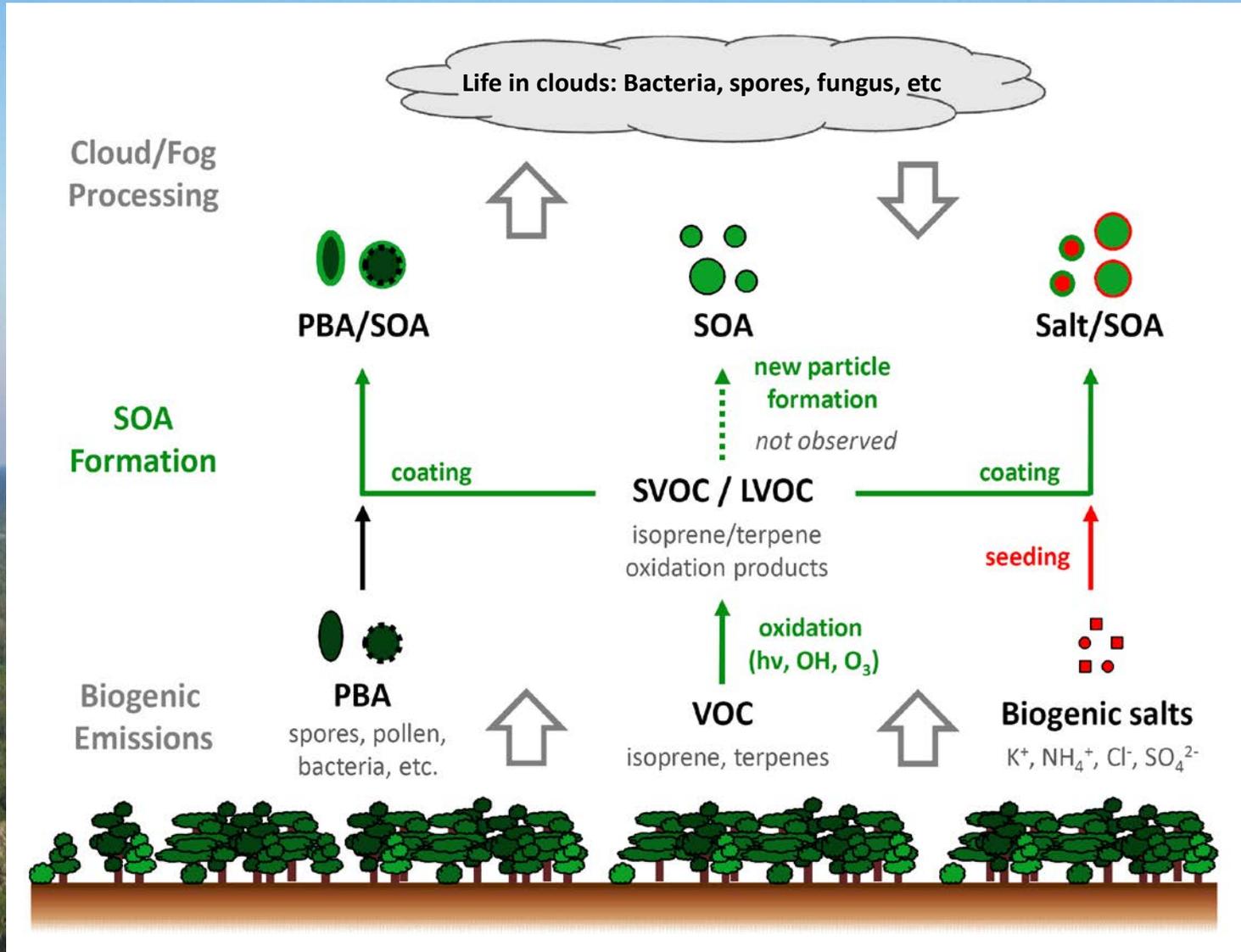


Aerosol Life Cycle Working Group Meeting
Fall 2013
SOA Breakout Session

Anthropogenic Influence on Climate-Relevant SOA Properties
Relevant to Climate Upscaling

The biology of the forest partially controls the chemistry and physics of the atmosphere in Amazonia



Strong interactions between forest biology, physics and chemistry of the atmosphere





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

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The direct and
indirect radiative
effects of biogenic
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Interactive Discussion

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 $+0.01 \text{ W m}^{-2}$ and -0.12 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 W m^{-2} and -0.77 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 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 W m^{-2} and -0.78 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.

The direct and indirect radiative effects of biogenic SOA

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Interactive Discussion



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Statement of Problem

Why are **model predictions** of **climate-relevant SOA properties** inconsistent with observations?

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SOA properties of two types

*Of the first type: **climate-relevant properties***

mass concentrations,
number-diameter distributions [which models predict number-
diameter distributions?],
optical properties [brown carbon?],
Kappa values [success]

*Of the second type: **process-relevant properties** (as needed *in*
models to predict climate-relevant properties *by* models)*

O:C elemental composition,
viscosity ,
kinetic rate constants

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)

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Outcome of Day's Work: Two Implementation Activities

1. "box model intercomparisons" **DETAIL (xx participants)**
2. "cooperative / multiple chamber studies" – (**xx participants**)

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Next Near-Term Steps (< 15 days)

1. People at meeting signed up to the different intellectual themes based on their interests. This to be complemented by sending out rapporteur report to ALC email list and doodle poll sent to be set up so that people can sign up to these areas. In signing up, people self identify to one of three areas (lab, model, field) to ensure good representation. *STM to do this activity.*
2. Leadership (2 to 3 people) in each group to be identified. *STM, JES, BFP to seek volunteers / make requests.*
3. Leadership will engage with folks in each area to define title, define topics, etc., as defined on previous slide (30 day time period)
4. Activities start and report back in March
5. (cross cutting for model intercomparisons and lab chamber /standards)

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Mid-Term Steps (< 2 months)

1. Leadership will engage with folks in each area to polish title (as needed) and further define and prioritize topics as bullet points.
2. Choose one bullet point as immediate priority.
3. Include consultation with cross-cutting implementation groups (i.e., model intercomparisons and cooperative / multiple chamber studies). [If judged as needed, an Executive Committee to form across all 3 intellectual and 2 implementation groups, and Committee to have monthly conference calls.]
4. Possibly start activities.
5. Report back in March meeting.

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Criterion / metric to judge if our SOA Focus Area is succeeding

We anticipate of the three intellectual themes represented by the three groups, 1 or 2 of these groups will show themselves as substantive success in regard to final point of previous slide, “Report back in March meeting.”

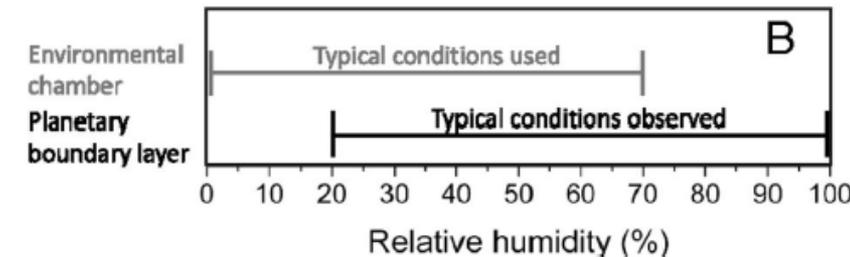
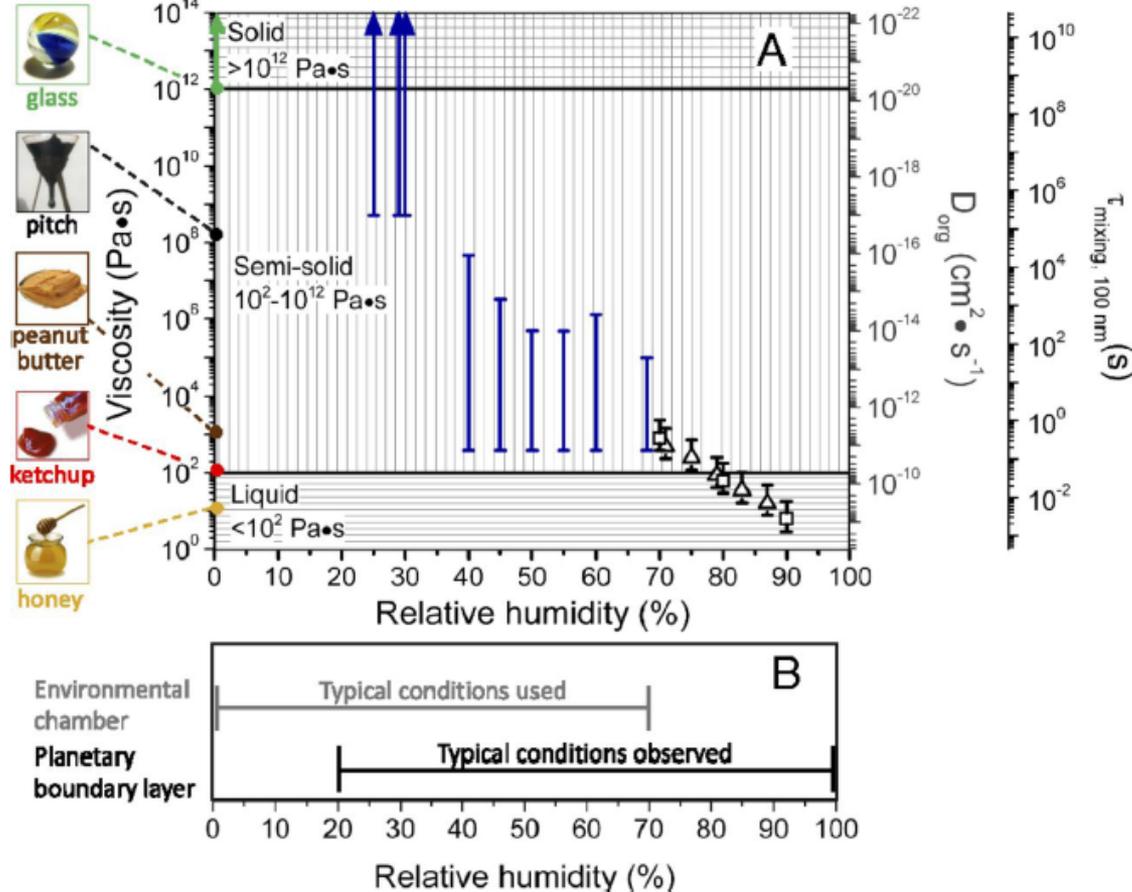
Substantive success should be that sum of group activity is greater than result of individual activities.

Having success of 1 or 2 groups in this regard will be a success for the SOA Focus Area as “the next step”.

Higher bar for ‘success metric’ to then be set at the March meeting.

1. “Phase/Viscosity”: The “What” Slide

(A) Summary plot of the SOM viscosities determined by a combination of the “bead-mobility” [black open squares and triangles for Harvard Environmental Chamber (HEC) and PNNL samples, respectively, where the black bars represent the 95% prediction intervals]...

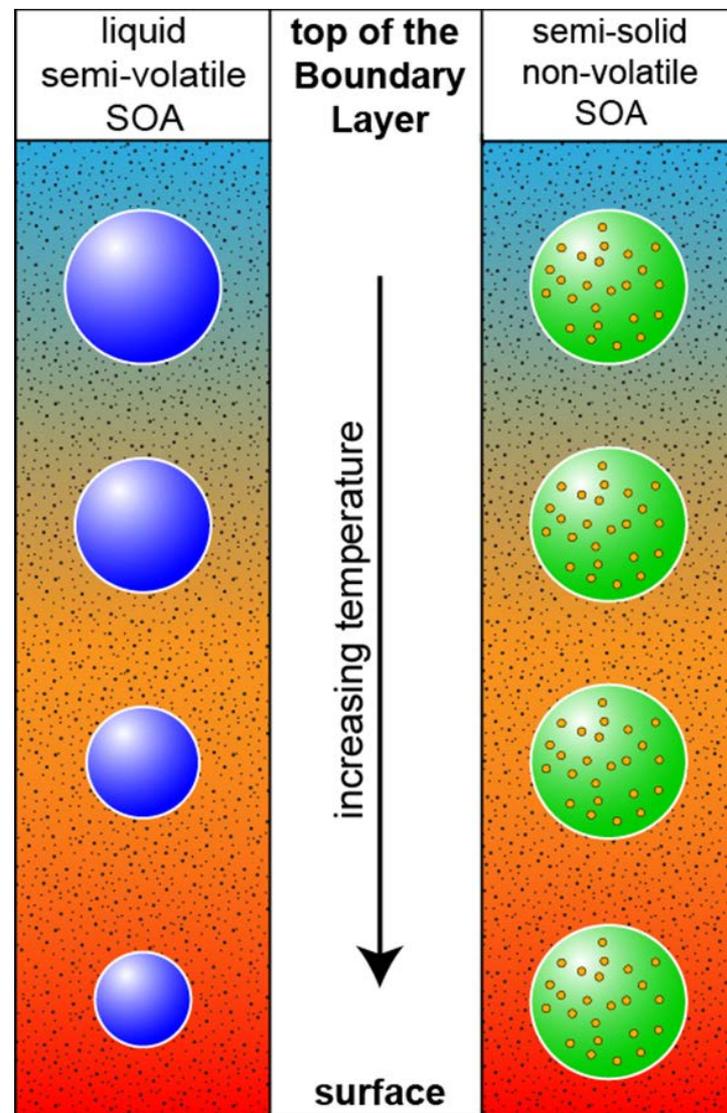


Renbaum-Wolff L et al. PNAS 2013;110:8014-8019

1. “Phase/Viscosity”

The “Why” Slide:

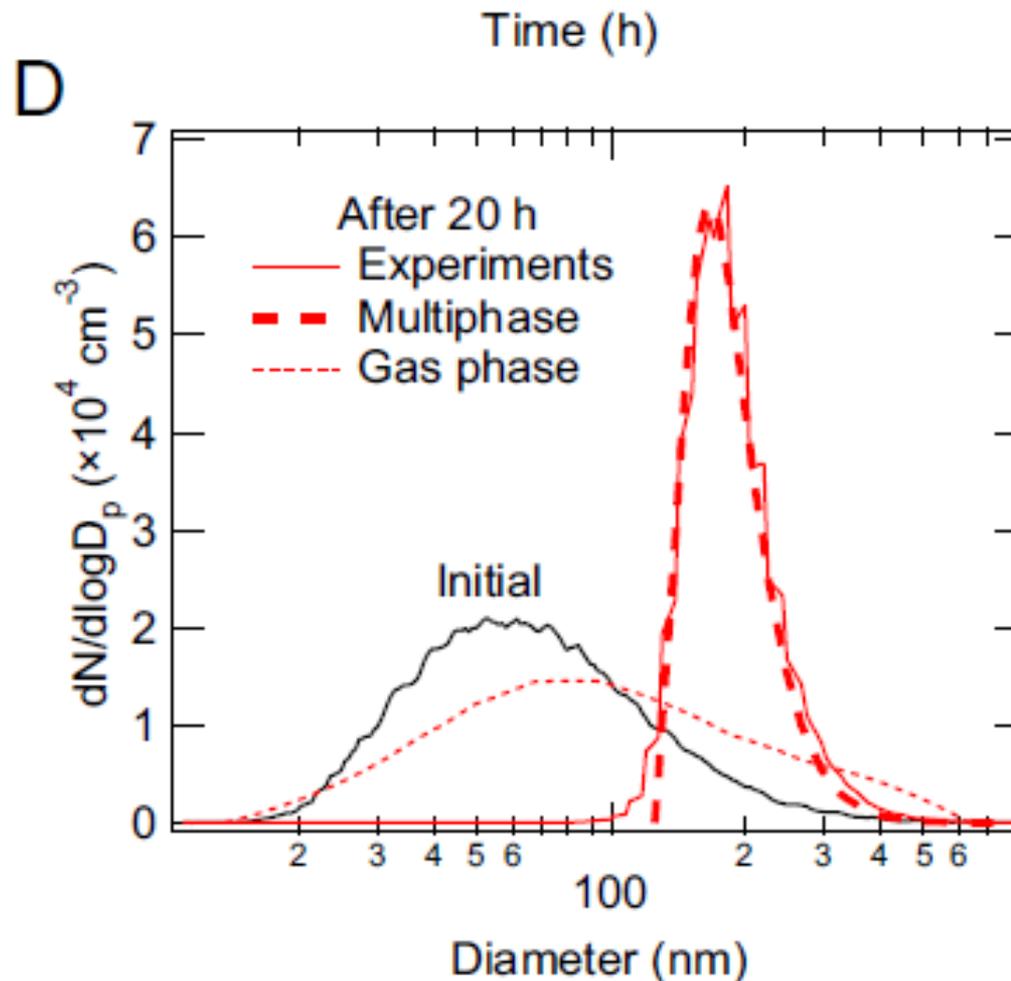
Just One Example: Getting both Mass Concentrations and Number-Diameter Distributions Right in the Models



2. “Growth Mechanisms”: The “What” Slide

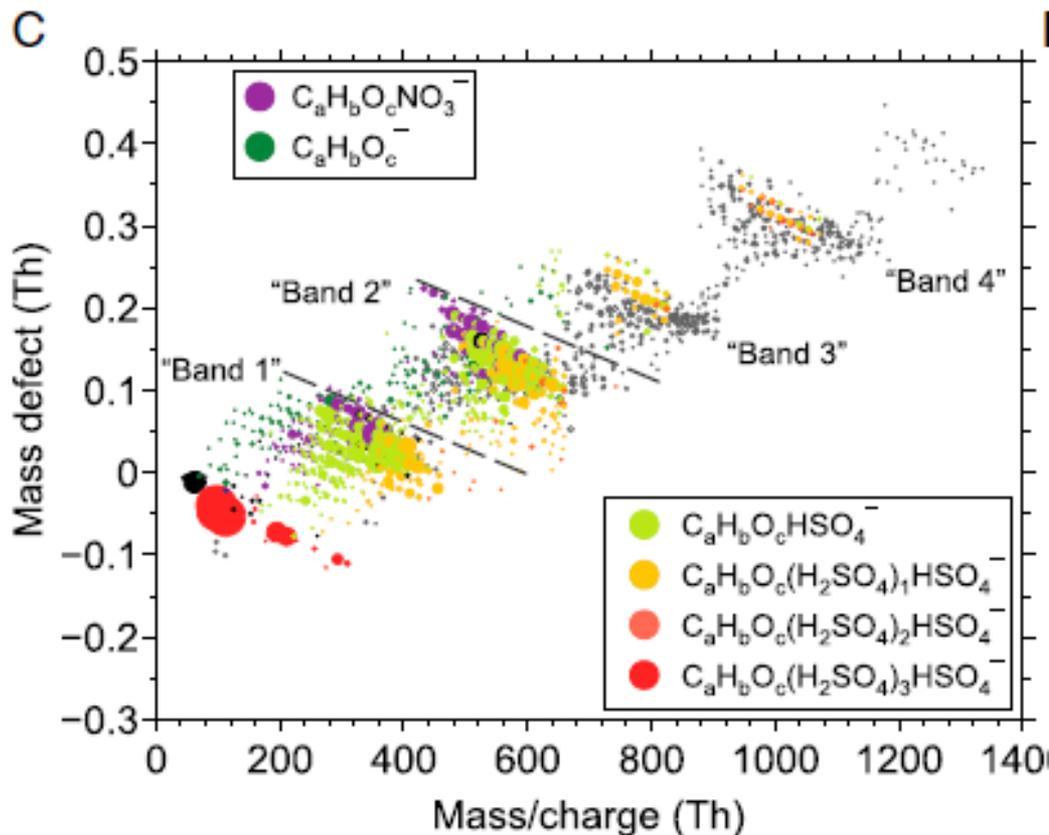
No.	Mechanism	Parameters Controlling Growth Dynamics
#1	Gas-phase diffusion limited condensation of effectively non-volatile organics	Aerosol surface area
#2	Absorption of semi-volatile organic vapors according to Raoult’s law or Henry’s law	Aerosol volume (& composition), Solute volatility, Phase state (viscosity, diffusivity)
#3	Mechanism #2 followed by particle-phase chemical reactions to form effectively non-volatile oligomers, organic salts, organosulfates, organic acids, etc.	Particle size (& composition), Solute Volatility, Phase state (viscosity, diffusivity), Particle-phase reactivity

(Just One) Example of **Why** We need to Make Progress As a Priority for this Intellectual Theme: *Getting the Number-Diameter Distribution of Atmospheric Particle Population Correct in Models*



3. “Sulfate as a trigger or regulator for SOA production & properties” *The “What” Slide:*

Mixtures of SO₂, ammonia, amines and terpenes create molecular clusters, which initiate new particle nucleation and nano-particle growth, conditions typical of much of the northern hemisphere.



Observations in the boreal forest show similar mixtures of these components in new particle formation events at very low ambient levels.

Does the current southern hemisphere (e.g., the Amazon), dominated by biogenic isoprene emissions, represent a valid model of particle nucleation, growth and CCN formation in pre-industrial low sulfate conditions?

3. “Sulfate as a trigger or regulator for SOA production & properties” *The “Why” Slide*

Current global aerosol is composed of roughly half sulfate by mass.

In the preindustrial atmosphere (1750), sulfate is thought to have been ~5x lower.

IPCC climate forcings depend on estimation of 1750 loadings that depend on models of a low sulfate (and ammonia?) atmosphere.

