Aerosol Phase State: Implementation in Air Quality Models and Implications on Aerosol Formation

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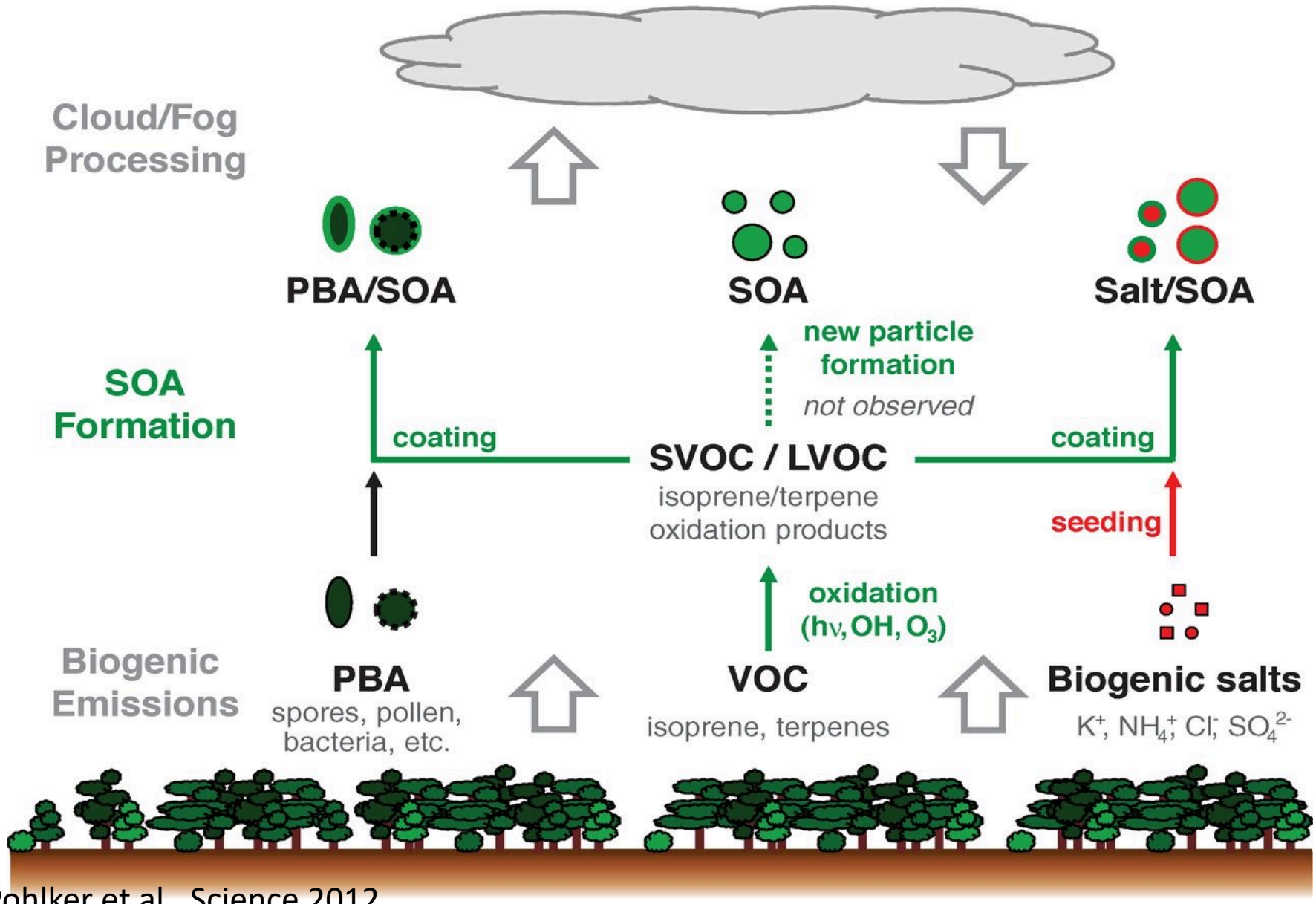
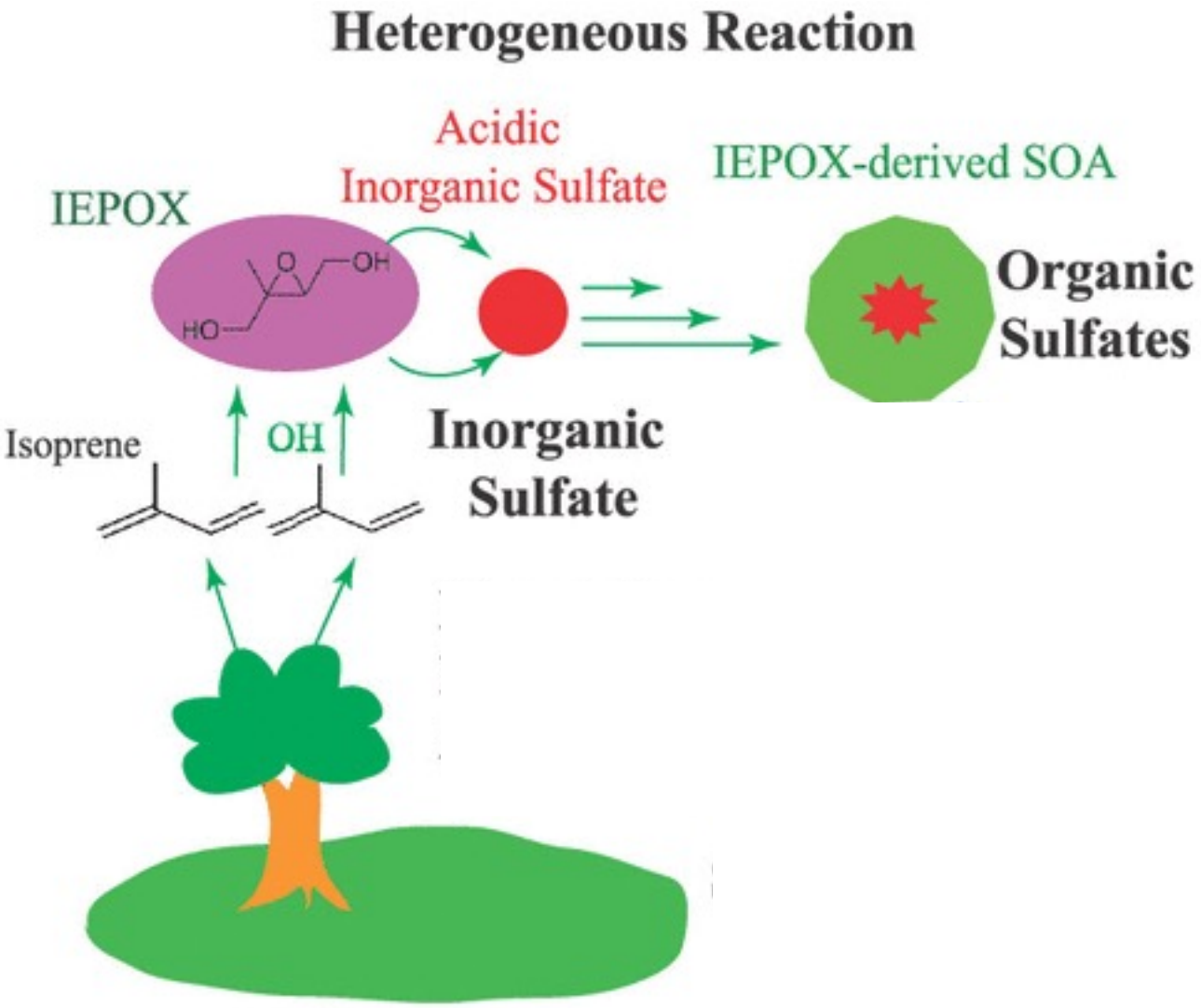
Secondary Organic Aerosols



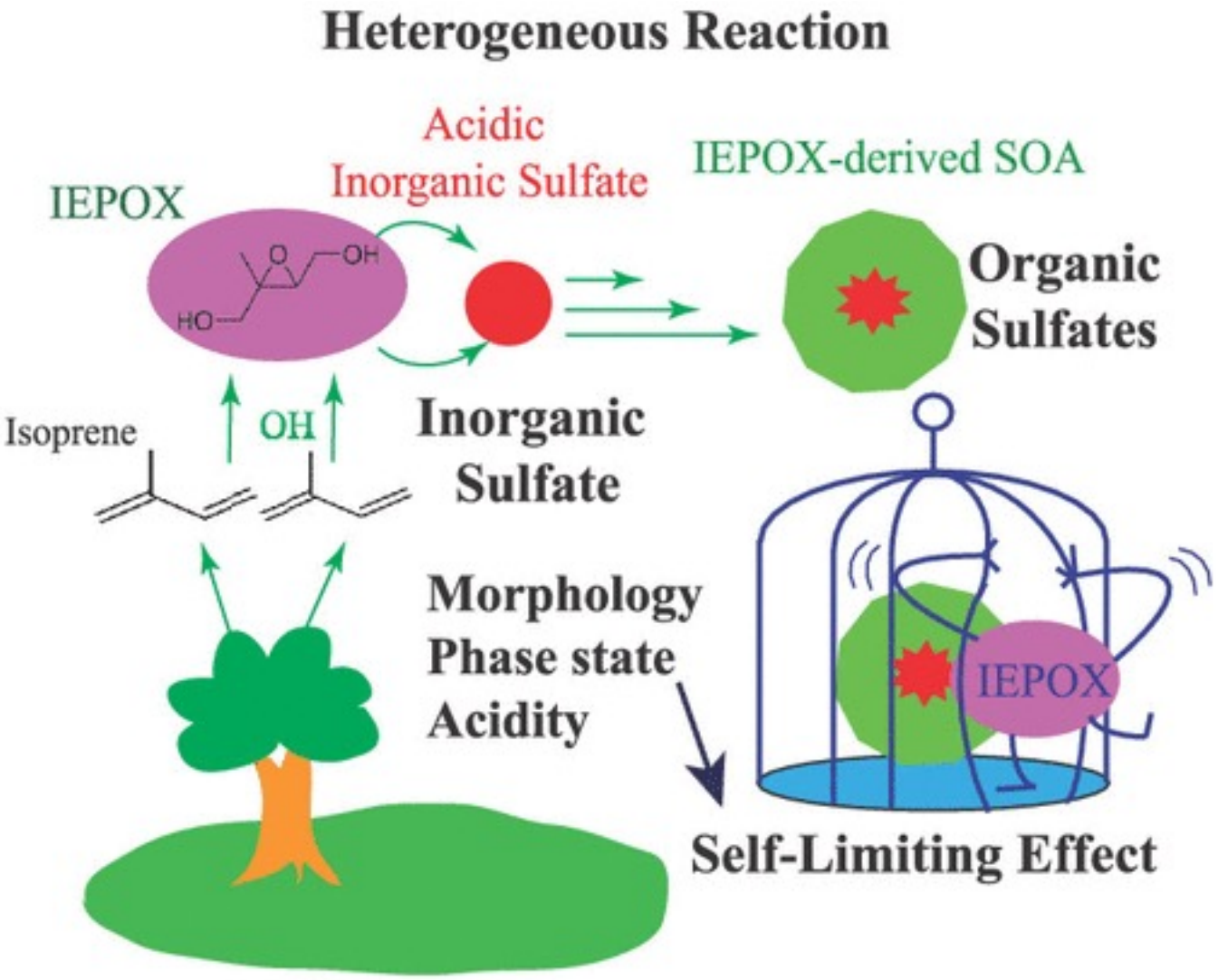
Pohlker et al., Science 2012

Isoprene SOA



Zhang et al., ACS Earth and Space Chemistry, 2019

SOA and Aerosol Phase State

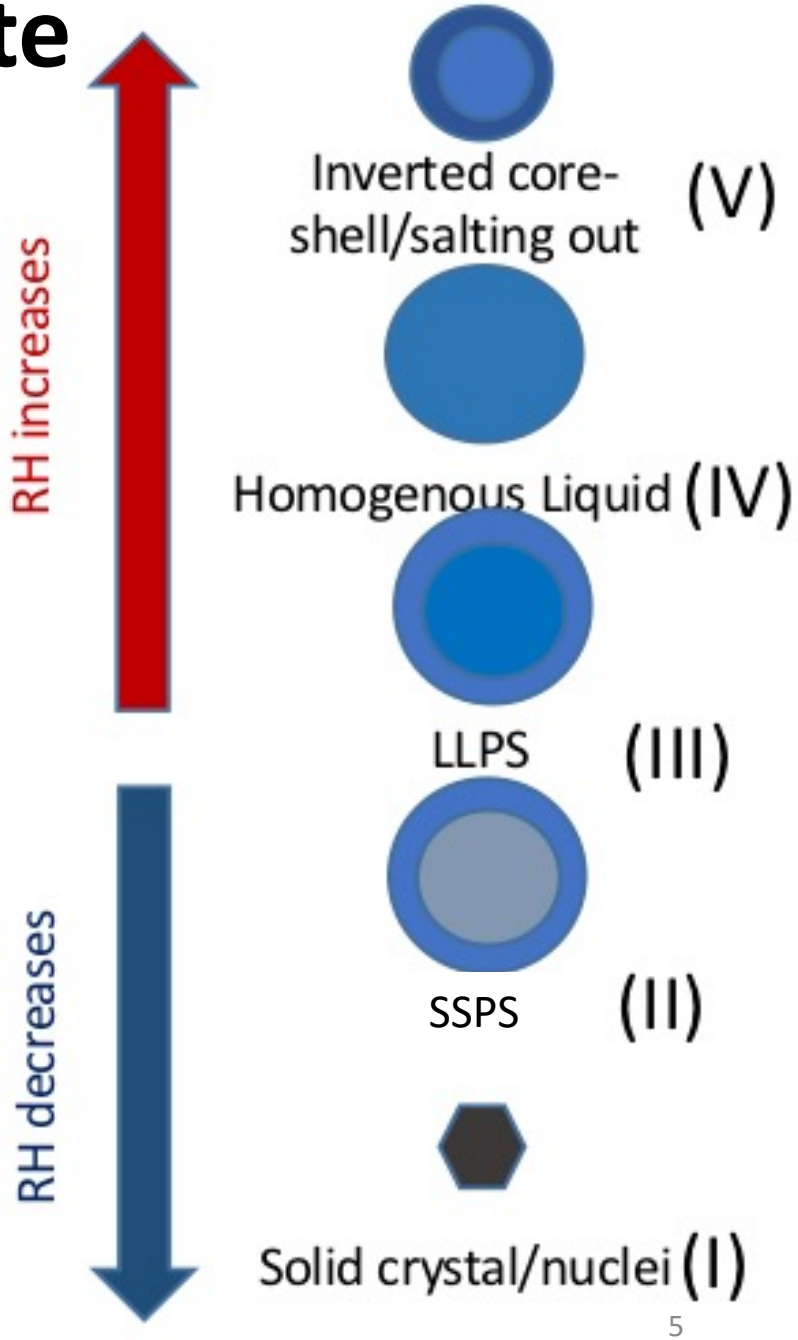


Zhang et al., ACS Earth and Space Chemistry, 2019

AQM Aerosol Phase State

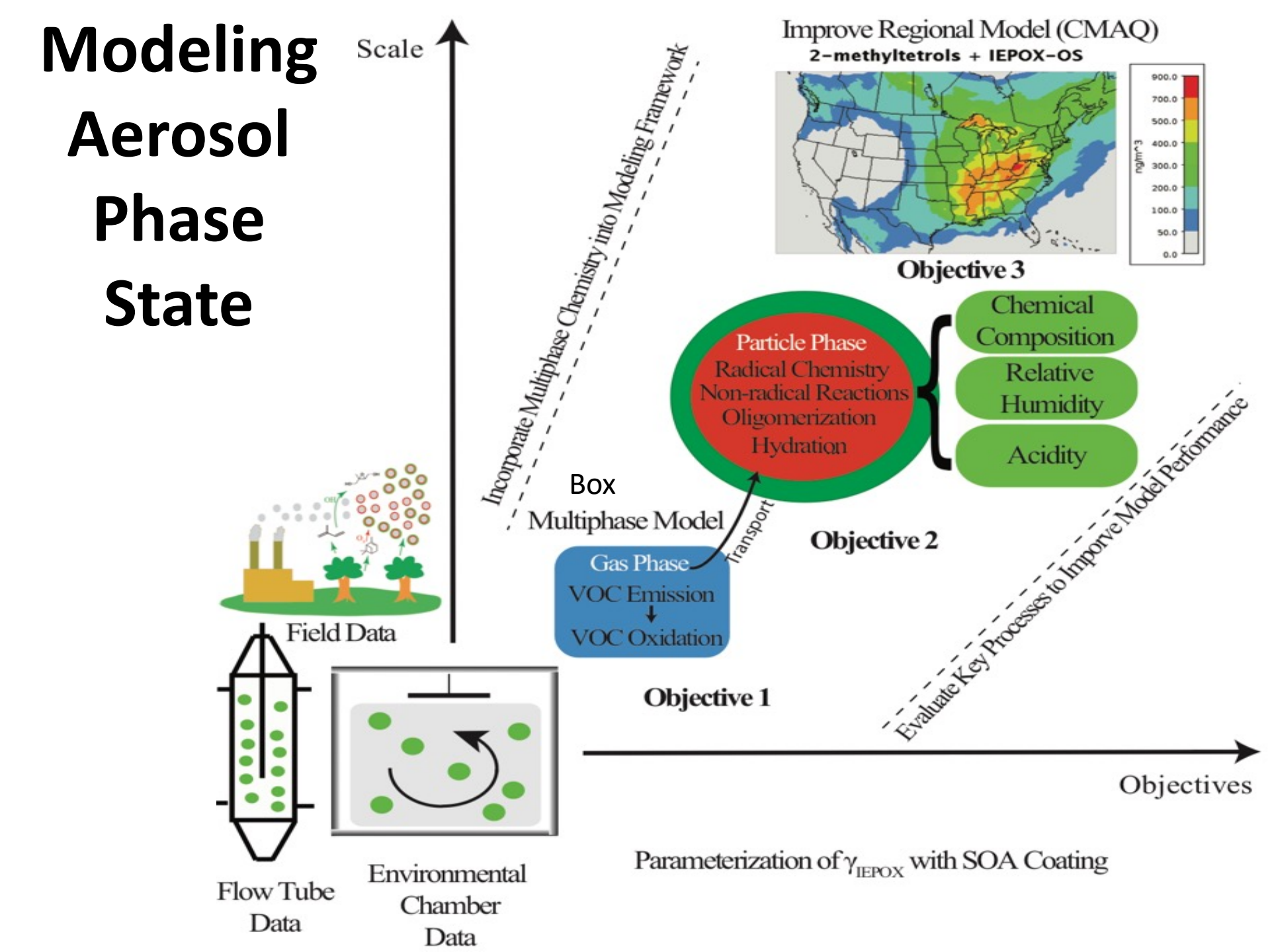
- Current AQMs assume Homogenous liquid

- Impacts
  - Radiative forcing
  - CCN
  - Long Range Transport (PAH)*
  - Multiphase chemistry Secondary Organic Aerosols



*Zhou et al., PNAS 2019

Modeling Aerosol Phase State



AQM Implementation

1. Determine the phase separation using Glass Transition Temperature and Separation Relative Humidity

2. If homogenous phase reverts to original kinetics equation in CMAQ

3. If cores-shell morphology modified kinetics (Gaston et al., 2014)
    1. Determine viscosity (Modified Vogel-Tamman-Fulcher Equation)
    2. Determine organic diffusion coefficient (Stokes Einstein)

Glass Transition Temperature

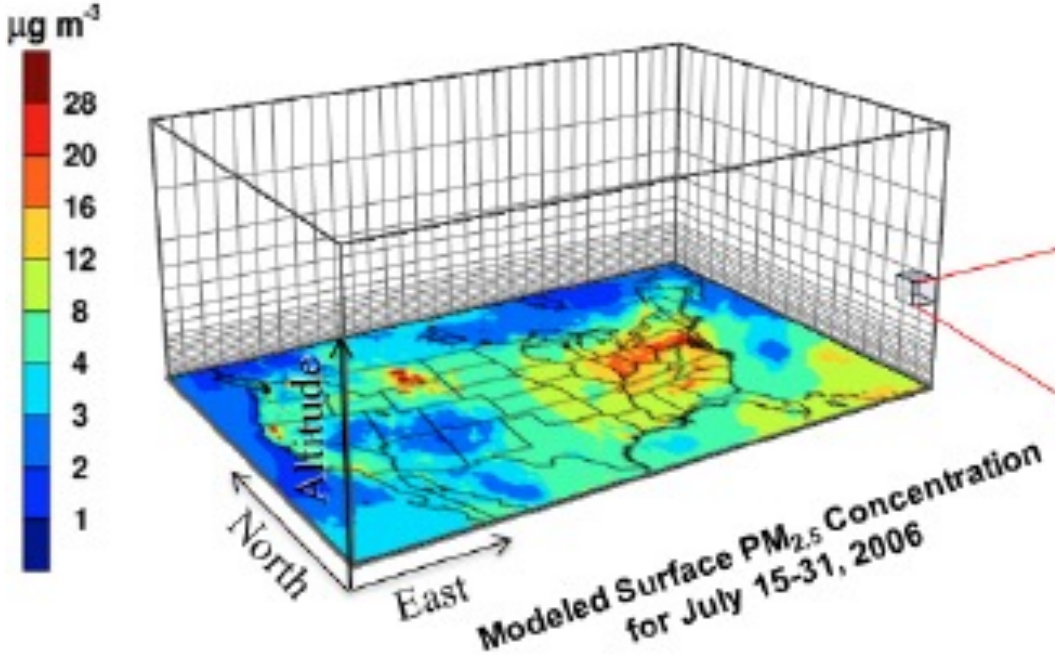
Used 179 organic species to fit a relationship between T_g, the molar mass (M), and O:C ratio

$$T_{g,x} \; or \; T_x = -21.57 + 1.51M_x - 0.0017M_x^2 + 131.4(O:C)_x - 0.25M_x(O:C)_x \qquad (4)$$

$$Where, \; M_x = \sum(w_{i,x}M_{i,x}) \; ;(O:C)_x = \sum\left(w_{i,x}(O:C)_{i,x}\right); \; w_{i,x} = \frac{Mass\ Concentration_{i,x}}{Total\ Mass\ Concentraion_x}$$

(Shiraiwa et al. 2017)

Air Quality Model Runs

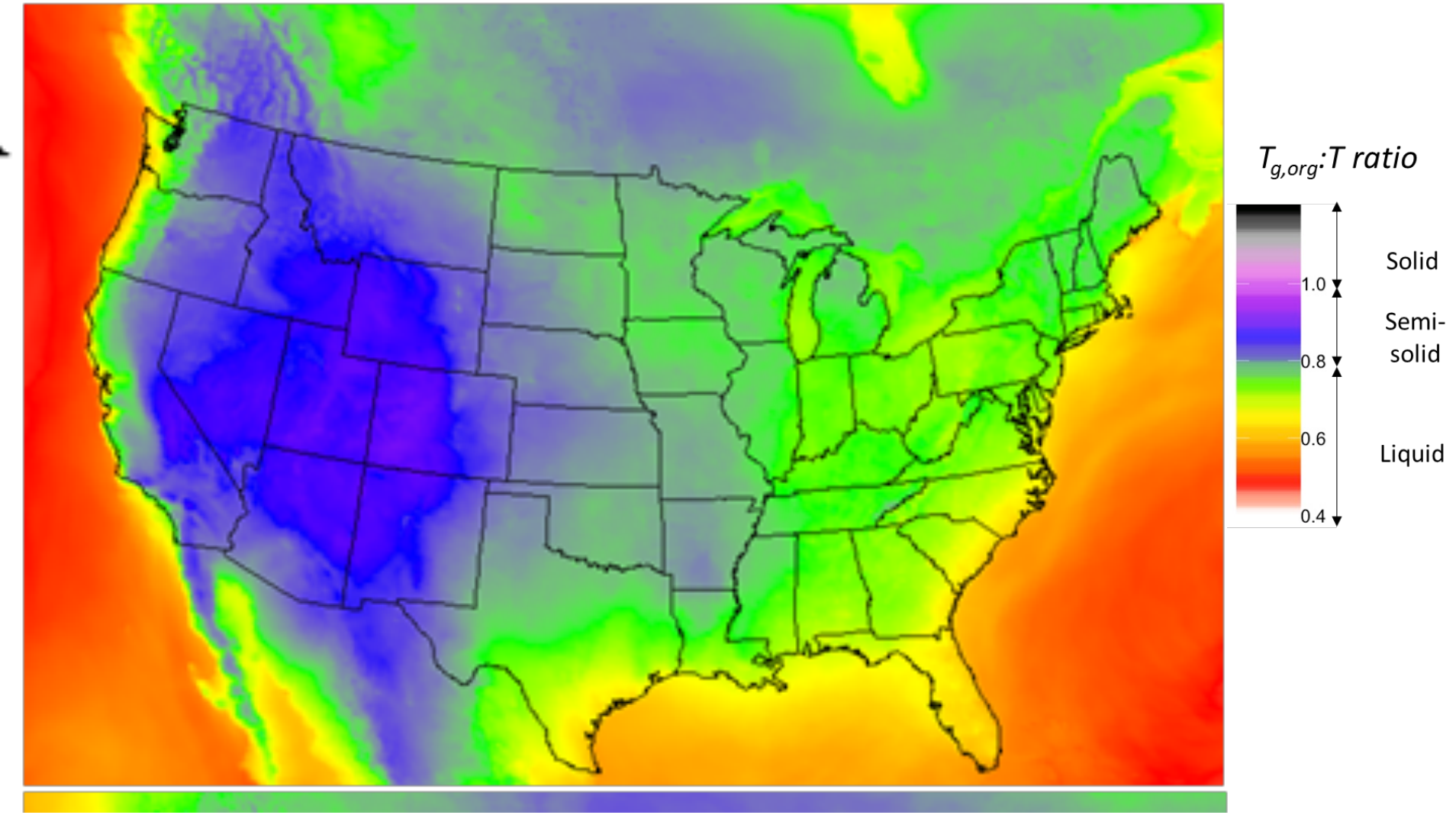


CMAQ
SOAS Campaign
June 1 - July 15, 2013

- **Implemented first phase state and impact on multiphase chemistry**

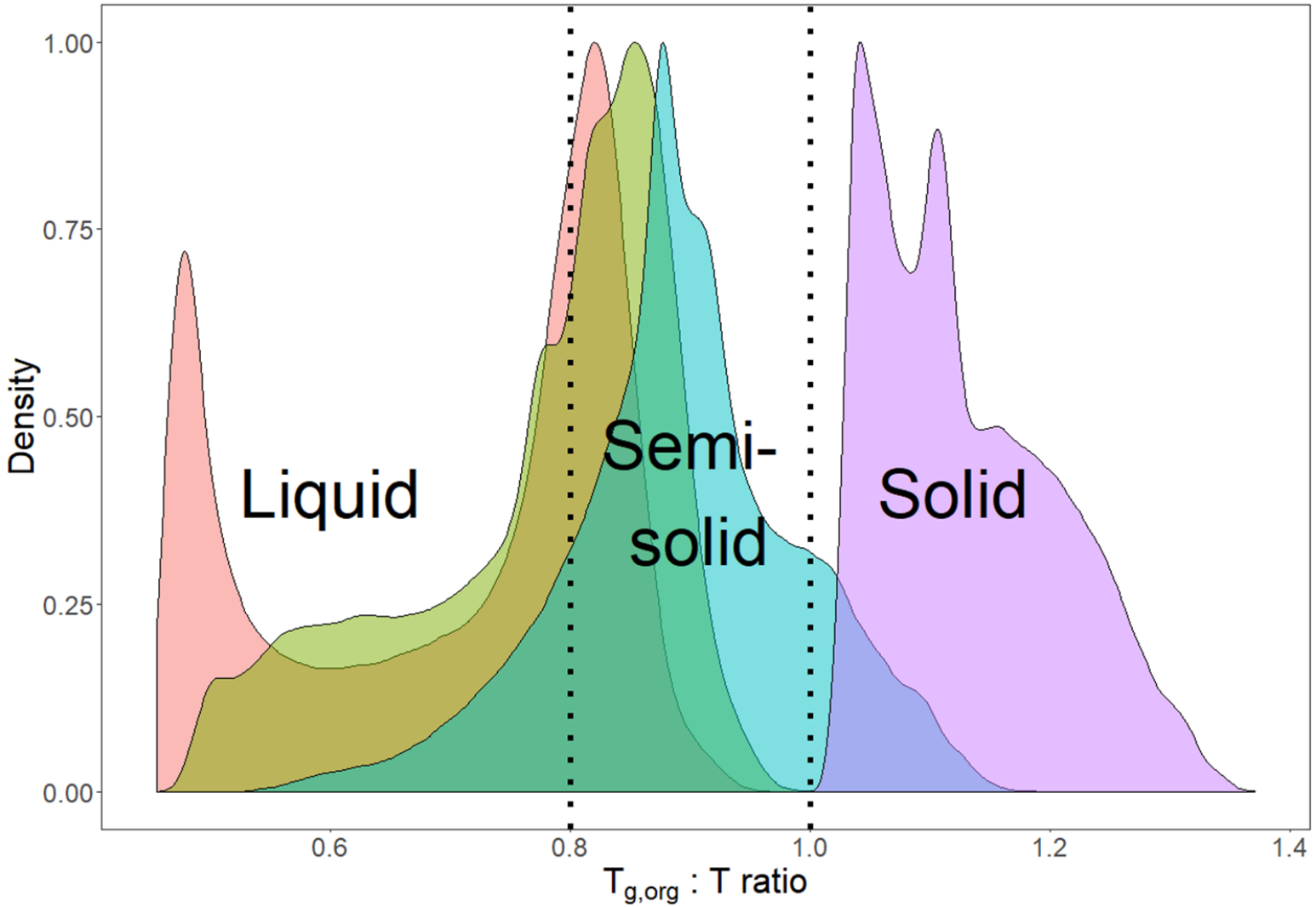
http://bioearth.wsu.edu/cmaq_model.html

Predicted Phase State

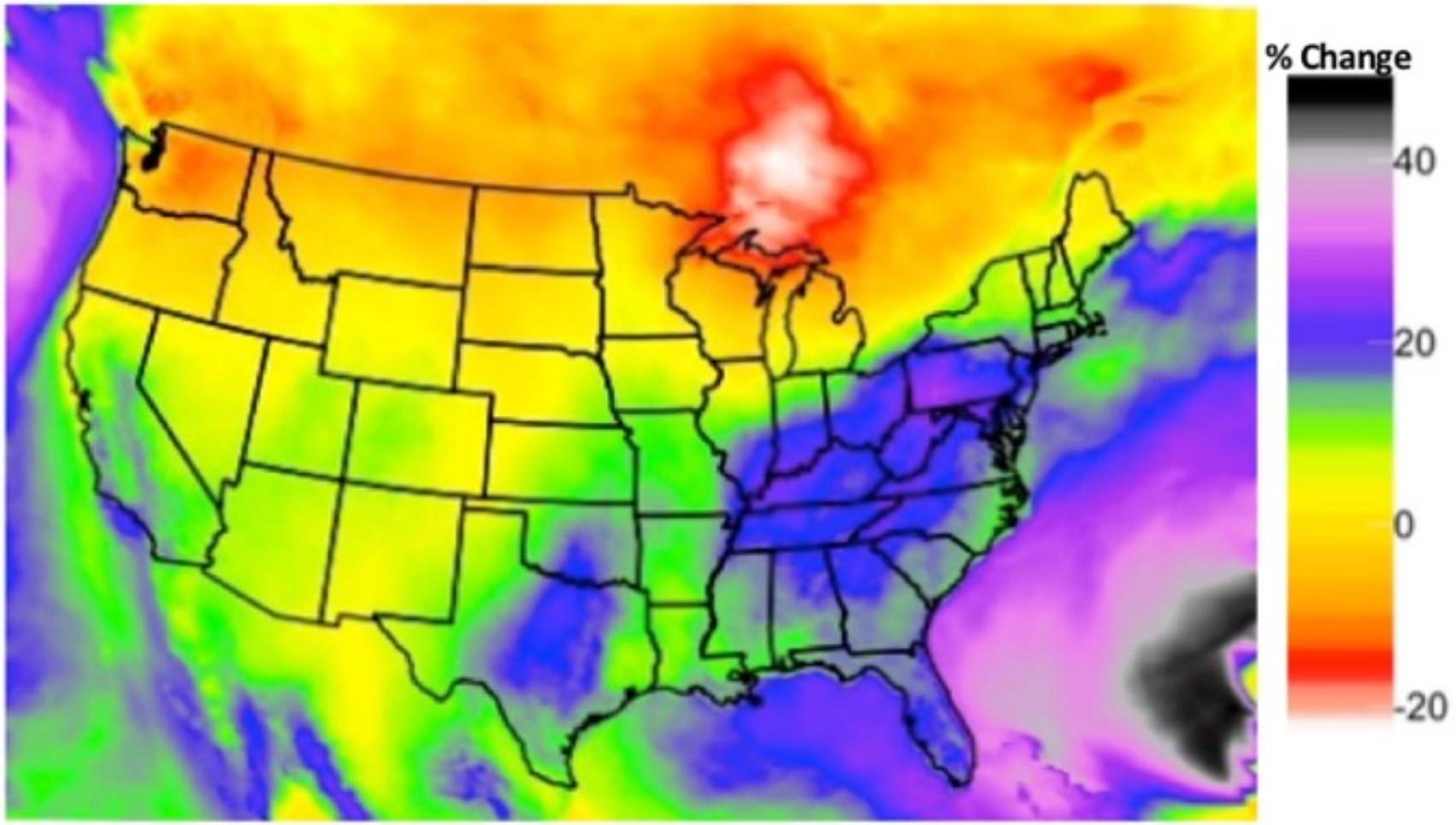


For all time steps and over the continental United States, the average glass transition temperature to ambient temperature ($T_{g,org}$:T) ratio

Predicted Phase State



Predicted Change in Biogenic SOA Mass



Future Work

- Update from CMAQv521 to CMAQ53
- Implementation and evaluation of multiple Tg equations (C*, mw, O:C, H:C)*
- Updated kinetics for formation of organosulfates implications on viscosity and PH
- Upper atmosphere evaluations
- Global Model Implementation

* Shiraiwa, Lin, Zhang

Conclusion

- **Phase Separation/State Matters**
  - Regional and Global Scale models must include phase state and expand to monoterpenes
  - Implications on climate, public health, long range transport
  - Need more field and laboratory data
- **Instrumented regional/global scale model**
  - Risk assessment
  - Exposure metrics
  - Measurement locations
  - Forecasting

Questions?

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