Cloud activation properties of organic aerosols observed during CalNex-LA and CARES

Fan Mei[1], Patrick Hayes[2,3], Amber Ortega[2,3], Jose-Luis Jimenez[2,3], Ari Setyan[4], Qi Zhang[4], Jonathan Taylor[5], James Allan[5], and Jian Wang[1]

1. Brookhaven National Lab, NY. 2.Cooperative Institute for Research in Environmental Sciences, CO. 3. University of Colorado, CO. 4. University of California, Davis, CA. 5. The University of Manchester, UK.

Introduction & Motivation

- Organic species are major components of atmospheric aerosol. Ambient aerosols often consist of hundreds of organic species, and their hygroscopicities (κ_{Org}) are not well understood.
- This incomplete understanding of κ_{Org} may lead to substantial uncertainty in simulated aerosol indirect effects (Liu and Wang, ERL, 2010).

Objectives

- Characterize the cloud activation properties of atmospheric organic aerosol.
- Examine the relationship between κ_{Org} and oxidation level of organics.
- Examine the impact of organics on droplet growth kinetics.

Measurements

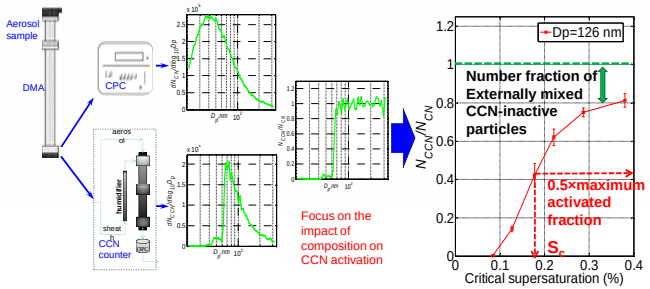
Sampling location and period

Table 1. Relevant measurements during CalNex-LA and CARES

Measurements	Instruments
Size-resolved CCN spectrum	Differential mobility analyzer and CCN counter (operated in series)
Aerosol composition (Non-refractory species)	High resolution time of flight aerosol mass spectrometer
Concentration of black carbon	7-wavelength aethalometer

Experimental setup

Measurements of size-resolved CCN spectrum



Data Analysis

Derivation of hygroscopicity of organics (κ_{Org})

Overall particle hygroscopicity:

$$k_{CCN} = \frac{4A^3}{27D_p^3 \ln^2 S_c}, \text{ where } A = \frac{4s_w MW_w}{RTr_w}$$

From size-resolved CCN spectrum

Organic hygroscopicity:

$$k_{Org} = \frac{1}{x_{Org}}(k_{CCN} - k_{NH_4NO_3}x_{NH_4NO_3} - k_{(NH_4)_2SO_4}x_{(NH_4)_2SO_4})$$

Volume fraction, derived from Aerosol Mass Spectrometer data

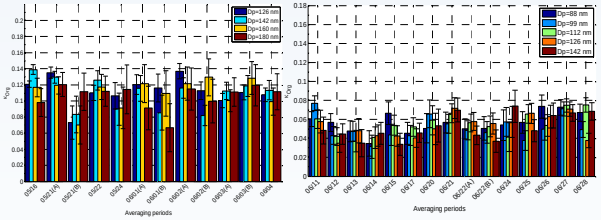
Uncertainty in derived κ_{Org}:

$$\sigma^2_{\kappa_{Org}} = \left(\frac{\kappa_{CCN} - \kappa_{Inorg}}{x^2_{Org}}\right)^2 \sigma^2_{x_{Org}} + \left(\frac{1}{x_{Org}}\right)^2 \sigma^2_{\kappa_{CCN}}$$

Results

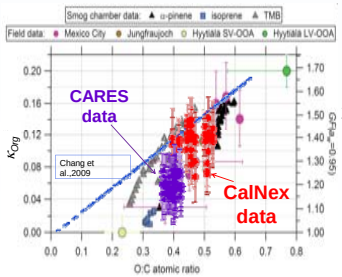
Derived organic hygroscopicity during the two studies

Both size-resolved CCN spectrum and chemical composition are averaged over periods of ~2 hours to increase counting statistics and signal to noise ratio. The following figure shows derived organic hygroscopicity during the averaging periods.



CalNex-LA: κ_{Org}=0.11±0.02 CARES: κ_{Org}=0.05±0.02

The relationship between κ_{Org} and the oxidation level of organics

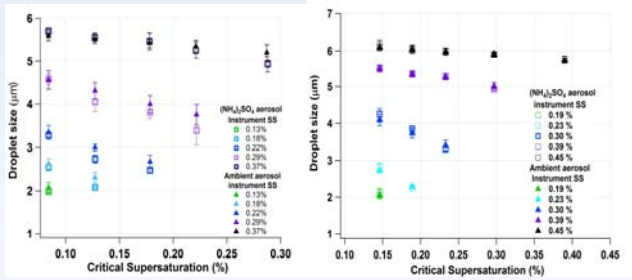


1. Organic hygroscopicity generally increases with increasing the oxidation level (i.e., atomic O:C ratio).
2. However, the large scatter in the data suggests that it may be difficult to develop a univocal parameterization of κ_{Org} based solely on the oxidation level.

Results

Impact of ambient organics on droplet growth kinetics

1. Film forming organic compounds may delay droplet growth by acting as a barrier for water condensation on particle surface (i.e., reduction of water accommodation coefficient).
2. During CalNex-LA and CARES, when exposed to the same supersaturation inside CCN counter, organic particles grew to the same droplet size as pure $(NH_4)_2SO_4$ particles with identical critical supersaturation, suggesting no influences of organic species on water accommodation coefficient.



CalNex-LA CARES

Conclusions

- The derived organic hygroscopicity for CCN activation are κ_{Org} = 0.11±0.02 during CalNex-LA and κ_{Org} = 0.05±0.02 during CARES, consistent with previous smog chamber studies.
- κ_{Org} generally increases with increasing oxidation level. However, the large scatter in the data suggests that it may be difficult to develop a univocal parameterization of κ_{Org} based solely on the oxidation level.
- Ambient organics observed during CalNex-LA and CARES do not inhibit droplet growth through reducing water accommodation coefficient at particle surfaces.

Acknowledgements

- Atmospheric System research program
- ARM climate research facility
- NOAA, CEC and Caltech
- Supporting from staff in BNL