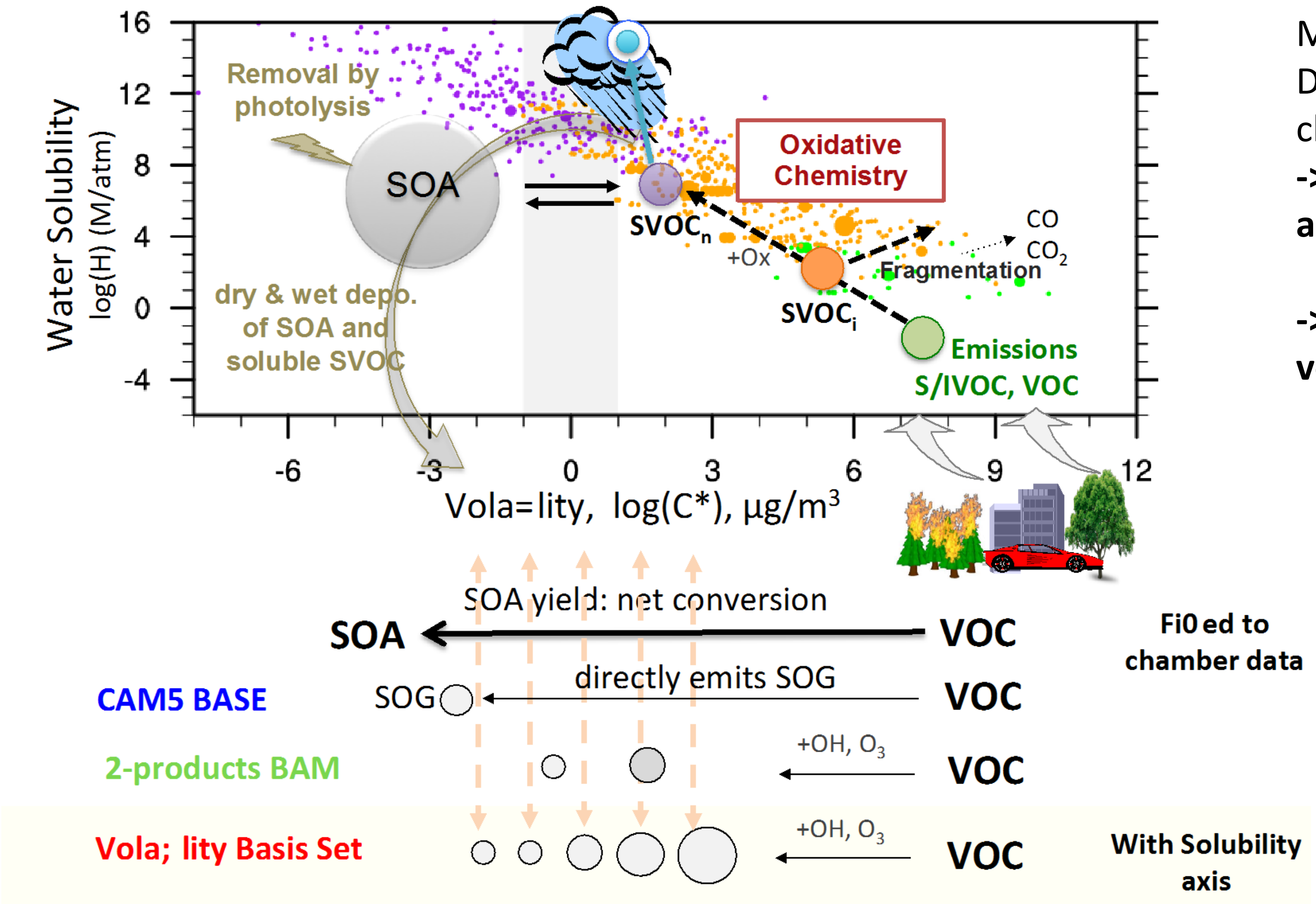
New SOA approach in CESM2 CAM5-Chem and WACCM

Simone Tilmes, Alma Hodzic, , Louisa Emmons
Francis Vitt, Manish Shrivastava, Christoph Knote

New SOA approach in CESM2 CAM5-Chem/ WACCM

Simplis=c ways of trea=ng the complex SOA lifecycle



More physical approach
Direct coupling to biogenic emissions changes from MEGAN
-> couples SOA formation to land use and climate change

-> only works in full chemistry version at this point

New SOA approach in CESM2 CAM5-Chem/ WACCM

CAM5-chem SOA scheme updated to include volatility basic set (VBS)

- 5 SOA bins with different volatility are included (Hodzic et al., 2016 ACP), implementation based on Shrivastava et al., 2014.
- 5 SOAG gas-phase intermediate volatile aerosol components
- 1 semi-volatile (SVOC) = 0.6* POM emissions
- 1 intermediate volatile precursor (IVOC); 0.2 *NMVOC emissions
- SOAG production different for biomass burning, fossil fuel and biogenic emissions -> three different categories are introduced
- 47 new species

WACCM simplified SOA Scheme including VBS

- Reduction in number of SOA variables
- 12 new species
- Significant reduction in computer time

New SOA approach: Production

Production:

- Yields are base on statistical oxidation model fitting of the wall corrected SOA yields (Zhang et al., 2014), partitioning dependent on characteristics of the composition (anthropogenic vs biogenic
- Using GECKO-A chemical mechanism to Identify S/IVOC yields (Hodzic et al., 2016)
- Aerosol uptake of GLYOXAL to SOAG0 (least volatile bin)

fossil fuel / biomass | biogenic (TERP, ISOP) | fossil fuel/ biomass (BENZ, TOL, XYL) | biogenic

Precursor	IVOC	TERP	ISOP	BENZ	TOL	XYL	SESQ
M_w (g mol^{-1})	189	136	68	78	92	106	204
$k_{OH@298\,K}$ (s^{-1})	1.34×10^{-11}	5.3×10^{-11}	10^{-10}	1.22×10^{-12}	5.63×10^{-12}	2.31×10^{-11}	5.3×10^{-11}
Log[C^*]	Mass yields at low NO_c						
< -2	0.315	0.093	0.012	0.007	0.371	0.395	0.270
-1	0.173	0.211	0.013	0.003	0.028	0.041	0.253
0	0.046	0.064	0.001	0.270	0.207	0.203	0.080
1	0.010	0.102	0.100	0.142	0.586	0.121	0.157
2	0.007	0.110	0.078	0.400	0.063	0.232	0.068
3	0.008	0.125	0.097	0.120	0.138	0.145	0.072
	Mass yields at high NOx						

We only consider low NOx at this point!
Only 5 bins considered.

Hodzic et al., 2016

New SOA approach: Removal

Removal:

- Dry and wet removal of gas phase semi volatile oxidized species (SOAG) strongly varies as a function of volatility
- Photolysis frequency of SOA based on the measured absorption of ambient aerosols (equivalent to 0.04% of NO_2 photolysis; biogenic photolysis may be assumed to be smaller (0.004% of NO_2)

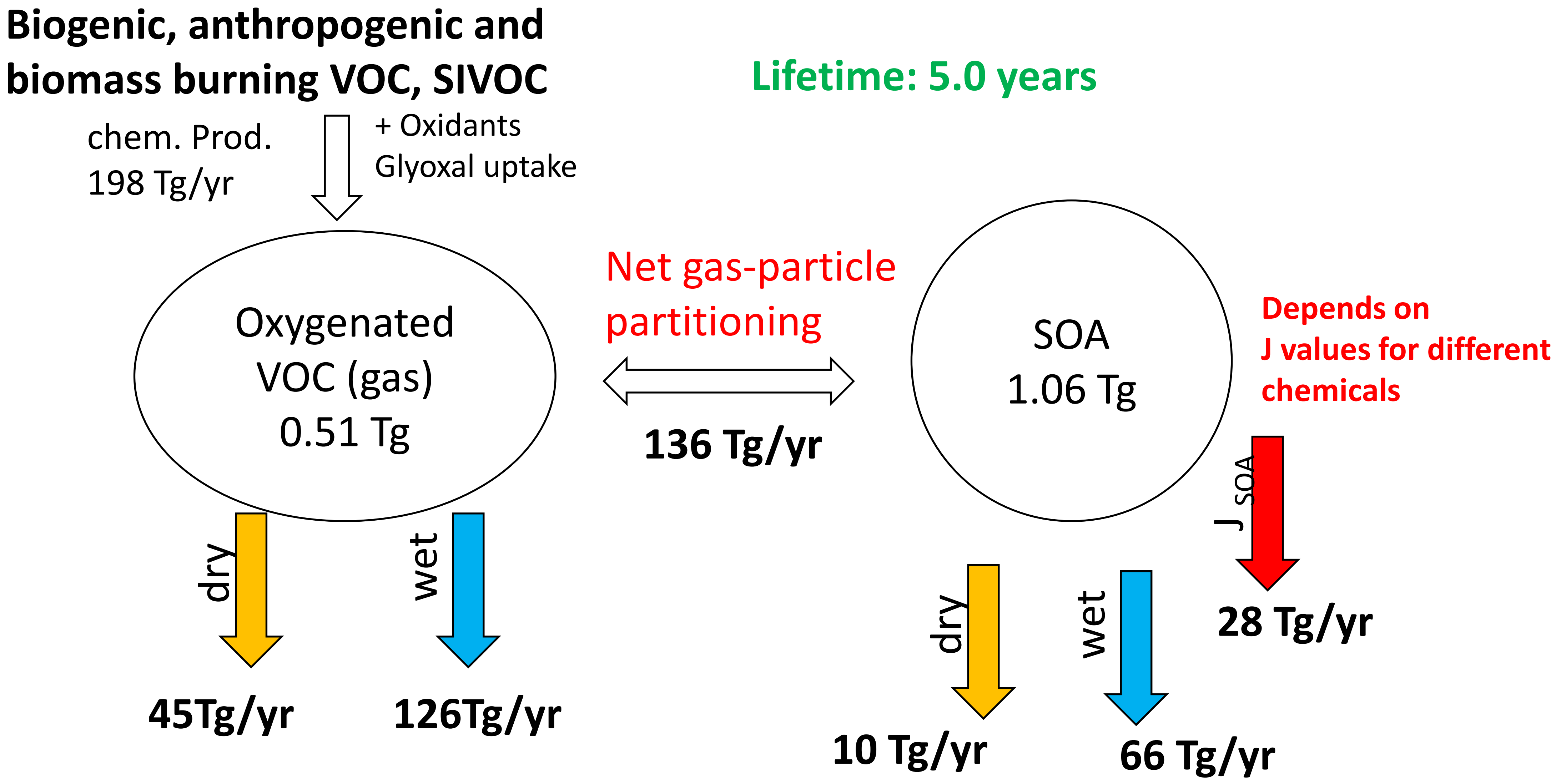
Table 2. Henry's law constants used in this study based on values reported in Hodzic et al. (2014). H^{eff} of n-alkanes is used for oxidation products of all anthropogenic precursors whereas H^{eff} of terpenes is used for those of biogenics. For products of IVOCs used in Table 1, we use $H^{eff} = 10^3$ M atm^{-1}.

Saturation concentrations (µg m^{-3})	0.01	0.1	1	10	100	1000
Anthropogenic: H^{eff} n-alkanes (M atm^{-1})	1.3×10^7	3.2×10^5	4.0×10^5	1.3×10^5	1.6×10^5	10^5
Biogenic: H^{eff} terpenes (M atm^{-1})	7.9×10^{11}	6.3×10^{10}	3.2×10^9	6.3×10^8	3.2×10^7	1.3×10^7

Hodzic et al., 2016

Most of it from biogenic emissions
-> strongly dependent on MEGAN emissions

VBS Budgets



Values very close to observational estimates!

F2000 CAM5chem test simulations 79 (5 years)

	CAM5-chem	VBS low photo	VBS high photo
ISOPRENE (TgN/yr)	513	518	523
SOAG BURDEN (Tg)	0.11	0.60	0.50
Chemical Production (Tg/yr)		197	198
SOAG DRY DEP (Tg/yr)		46.8	44.1
SOAG WETDEP (Tg/yr)		-135.7	-124.7
SOA BURDEN (Tg)	1.301	1.75	1.05
SOABB BURDEN (Tg)		0.11	0.11
SOAFF BURDEN (Tg)		0.34	0.34
SOABG BURDEN (Tg)		1.30	0.60
SOA Formation (Tg/yr)	105.0	123.4	136.2
SOA DRY DEP (Tg/yr)	16.1	10.9	9.9
SOA WETDEP (Tg/yr)	-88.7	-80.8	-66.4
SOA Photolysis (Tg/yr)		16.4	28.7
SOA LIFETIME (days)	4.5	7.0	5.0
POM BURDEN (Tg)	0.89	0.71	0.72
POM LIFETIME (days)	7.4	5.9	6.0
BC BURDEN (Tg)	0.12	0.10	0.10
BC LIFETIME (days)	6.2	5.3	5.3

- SOA VBS results in a reduction of POM and BC (increased aerosol number speed up aging of POM and BC)
- net gas-phase partitioning (~136 TgC/yr), in agreement with observations
- SOA VBS Lifetime increased due to reduced wet deposition

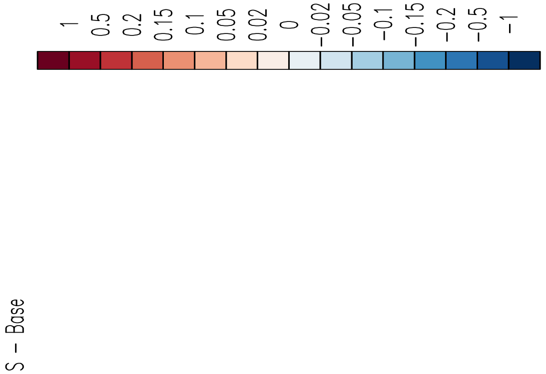
F2000 CAM5chem test simulations (2deg, 10 years)

CAM5 SOAG, 0.94 TgC/yr

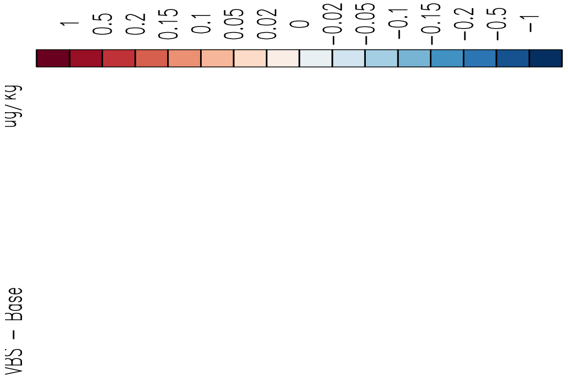
CAM5 SOAG 1.5, 1.3 TgC/yr

VBS SOA, 1.0 TgC/yr (low photolysis)

VBS minus CAM5



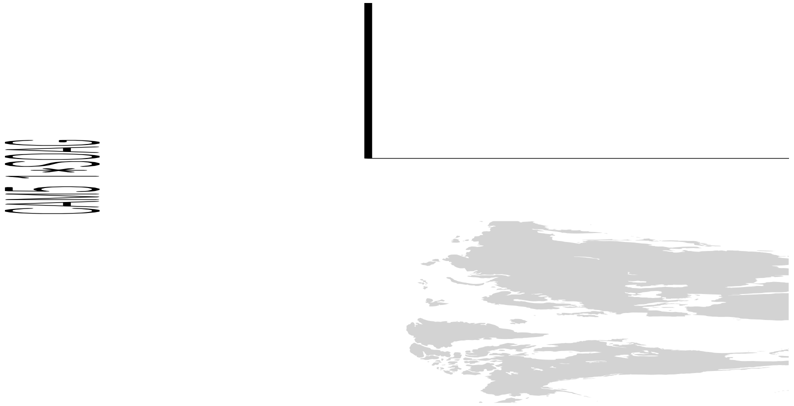
VBS minus CAM5 1.5 SOAG



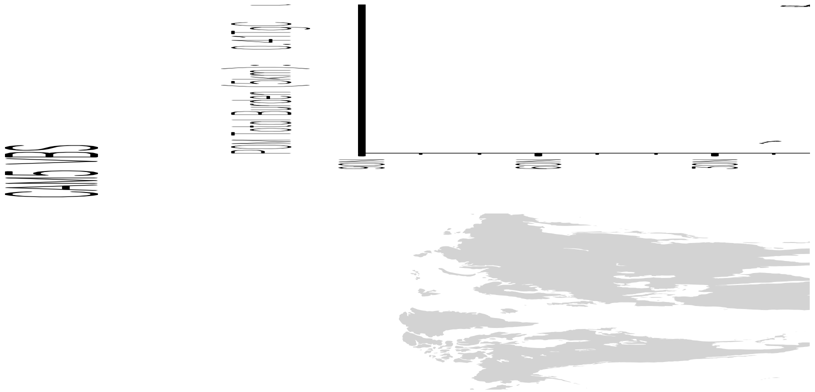
- **Formation of SOA slower due to chemical processing -> reduced SOA near surface, more SOA in upper Tropics**
- **Removal processes included for SOAG (deposition) and SOA (photolysis) -> reduced values in high latitudes**

F2000 CAM5chem test simulations (2deg, 10 years)

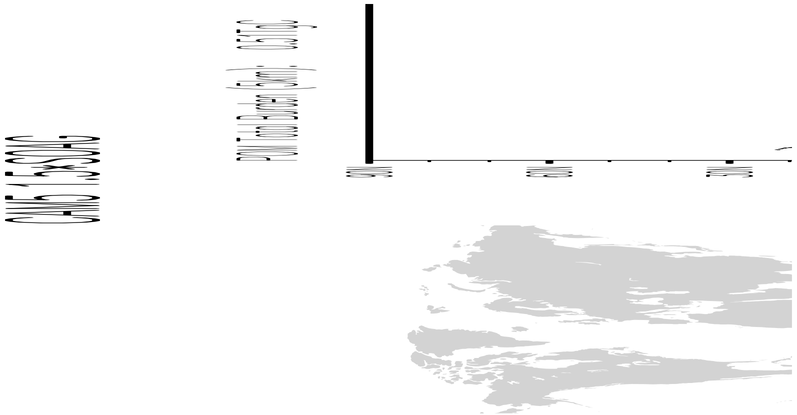
CAM5 Chem, 0.94 TgC/yr



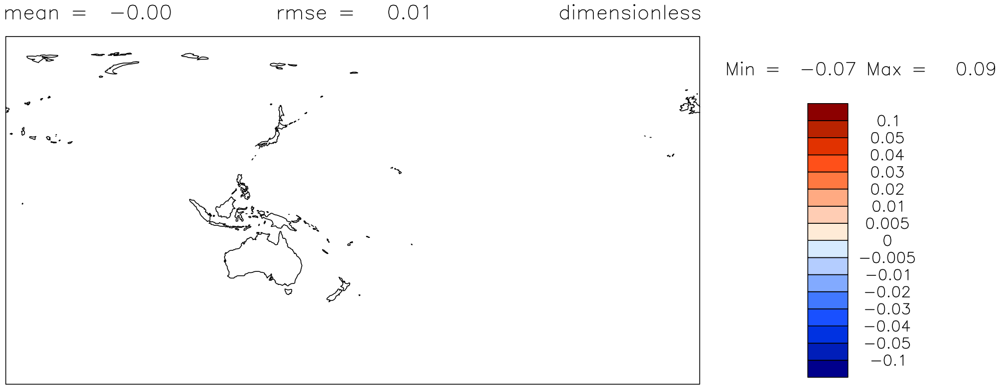
VBS SOA, 1.0 TgC/yr



CAM5 Chem SOAG 1.5, 1.3 TgC/yr



AOD Changes: VBS SOA minus CAM5 CAM 1.5*SOAG



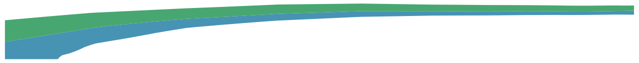
- VBS SOA approach results in more SOA over land, less over ocean
- Increased formation over polluted regions (South East Asia)

-> Changes in AOD over East Asia, reduction over the US and the ocean

Specified Dynamics CAM5chem test simulations 2011-2013

VBS SD-2012-13, Burden: 1.48 TgC

VBS F2000, Burden 1.0 TgC (earlier simulation)



- Large difference in isoprene emissions between SD and FR (450 Tg/yr vs. 550 Tg/yr)

-> increases in biogenic SAOG and SOA burden (biogenic emissions largest contributor); changes are due to clouds, leaf area index etc.

-> **testing with newest changes for CAM5 Chem SD and CLM in progress**

New SOA approach: Simplification for WACCM

- Merge 3 different categories (biomass burning, fossil fuel and biogenic) SOAG into one category
- Average Henry's law coefficient
- Estimate photolysis of SOA -> possibility to adjust the scheme to get closer agreement

Cost reduction:

SD CAM5-chem 2 degrees: ~5500 Core hours/yr

SD CAM5-chem 2 degrees VBS: ~6500 Core hours/yr ~20% increase in costs

SD CAM5-chem 2 degrees VBS simple: ~5700 Core hours/yr ~ 4% increase in costs

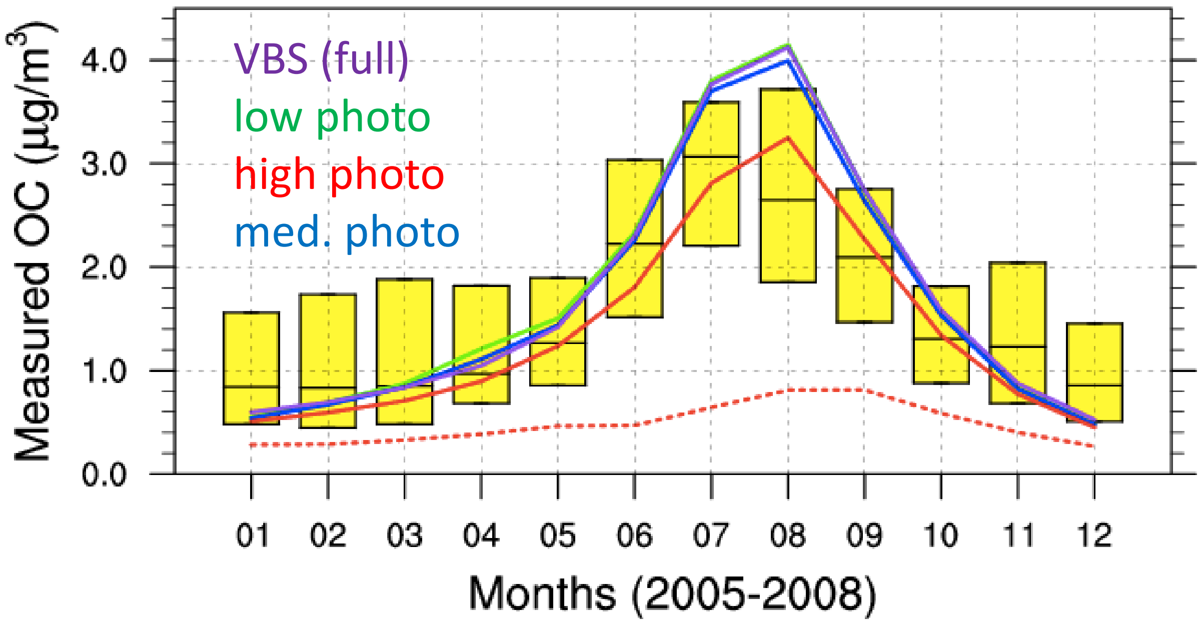
SOA Sensitivity Studies Specified Dynamics 2deg, 2011-2013, Simple Approach

VBS bg low photo

VBS low photolysis (0.004)

10 5 2 1 0.5 0.2 0.1 0.05 0.02 0.01 0.005 002

IMPROVE Comparison (20% low in summer)



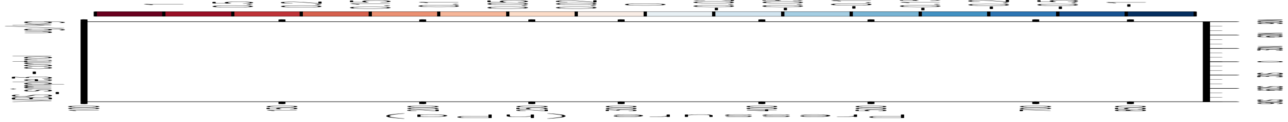
VBS high photolysis (0.04)

VBS med. photolysis (0.008)

VBS low photolysis

VBS high photolysis

VBS med. photolysis



SOA Burden (TgC)

VBS: 1.48

Simple low photolysis: 1.76

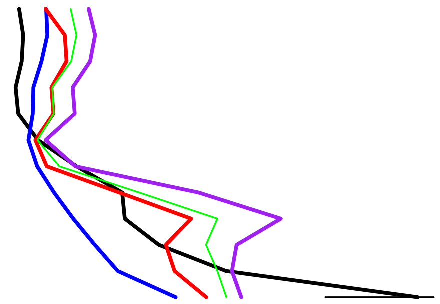
Simple high photolysis: 0.83

Simple medium photolysis: 1.51

Photolysis is based on only on a few measurements

-> can be used as tuning factor

Comparison to SEACRS Observations over the US (2013)



CAM5 chem
VBS bg low photo
VBS bg high photo
VBS lower ISOP

- Good agreement of VBS schemes at the surface.
- Important factors in higher altitudes: isoprene emissions and photolysis

Next Steps for Testing VBS for SOA for CESM2

- SD CAM5chem and WACCM simulations between 2005-2013 to compare to other aircraft campaigns
- Test full VBS version vs. simplified version in WACCM
- FHIST WACCM to identify differences in ISOPRENE emissions

-> finalize tuning for CMIP6 runs

- 1850 WACCM VBS vs. VBS simplified and to test aerosol impact
- FHIST WACCM 1850-2010 VBS simplified compare to no VBS simulation