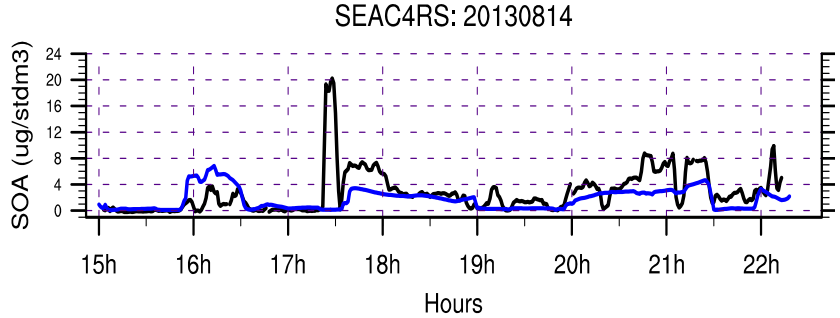
Ongoing work: Model evaluation at high resolution with campaign data

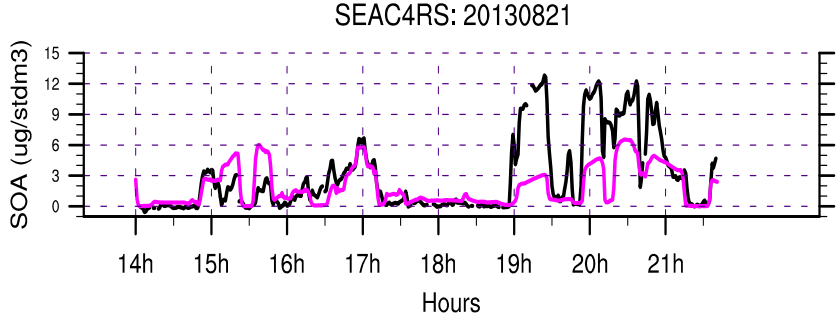


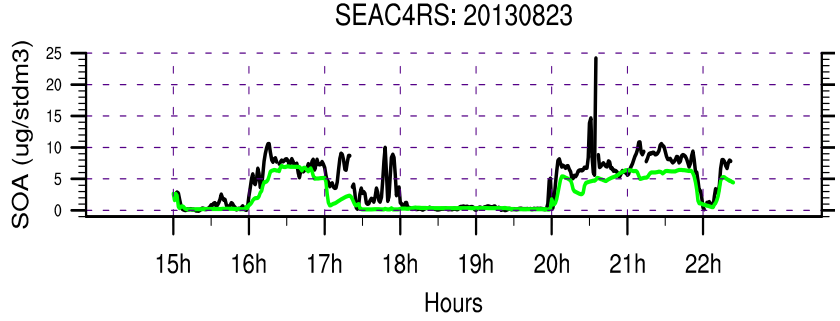










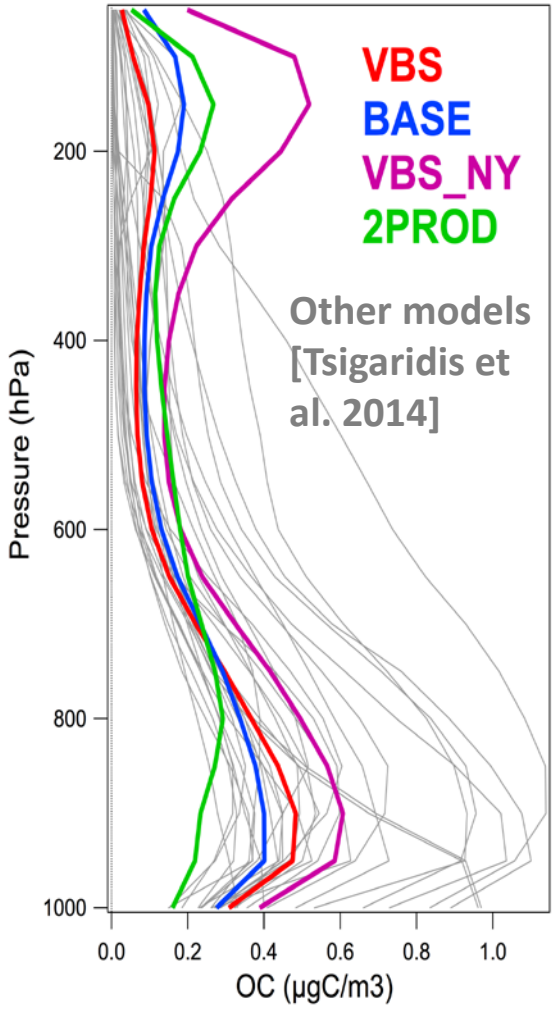


Summary and future work

Added a flexible VBS framework into CAM5 that includes updated yields and photolysis removal rates based on recent lab. data, and explicit chemistry

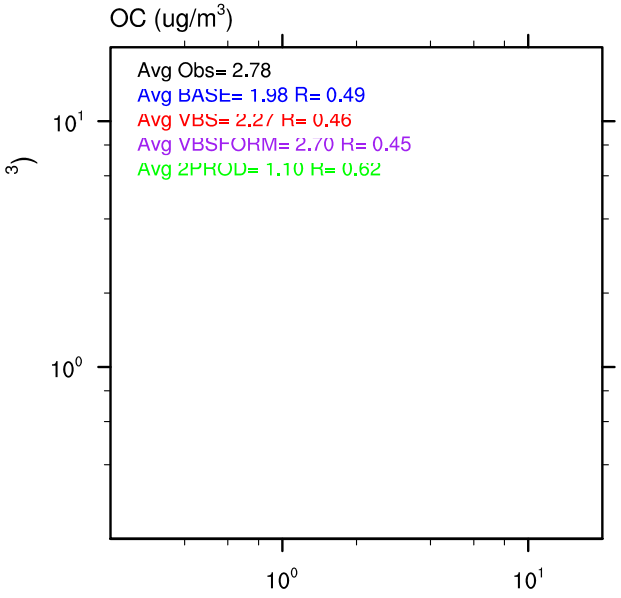
The new SOA approach leads to

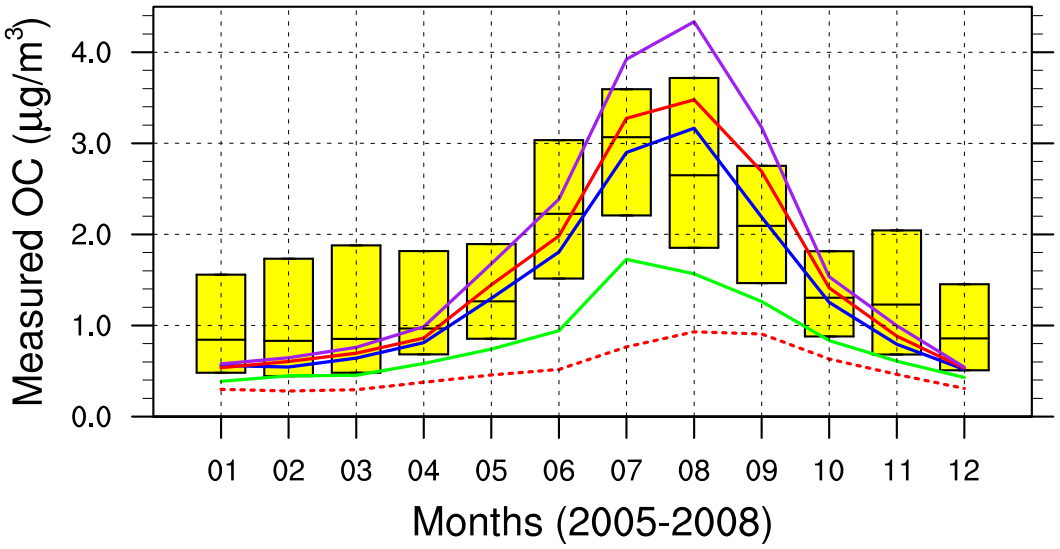
- a more dynamic system (higher production, and higher removals),
- larger concentrations in the near source regions, and lower concentrations in the upper troposphere,
- Dominance of biogenic sources over BB
- Improved agreement with profile data

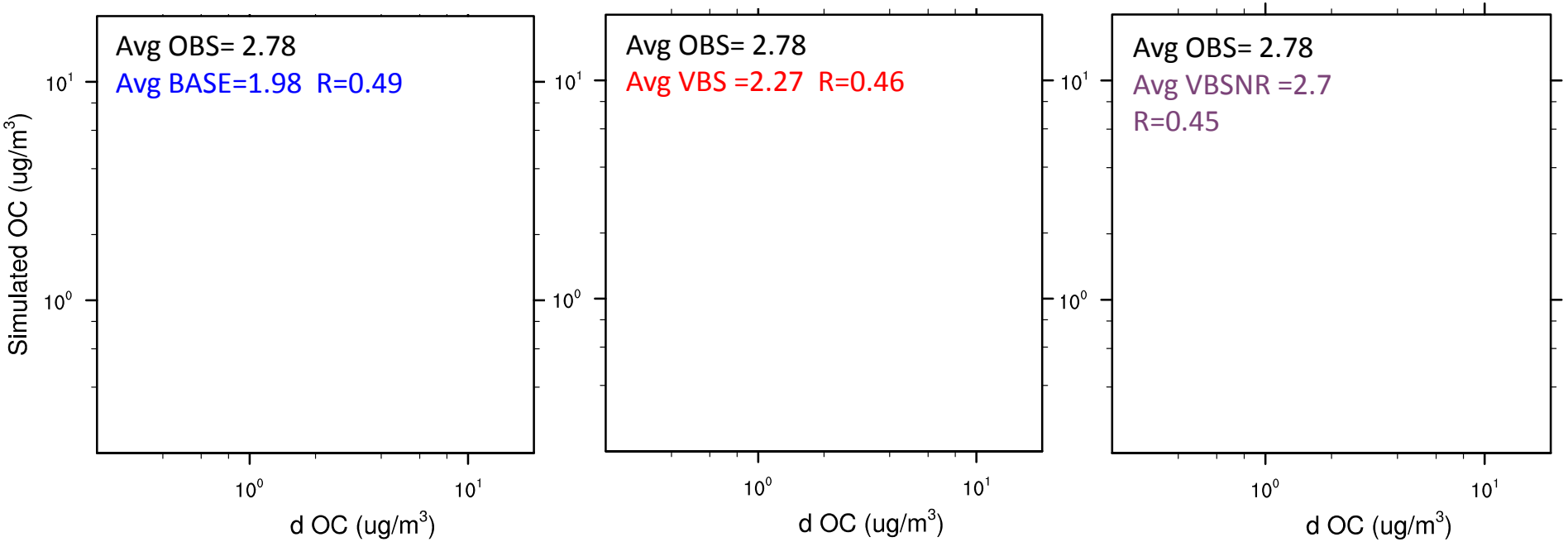


- Examine vertical gradients in OA with upcoming ATOM
- Revise Isoprene chemistry, and cloud chemistry
- Work on reducing the number of species for climate runs to 10 SOA, and 5 gas species.

Model evaluation with the U.S. IMPROVE data (2005-2008)







Results: Annual average SOA column burden

BASE SOA burden=0.66 Tg

VBS_NY SOA burden=1.78 Tg

VBS SOA burden=0.78 Tg

2PROD SOA burden=0.93 Tg

VBS vs. BASE:

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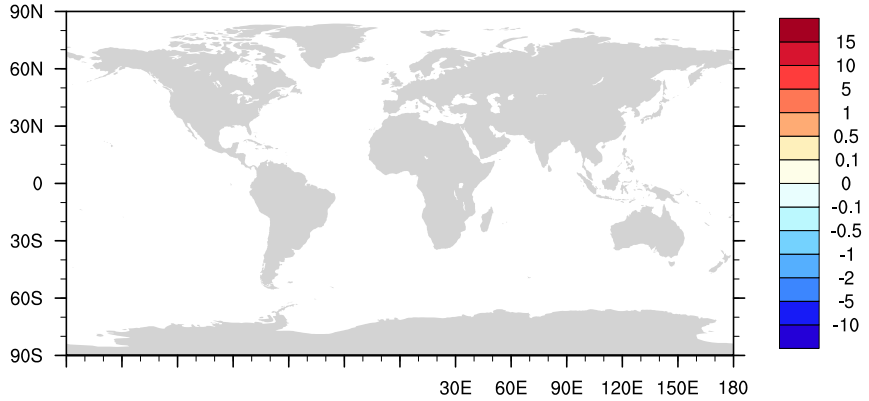
Results: Annual average SOA column burden

Changes due to updated SOA production

Changes due to updated removals

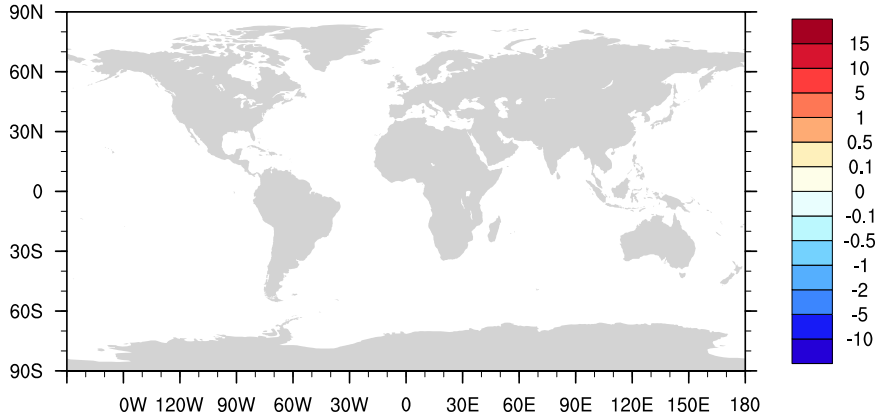
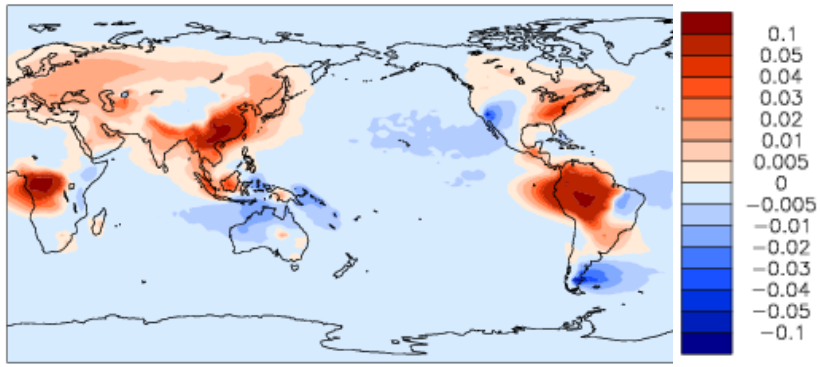
VBS NY - BASE

VBS – VBS NY



The overall difference

VBS - BASE



AOD 550nm: VBS - BASE