F: Possible next-generation aerosol developments in the ACME global model

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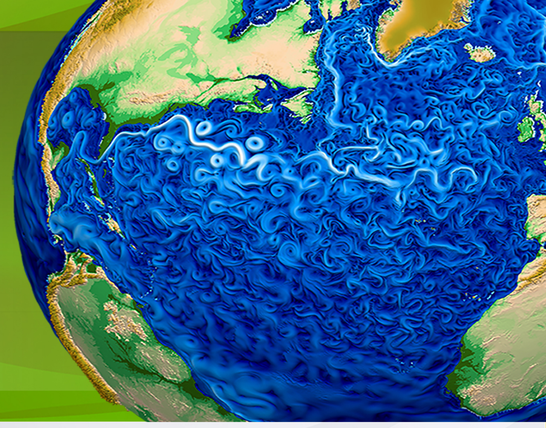


Hypothesis

New and advanced aerosol parameterizations will largely change the radiative forcing of aerosols simulated by current generation global climate models (GCMs) since key sources and processes that govern climate-relevant aerosol populations are "missing" in these GCMs.

Secondary organic aerosols (SOA) are often the dominant components of fine aerosols at many locations globally, but are also the least understood. Current climate models have very crude representations of SOA particles , which do not account for :

- Major fraction of semi-volatile/low volatility SOA precursor emissions ("missing" sources)
- Multigenerational chemistry of gas-phase SOA precursors and related key source and sink terms (functionalization and fragmentation reactions)
- Particle-phase processes governing the volatility, loadings, lifetimes and evolution of SOA

Recently, in Shrivastava et al. (2015), we demonstrated that replacing the previous crude SOA treatments with more advanced treatments that account for above listed processes, causes large increases in simulated aerosol burdens, lifetimes and direct radiative forcing compared to previous models. These new treatments dramatically improve agreement with a suite of surface, aircraft and satellite measurements

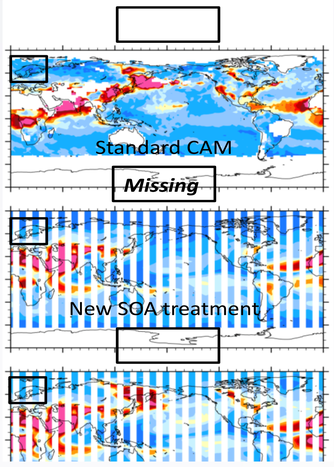
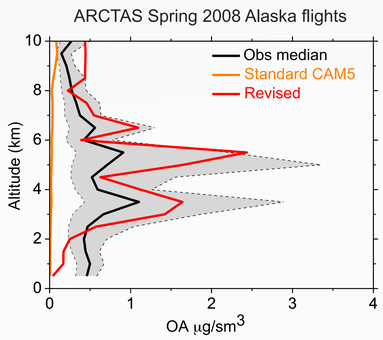
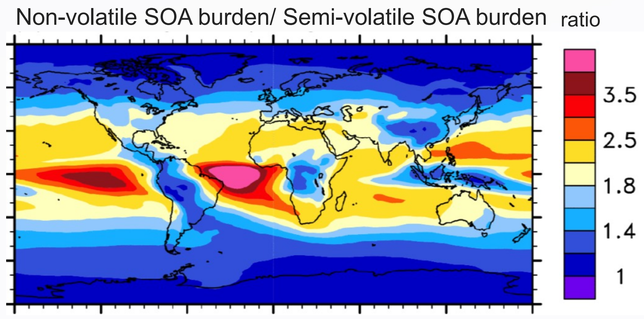


Figure on left: Compared to MODIS Aerosol Optical Depth (AOD) measurements, standard CAM severely underpredicts AOD, mainly over biomass burning regions of Eastern Russia, Northern Canada and Central Africa. In comparison, our revised SOA treatment shows much better agreement with MODIS AOD, mainly due to large simulated contribution of organic aerosols (OA) from biomass burning.

Figure on right: Standard CAM formulation (orange line) completely fails to simulate SOA corresponding to aircraft measurements during the ARCTAS 2008 field campaign. New SOA treatments show much better agreement with measured vertical profile of OA over the North American Arctic and sub-Arctic regions.





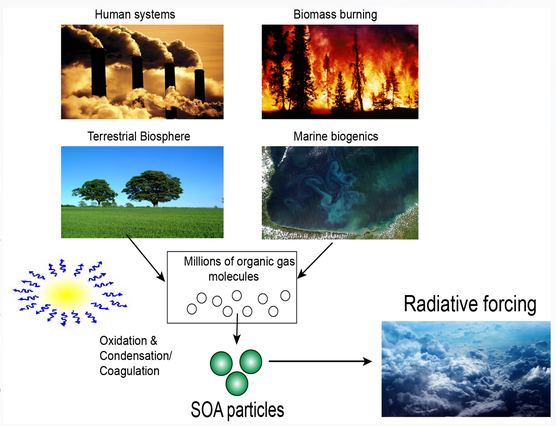
Recent measurements suggest that contrary to traditional model assumptions treating SOA as semi-volatile liquid-like solutions, ambient SOA has very low "effective volatility" (Vaden et al. 2011). We investigated implications of treating SOA as "effectively non-volatile". The ratio of the new non-volatile SOA to previous semi-volatile SOA treatment (used in most global models) varies by a factor of 2-5. Larger differences are seen over regions of continental outflow over the oceans, strongly suggesting that findings related to low volatility of SOA have large implications on simulated SOA loadings and cloud condensation nuclei (CCN) concentrations mainly over cleaner marine regions.

Developments Required

Future aerosol developments that ACME should consider

SOA particles are created by complex multiscale interactions between human systems, biomass burning, terrestrial biosphere and marine biogenic emissions linked by physical and chemical processes in the atmosphere. While new treatments of SOA largely improves global model predictions compared to measurements, these new treatments are computationally expensive. We suggest the following developments as a part of future directions:

1. Implementation of a reduced-tracer and computationally efficient version of these new SOA treatments within the ACME model for long-term climate simulations
2. New treatments of organic aerosols related to biomass and biofuel burning, terrestrial biogenics and fossil-fuel, which explicitly account for gas-phase multigenerational chemistry, low volatility-SOA and missing precursor emissions
3. Implementation of a new nucleation mode within the modal aerosol model (MAM) to explicitly represent freshly nucleated particles and their growth from 1 to 10 nm
4. Including the role of low volatility organic vapors during the nucleation and growth of organic aerosols
5. Investigating the absorbing effects of brown carbon OA
6. New SOA formation mechanisms due to aging of marine isoprene and terpene emissions (not currently included in most atmospheric models)
7. Conducting coupled land-ocean-atmosphere simulations to investigate the impacts of these new multi-scale interactions on direct/indirect forcing and future climate change



Expected Impact

Above suggested "missing" multi-scale interactions between human systems and terrestrial and marine biosphere are expected to change how we think about aerosols and their associated climate forcing

In addition following are expected immediate impacts on aerosol predictions by the climate model:

1. Large improvements in simulated SOA loadings and associated radiative forcing as demonstrated by our recent study (Shrivastava et al. 2015)
2. Impacts on simulated indirect forcing of aerosols due to inclusion of "missing" precursors, new multigenerational chemistry treatments, and proposed new nucleation mode explicitly accounting for the role of organic vapors on new particle formation and growth
3. Significantly improved representation of absorbing effects of brown carbon OA, especially in regions affected by biomass burning
4. Large impacts on climate due to new treatments of aging of marine biogenic emissions and also increased loadings and lifetimes of SOA corresponding to continental outflow over marine environments, where cloud albedo is highly sensitive to CCN concentrations

References:

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2. Vaden TD, Imre D., Beranek J., **Shrivastava M.**, Zelenyuk. A. "Evaporation Kinetics and Phase of Laboratory and Ambient Secondary Organic Aerosol." *Proceedings of National Academy of Sciences*, 2190-2195, 2011.

ACME Accelerated Climate Modeling for Energy

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