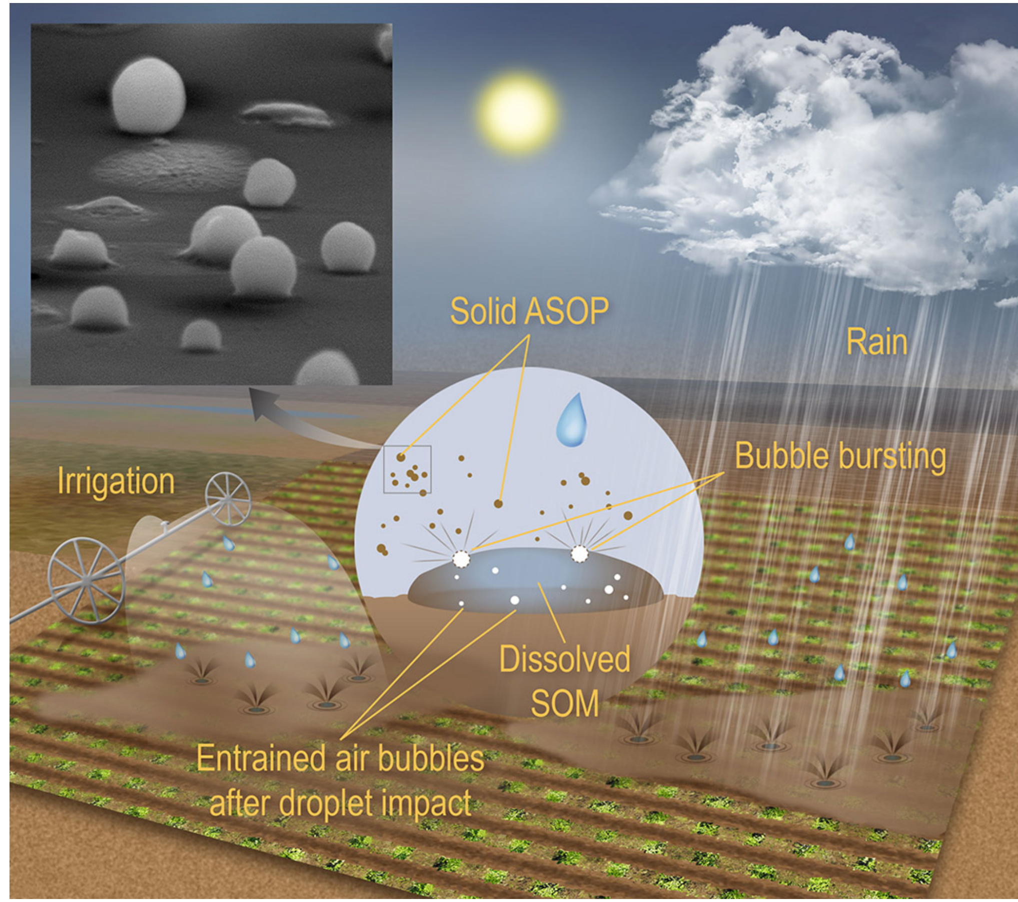
Aerosol Life Cycle Working Group Updates

Jian Wang

Airborne Soil Organic Particles Generated by Precipitation

Laskin, Gilles *and co-workers*
Nature Geoscience, (2016)
doi:10.1038/ngeo2705.



- Raindrop impaction of soils generates airborne solid organic particles
- Solid organic particles from soils may be a widespread phenomenon in the areas of agricultural lands and grasslands

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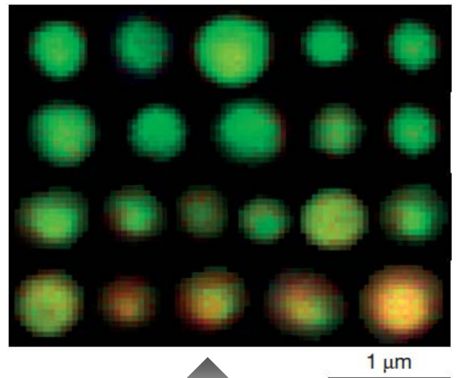
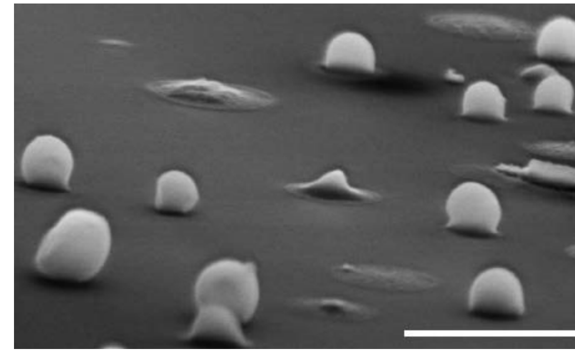
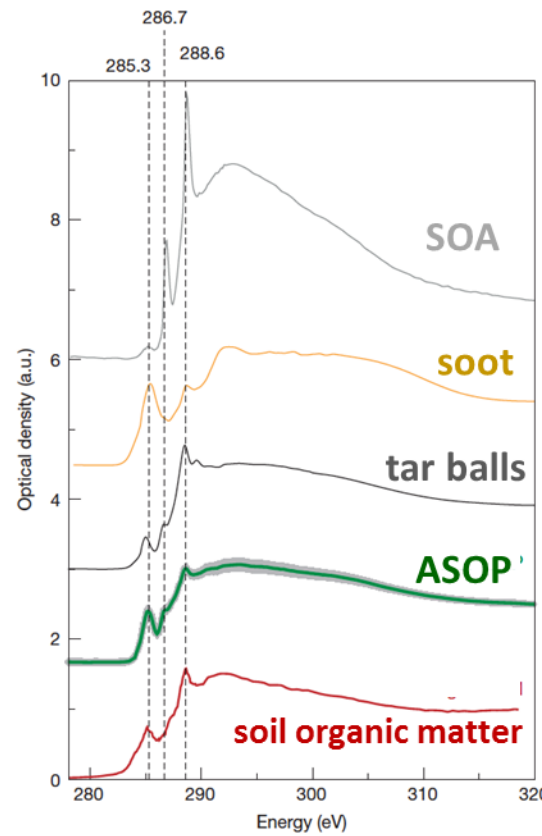




- **Particles are homogeneous mixtures of Elemental Carbon and Organic Carbon in amorphous solid (glassy) phase**
- **Particle composition is soil organic matter**
- **Physico-chemical properties of particles indicate that they have important impacts on cloud formation and efficiently absorb solar radiation**



Poster #17

Anthropogenic - Biogenic (A-B) Interactions Observed in CARES

Questions: Are high OA events uniquely associated with:

High concentrations of A and B? **Yes**[1]

A synergistic interaction leading to extra OA from A and B?

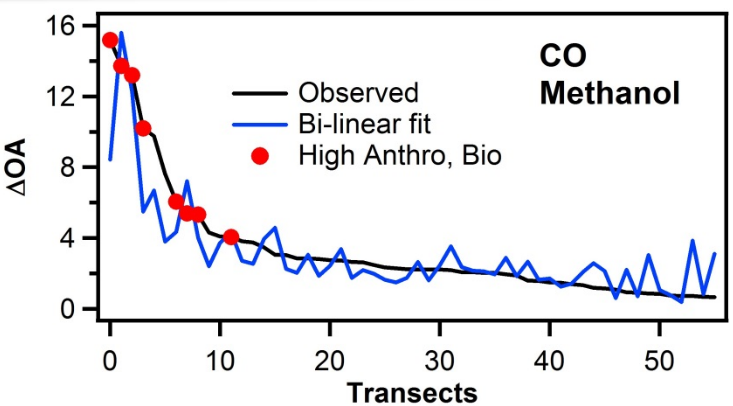
If Yes to 2nd bullet, then A emissions can have a disproportionate influence in forming OA from larger pool of B emissions.

Found[2]: CH_3OH is a better tracer of B than short-lived isoprene, MVK+MACR[3]

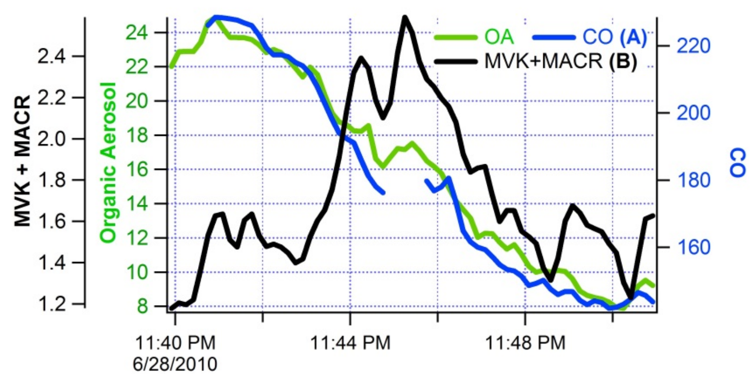
Plume OA best correlated with CO. Background OA best correlated with CH_3OH

High OA only occurred with high A and high B in large part due to a high temperature pollution episode, in which OA responded linearly to A and B.

Bi-linear model (no A-B) captures high OA events



Highest OA Transect.

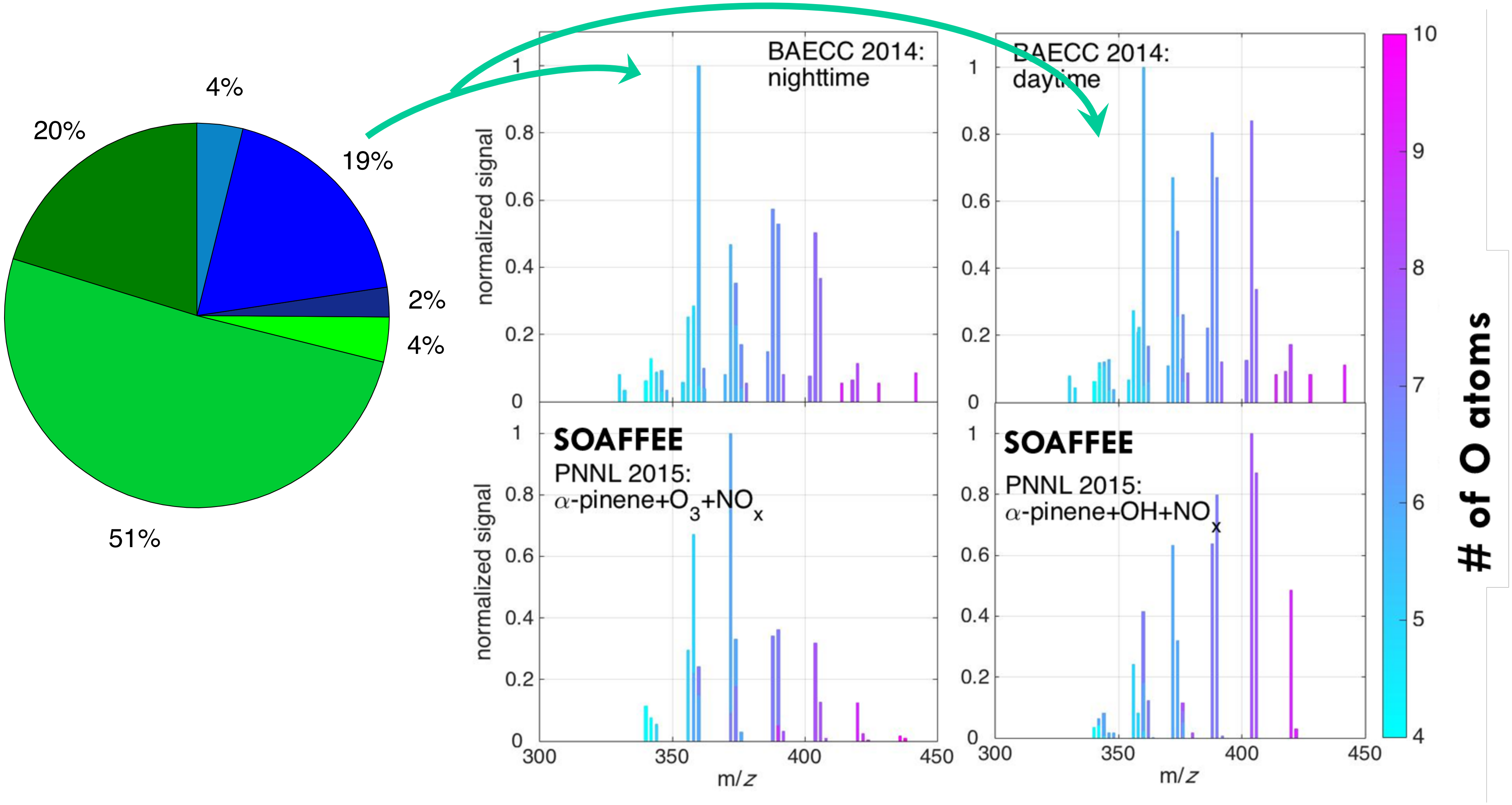


r^2 OA-A=0.99

r^2 OA-B = 0.00

[1] Setyan et al ACP; 2012; Shilling et al ACP 2013 [2] Kleinman et al ACP 2016 [3] suggested by J. Shilling

Monoterpene nitrates → 20% of SOA



SOAFFEE results used to support hypotheses in Lee et al PNAS 2016, Lopez-Hilfiker et al ES&T 2016, plus others in preparation

Prediction of cloud condensation nucleus activity for organic compounds using functional group contribution methods

Approach:

Implemented functional group contribution models to estimates rational activity coefficients and molar volume.

Evaluated the model against a experimentally determined sensitivity of functional groups on CCN activity.

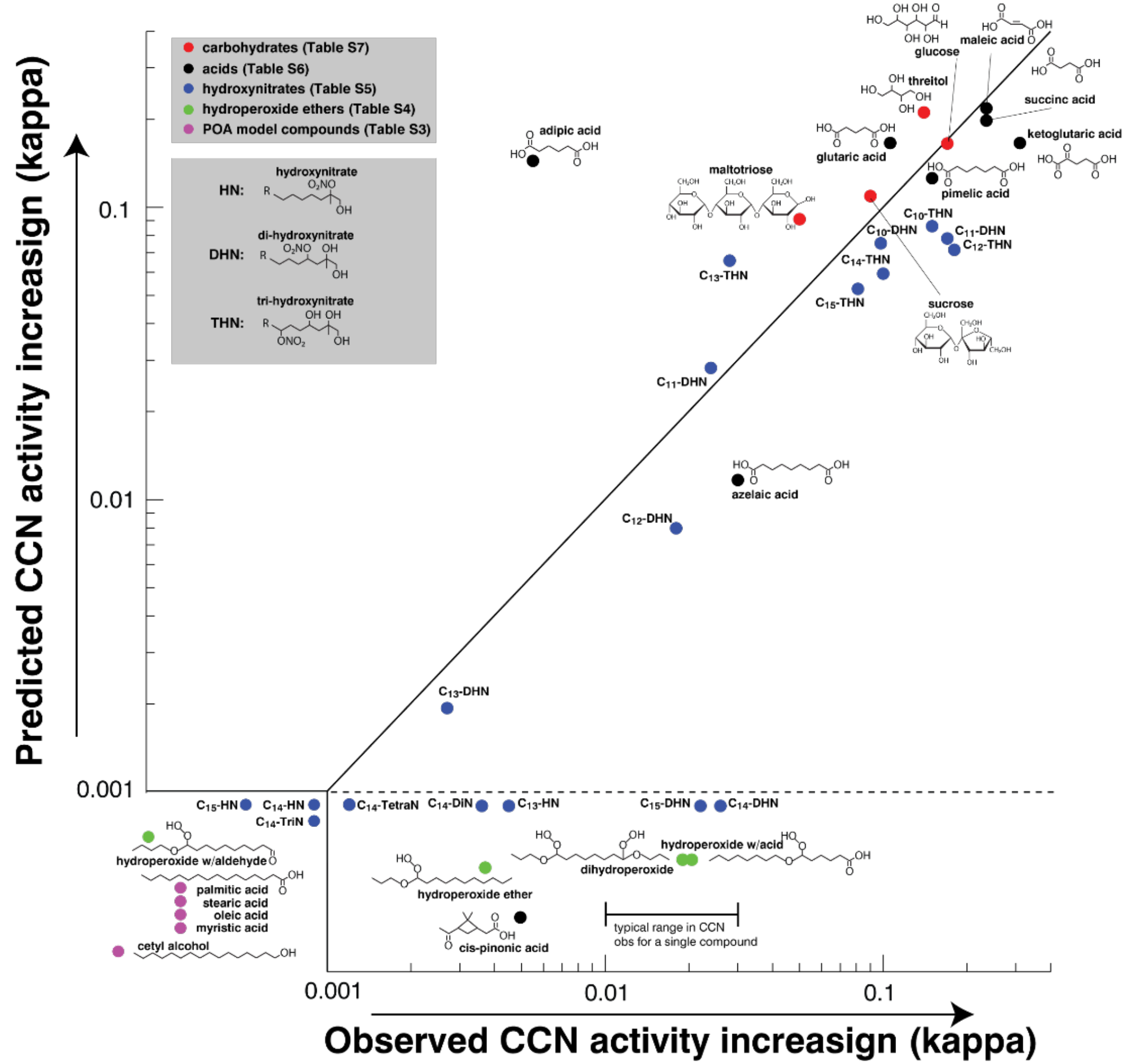
Assigned numerical value to relative effectiveness of functional group to predict CCN activity.

Results:

Numerical model prediction of an organic compounds' contribution to the CCN activity of a particle.

Significance and Impact:

Enable mechanistic modeling of the effect of organic aerosol chemical transformations on cloud properties.



Work was performed at NC State University

NC STATE UNIVERSITY
Colorado State University
University of Colorado Boulder

Petters, M.D. , S. M. Kreidenweis, and P. J. Ziemann. 2016. *Geoscientific Model Dev 9*, 111-124, doi:10.5194/gmd-9-111-2016

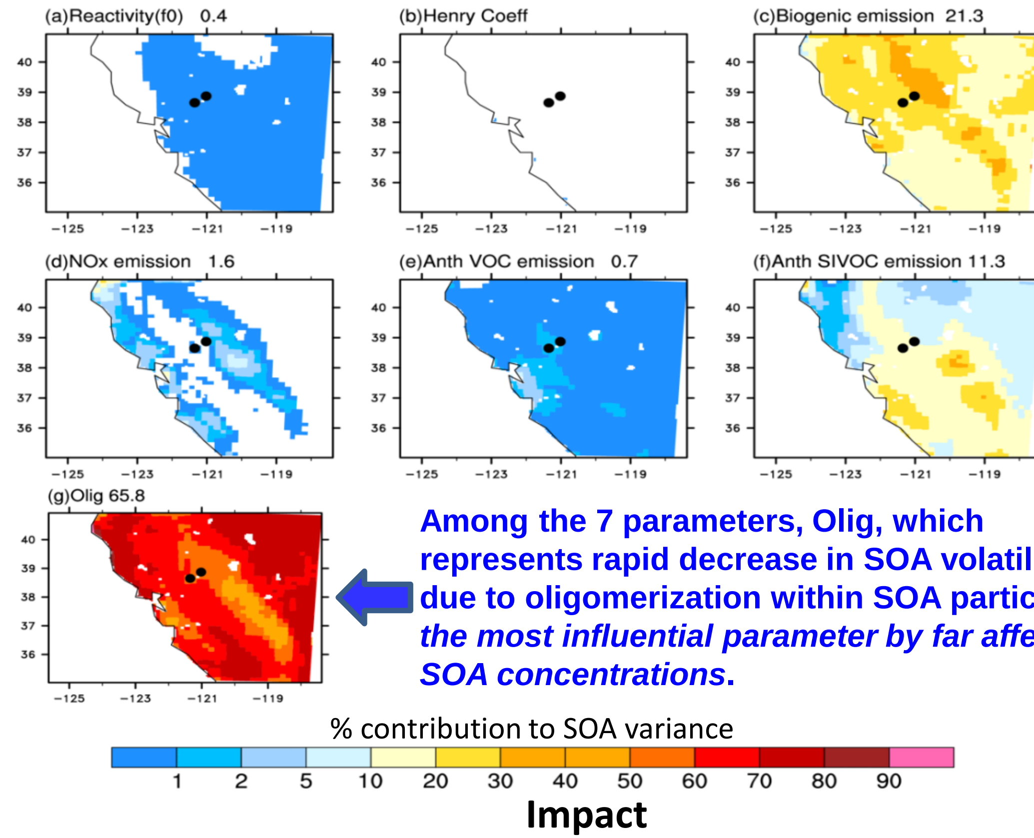
Sensitivity Analysis of SOA Loadings during CARES 2010

Objective

- Investigate the sensitivity of simulated secondary organic aerosols (SOA) to seven selected model parameters during CARES 2010 field study

Approach

- Performed a 250-member ensemble of simulations using the Weather Research and Forecasting Model coupled to chemistry (WRF-Chem)
- Varied all seven parameters simultaneously and used a quasi-Monte Carlo sampling approach to sample the high-dimensional parameter space
- Used a Generalized Linear Model (GLM) to estimate individual parameter and parameter interaction contributions to SOA variance, and their statistical significances.

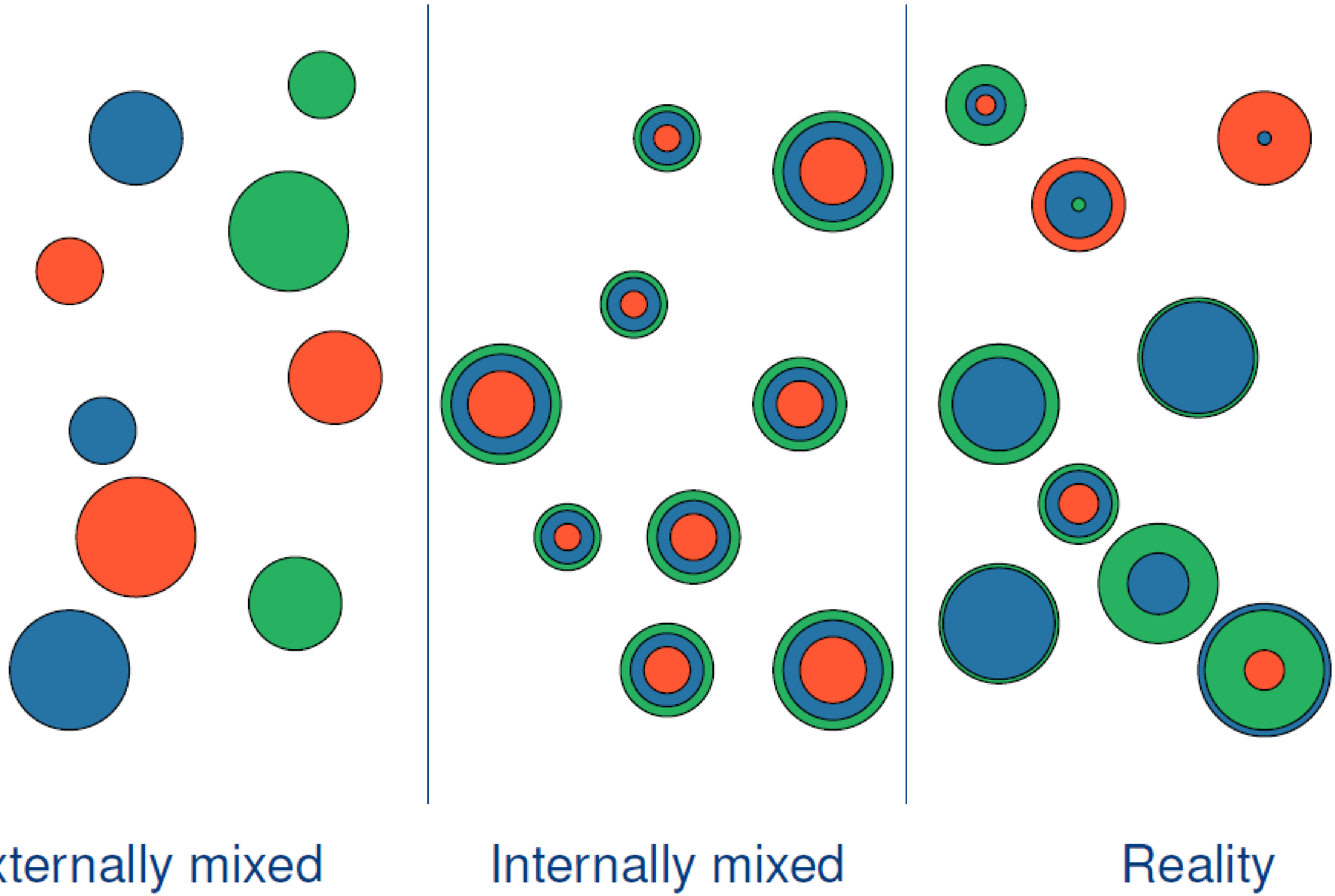


Among the 7 parameters, Olig, which represents rapid decrease in SOA volatility due to oligomerization within SOA particles, *is the most influential parameter by far affecting SOA concentrations*.

- Rapid volatility decrease due to chemistry within SOA particles is shown to be important, but neglected by most models
- Particle-phase chemistry is also shown to change the dominant source types of SOA: biogenic versus anthropogenic
- This statistical approach is promising for ranking the most important processes/parameters affecting SOA

Shrivastava M, C Zhao, RC Easter, Y Qian, A Zelenyuk, JD Fast, Y Liu, Q Zhang, and A Guenther. "Sensitivity Analysis of Simulated SOA Loadings using a Variance-Based Statistical Approach." *Journal of Advances in Modeling Earth Systems*. DOI: 101002/2015MS000554, 2016.

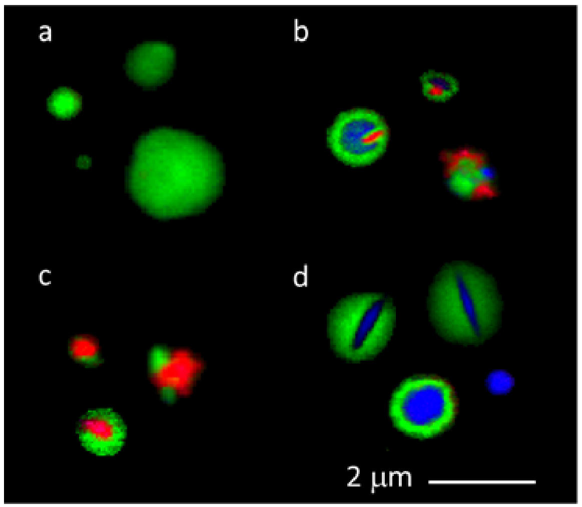
Describing the aerosol mixing state



Courtesy of Nicole Riemer

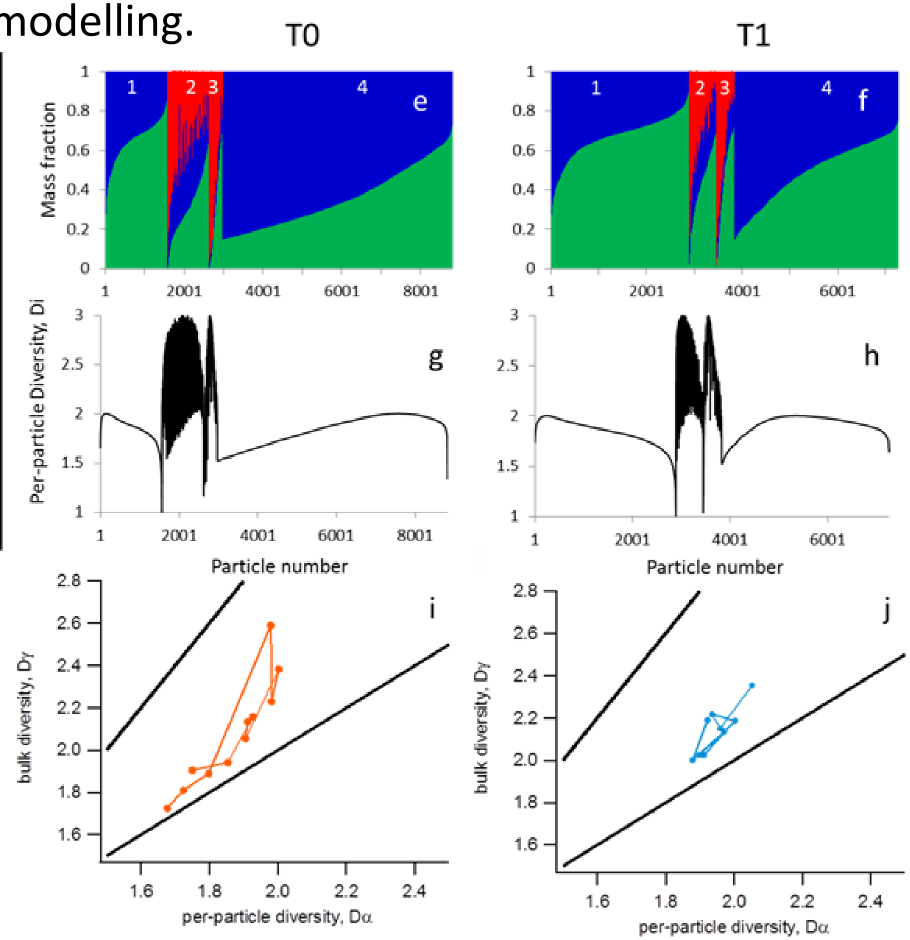
Constraining Mixing State using Entropy Metrics and Spectromicroscopy

- **Objective:** To quantify the range of mixing states during CARES.
- Spectromicroscopy is used to obtained single particle chemical composition
- **Impact:** STXM and SEM provided carbon based and high Z mixing states respectively providing constraints for process modelling.



Organic (OC)
Inorganic (IN)
Soot/Elemental Carbon (EC)

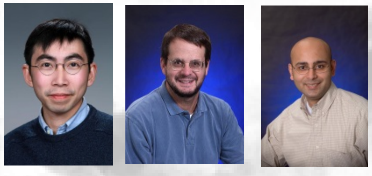
Type 1: OC
Type 2: OCBCIN
Type 3: OCBC
Type 4: OCIN



O'Brien, R. E., Wang, B., Laskin, A., Riemer, N., West, M., Zhang, Q., Sun, Y., Yu, X.Y., Apert, P.A., Knopf, D.A., Gilles, M. K., and **Moffet, R.C.** (2015), J. Geophys. Res. Atmos., *120,*

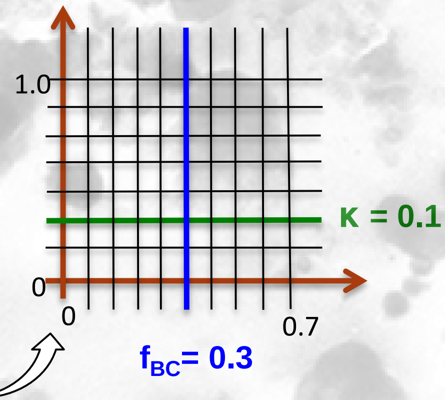
New Black Carbon (BC) Mixing State Model

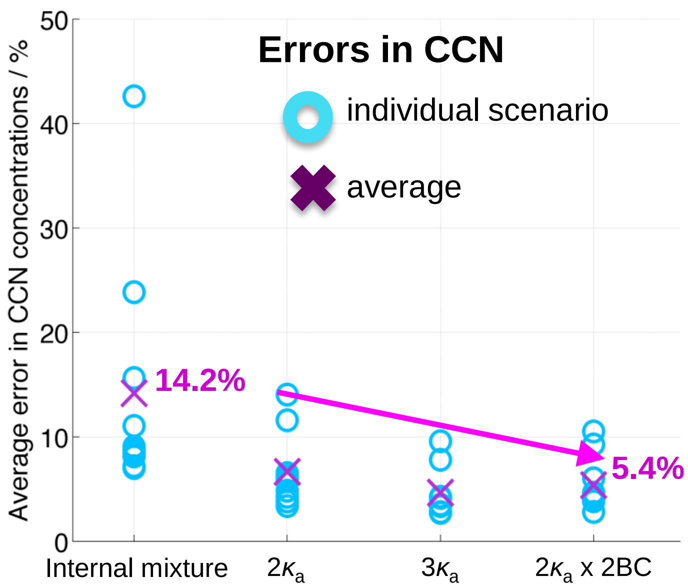
Ching et al. JGR, 2016 *(in revision)*

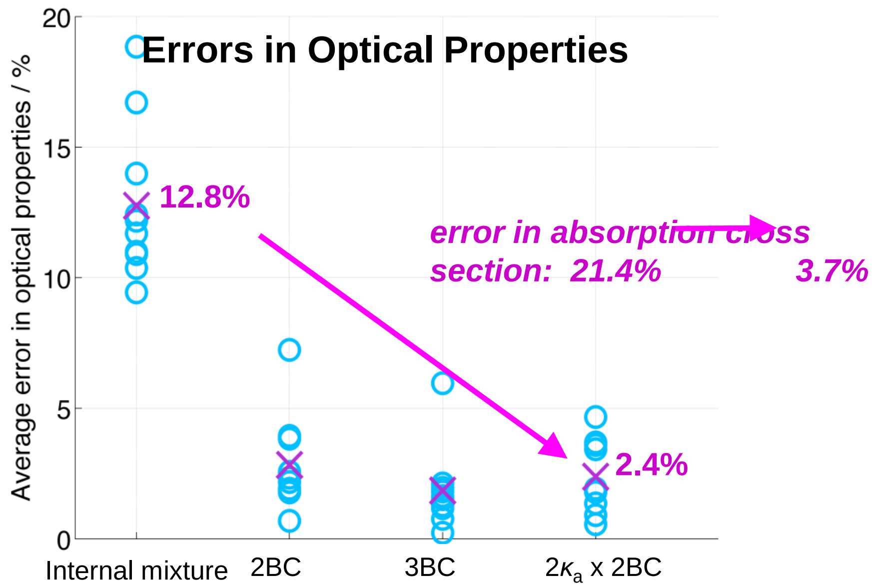


Internal mixture assumption for BC in can lead to large errors in the predicted CCN concentrations and optical properties. A systematic approach is used to develop a novel 3-dimensional mixing state representation for aged BC particles, that is different from "brute force" approaches or ad-hoc choices of bin boundaries.

- 10 scenarios simulated by MOSAIC-Mix
- Over 2000 combinations of f_{BC} and κ boundaries tested
- Optimum f_{BC} and κ bin boundaries located, that give the **minimum overall error**



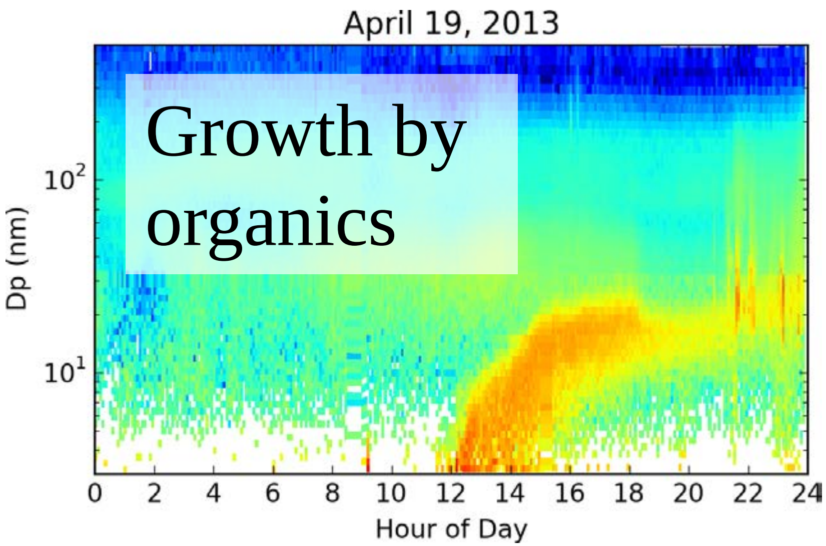


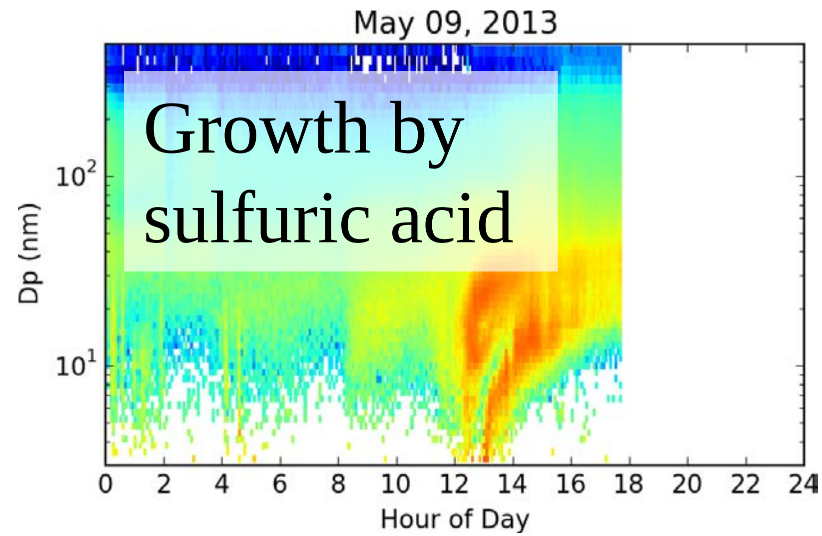


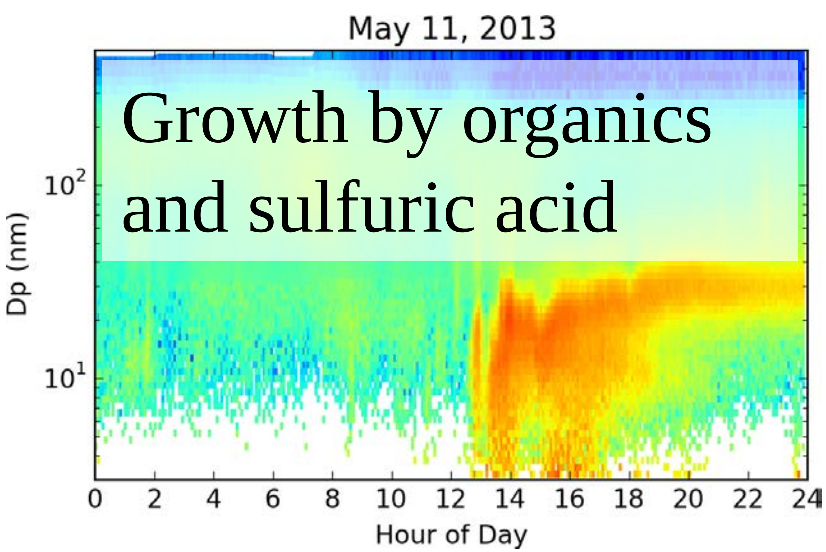
- A simple ***2 f_{BC} x 2 κ bins* achieves smaller total error** (CCN + optical) than either 2 *or 3 f_{BC}* bin-only or 2 *or 3 κ* bin-only configurations.
- In the near future, MOSAIC-Mix will be implemented in WRF-Chem to evaluate its performance using field measurements and assess the impact of BC mixing state on direct and indirect radiative forcing.

Multiple new-particle growth pathways observed at SGP

Hodshire, A. L., et al.:, *Atmos. Chem. Phys. Disc.*, 2016.



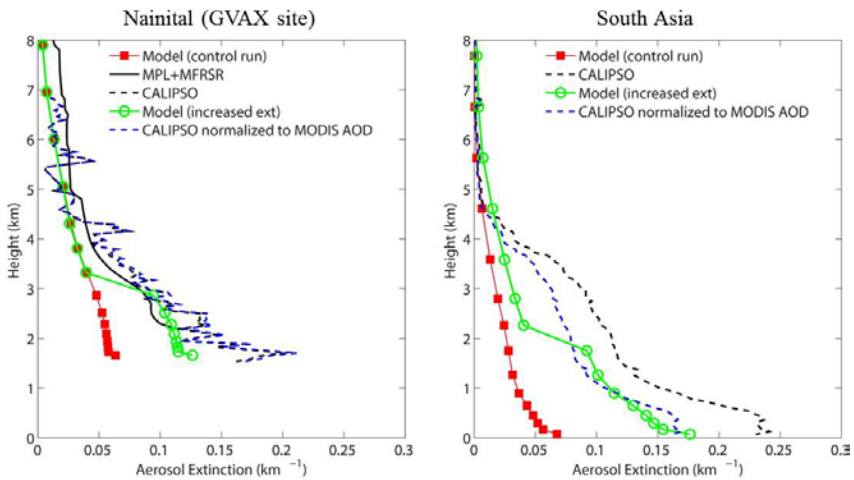




- 3 days with similar growth rates at SGP
- Each day had different growth mechanism
- Determined through 4 state-of-art instruments and modelling

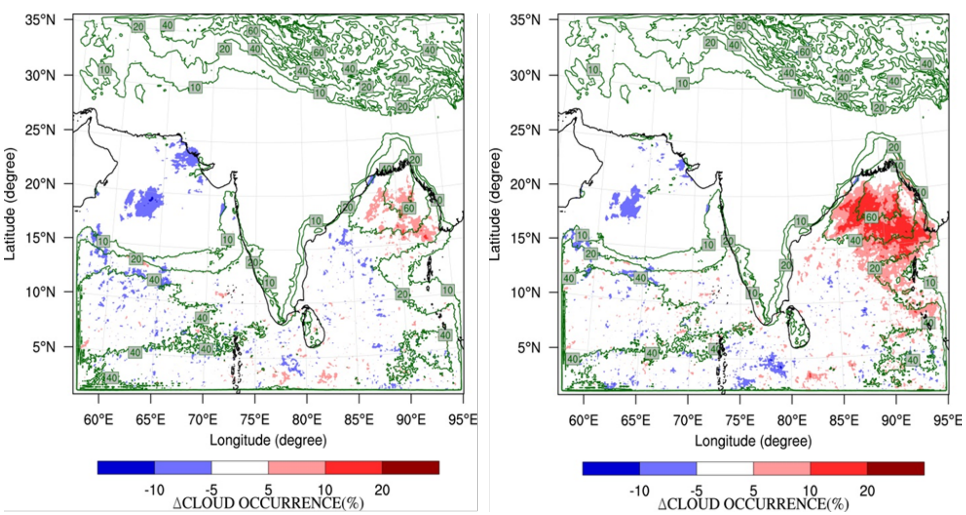
Radiative and thermodynamic responses to uncertainty in aerosol extinction profiles

Journal reference: Feng, Y., V. R. Kotamarthi, R. Coulter, C. Zhao, and M. Cadeddu, Radiative and Thermodynamic Responses to Aerosol Extinction Profiles during the Pre-monsoon Month over South Asia, *Atmos. Chem. Phys.*, 16, 1–18, doi:10.5194/acp-16-1-2016, 2016.



The WRF-Chem (red) underpredicts aerosol extinctions in lower atmosphere compared with the AMF1 MPL (solid black) and satellite remote sensing (dashed blue and black) during GVAX. Doubling the modeled aerosol extinctions below 850 hPa (green) leads to better agreement with observed profiles.

The corresponding percent changes in cloudiness due to aerosols is also shown for the control run (left) and for aerosol profiles adjusted to observations (right). The green contours indicate the predicted cloudiness (%) in the absence of aerosols.



Recent field campaigns/collaborations

Biomass Burn Observation Project (BBOP)

Larry Kleinman, Art Sedlacek et al.

Objective:

- Investigate the **evolution** of chemical, hygroscopic, microphysical, and optical properties of biomass burn aerosols in the near field

Results/ongoing work:

- Collier, S., S. Zhou, T. Onasch, D. Jaffe, L. Kleinman, A. J. Sedlacek, N. Wigder, J. Hee, E. Fortner, J. Shilling, D. Worsnop, R.J. Yokelson, C. Parworth, X. Ge, J. Xu, Z. Butterfield, D. Chand, M.K. Dubey, M. Pekour, S. Springston, and Q. Zhang, Aerosol Emissions Influenced by Wildfire Combustion Efficiency in the Western U.S., ES&T (*Under review*)
- Sedlacek III, A. J., L. I. Kleinman, W. P. Arnott, T. Onasch, S. R. Springston, S. Smith, and S. Oatis, Attribution of Aerosol Light Absorption in Wildfires (*manuscript in preparation*)

Recent field campaigns/collaborations

GoAmazon 2014/5

Scot’s presentation this morning.

Recent field campaigns/collaborations

Biogenic Aerosols - Effects on Clouds and Climate

Tuukka Petäjä et al.

Objective:

- Quantify the importance of new particle formation to a) CCN budget, b) observed cloud properties.

Results/ongoing work:

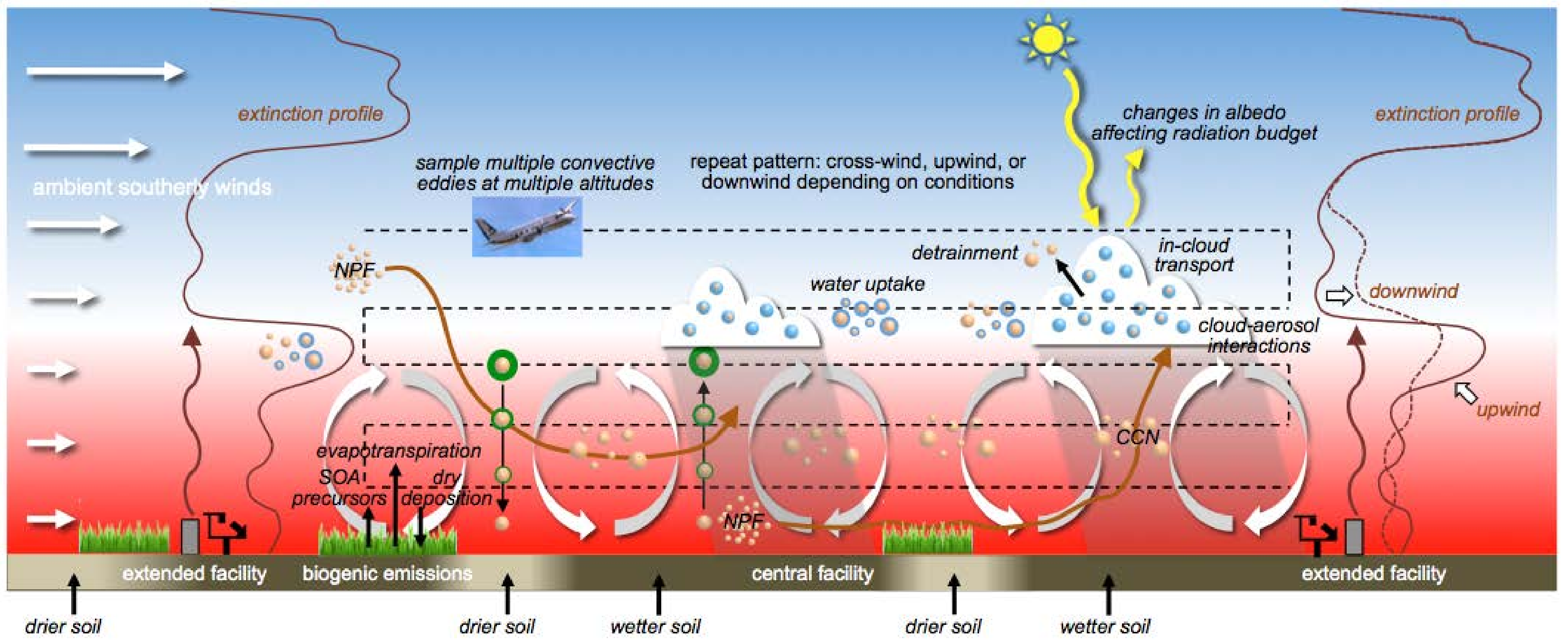
- Petäjä, T. et al., BAECC A field campaign to elucidate the impact of Biogenic Aerosols on Clouds and Climate BAMS (in press)
- Petäjä, T., Tabakova, K., O'Connor, E., Moisseev, D., Sinclair, V. et al. (2016) Influence of secondary aerosol formation on cloud properties in Boreal environment (in preparation).

Holistic Interactions of Shallow Clouds, Aerosols, and Land-Ecosystems (HI-SCALE)

Phase 1: April 24 – May 20, Phase 2: August 28 – September 23

Jerome Fast and Larry Berg et al.

Approach: Couple G-1 aircraft measurements and supplemental surface measurements with the SGP "mega-site" data to quantify the influence of inhomogeneities in land use, vegetation, soil moisture, convective eddies, and aerosol properties on the evolution of shallow clouds as well as the feedbacks of cloud radiative effects on heat, moisture, and momentum fluxes and on aerosol photochemical processes.



Science Questions: Aerosol

- What controls **particle growth** from ultrafine to accumulation mode size?
- How do **new particle formation**, **secondary organic aerosol** formation, and aerosol growth contribute to **CCN concentration**? Do variations in aerosol microphysical properties in the boundary layer contribute to gradients in CCN?
- What is the relative contribution of **anthropogenic, biogenic, and biomass burning** sources of aerosols from both local sources and long-range transport and their effect on cloud properties over the year?
- Can **Large Eddy Simulations** (LES) adequately capture the observed temporal and spatial variability of surface fluxes, boundary layer mixing, aerosol and CCN properties, cloud-aerosol interactions, and cloud properties over the SGP site?
- How can the aircraft data coupled with LES modeling and routine ARM measurements be used to develop **new parameterizations** of sub-grid scale variability associated with boundary layer turbulence and shallow clouds?

coupled with cloud lifecycle and land-atmosphere-cloud interactions

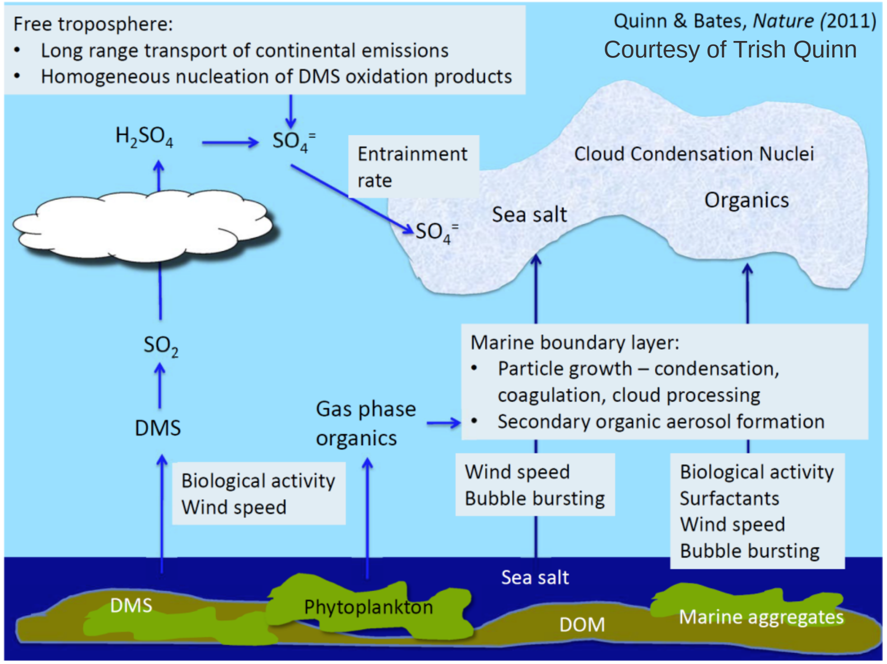
Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA)

Jian Wang, Xiqun Dong, Rob Wood et al.,

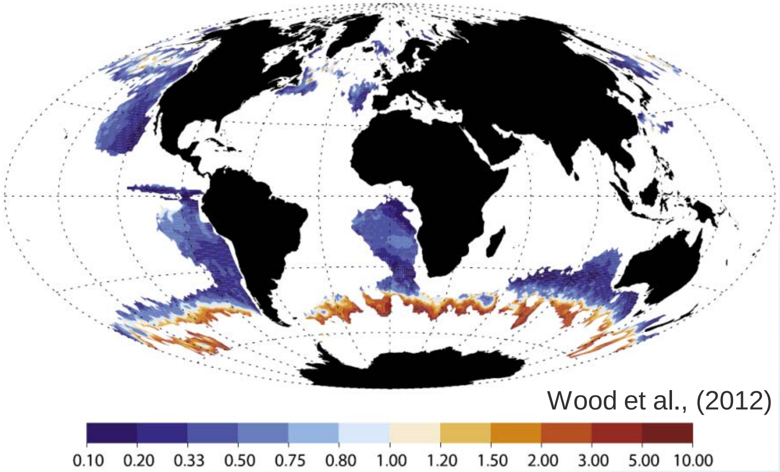
1st IOP (summer): 80 flight hours, June 15-July 25, 2017

2nd IOP (winter): 80 flight hours, January 11 – February 20, 2018

Budget of Aerosol and CCN in Marine Boundary Layer



In the subtropics and tropics, the majority of the CCN likely originate from the FT



Ratio of CCN flux from surface to that due to FT entrainment

- What are the contributions from different sources, including sea spray aerosol, long-range transport, and new particle formation? What are the **seasonal variations of the characteristics and contributions of various sources**?
- How does **removal of CCN by droplet coalescence** control the CCN population in the MBL **for representative cloud regimes**?