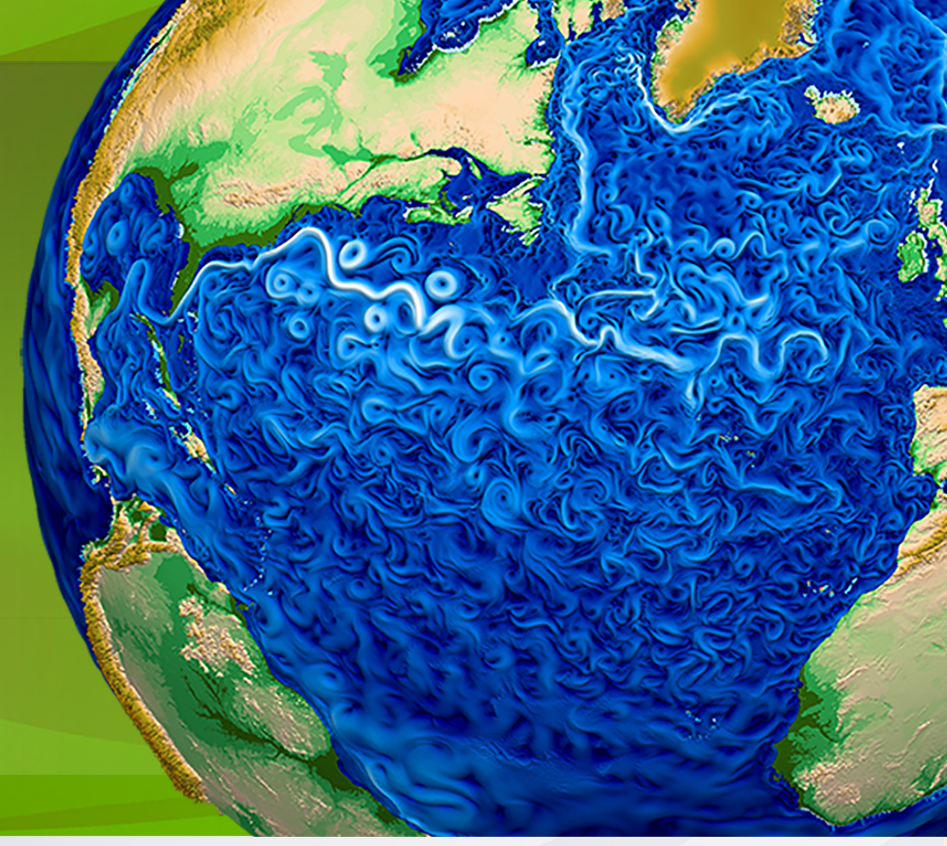

F: Coordinating Natural Aerosol Simulations: A Unique ACME Capability

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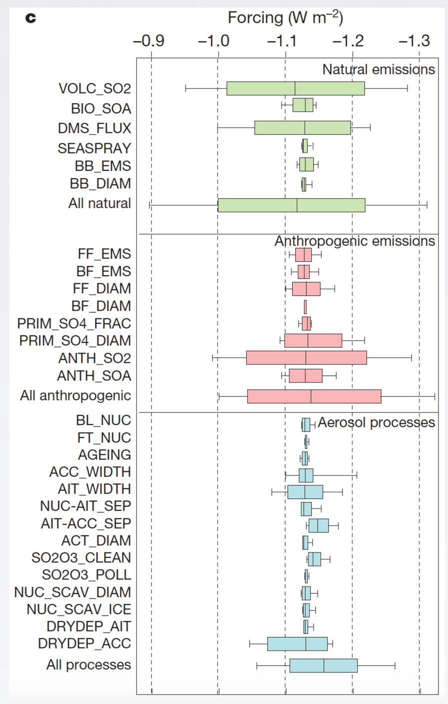
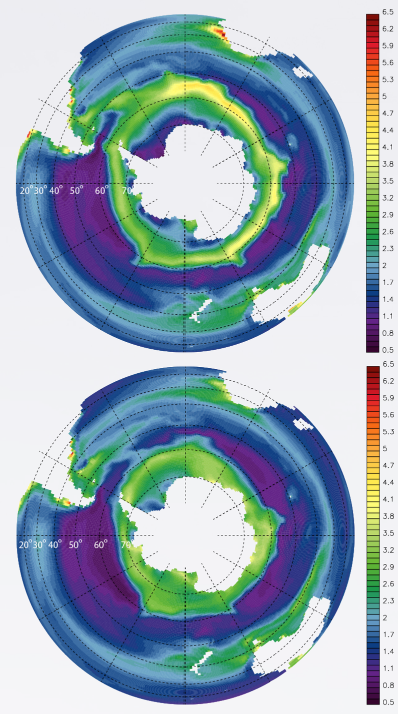
Hypothesis

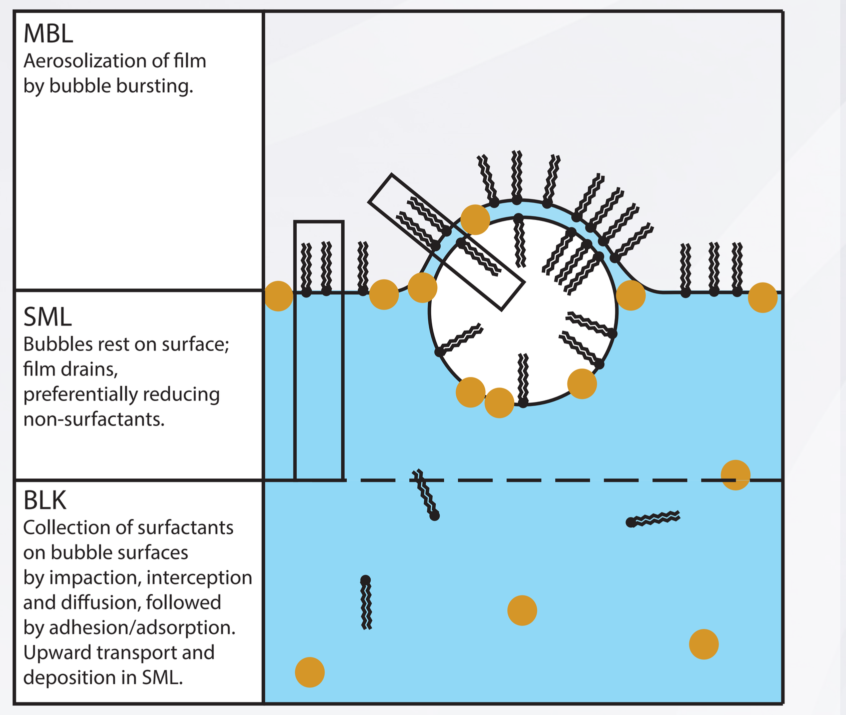
Carslaw and company may be right: Uncertainties in natural aerosol emissions are dominant over the anthropogenic (Nature 503, 2013)

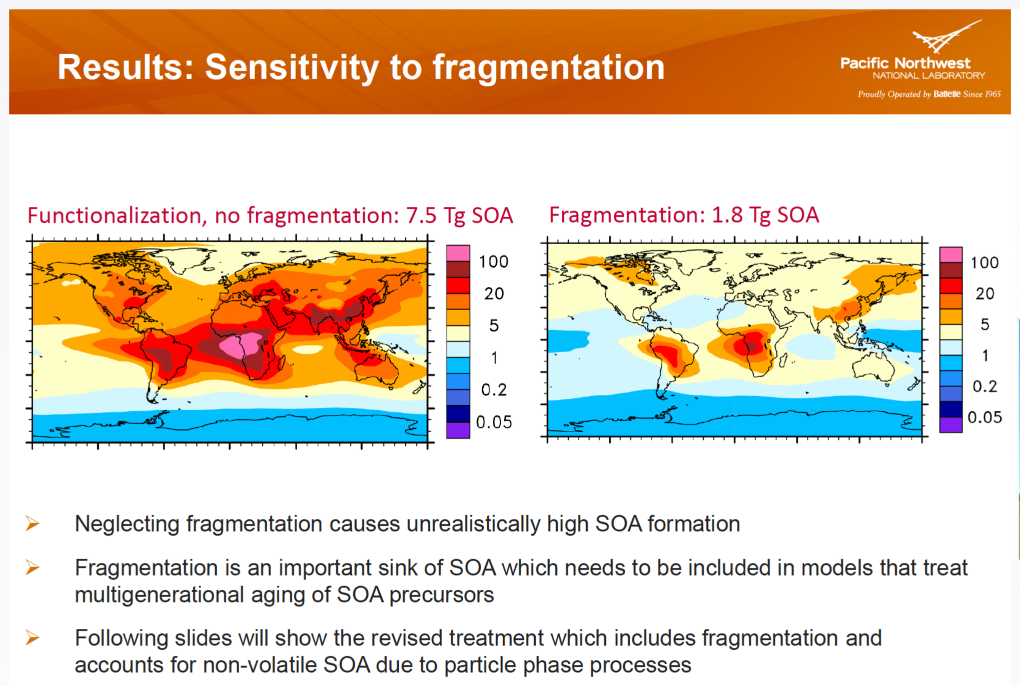
Carslaw et al. have argued eloquently and recently (2013) that a major source of uncertainty in our near term understanding of the Earth System lies hidden in the natural background aerosol. The major points may be distilled as follows: Variation deriving from our lack of comprehension of background emissions/effects is so large that even the poorly known anthropogenic contribution is overwhelmed. The pristine-remote aerosol system is complex, involving detailed marine, terrestrial and atmospheric organic chemistries and also their coupling to inorganics through multiphase processes. But due to a confluence of legacy capabilities and new collaborations, the Department of Energy is now uniquely positioned to disentangle some of the critical channels. We overview detailed mechanisms under development for ACME in the areas of marine-terrestrial POA-SOA emissions (P= Primary and S=Secondary Organic Aerosol respectively). Interactions with other particle classes are included plus the ensuing atmospheric, aerosol and cloud chemical processing. Taken together, current efforts distributed across the complex promise to elucidate major geochemical influences on cloud structure and radiation transfer. Our presentation ranges from oceanic biopolymers in spray, to biomass burning byproducts to terrestrial vegetation emissions of volatile carbon. We propose an orchestrated effort spanning the laboratories, to apply ACME as a tool for quantification of the implied uncertainties. Although dust is less directly related to the radiation budget, we will further extend development and simulations to the chemistry of mineral particles including ocean fertilization. Our concepts follow logically from the commitment of ACME to soon release marine ecodynamics and carbon cycling, since together these are major drivers on the ocean side. All current simulation themes of DOE Earth System modeling are impacted. Connections with overall global biogeochemistry are clear, plus aerosol-cloud relations play into hydrological cycling and the stability of land ice.

Top Row: Natural aerosol indirect effect uncertainties are a key issue in the study of climate sensitivity. DOE supports dynamic DMS (year 2000 & 2100 nM), plus also the upper ocean macromolecules, including proteins, polysaccharides, lipids, mixtures, amino sugars and the humics









Bottom Row: Also at the various labs -atmospheric organic chemistry, several approaches to vegetative emission, dust evolution.

Developments Required

Many ACME components already in place:

Marine sulfur cycle –LANL and LLNL have developed dynamic, ecostructural models for the oceanic distribution of dimethyl sulfide and its precursors. Order one dozen phyto-classes are treated for their organosulfur content with resolved nutrient, cryological, acidification and other stressors. Gas phase atmospheric photochemical processing is handled in the superfast mechanism of LLNL.

Marine primary organics –PNNL and LANL have developed a first representation for the individual marine macromolecules which become primary organic aerosol via bubble breaking. Traditional DOC is resolved in the systems model ocean into at least half a dozen polymer families. Global Langmuir monolayer simulations at the air(bubble)-water interface provide organic sourcing to the marine aerosol.

Terrestrial SOA -PNNL simulates the chemistry of large organics emanating from biomass burns and other sources. Sophisticated handling of volatility, oligomerization and fragmentation translates to many dozens of tracers, but it is possible to streamline to a numerical size comparable to the Superfast. Early results suggest strong effects on cloud condensation nucleus numbers in continental plumes.

VOC –Contributions from PNNL and ORNL. Terrestrial vegetation releases isoprene, terpenes and other volatile organics into and from the canopy. Effects can be characterized by MEGAN2 bio-emissions linked to dynamic plant functional types in the ACME Land Model (ALM). Twenty compound classes are now used to represent 150 compounds. Atmospheric organic chemistry courtesy PNNL and LLNL.

***Needs*: Such emissions are only partially integrated into the multi-modal CAM aerosol and links to the ACME land models require further development. Dust processing capabilities exist within the DOE complex and will be drawn in. With proper coordination, ACME could be used to reduce large uncertainties in AID and geochemical feedbacks.**

Expected Impact

Already achieved:

Our team has already reduced marine DMS uncertainties through detailed ecostructural calculations, while simultaneously revealing a strong nonlinear dependence of Arctic sea ice on biogenic release. The first ever global macromolecular surfactant and spray flux calculations have been reported. Volatility basis sets and reaction pathway parameterizations have been tested for the biomass burning aerosol. Dynamic vegetation release of VOC by distinct plant types can soon be coupled. Dust iron cycling is being explored for implementation.

***ACME Application*: We will simulate the collective natural aerosol system in a unified manner while eliminating, reducing or elucidating a major set of Earth System uncertainties –those obscuring climate sensitivity analysis through aerosol indirect effects. Our project unifies the laboratories and results will be unique to ACME. Most of the above mentioned aerosol emission and processing capabilities are available only inside the complex. Connections are readily cited with the current major DOE simulation themes 1) hydrological cycling (through aerosol-cloud dynamics), 2) biogeochemistry (since all sources fall into this category), plus 3) land ice decomposition (clouds and precipitition). In fact we will lobby for a new aerosol theme in V2.**

Publications:

Available on request regarding ACME specific developments in marine reduced sulfur cycling, the biomacromolecules as surfactants and POA components, VBS and fragmentation in organic-to-particle conversion, emission sets for volatile organics from vegetation, DOE land models, dust iron chemistry.