Aerosol Life Cycle Working Group

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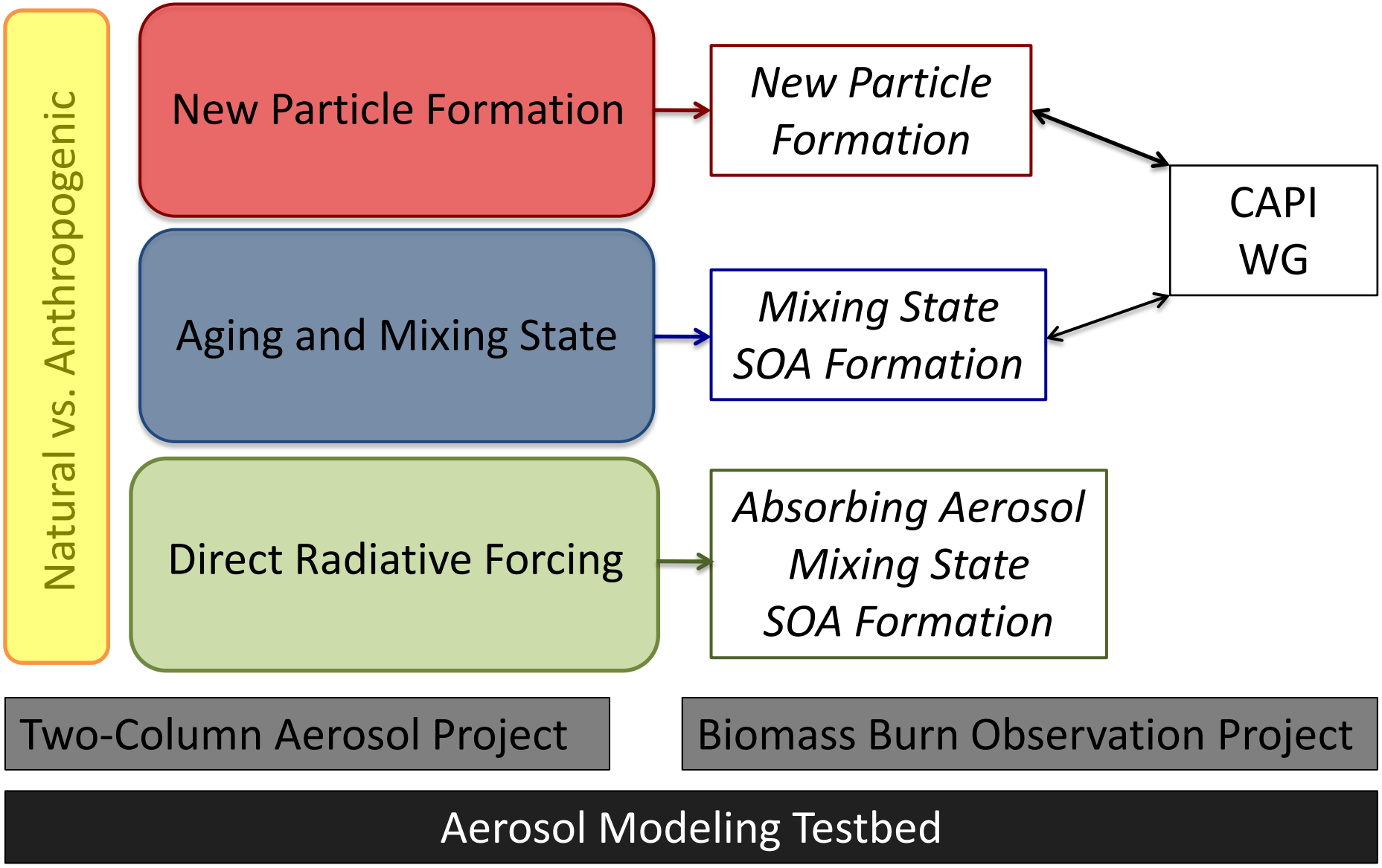
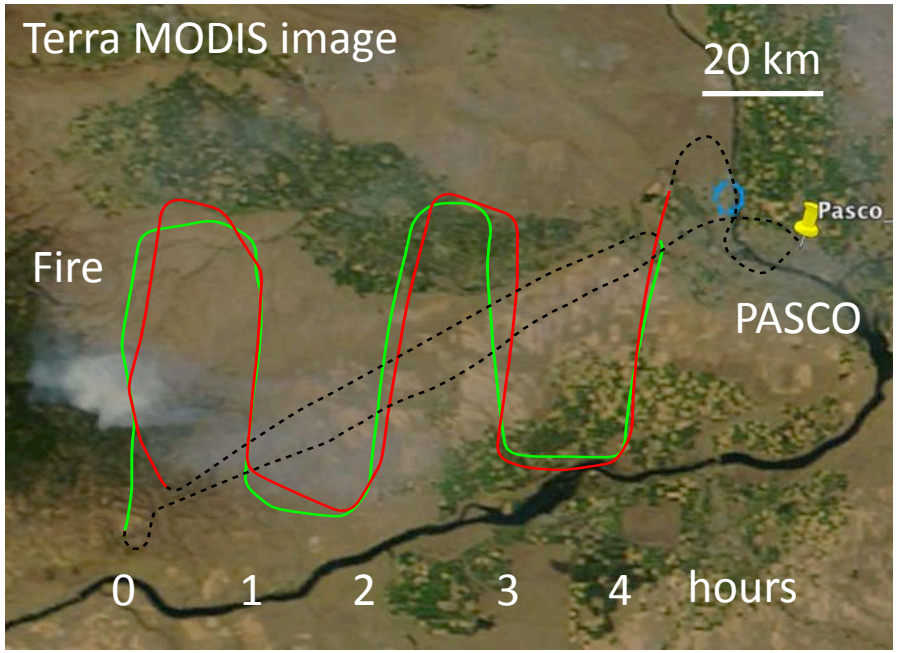
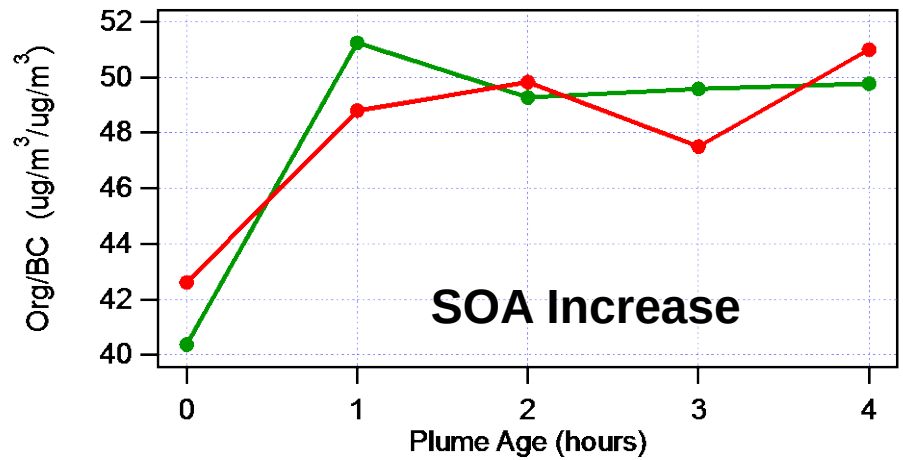


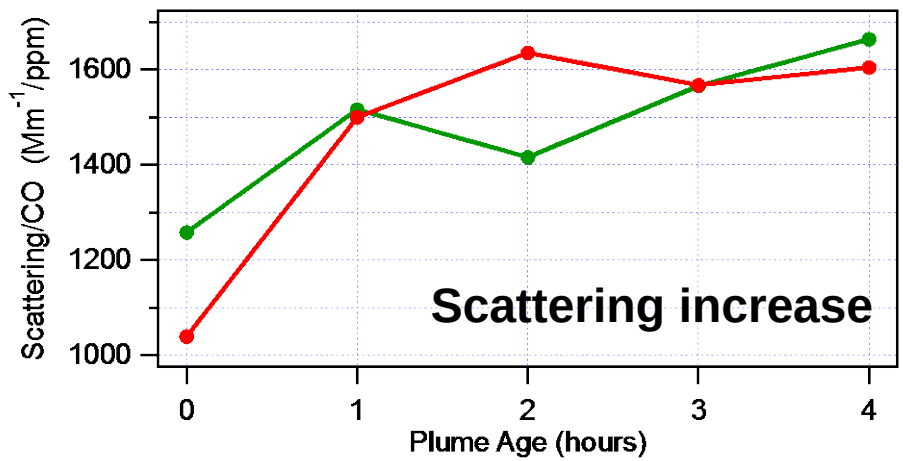
BBOP: Near-Field Evolution of Smoke Aerosol Properties

"28 Mile Marker" Fire sampled at source and 1, 2, 3, 4 hour downwind

Targeted 07-26-2013







Repeatable

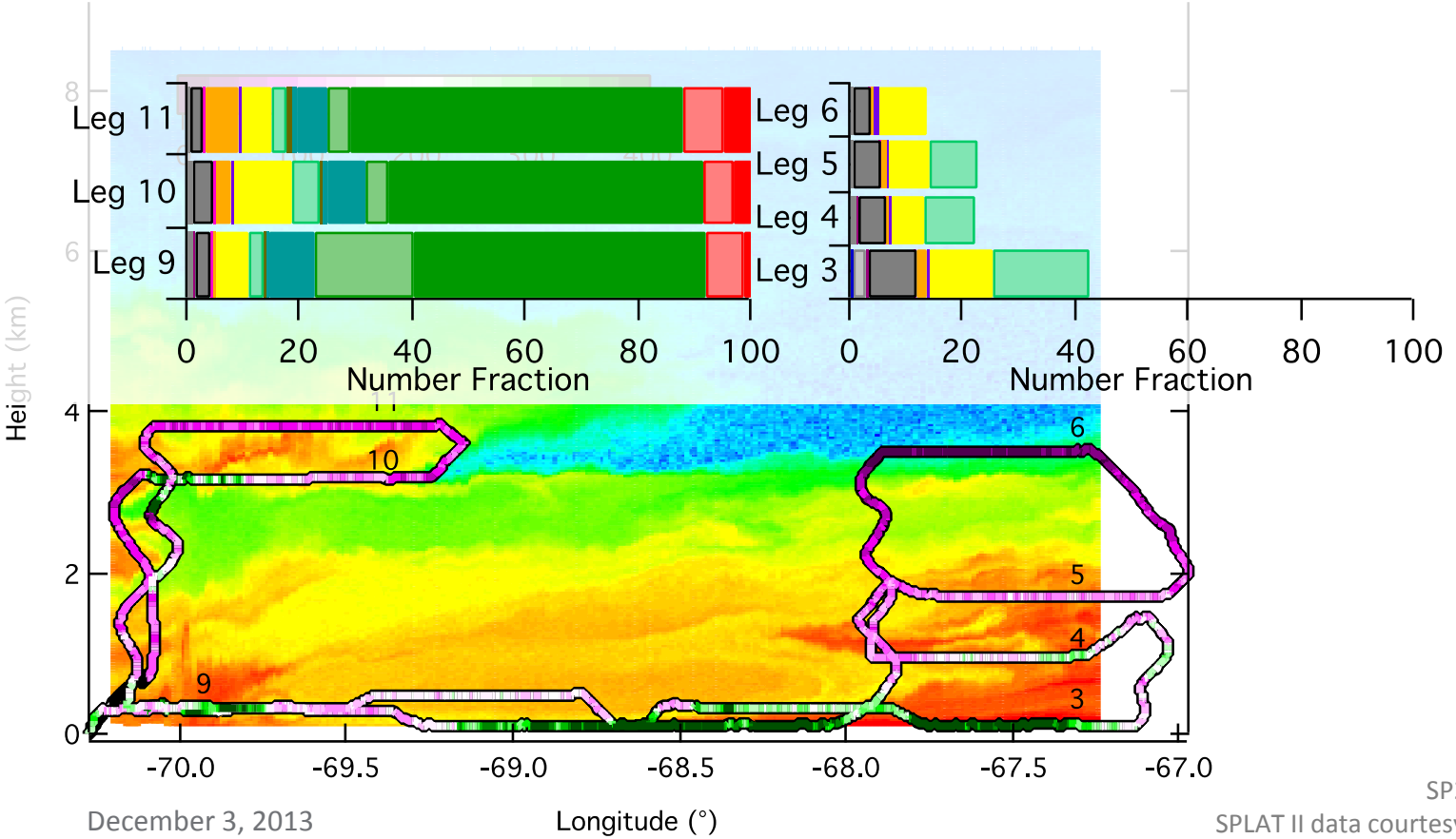
Rapid increase within 1st hour in SOA (25%) and scattering (50%)

Soot Particle-AMS, Onasch

TCAP

Aerosol Mixing State

Relevant measurements: Chemical composition [SPLAT II, SP2, MIT CCN-PCVI-AMS (Phase 2 only)]

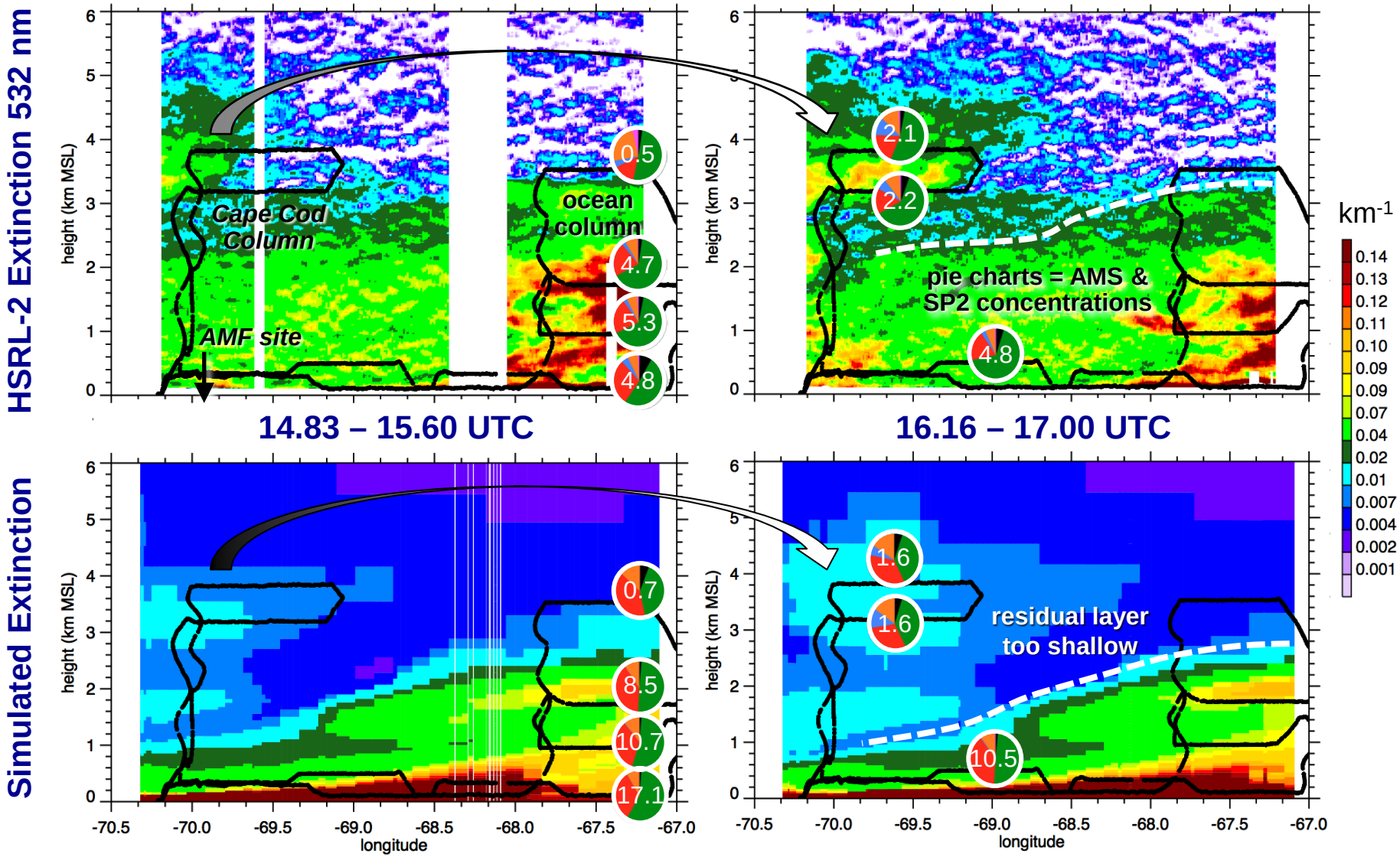


December 3, 2013

SP2 data courtesy of A. Sedlacek
SPLAT II data courtesy of A. Zelenyuk and J. Wilson
HSRL-2 data courtesy of R. Ferrare, C. Hostetler et al.

Aerosol Modeling Testbed at TCAP

TCAP: B-200 and G-1 July 17 Flight



Mixing State Focus Group

- **Population mixing state:** the distribution of chemical compounds across the particle population,
- **Morphological mixing state**: the distribution of chemical compounds within and on the surface of each particle.

Scientific Questions

- Q1: What is the impact of mixing state on the climate-relevant properties of aerosol particles?
- Q2: What mixing state information should be included in models that quantify aerosol climate impacts?
- Q3: What mixing state information should be measured in the field and in the lab?
- Q4: How can we connect measurements (lab and field) to each other and to modeled mixing state information?

Mixing State Focus Group
Overview of Deliverables

funded

D1: Mixing state metrics from microscopy
Moffet, Gilles, Laskin, Sellon

D2: PartMC-3D and error quantification
West, Riemer

D3: Error bounds for mixing state modeling in GCM
McGraw

D4: Development of MOSAIC-mix
Zaveri, Easter, Fast

D5: Characterize single-particle morphology in different environments
Subramanian, Mazzoleni

D6: Chamber experiments and PR modeling of Soot Aging
Shilling, Zaveri, Zelenyuk, Sedlacek,

D7: Biomass burning mixing state analysis during GVAX with WRF-Chem
Feng, Kotamarthi

proposed

D8: Validating PR model simulations with observations from CARES and TCAP
Riemer, Fast, Zaveri, West

D9: Improve models of surface tension to improve representation of morphology
Anthony Wexler, Simon Clegg, Cari Dutcher

D10: Chamber experiments and PR modeling to study mixing state
Davidovits, Lambe, Lewis, Onasch, Sedlacek

D11: Including morphology information in PartMC-3D for improved radiative forcing estimates
Scarnato, Mazzoleni, Riemer

planned

D12: Single-particle instrument intercomparison
TBD

D13: Mixing state modeling obstacle course
Riemer, West

D14: Mixing state analysis and modeling during BBOP
Adachi, Buseck, Onasch, Sedlacek

D15: BC/BrC Mixing state as a function of age: Laboratory (FLAME, BC) to Field (Clearflo, CARES, BBOP?) for Model
Dubey, Mazzoleni, Cappa, Aiken, Donahue, Zaveri, Fast, Feng

Table 3: Readiness of Tools

	Population mixing state		Morphological mixing state	
	Readiness level	Deliverables	Readiness level	Deliverables
Theory/Metrics	high	D1, D6, D9	low	D9
PRM	high	D2, D6, D8, D10, D11, D13	low	D11
RM	low	D4, D7, D13	low	
GCM	low	D3, D4, D13	low	
SP2	medium	D5, D6, D8, D10, D12	medium	D6, D10
Microscopy	medium	D1, D8, D12	high	D6, D5
Single-particle mass spetrometry	high	D8, D12	low	

Table 3: Readiness of tools to represent population mixing state and morphological mixing state, and the deliverables that address each aspect. See Section 5.1 for descriptions of deliverables D1-D13. Despite the fact that many tools by themselves have a high readiness level, the output of these tools is typically not directly comparable (see Table 2). The difficulties in comparison is a primary bottleneck in understanding mixing state. The entries under "readiness level" are color-coded according to high level of readiness (green), medium level or readiness (yellow), and low level of readiness (red). The entries under "deliverables" are color-coded according their status of funding (funded, proposed, planned).

Table 2: Connections between Different Tools

	Theory/ Metrics[1]	PRM[2]	GCM[3]	SP2[4]	Micros-copy[5]	SP mass spectro-metry[6]	Remote sensing[7]	Bulk measure-ments[8]
Theory/ Metrics[1]		high	low	medium	medium	low	low	low
PRM[2]	high		low	medium	low	low	low	high
GCM[3]	low	low		low	low	low	high	medium
SP2[4]	medium	medium	low		low	low	low	high
Micros-copy[5]	medium	low	low	low		low	low	medium
SP mass spetro-metry[6]	low	low	low	low	low		low	medium
Remote sensing[7]	low	low	high	low	low	low		high
Bulk measure-ments[8]	low	high	medium	high	medium	medium	high	

Table 2: Assessment of current abilities to connect data and outputs amongst different tools. The lack of comparable mixing state outputs between many tools is a key bottleneck in our ability to understand mixing state impacts.

Bottleneck:

Lack of comparable mixing state outputs between many tools

ALWG: Absorbing Aerosol Breakout

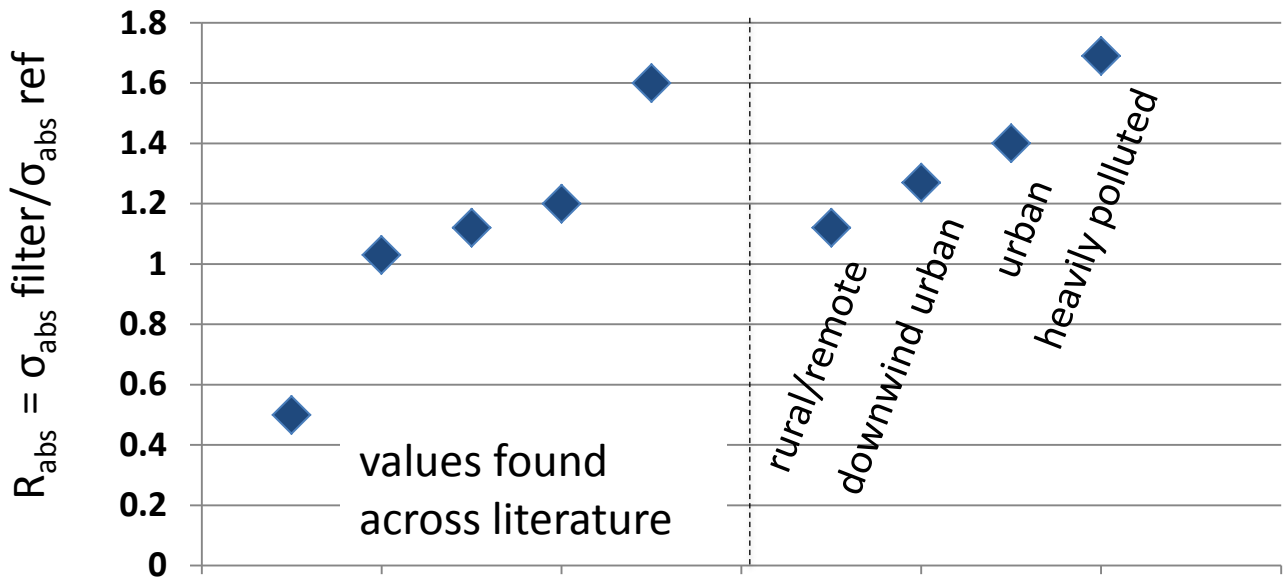
Direct Radiative Forcing by Aerosol

$$F = \tau \times AFE$$

$$AFE = f(\omega, g)$$

OA mass (µg m^{-3})
- rural/remote = 0-2.5
- downwind urban = 2.5-5.0
- urban = 5.0-12.5
- heavily polluted = >12.5

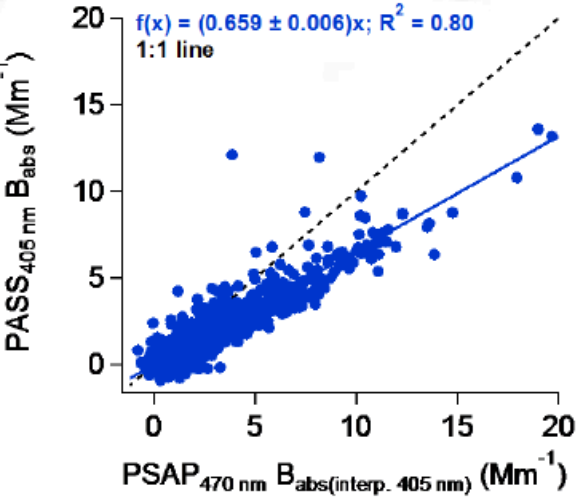
Consistency in measurement approaches is lacking

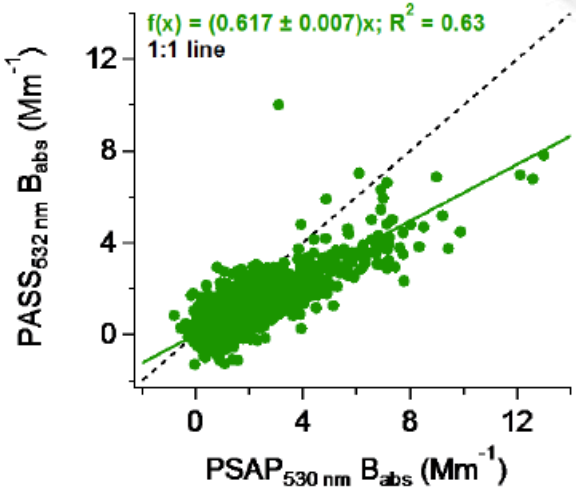


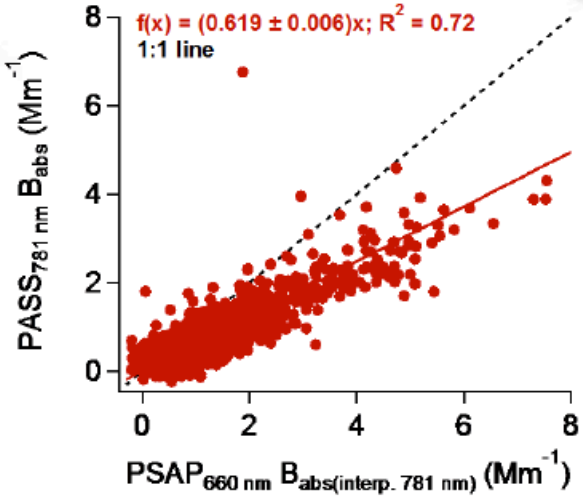
Lack et al. 2008

Absorption: PASS vs PSAP and CLAP

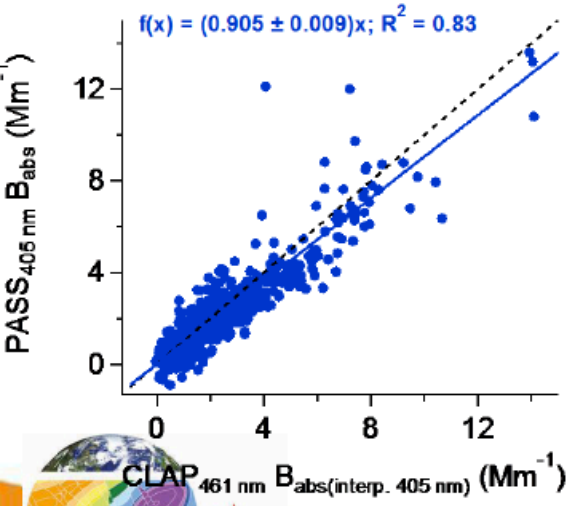
- PSAP

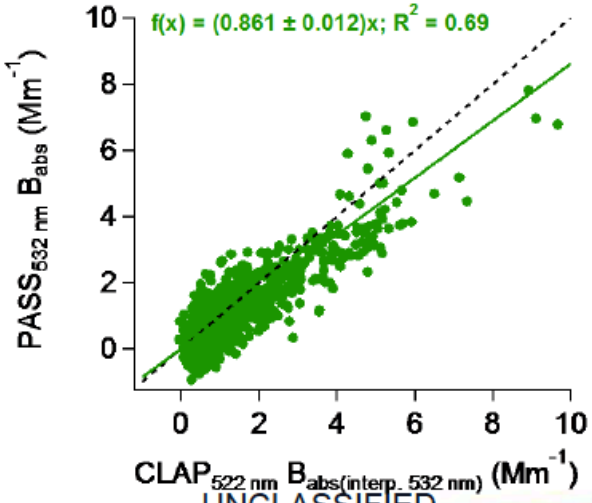


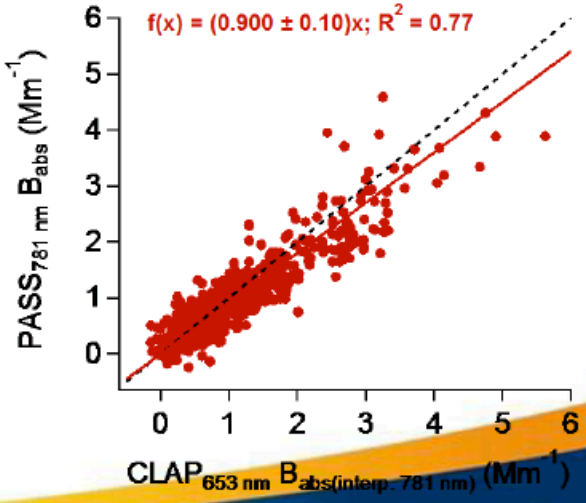




- **CLAP (shorter time period)**







Aerosol Absorption Measurements Inter-Comparison

IOP planning discussion

Objective

Promote better understanding of the advantages and limitations, specifically with respect to measurement uncertainty, of different approaches to in situ aerosol light absorption measurements.

Outcome

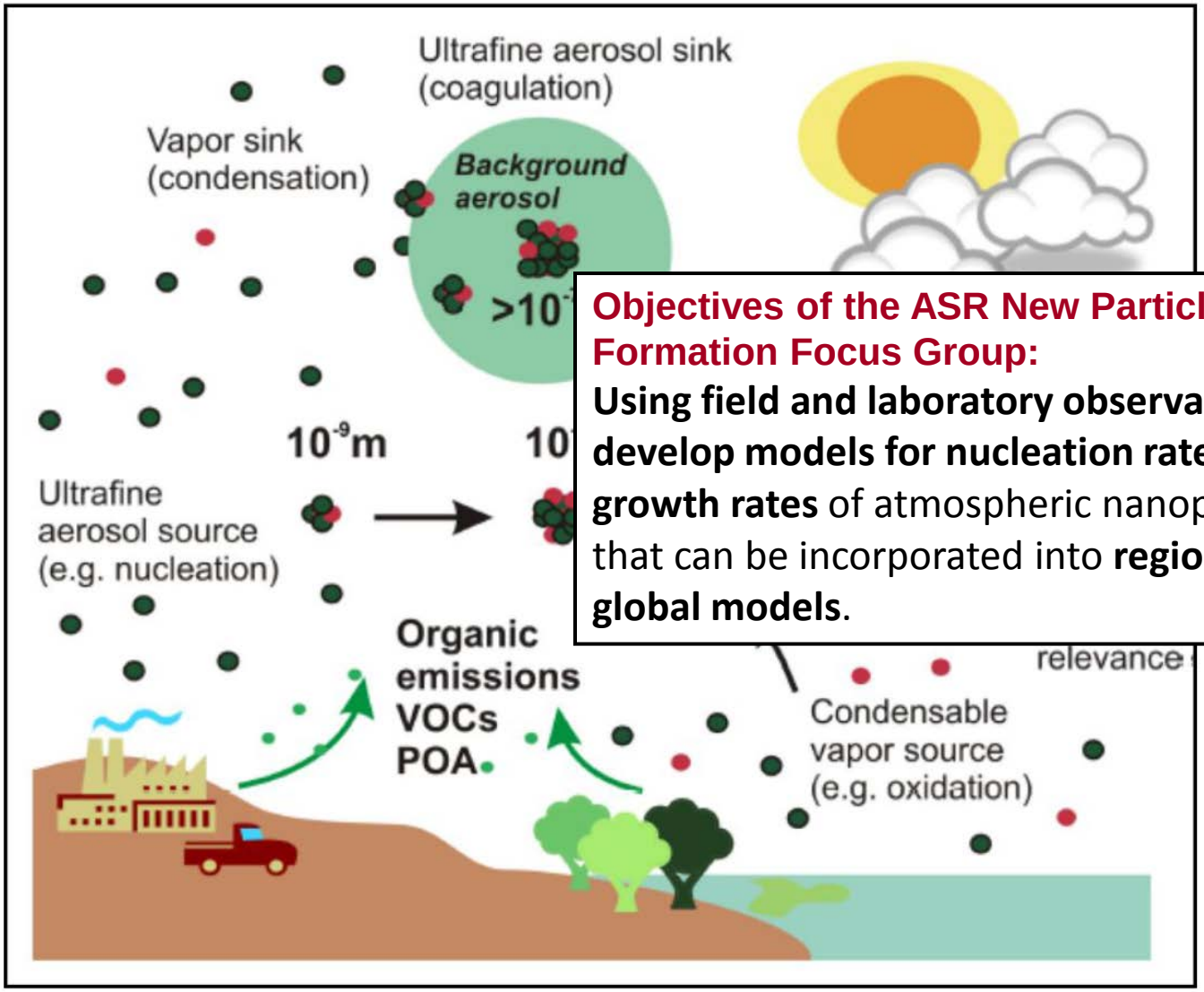
Report accuracy and precision for instruments used to measure aerosol optical properties.

Provide a basis of determination for investment in and deployment of particular instruments for continuous, long-term operation at ARM-like fixed sites.

Goals for this discussion

- Why?
- What?
- Where?

Background: New Particle Formation (Jim Smith and Chongai Kuang)



Objectives of the ASR New Particle Formation Focus Group:

Using field and laboratory observations, develop models for nucleation rates and growth rates of atmospheric nanoparticles that can be incorporated into **regional and global models**.

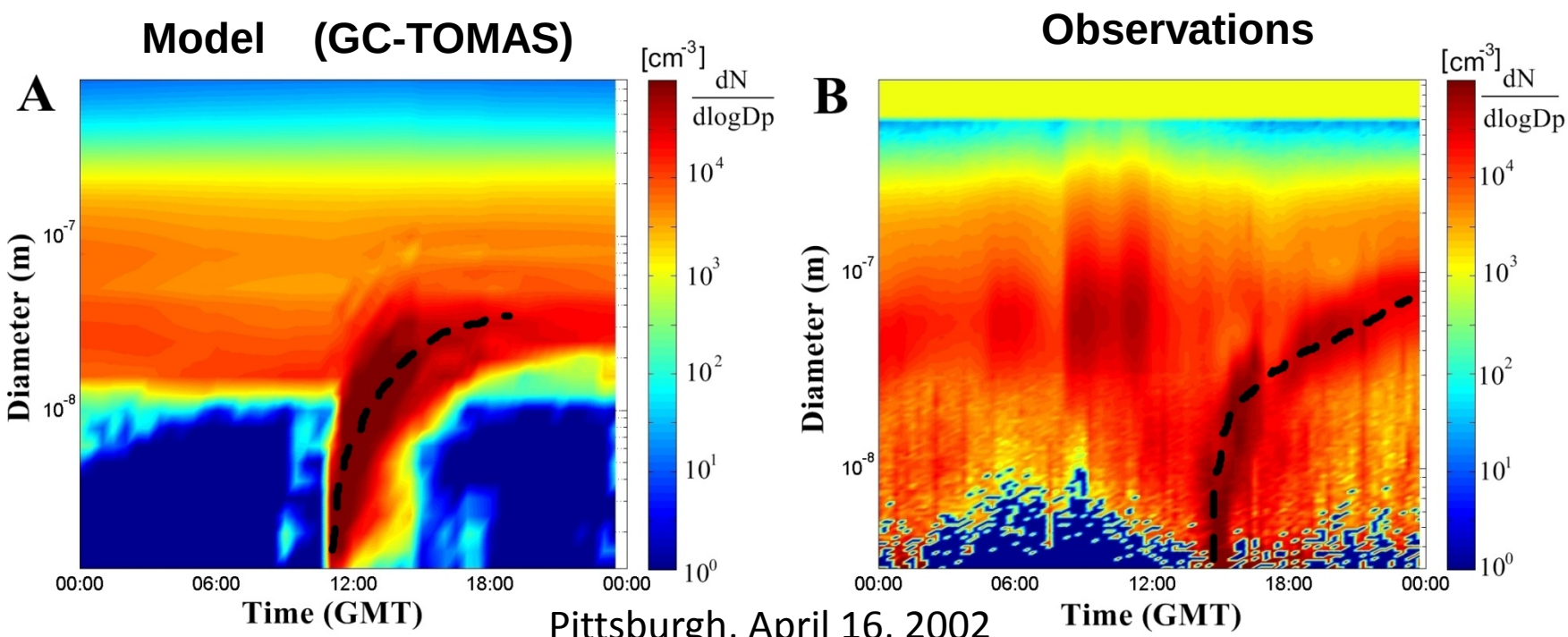
Modeling new particle formation and growth

Several presentations regarding global modeling of new particle formation:

- models (e.g., GEOS-Chem + TOMAS) are ready for mechanisms for nucleation/growth
- currently each model has “shortcuts” in representing growth rates and contribution of organics.
- the rapid time scales for particle formation require changes to models (e.g., MAM + CAM5)

Also progress in process-level modeling of growth:

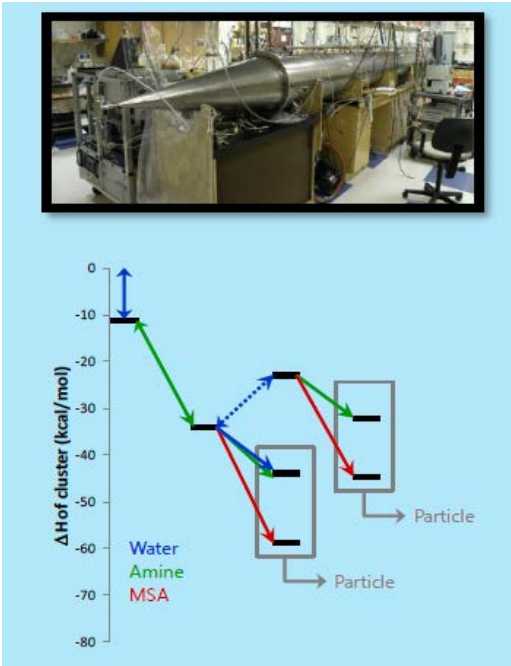
- Initial growth of sub-10 nm particles (which is the most poorly understood process
- growth rates due to reactive and non-reactive uptake and their impacts on observed growth rates.

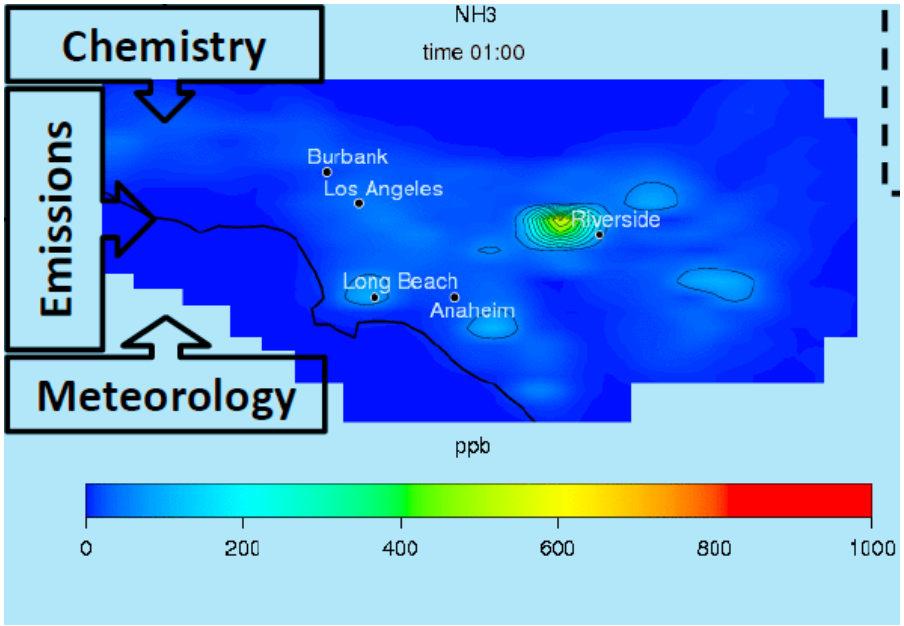


Laboratory studies

The focus of lab studies has been on understanding and modeling nucleation and the first steps in growth (UMN and UC Irvine):

- Recent lab studies include work at UC Irvine to understand and model nucleation in the MSA+amine+H2O system and incorporate into regional model.





lab studies and mechanism … leads to a regional model of the role amines+organosulfates

Field Observations

Recent field studies with observations of new particle formation:

- CARES (Central Valley, CA, 2010)
- NPFS (Southern Great Plains, 2013)
- ALC (Aerosol Life Cycle Study (BNL, 2011)

Planned for 2014:

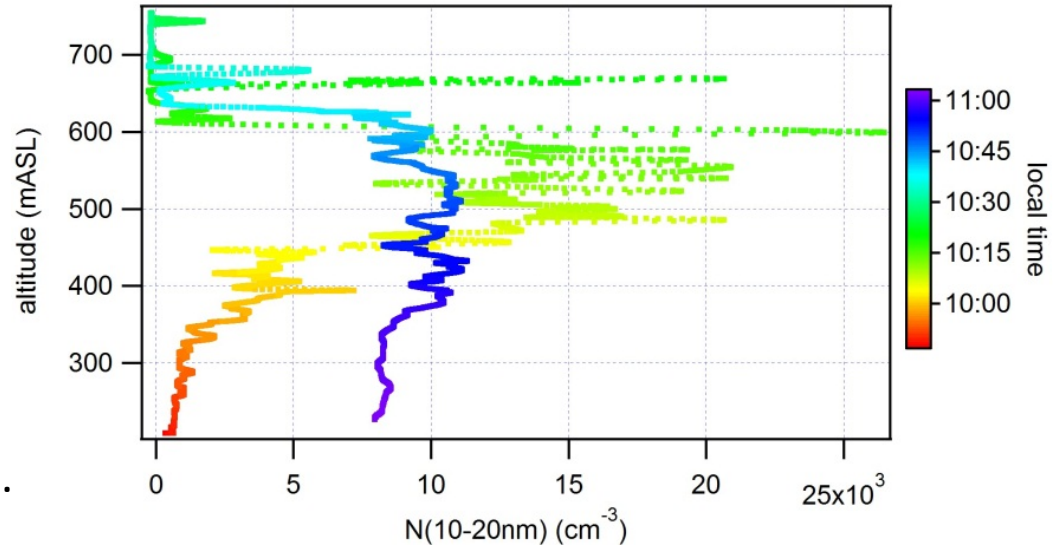
- GoAmazon 2014 IOPs
- BAECC (ARM Mobile Facility deployment, Hyytiälä, Finland Feb – Oct 2014

tethered blimp during NPFS

ARM Long-term observations: Newly procured instrumentation allows extension of existing measurements to quantification of nucleation and growth

- SGP central facility will receive a trace SO2 analyzer and a nano-Scanning Mobility Particle Sizer.
- This will bring new data products to ARM relating to particle formation.

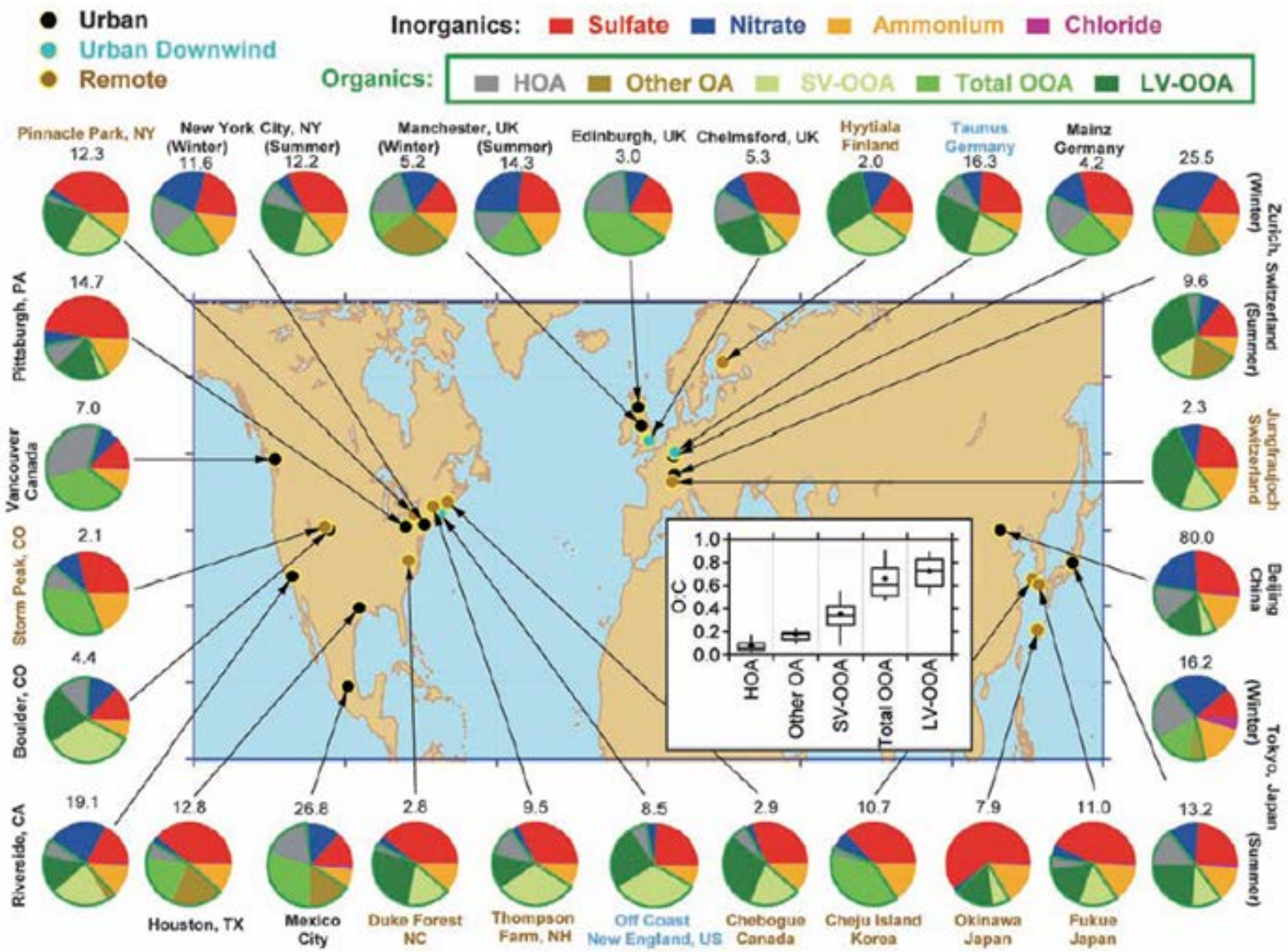
10-20 nm diameter particle concentration profiles



Anthropogenic Influence on Climate-Relevant SOA Properties

Scot Martin and John Shilling

Including mass concentrations, number-diameter distributions



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Atmospheric Chemistry and Physics Discussions
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This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

The direct and indirect radiative effects of biogenic secondary organic aerosol

C. E. Scott[1], A. Rap[1], D. V. Spracklen[1], P. M. Forster[1], K. S. Carslaw[1], G. W. Mann[1,2], K. J. Pringle[1], N. Kivekäs[3], M. Kulmala[4], H. Lihavainen[3], and P. Tunved[5]

[1]School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK
[2]National Centre for Atmospheric Science, University of Leeds, Leeds, LS2 9JT, UK
[3]Finnish Meteorological Institute, Erik Palmenin aukio 1, 00560 Helsinki, Finland
[4]Department of Physics, University of Helsinki, P.O. Box 64, 00014, Finland
[5]Department of Applied Environmental Research, Stockholm University, Svante Arrhenius Väg 8c, 10691 Stockholm, Sweden

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Correspondence to: C. E. Scott (pm08c2s@leeds.ac.uk)

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Abstract

We use a global aerosol microphysics model in combination with an offline radiative transfer model to quantify the radiative effect of biogenic secondary organic aerosol (SOA) in the present day atmosphere. Through its role in particle growth and ageing, the presence of biogenic SOA increases the global annual mean concentration of cloud condensation nuclei (CCN; at 0.2 % supersaturation) by 3.6–21.1 %, depending upon the yield of SOA production, and the nature and treatment of concurrent primary carbonaceous emissions. This increase in CCN causes a rise in global annual mean cloud droplet number concentration (CDNC) of 1.9–5.2 %, and a global mean first aerosol indirect effect (AIE) of between $+0.01\,\mathrm{W\,m^{-2}}$ and $-0.12\,\mathrm{W\,m^{-2}}$. The radiative impact of biogenic SOA is far greater when it also contributes to particle nucleation; using two organically mediated mechanisms for new particle formation we simulate global annual mean AIEs of $-0.22\,\mathrm{W\,m^{-2}}$ and $-0.77\,\mathrm{W\,m^{-2}}$. The inclusion of biogenic SOA substantially improves the simulated seasonal cycle in the concentration of CCN sized particles observed at three forested sites. The best correlation is found when the organically-mediated nucleation mechanisms are applied, suggesting that the AIE of biogenic SOA could be as large as $-0.77\,\mathrm{W\,m^{-2}}$. The radiative impact of SOA is sensitive to the presence of anthropogenic emissions. Lower background aerosol concentrations simulated with anthropogenic emissions from 1750 give rise to a greater fractional CCN increase and a more substantial indirect radiative effect from biogenic SOA. Consequently, the anthropogenic indirect radiative forcing between 1750 and the present day is sensitive to assumptions about the amount and role of biogenic SOA. We also calculate an annual global mean direct radiative effect (DRE) of between $-0.08\,\mathrm{W\,m^{-2}}$ and $-0.78\,\mathrm{W\,m^{-2}}$ in the present day, with uncertainty in the amount of SOA produced from the oxidation of biogenic volatile organic compounds (BVOCs) accounting for most of this range.

ACPD
13, 16961–17019, 2013

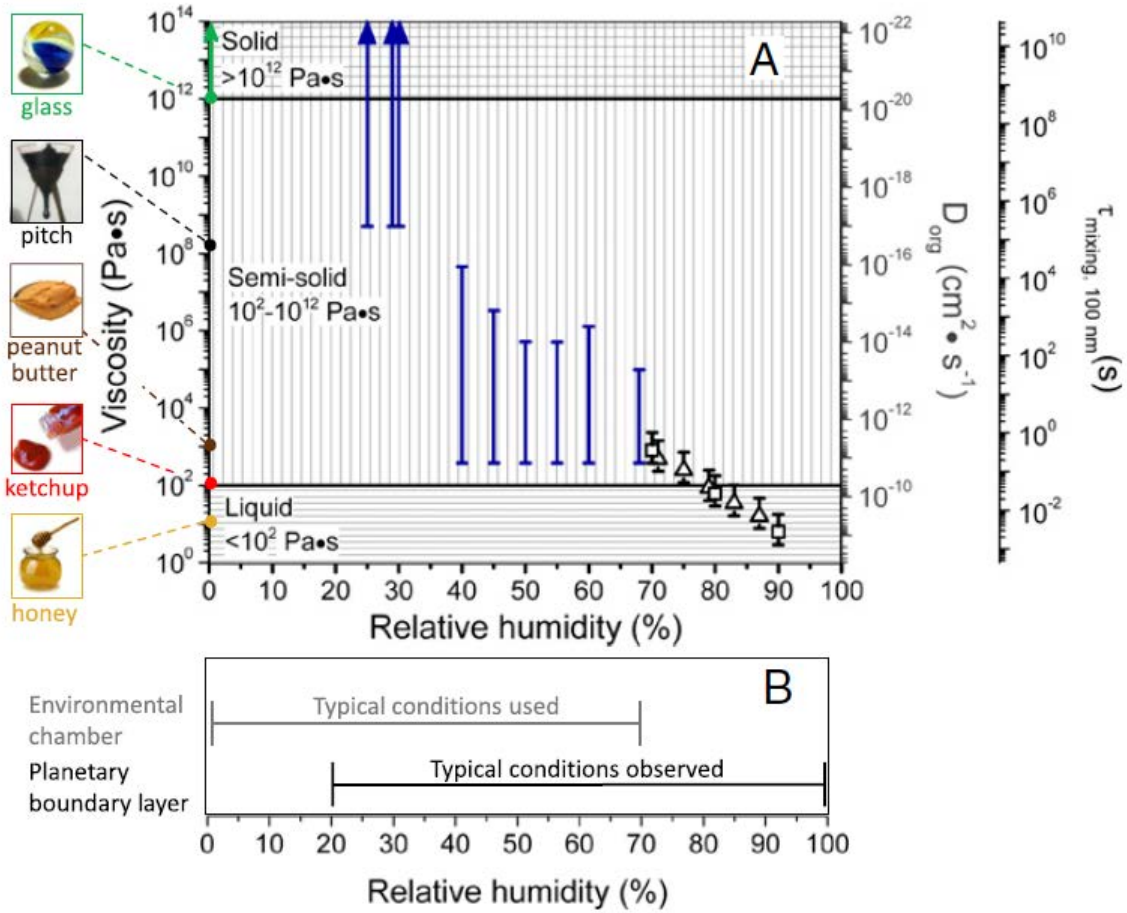
Aerosol Life Cycle Working Group Meeting
Fall 2013
SOA Breakout Session

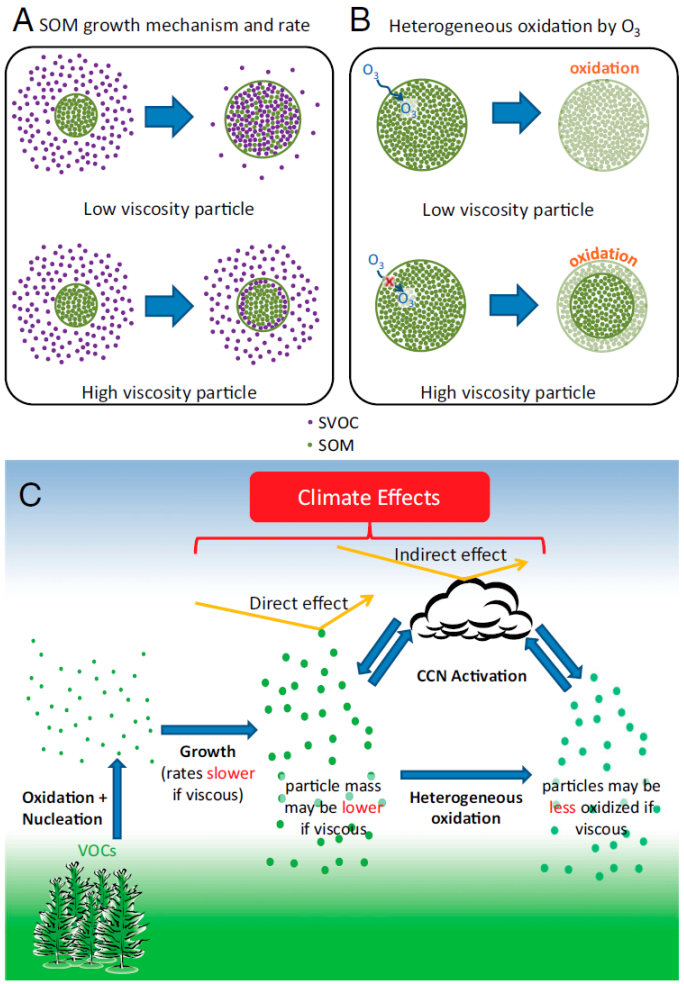
Outcome of Day's Work: Three Intellectual Themes

These group names are still to be polished, i.e., still a working document. Nevertheless, group activity had terrific success in defining its goals around three themes.

1. "viscosity/phase" – (15 participants so far)
2. "growth mechanisms" – (14 participants) (with particle chemistry as an emphasis point)
3. "sulfate as a trigger or regulator for SOA production & properties" – (12 participants)

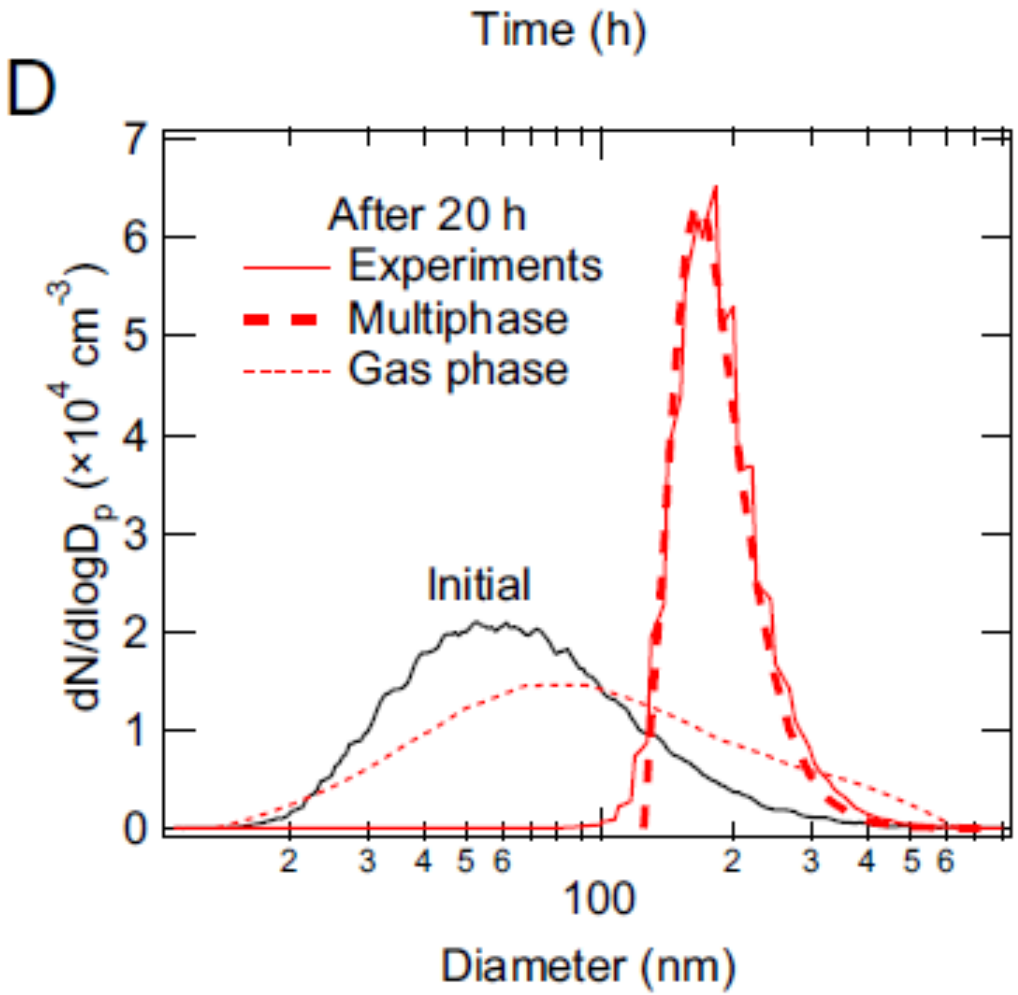
1. “Phase/Viscosity”:





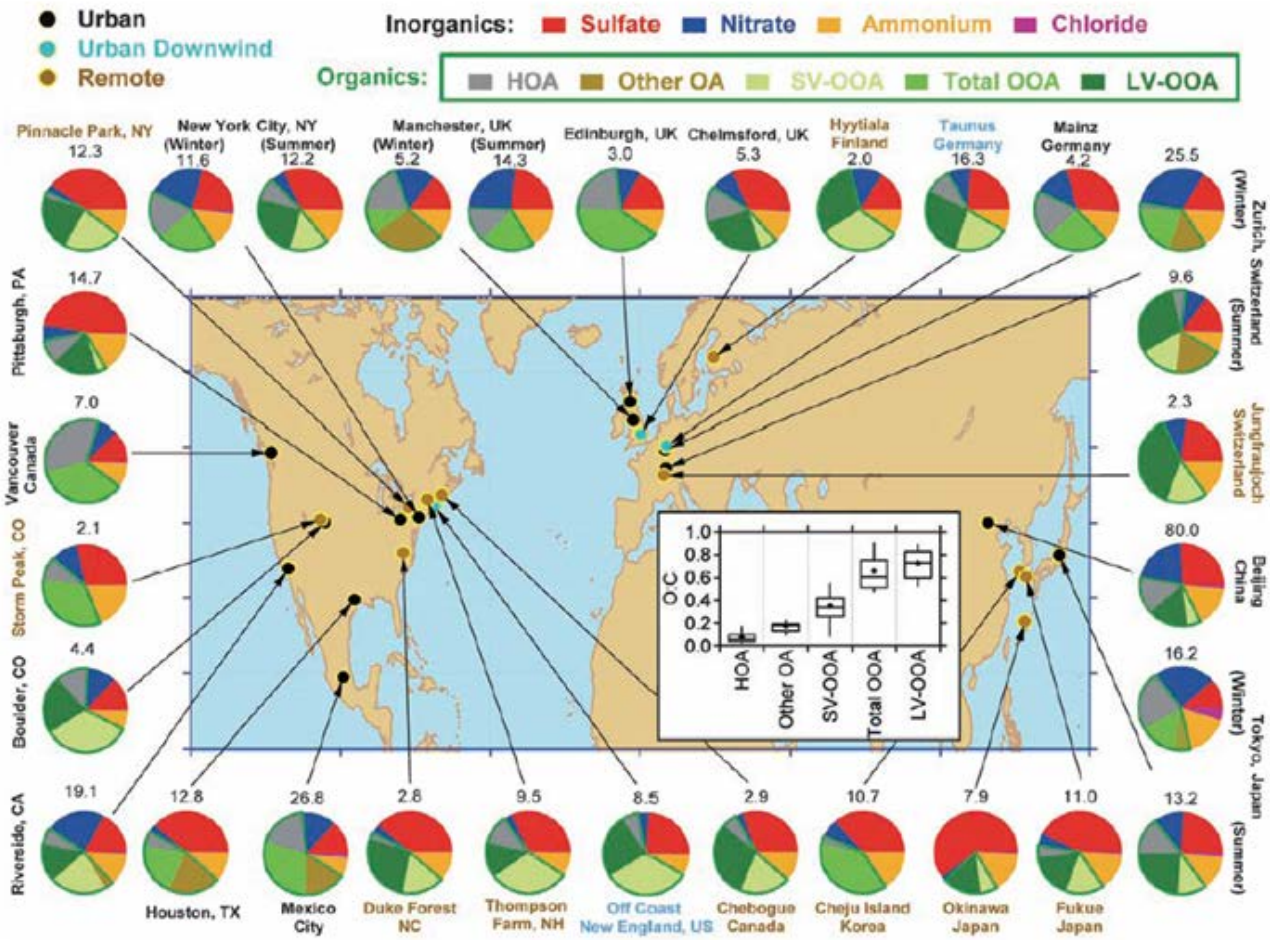
2. Growth Mechanisms

Why? *Getting the Number-Diameter Distribution of Atmospheric Particle Population Correct in Models*



3. "<u>Sulfate</u> as a trigger or regulator for SOA production & properties"

In the preindustrial atmosphere (1750), sulfate mass loading is thought to be much lower.

IPCC climate forcings depend on estimation of 1750 loadings that depend on models of a low sulfate atmosphere.

Sulfate triggers and regulates SOA production rates & properties from highly oxygenated molecules.