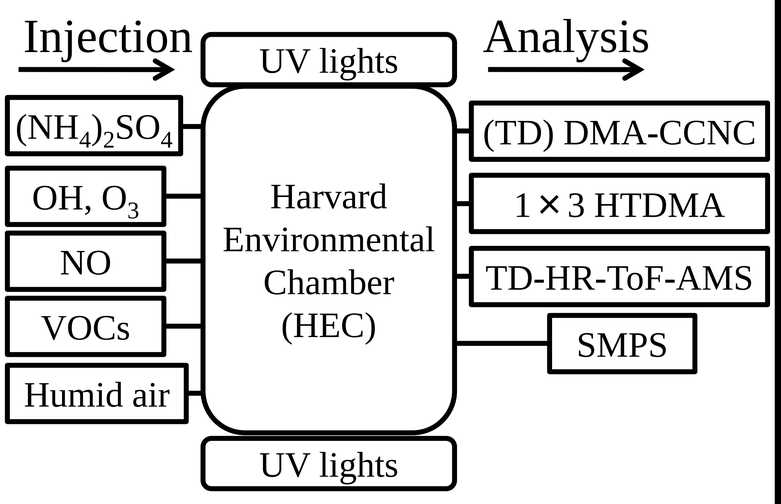
CCN Activity and Hygroscopicity of Secondary Organic Material

Scot T. Martin, Mikinori Kuwata, and Mackenzie L. Smith

School of Engineering and Applied Sciences, Harvard University, Cambridge, MA (scot_martin@harvard.edu)

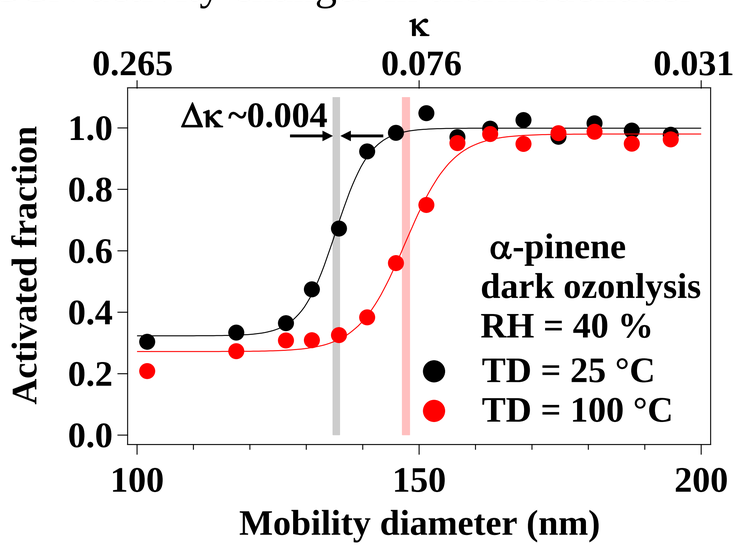
Harvard Environmental Chamber



The chamber was operated under a condition of continuous flow. This feature was important for the study of atmospherically relevant concentrations (e.g., < 1ppbv α-pinene).

CCN relationships: O:C ratios and thermodenuder (TD) results

CCN activity changes in thermodenuder

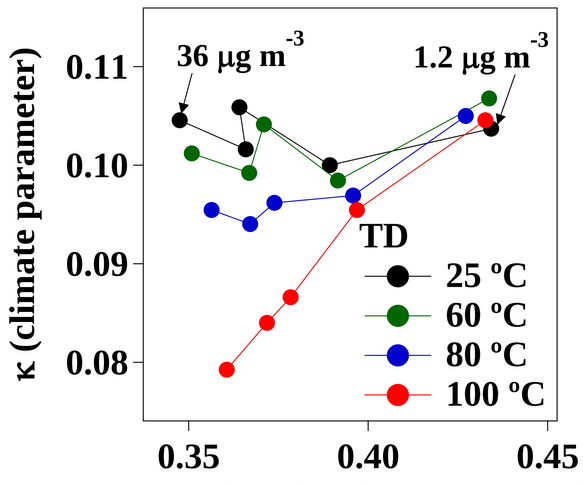


κ values for α-pinene secondary organic material decreased at elevated temperatures.

Why?

Ref: *Petters and Kreidenweis (2007)*

Model development

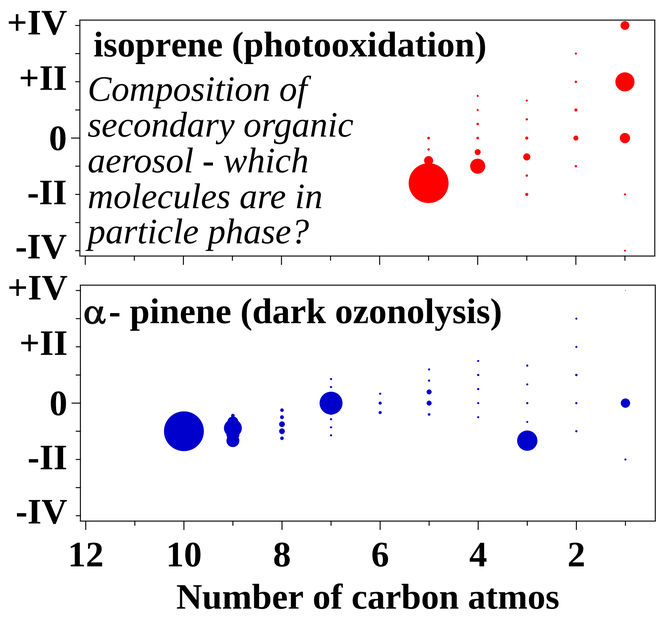


Oligomer formation in the organic material decreased κ, indicating aging effects on CCN activity. Materials at lower loadings were more oxidized.

Ref:*Shilling et al.(2008, 2009), Kuwata et al., in preparation*

Theory

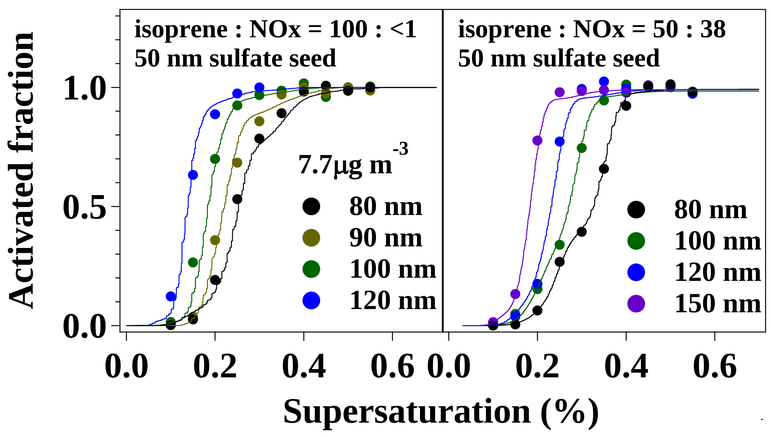
Gas-Phase Reaction Mechanism: Products



Isoprene SOA produces smaller molecules. Carbon number decreases by oxidation.

CCN mixing rules for organic-inorganic compositions

Modeling of CCN activation curves



We could model the activation curves using a single set of parameters regardless of different organic mass fractions and reaction conditions. Generalization of this result makes the treatment of organic material in climate models more feasible.

Model development

$$1 + \frac{S}{100} = \frac{d_{aq}^3 - d_{geo,dry}^3}{d_{aq}^3 - d_{geo,dry}^3 \left(1 - \sum_{k \in \{AS,org\}} \varepsilon_k \kappa_k\right)} \exp\left(\frac{4\sigma V_{m,w}}{RT d_{aq}}\right)$$

Parameters	Value	Unit
Surface tension, σ	0.0725	N m^{-1}
Effective molar volume, $V_{m,\,org}$	180	cm^3 mol^{-1}
Effective van't Hoff factor, i_{org}	1	
Effective soluble fraction, ω_{org}	1	
Hygroscopicity parameter*, κ_{org}	0.10	
Hygroscopicity parameter*, κ_{AS}	0.53	

*$\kappa_k = \omega_k i_k (V_{m,\,org})^{-1} V_{m,w}$

Ref:*King et al. (2007, 2009, and 2010)*

Conclusions

Thermodenuder experiments show that oligomer formation is responsible for the decrease in CCN activity. This magnitude depends on the O:C ratios.

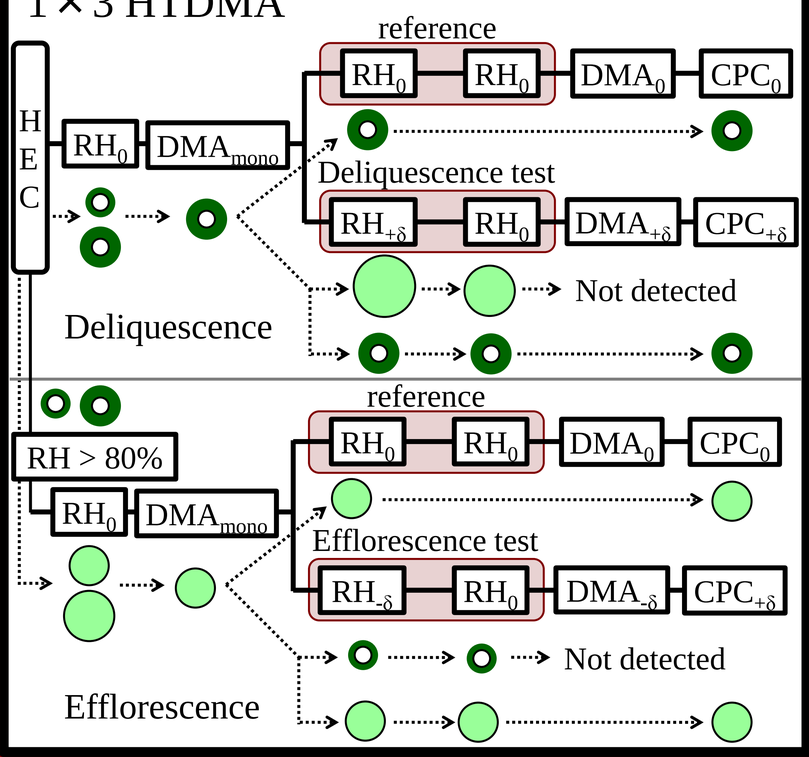
CCN activity for organic-inorganic mixed particles is well predicted assuming a simple mixing rule and κ~ 0.1.

The gas-phase reaction mechanism predicts that molecular weights of isoprene SOA compounds are smaller and hence more water soluble than those of α-pinene SOA compounds. Even so, κ values for particles of these materials are similar.

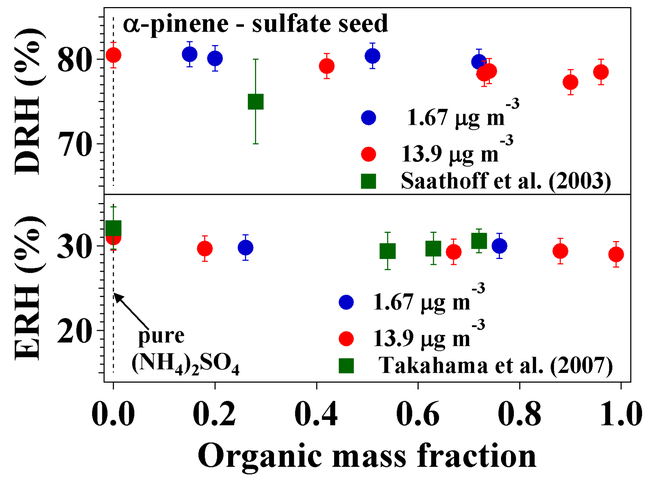
Secondary organic material produced from the dark ozonolysis of α-pinene minimally affects the DRH and ERH of ammonium sulfate.

Hygroscopicity and phase transitions

1 × 3 HTDMA



Model development



Both DRH and ERH of $(NH_4)_2SO_4$ do not change by coating with secondary organic material. Generalization of this result makes the treatment of organic material in climate models more feasible.

Ref: *Smith et al., in preparation*

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