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PNNL Improvements to MAM

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MAM4

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No.	Variables or Species	Gas-Phase	Accum.	Aitken	Primary Carbon	Coarse
1.	Number		X	X	X	X
1.	BC		X		X	
2.	POM		X		X	
3.	SOA	X	X	X		
4.	SO4	X	X	X		X
5.	NaCl		X	X		X
6.	Dust		X	X		X

X = Standard CAM5-MAM3 X = CAM5-MAM4

Total Number of Species = 21

Convective Transport and Scavenging

- Cloud-borne aerosol in convective clouds
- Vertical transport and wet removal treated simultaneously
- Improves vertical distribution of the aerosol and aerosol concentrations in remote regions
- Increases cloud liquid water path, strengthens SWCF
- Candidate for CAM5.4

H. Wang et al., GMD, 2013

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Advancing treatment of secondary organic aerosols in CESM using the Volatility Basis Set - MAM approach

MANISH SHRIVASTAVA

Pacific Northwest National Laboratory

JGR-Atmospheres, revised

Acknowledgement: DOE SciDAC ACES4BGC project

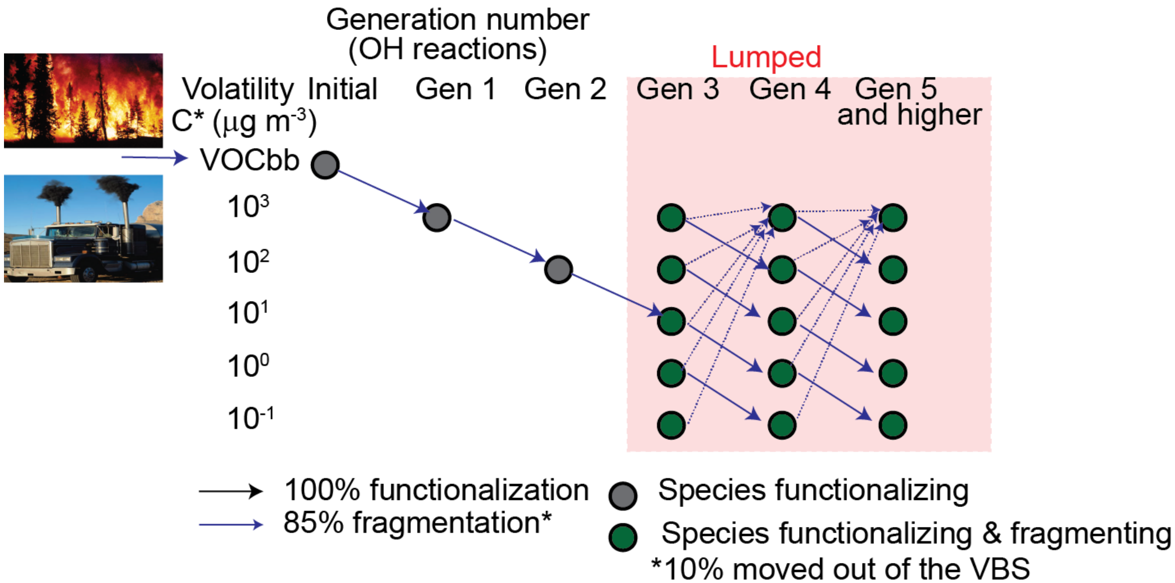
Methodology

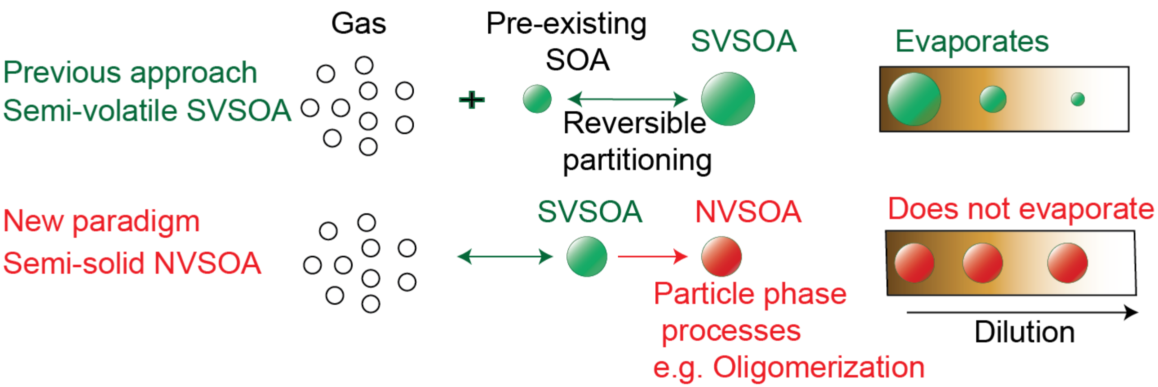
- CAM5 (1.9× 2.5), nudged to ERA-Interim reanalysis
- Mozart chemistry coupled to modal aerosol module (MAM3)
- 62 SOA species (42 SOA particle species and 20 gas phase organic species) + 104 gas-phase tracers from Mozart chemistry
- Compare 4 different treatments of SOA particles:

1. Standard CAM treatment (SOAG, SOA, no chemistry)
2. Semi-volatile liquid-like SOA formed by photochemistry: No fragmentation
3. Semi-volatile SOA with gas-phase fragmentation
4. Non-volatile semi-solid SOA with gas-phase fragmentation

- 2 and 3: Effects of gas-phase chemistry: fragmentation reactions
- 3 and 4: Effects of phase-state of SOA: liquid vs. semi-solid

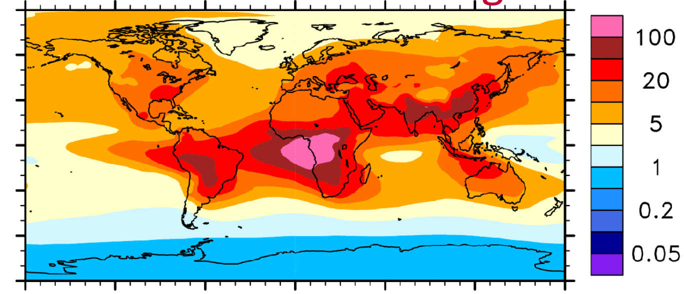
VBS-MAM SOA treatment: Effects of fragmentation and semi-solid SOA



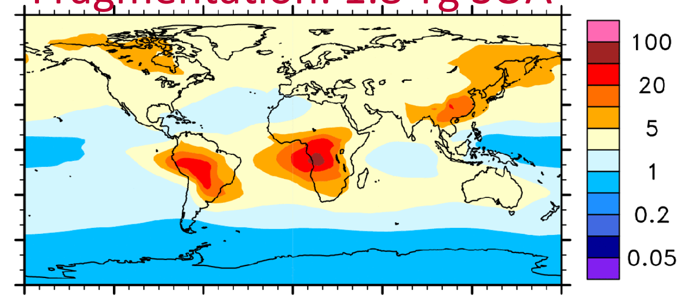


Standard CAM : 1.1 Tg SOA

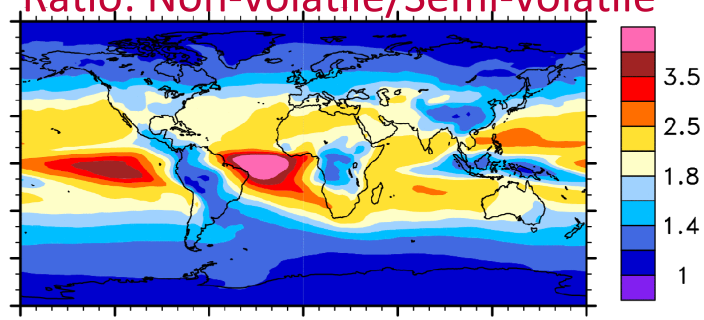
Functionalization: 7.5 Tg SOA



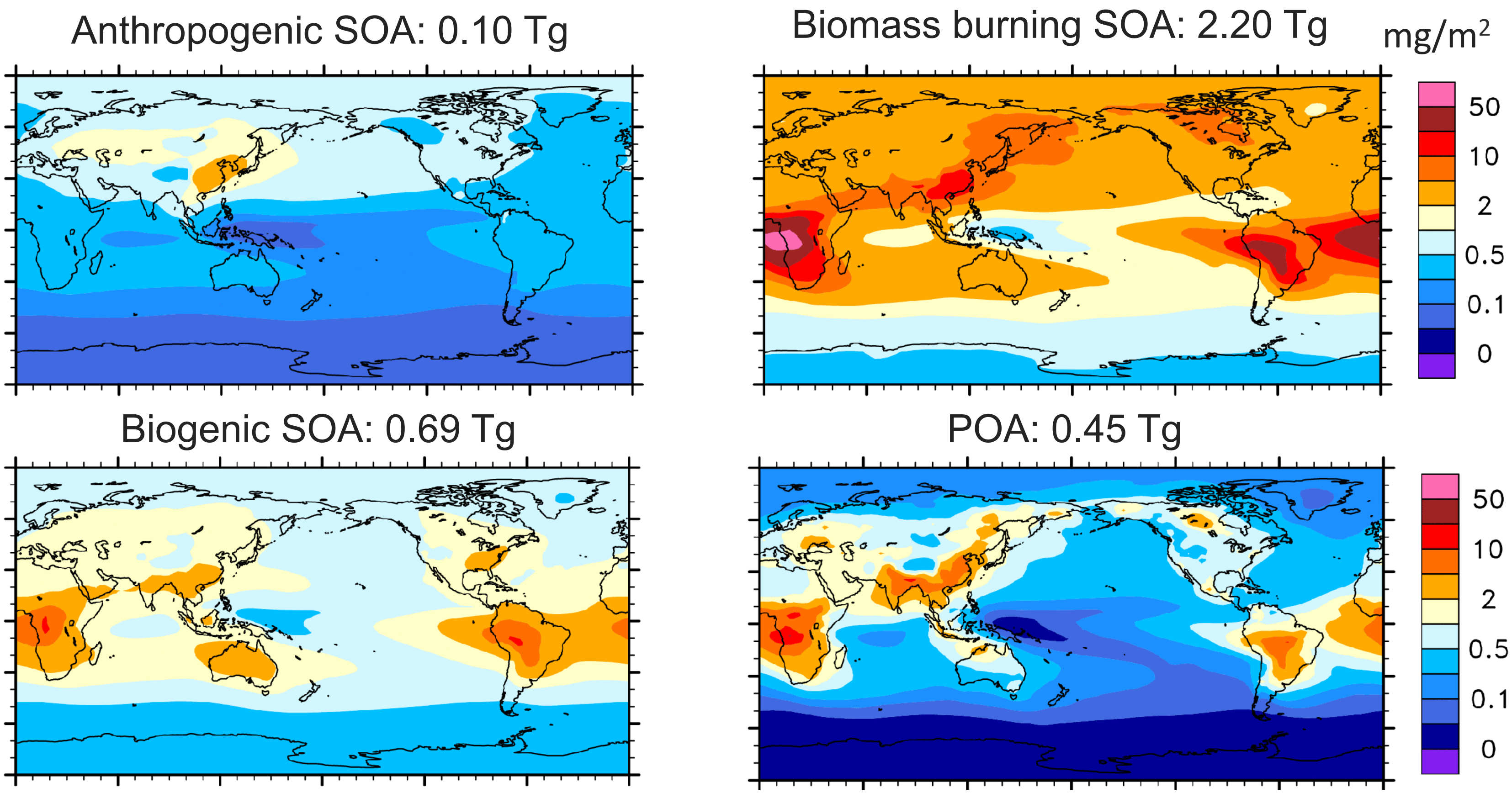
Fragmentation: 1.8 Tg SOA



Ratio: Non-volatile/Semi-volatile



SOA Source contributions: Fragmentation and non-volatile SOA



- Biomass burning is the largest source of SOA globally
- SOA from either biomass burning or biogenic sources is much larger than anthropogenic SOA

Simulated and measured vertical profile of OA: Aircraft observations

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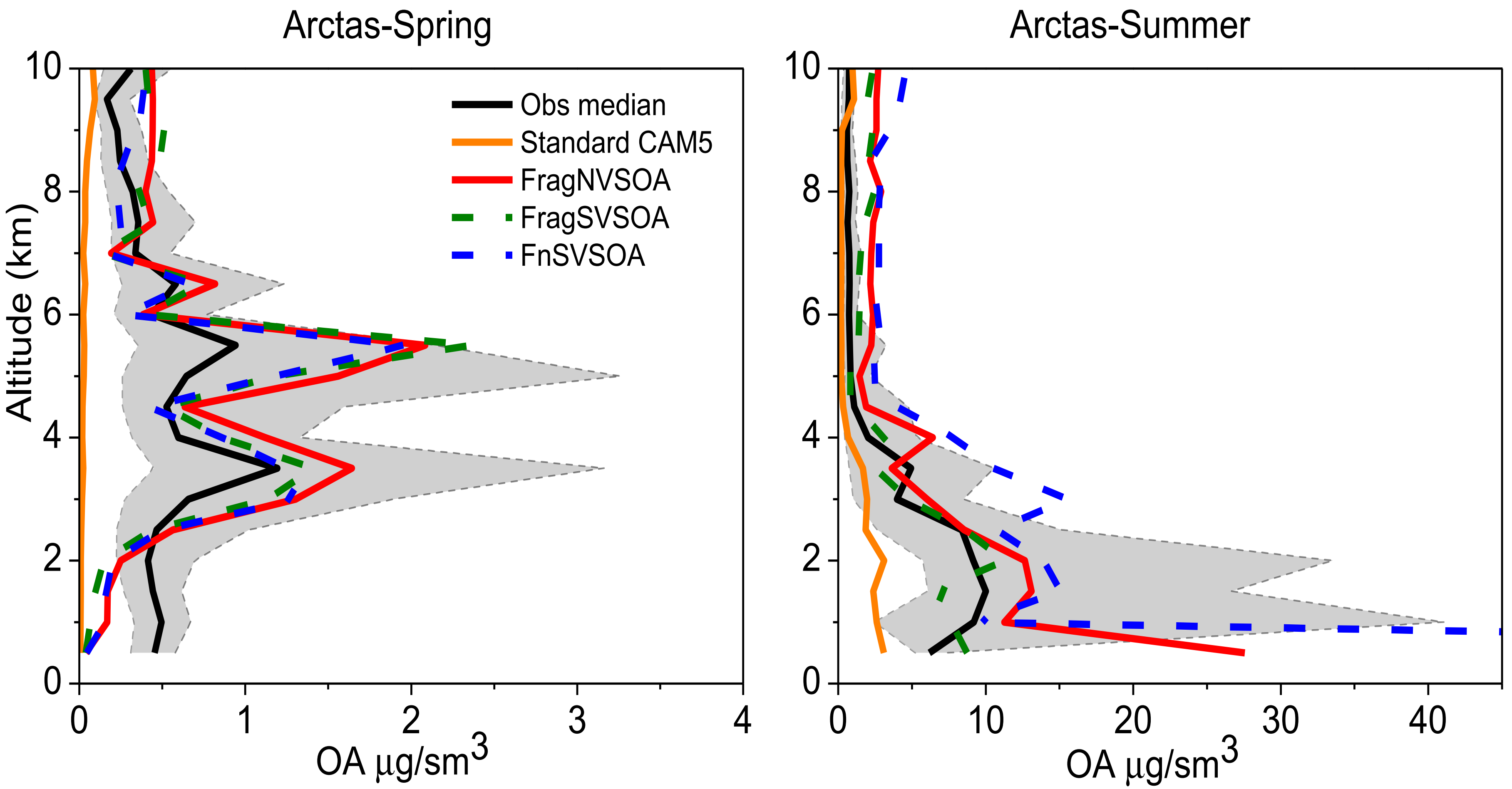
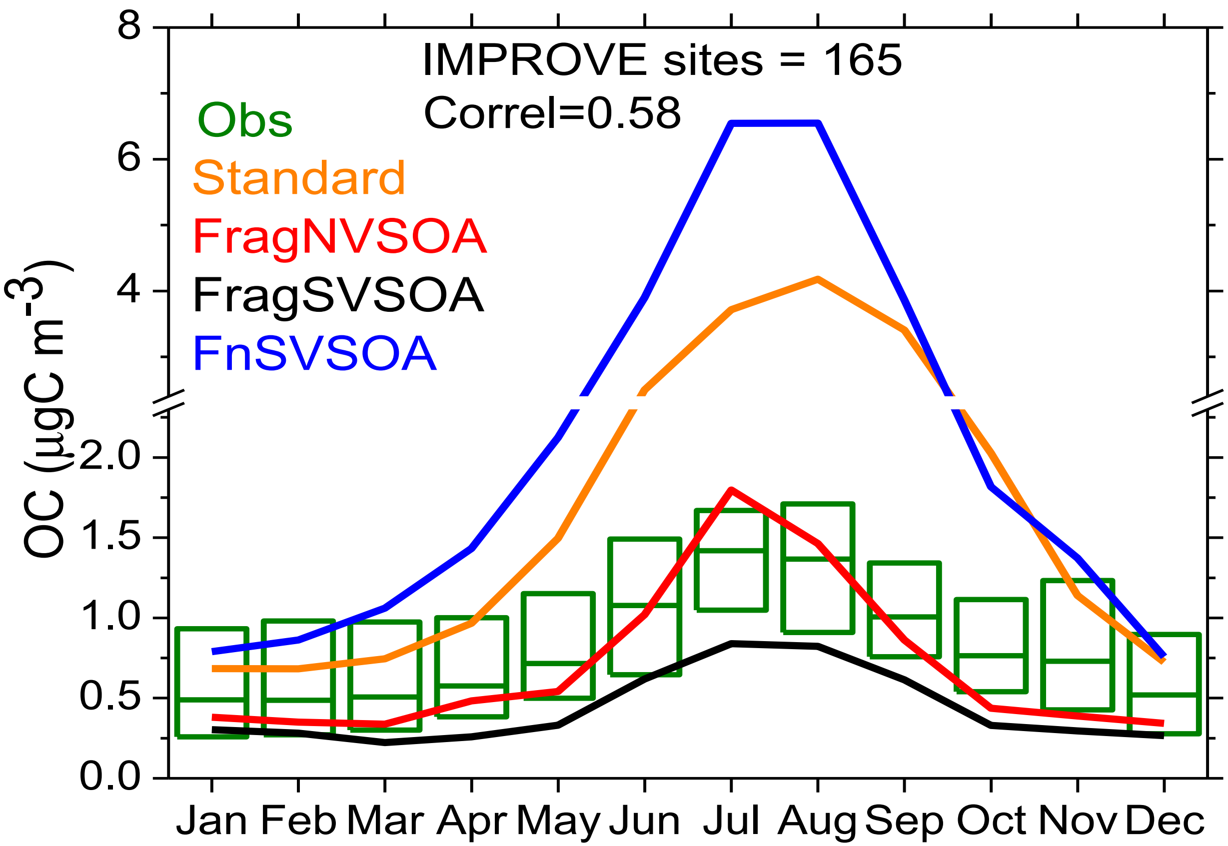


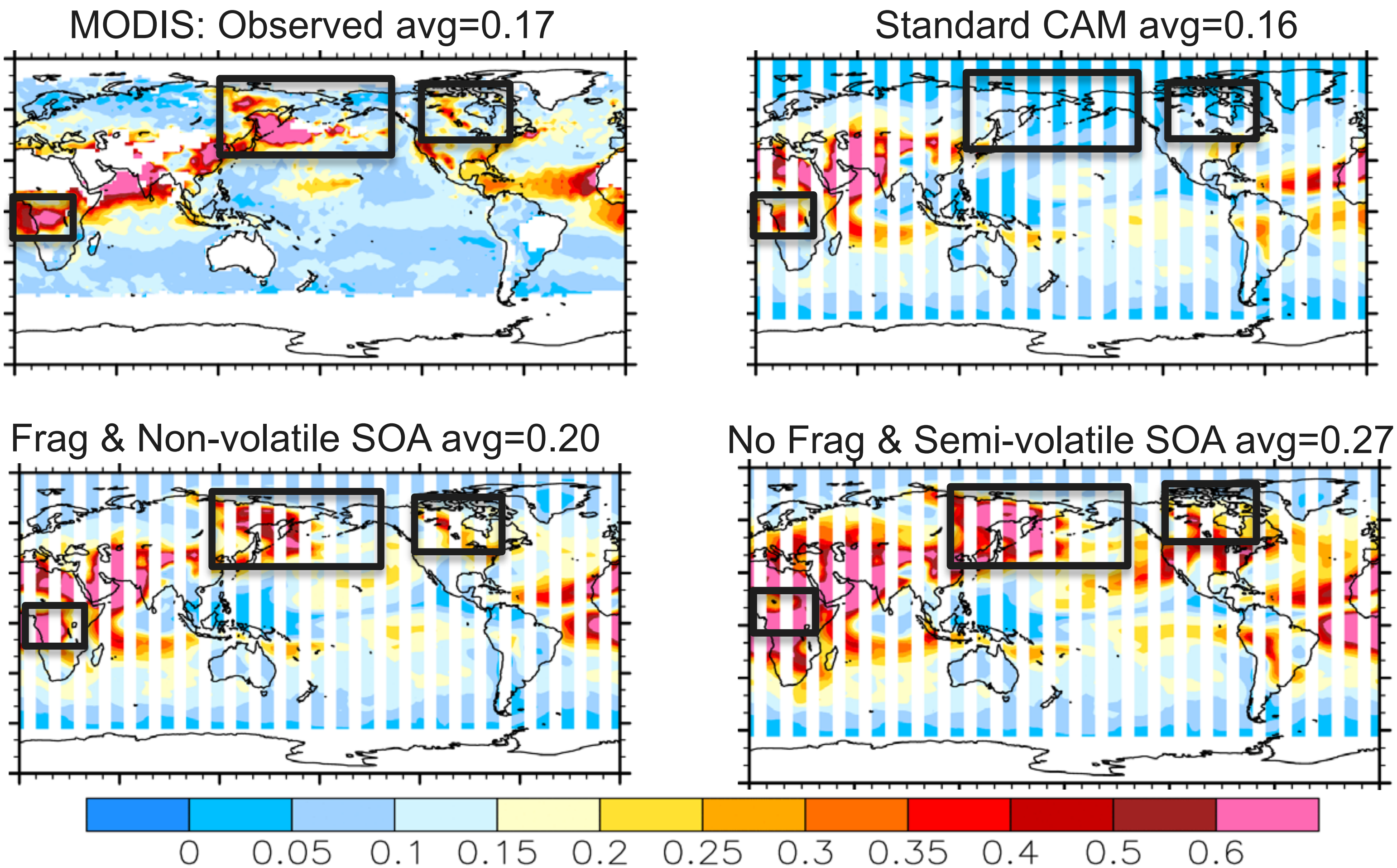
- Standard CAM underpredicts OA by orders of magnitude
- Revised model treatments significantly improve OA predictions mainly due to contributions from biomass burning SOA, missing in standard CAM

Evaluation with U.S. IMPROVE network OC measurements (2007-2011)



- Standard CAM treatment overestimates OC
- Revised treatment with non-volatile NVSOA and fragmentation (red) shows much better agreement

Measured and simulated AOD: MODIS July 2008



- Standard CAM underpredicts AOD over Eastern Russia, Northern Canada and Central Africa
- Revised treatments shows much better agreement

Direct radiative forcing (DRF) of SOA

Studies	DRF (W m^{-2})
Aerocom Phase II Intercomparison experiments (Myhre et al. 2013)	-0.01 to -0.21 (mean: -0.06)
Spracklen et al. 2011	-0.26±0.15 (anthropogenic controlled SOA)
This study: Fragmentation & Non-volatile SOA	TOA: -0.50, Surface: -1.3
This study: Fragmentation and Semi-volatile SOA	TOA: -0.26, Surface: -0.71

- This study: DRF of SOA at the high end of previous estimates
- Difference between surface and top of atmosphere (TOA) DRF is mainly due to the absorbing component of SOA
- Although SOA has ~40x lower imaginary refractive index compared to BC, the large SOA loadings result in SOA DRF comparable to BC

Conclusions

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- Multi-generational aging of organic vapors increases SOA concentrations throughout the domain
- Fragmentation reduces SOA concentrations by a factor of 2-3
- Treating SOA as non-volatile semi-solid (glassy) increases SOA concentrations compared to its traditional semi-volatile treatment
- Biomass burning is the largest global source of SOA
- Fragmentation and non-volatile SOA shows much better agreement with measurements compared to Standard CAM treatment
- Simulated direct radiative forcing of SOA is at the higher end of previous global model estimates
- Absorbing component of SOA is comparable to black carbon (BC) due to much higher simulated SOA loadings
- Effects of absorbing SOA expected to be even larger downwind of biomass burning regions
- Improved process-level SOA representation: key for improving CCN predictions

Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) in CAM5

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MOSAIC Processes

- Gas-phase chemistry treated by MOZART photochemical mechanism
- Fully dynamic partitioning of sulfate, nitrate, chloride, and ammonium amongst different modes via reversible and irreversible reactions
- Equilibrium water content calculated using the ZSR mixing rule
  - Polynomial fits for water due to inorganic salts
  - Kappa-Kohler theory for water due to organics
  - Kelvin effect taken into account at high RH
- Particles assumed to be completely deliquesced at RH > 35%
  (water hysteresis will be activated after implementing organic-inorganic interactions)

Reversible Gas-Particle Reactions

gas-solid

$NH_4Cl(s) \leftrightarrow NH_3(g) + HCl(g)$

$NH_4NO_3(s) \leftrightarrow NH_3(g) + HNO_3(g)$

gas-liquid

$NH_3(g) \leftrightarrow NH_3(aq)$

$HNO_3(g) \leftrightarrow H^+(aq) + NO_3^-(aq)$

$HCl(g) \leftrightarrow H^+(aq) + Cl^-(aq)$

liquid-liquid

$H_2O(aq) + NH_3(aq) \leftrightarrow OH^-(aq) + NH_4^+(aq)$

$H_2O(aq) \leftrightarrow H^+(aq) + OH^-(aq)$

$HSO_4^-(aq) \leftrightarrow H^+(aq) + SO_4^{2-}(aq)$

Zaveri et al., JGR, 2008

Irreversible Heterogeneous Reactions

Reactions With $H_2SO_4(g)$

$CaCO_3(s) + H_2SO_4(g) \rightarrow CaSO_4(s) + H_2O(g)\uparrow + CO_2(g)\uparrow$

$CaCl_2(s,l) + H_2SO_4(g) \rightarrow CaSO_4(s) + 2HCl(g)\uparrow$

$Ca(NO_3)_2(s,l) + H_2SO_4(g) \rightarrow CaSO_4(s) + 2HNO_3(g)\uparrow$

$2NaCl(s,l) + H_2SO_4(g) \rightarrow Na_2SO_4(s,l) + 2HCl(g)\uparrow$

$2NaNO_3(s,l) + H_2SO_4(g) \rightarrow Na_2SO_4(s,l) + 2HNO_3(g)\uparrow$

Reactions With $HNO_3(g)$

$CaCO_3(s) + 2HNO_3(g) \rightarrow Ca(NO_3)_2(s) + H_2O(g)\uparrow + CO_2(g)\uparrow$

$CaCl_2(s) + 2HNO_3(g) \rightarrow Ca(NO_3)_2(s) + 2HCl(g)\uparrow$

$NaCl(s) + HNO_3(g) \rightarrow NaNO_3(s) + HCl(g)\uparrow$

Reactions With $HCl(g)$

$CaCO_3(s) + 2HCl(g) \rightarrow CaCl_2(s) + H_2O(g)\uparrow + CO_2(g)\uparrow$

Reactions With $NH_3(g)$

$NH_4HSO_4(s) + NH_3(g) \rightarrow (NH_4)_2SO_4(s)$

$(NH_4)_3H(SO_4)_2(s) + NH_3(g) \rightarrow 2(NH_4)_2SO_4(s)$

$2NaHSO_4(s) + NH_3(g) \rightarrow Na_2SO_4(s) + NH_4HSO_4(s)$

Variables and Species in Standard CAM5-MAM7

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No.	Variables or Species	Gas-Phase	Accum.	Aitken	Primary Carbon	Fine Seasalt	Coarse Seasalt	Fine Dust	Coarse Dust
1.	Number		X	X	X	X	X	X	X
2.	BC		X		X				
3.	POM		X		X				
4.	SOA	X	X	X					
5.	SO4	X	X	X		X	X	X	X
6.	NH4	X	X	X		X	X	X	X
7.	NaCl		X	X		X	X		
8.	Dust							X	X
		3	7	5	3	4	4	4	4

Total Number of Species = 34

Variables and Species in CAM5-MAM7-MOSAIC

No.	Variables or Species	Gas-Phase	Accum.	Aitken	Primary Carbon	Fine Seasalt	Coarse Seasalt	Fine Dust	Coarse Dust
1.	Number		X	X	X	X	X	X	X
1.	BC		X		X				
2.	POM		X		X				
3.	SOA	X	X	X					
4.	SO4	X	X	X		X	X	X	X
5.	NH4	X	X	X		X	X	X	X
6.	NO3	X	X	X		X	X	X	X
7.	Cl	X	X	X		X	X	X	X
8.	Na		X	X		X	X		
9.	Dust							X	X
10.	Ca							X	X
11.	CO3							X	X
12.	MOZART Trace Gases	114							
		119	9	7	3	6	6	8	8

X = Standard CAM5-MAM7 X = CAM5-MAM7-MOSAIC Total Number of Species = 166

Variables and Species in CAM5-MAM4-MOSAIC

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No.	Variables or Species	Gas-Phase	Accum.	Aitken	Primary Carbon	Coarse
1.	Number		X	X	X	X
1.	BC		X		X	
2.	POM		X		X	
3.	SOA	X	X	X		
4.	SO4	X	X	X		X
5.	NH4	X	X	X		X
6.	NO3	X	X	X		X
7.	Cl	X	X	X		X
8.	Na		X	X		X
9.	Dust		X	X		X
10.	Ca		X	X		X
11.	CO3		X	X		X
12.	MOZART Trace Gases	114				
		119	9	7	3	9

X = Standard CAM5-MAM4   X = CAM5-MAM4-MOSAIC   Total Number of Species = 150