Kinetics of CCN activation and Droplet growth observed in recent field campaigns

Fan Mei (fan.mei@pnnl.gov) and Jian Wang (jian@bnl.gov)

Pacific Northwest NATIONAL LABORATORY

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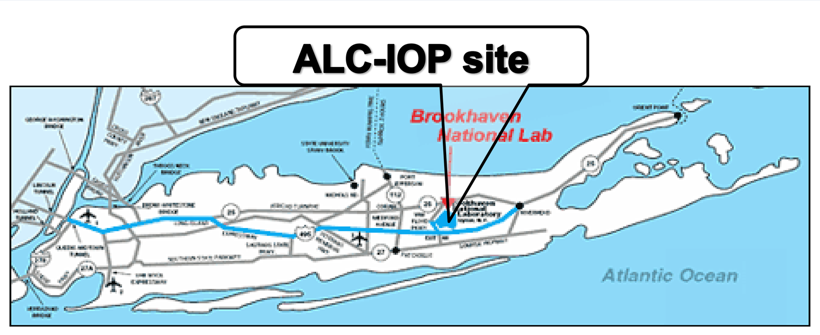
Introduction & Motivation

- Aerosol indirect effects, which describe the influences of aerosol on climate through modifying cloud properties, remain the most uncertain components in forcing of climate change over the industrial period.
- The formation of cloud droplets from aerosol particles is kinetically controlled by the availability of water vapor, equilibrium water vapor pressure above the growing droplet surface, and both the gas phase and aerosol phase mass transfer resistances.
- It has been hypothesized that the formation of surface organic films or the delay in dissolution of solute could significantly delay the growth of cloud droplets, therefore influence the cloud formation, life time and precipitation.
- In this study, we examine the droplet growth kinetics using data collected in three recent field campaigns.

Sampling Locations and Periods

http://pubs.acs.org/cen/coverstory/89/8906cover.html



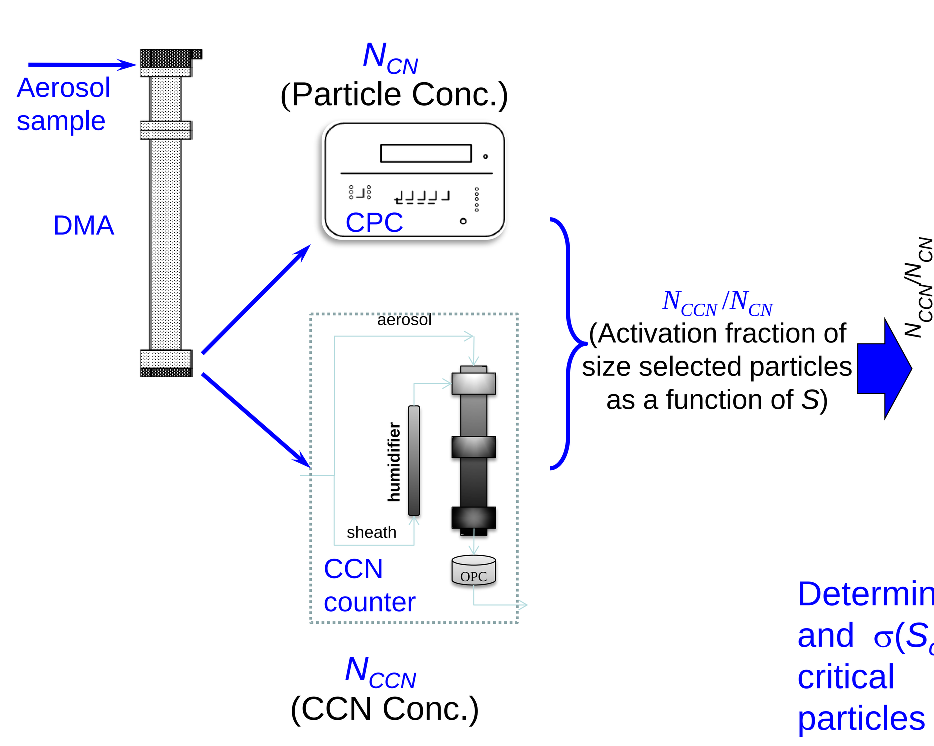
CalNex-LA site
•05/16/2010 -- 06/05/2010

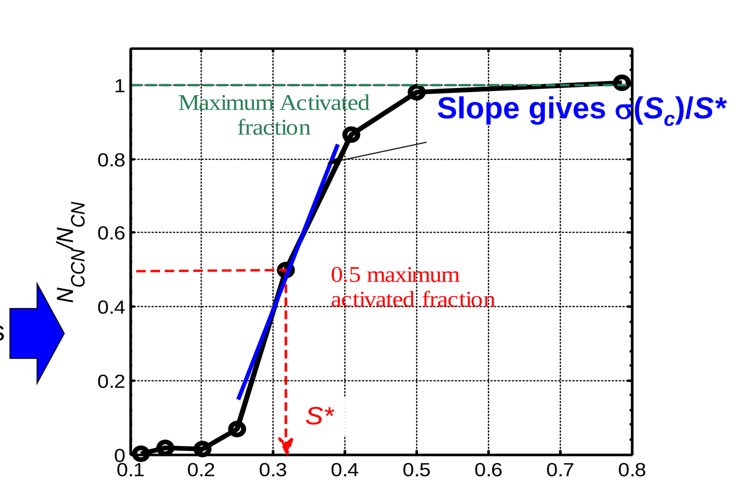
CARES site
•06/10/2010 -- 06/28/2010

ALC-IOP site
•07/04/2011 -- 08/15/2011

Experimental setup

Measurements of size-resolved CCN spectrum





Determine characteristic supersaturation S^* and $\sigma(S_c)/S^*$, which reflects heterogeneity in critical supersaturation of size selected particles (e.g., due to difference in particle composition)

General Approach

➢Droplet growth inside CCN counter

Cloud droplet growth rate inside CCN counter ⇨

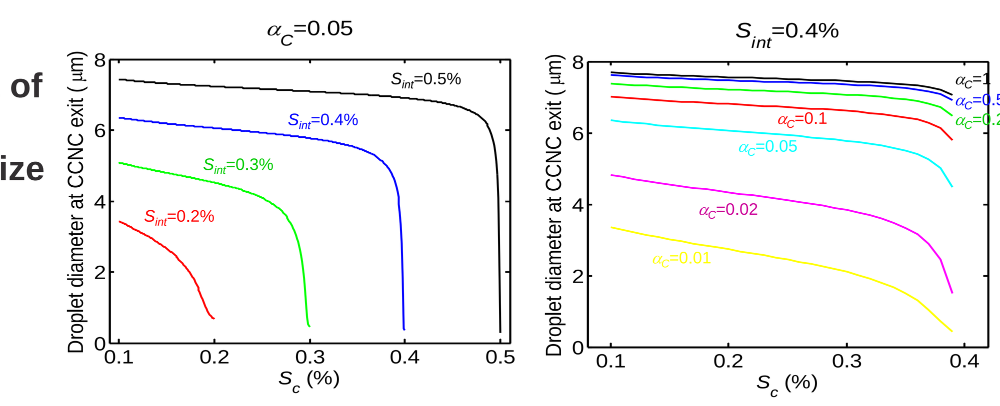
$$\frac{dD}{dt}=\frac{S_{int}-S_{eq}(D,S_c)}{D\cdot G(\alpha_C)}$$

Instrument supersaturation

Equilibrium S (S_{eq}) over growing droplet, a function of D and S_c of CCN

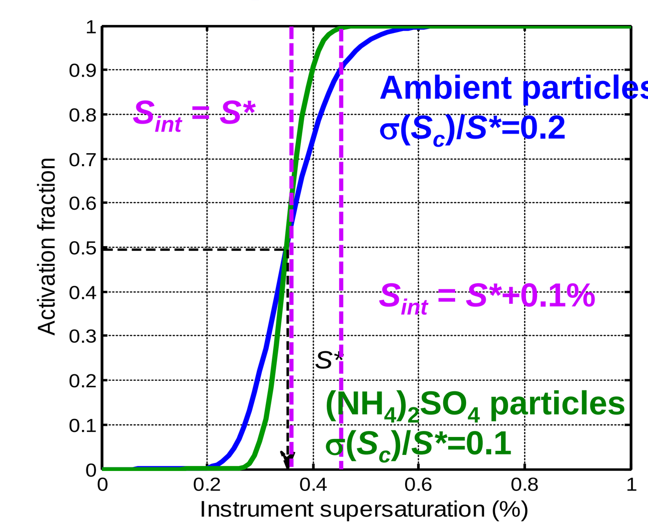
Mass transfer resistance is a function of water accommodation coefficient α_c

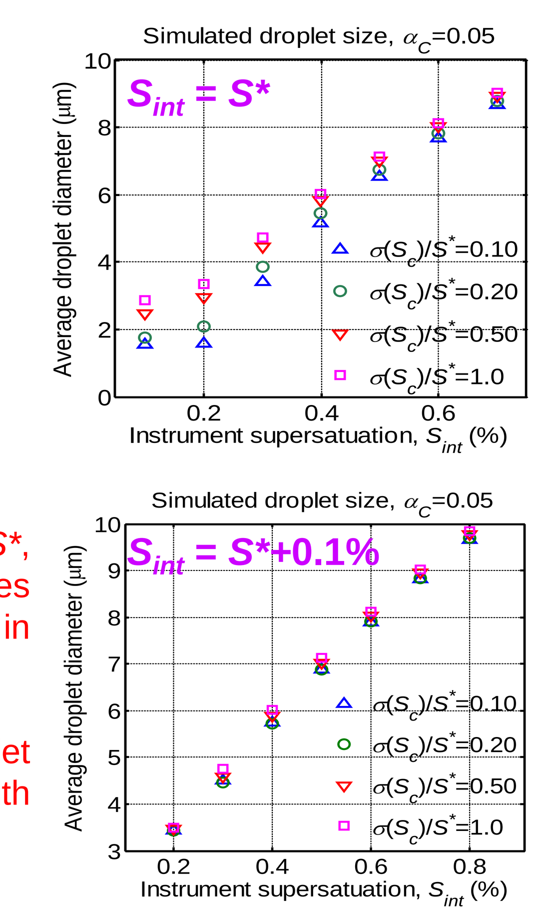
- **Use $(NH_4)_2SO_4$ particles (no organic film) as standards.**
- **To isolate the potential impact of α_c on droplet growth rate, we need to compare the droplet size of ambient particles to that of $(NH_4)_2SO_4$ particles with the same S_c, and exposed to the identical S_{int}.**



Results

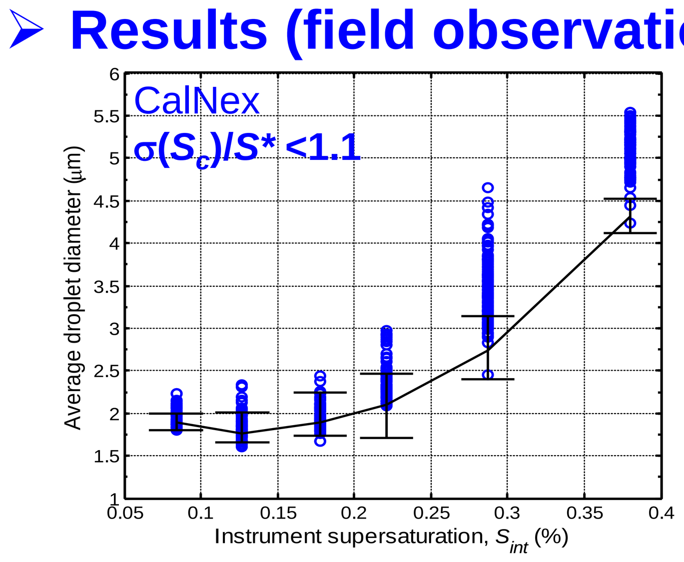
➢Results (simulations of droplet growth)

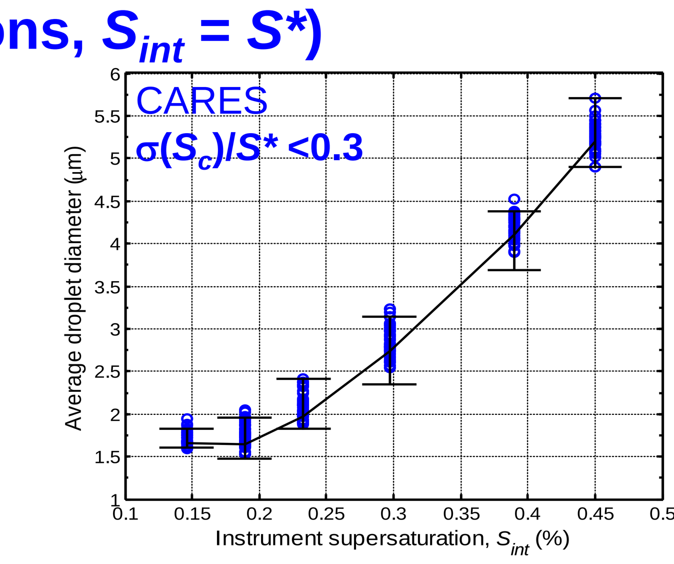




- When S_{int} = S^*, for aerosols with identical S^*, but different $\sigma(S_c)/S^*$, the average droplet sizes are different, which may mask the difference in droplet sizes due to potential growth delay.
- When S_{int} = S^*+0.1%, the average droplet sizes are essentially the same for aerosols with identical S^*, but different $\sigma(S_c)/S^*$.

➢ Results (field observations, S_{int} = S^*)

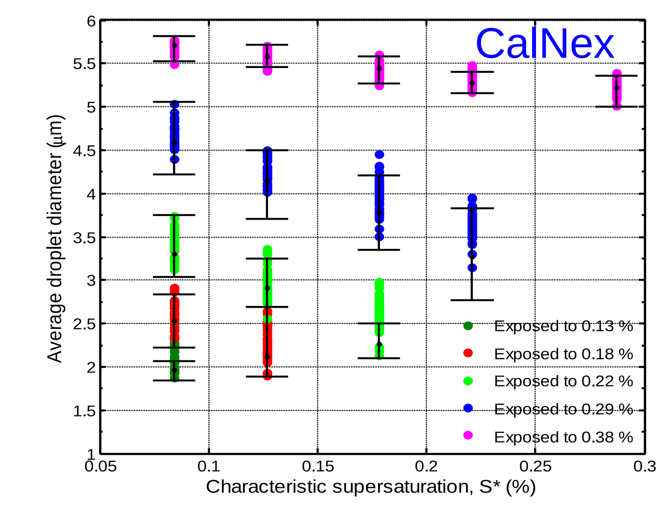


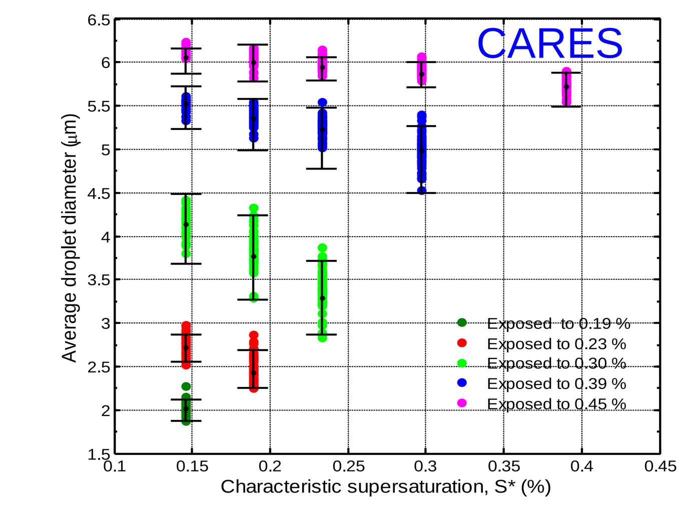


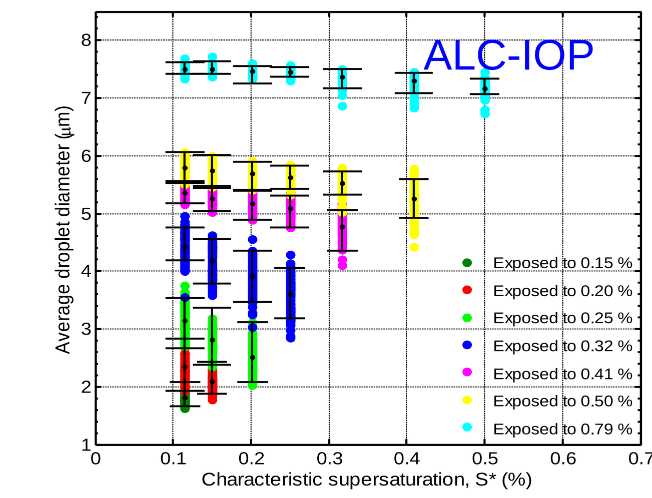
- When S_{int}=S^*, ambient particles sampled at the CalNex-LA site often grew to substantially larger average sizes (blue circles) than $(NH_4)_2SO_4$ particles with the same S_{int} and S^* (indicated by black line with uncertainty). This is due to much larger values of $\sigma(S_c)/S^*$ for ambient particles and is consistent with the simulation above. This also suggests that examining droplet sizes under S_{int} > S^* is a better approach to identify potential delay in droplet growth.
- For particles sampled during CARES, $\sigma(S_c)/S^*$ was much lower, and the average droplet sizes were mostly in agreement with those of $(NH_4)_2SO_4$ particles.

Results

➢ Results (field observations, S_{int} > S^*)







For particles from various representative sources sampled during the three field campaigns, the average droplet sizes were essentially the same as those of $(NH_4)_2SO_4$ calibration aerosol (with the same S_{int} and S^*), suggesting no significant delay in droplet growth.

Conclusions

- When S_{int} = S^*, the average droplet size for size-selected ambient particles with the same S^* and α_C is larger than that for $(NH_4)_2SO_4$ particles (due to higher $\sigma(S_c)/S^*$).
- This may mask reduced droplet growth due to lower α_C value (i.e, formation of compressed organic film).
- Comparing the droplet sizes at S_{int} > S^* + 0.1% allows us to better isolate the impact of α_C on average droplet size and identify potential slow droplet growth.
- By comparing the average droplet sizes of size-selected ambient particles to those of $(NH_4)_2SO_4$ calibration particles (for the same S_{int} and S^*), we show that there were no significant delay in droplet growth for the aerosol particles from a variety of sources observed during the three field campaigns.

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ASR Atmospheric System Research