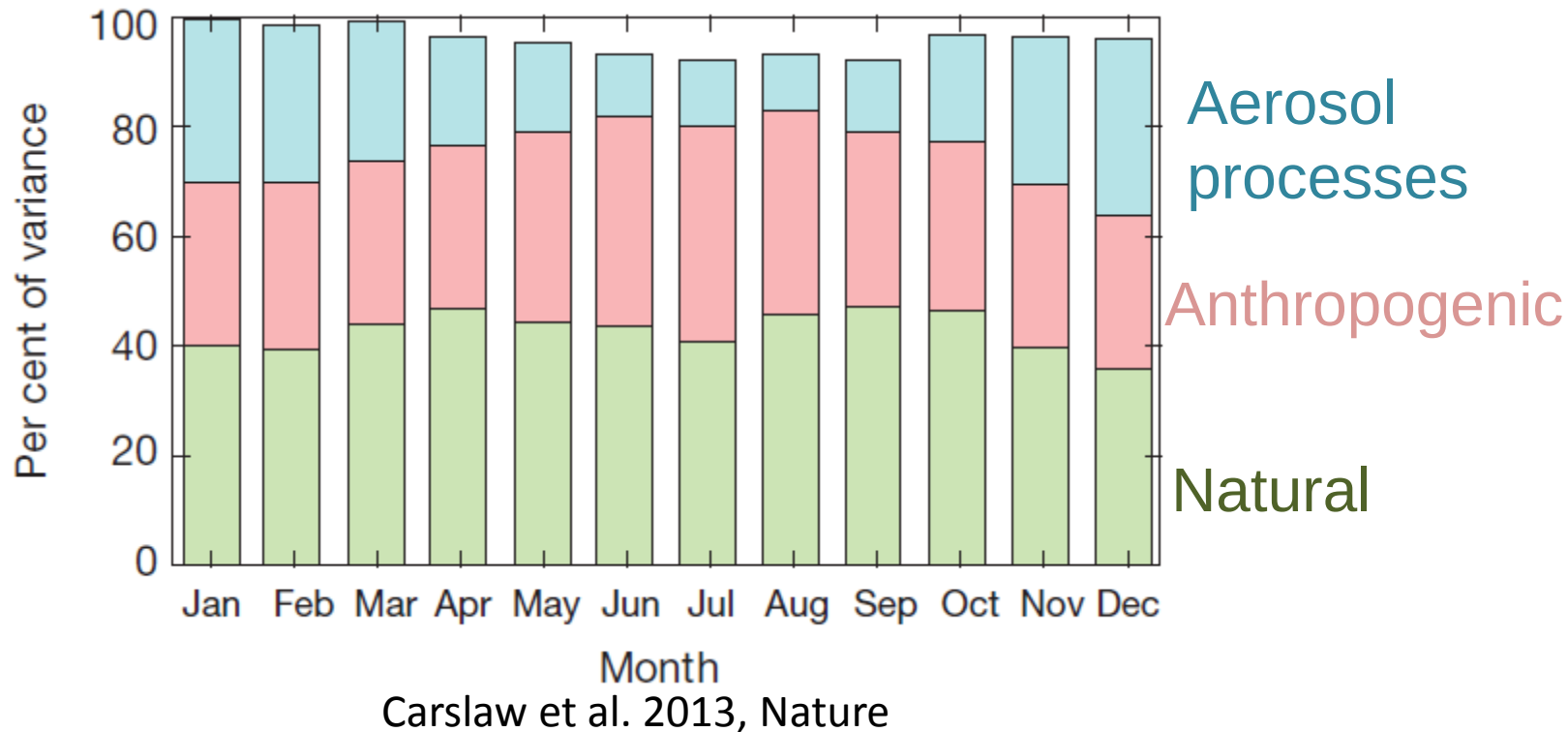
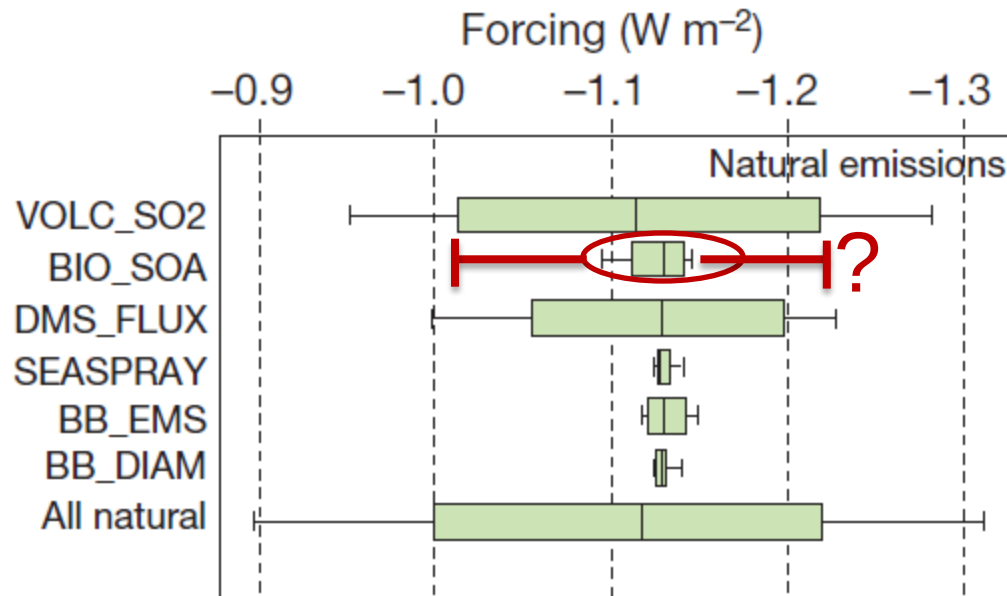


Aerosol Indirect Forcing: Highly sensitive to natural emissions during pre-industrial times



Natural: Volcanic SO₂, marine DMS emissions and biogenic SOA: largest contributors to the variance

Variance related to biogenic SOA



Carslaw et al. 2013, Nature

- SOA formation yields highly uncertain under low sulfate conditions

Hypothesis: Variance associated with biogenic SOA in pre-industrial times is even more uncertain because of less understood SOA chemical mechanisms

If Aerosol Indirect Forcing Uncertainty is not enough motivation

- As emissions control technologies and standards evolve, sulfate, nitrate and ammonia emissions would be reduced
- Important to understand how SOA chemistry and its phase and properties would change in the future
- Key to understanding future changes in aerosol-cloud interactions

Science questions

1. To what extent are sulfur and NO_x necessary for nucleation of new particles?
2. How does reduction of sulfate, nitrate and ammonia affect SOA formation? (relevant to pre-industrial times and the future)
3. How do the chemistry, growth and properties (phase, viscosity, hygroscopicity) of SOA change in the presence of sulfate? (It is well known that SOA chemistry is different under high and low NO_x conditions, but much less is known about sulfate)
4. Under what conditions does the presence and absence of inorganics affect CCN formation, and how does this relate to the phase state of SOA? (e.g. glassy SOA coating a sulfate or nitrate core could prevent water from seeing the sulfate, thus reducing water uptake by the inorganic core)

Laboratory experiments

- Near term goals
 - Investigate how inorganic seeds like sulfate and nitrate affect SOA formation yields under different conditions of T and RH (measurements team?)
 - Explore different sulfate-NO_x-ammonia regimes for SOA formation that would be relevant for the preindustrial times and the future
- Long term goals
 - Lab-intercomparison experiments: Study the effects of seeds on SOA yields from different smog chambers to understand the measurement uncertainty

Field experiments

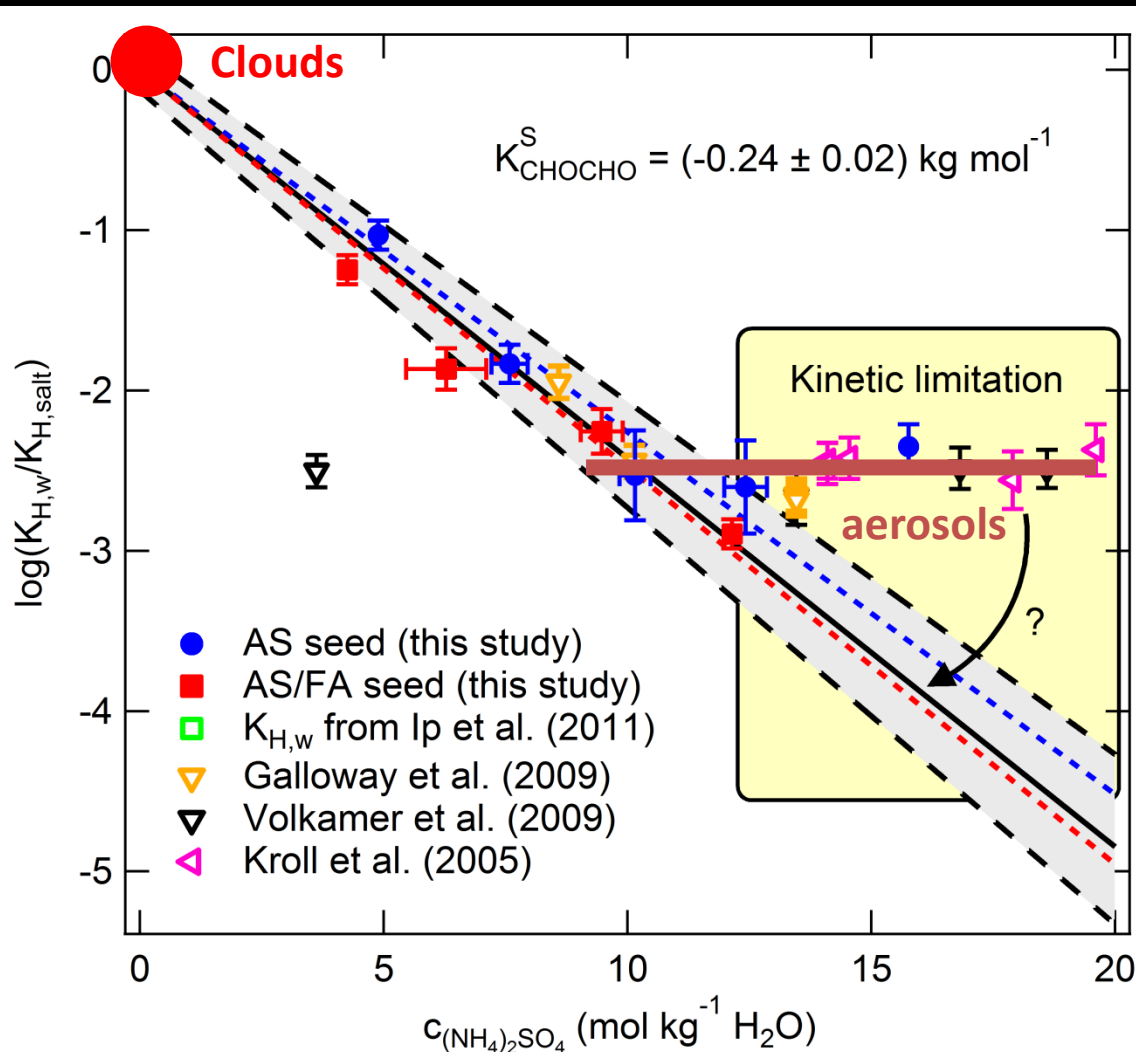
- Near term goals: Analyze recent field campaign data (BAECC, CARES, GoAmazon) to understand how variation of inorganic components affects SOA properties
- Long term goal: Planning future field campaign based on identified knowledge gaps
 - Results from past field studies in the Amazon: Isoprene oxidation products are not good nuclei for SOA formation
 - But terpene oxidation products are potentially much more important for nucleation than isoprene
- Question: How important is the role of sulfate or nitrate seeds in SOA formation within regions dominated by terpenes such as boreal forests?
- Potential future campaign: Terpene equivalent of GoAmazon

Modeling activities

- Process based box models to understand how the chemical and physical kinetic processes governing SOA nucleation and growth change due to presence of inorganics as sulfate or nitrate
- Role of aqueous phase chemistry (e.g. glyoxal salting-in triggered by particulate sulfate) (Rainer Volkamer)
- Derive parameterizations that can capture the effects of these processes for use in chemical transport models
- Regional models such as WRF-Chem to investigate role of sulfate on SOA and evaluate with field campaigns such as TCAP and CARES
- Global models: Investigate how SOA formation changes between pre-industrial, present day and the future
- Investigate resulting effects on aerosol indirect forcing through aerosol-cloud interactions

Salting-in triggered by particulate SO_4^{2-}

Setschenow (1889): $\log \left(\frac{K_{H,w}}{K_{H,salt}} \right) = K_i^S = (-0.24 \pm 0.02) \times c_{AS}$



Example: Glyoxal – similar salting-in likely applies also to other highly oxygenated molecules.

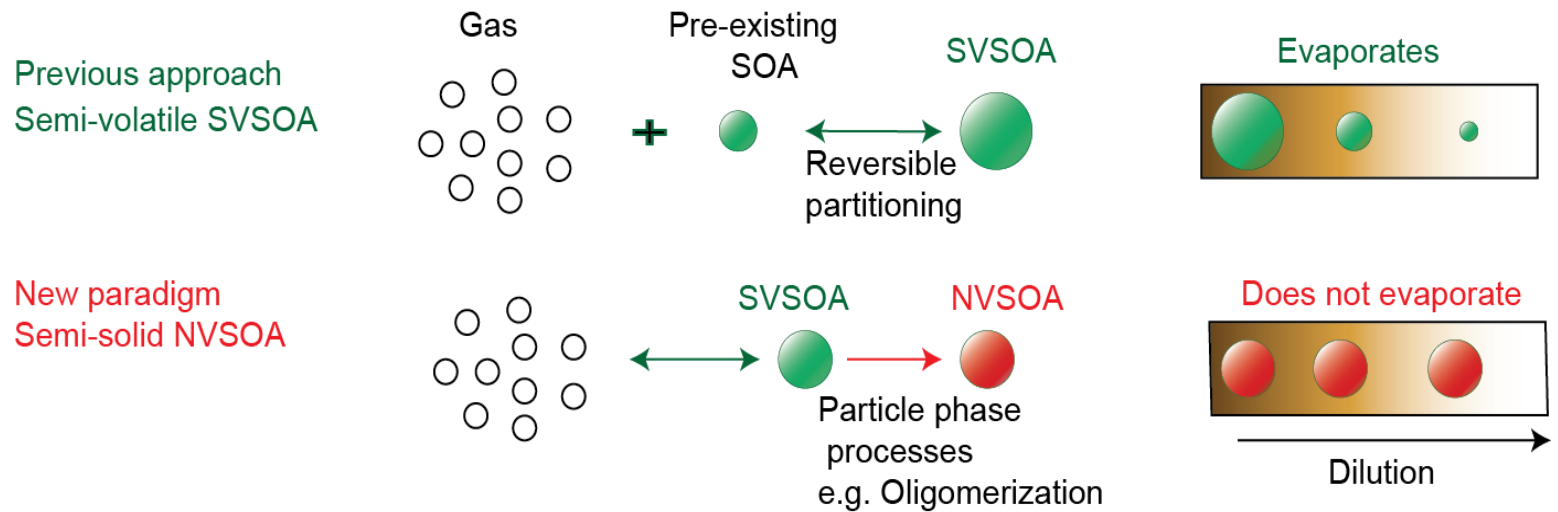
Sulfate triggers and regulates SOA production rates & properties from highly oxygenated molecules (here glyoxal).

Sulfate is higher now than in pre-industrial times.

Kampf et al., 2013, ES&T
Waxman et al., 2013, GRL
Slide courtesy: Rainer Volkamer

Near-term tangible outcomes

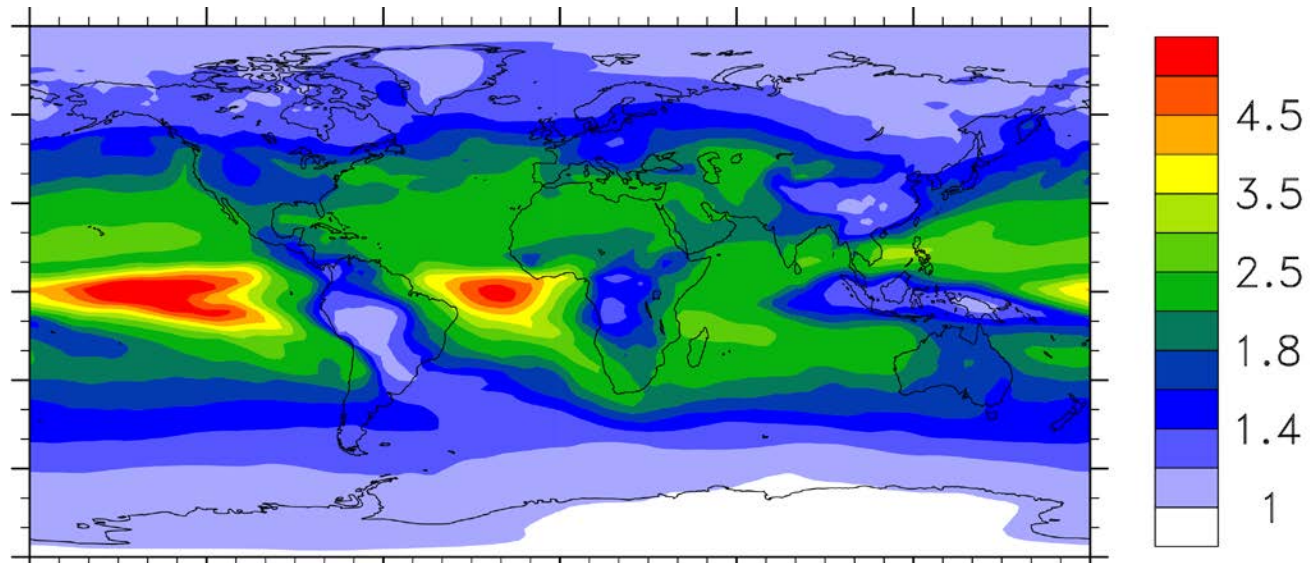
1. Writing a white paper documenting the science them questions and means to address them in terms of modeling and measurements
2. Possible journal article: Perspectives on role of inorganic seeds in SOA formation
3. Identifying the what collaborative efforts could be conducted among various focus groups (new particle formation and growth, phase-viscosity) towards laboratory measurements, field campaign proposals and modeling



Current SOA yields: presence of sulfate seeds

Semi-solid SOA effects: Global model CESM

Ratio: Non-volatile/Semi-volatile Mean: 1.64



M. Shrivastava et al. 2013