Organic Coating of Aerosol Particles Observed during ACTIVATE and its Potential Impact

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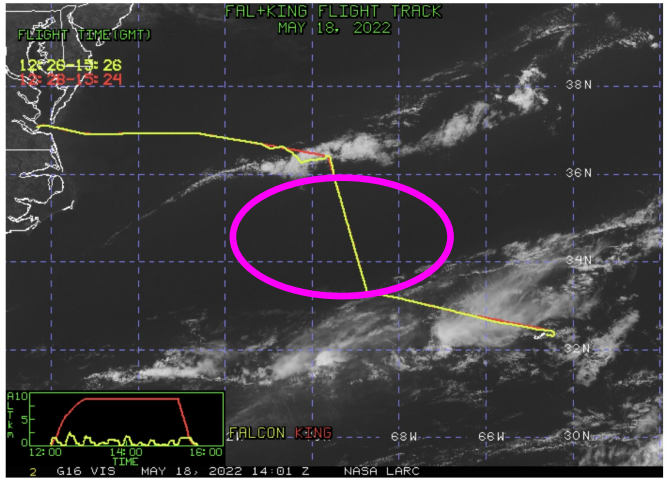
Two aerosol samples collected during three flights

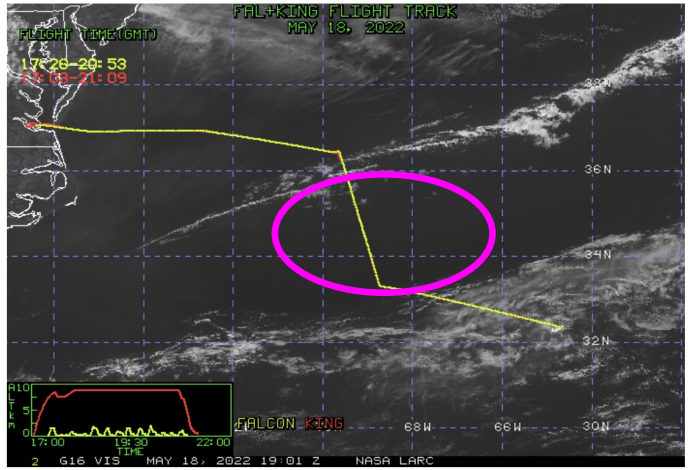
RF156 (5/18/22, AM)

Visible Image



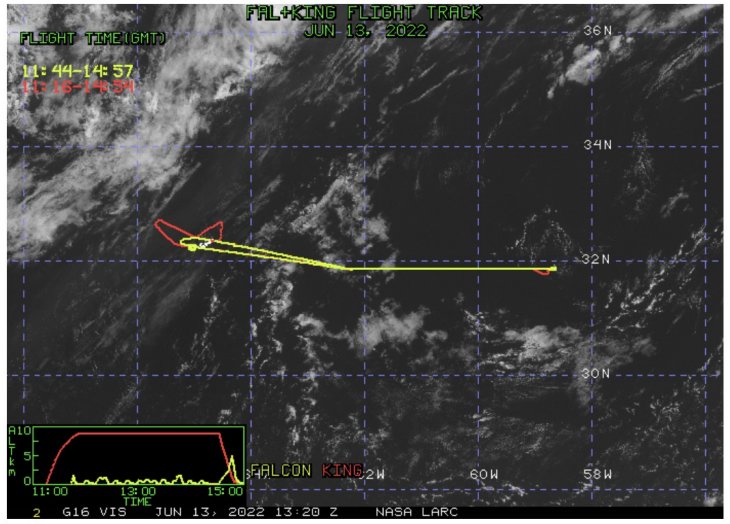
RF157 (5/18/22, PM)

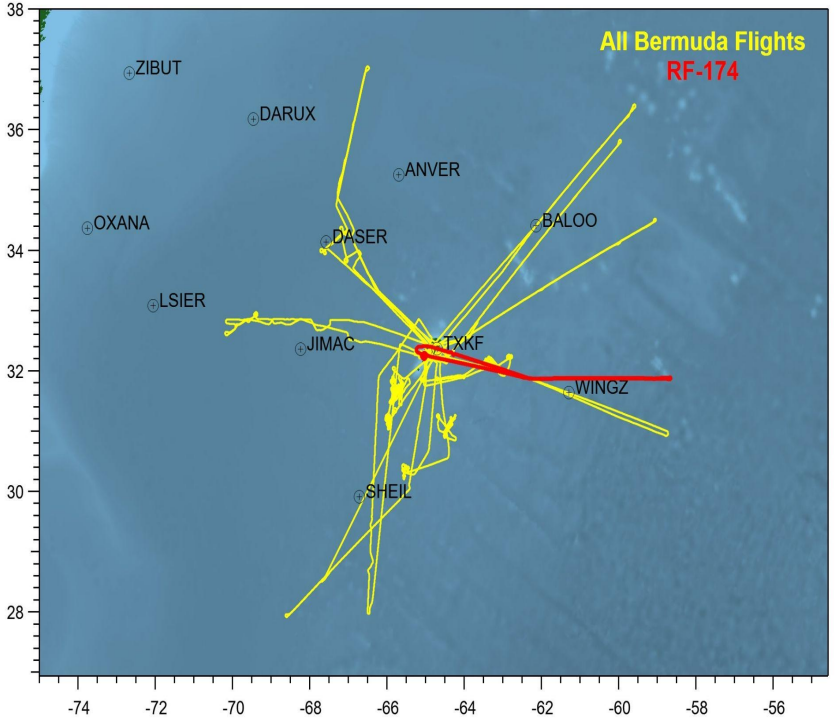
Visible Image



RF174 (6/13/22)

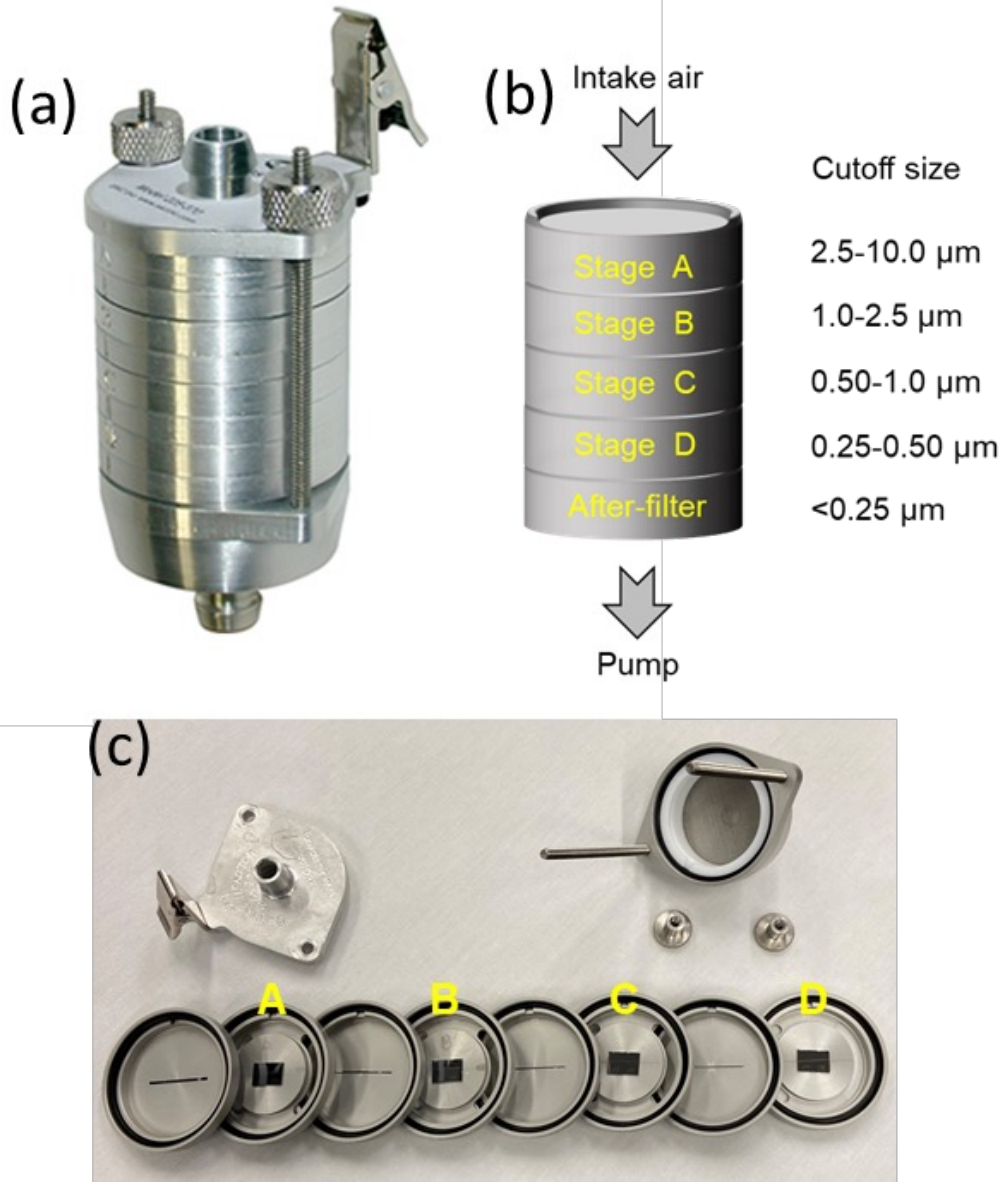
Visible Image





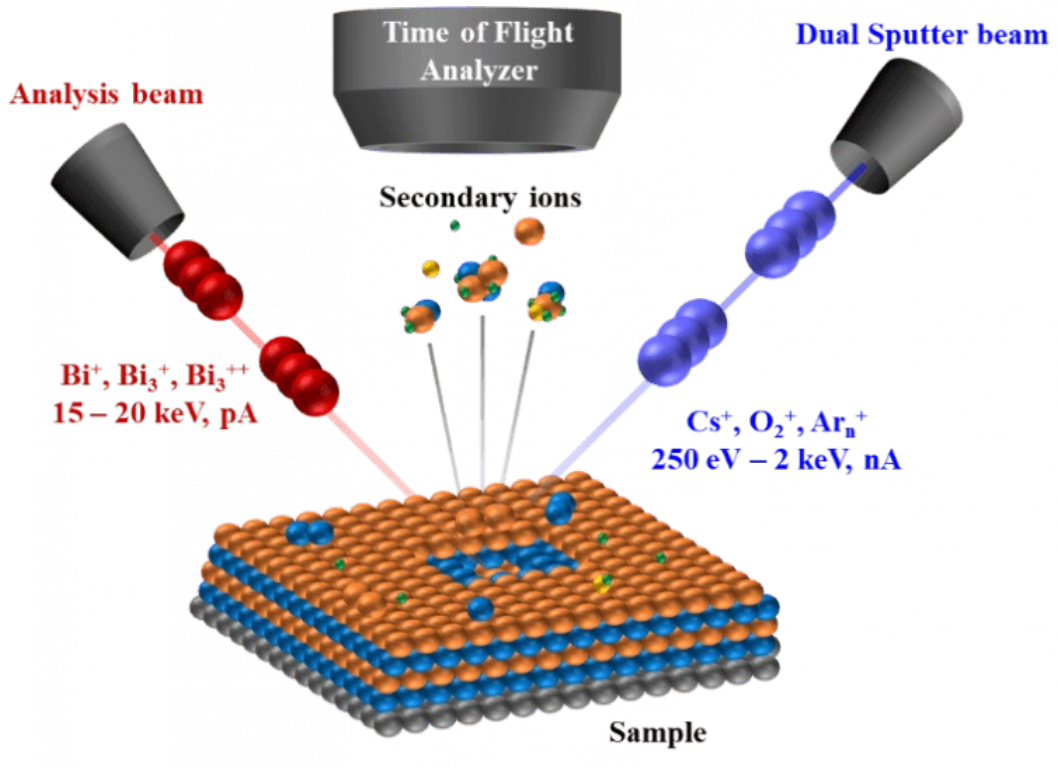
Aerosols collected using cascade impactor for chemical imaging analysis

- Cascade impactor with 4 stages and decreasing cutoff diameters collecting particles on 5x7mm silicon wafers (**F. Mei**)
- Used for Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) surface analysis (**Z. Zhu**)



ToF-SIMS surface spectrometry and imaging

- ToF-SIMS is used to obtain elemental composition and molecular information on a surface and in-depth (i.e., 3-D chemical composition)
- Localized information with a beam spot size of about 100 nm and sputtering depth of 5-10 nm

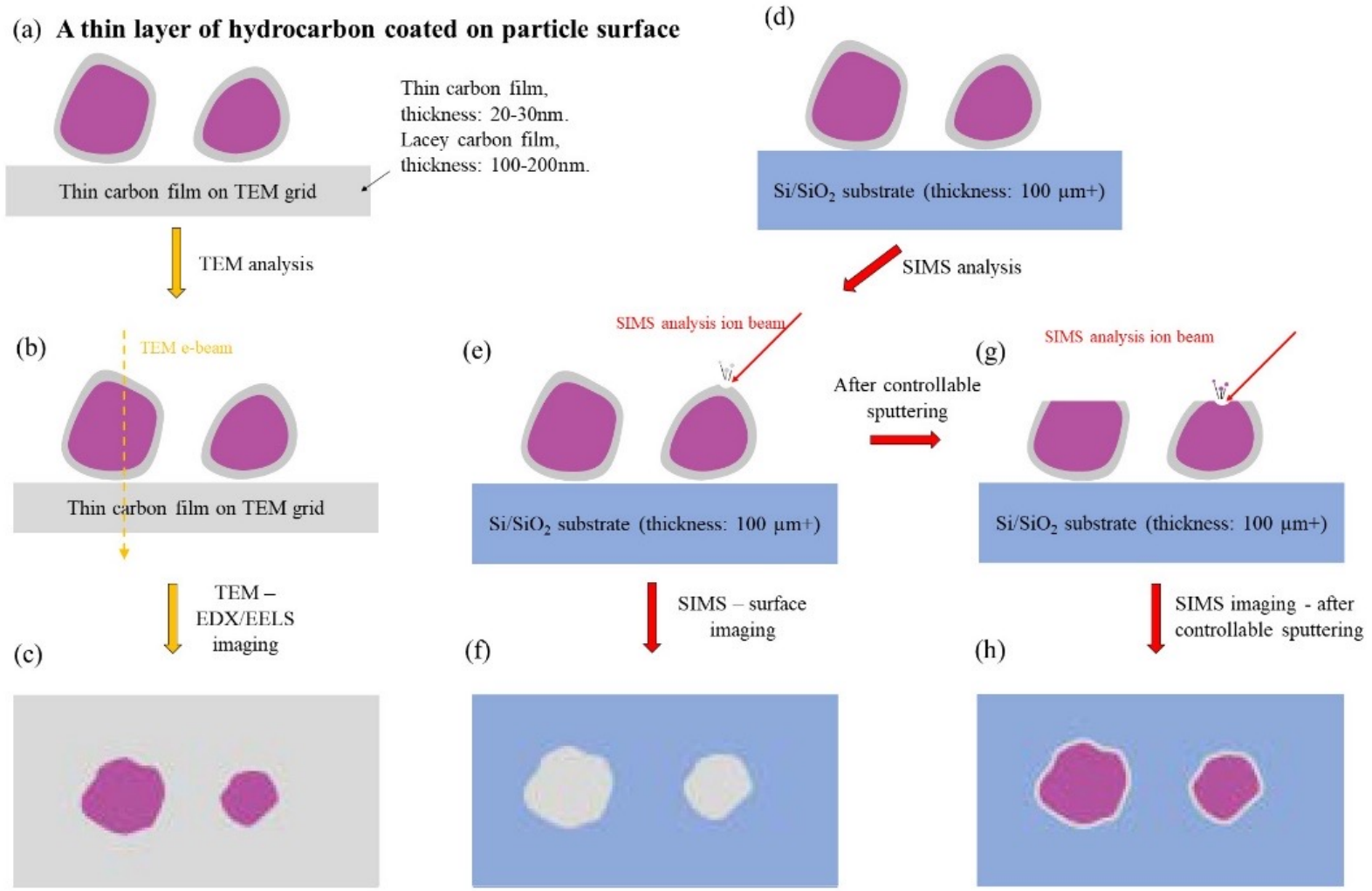


Schematic from Rice University

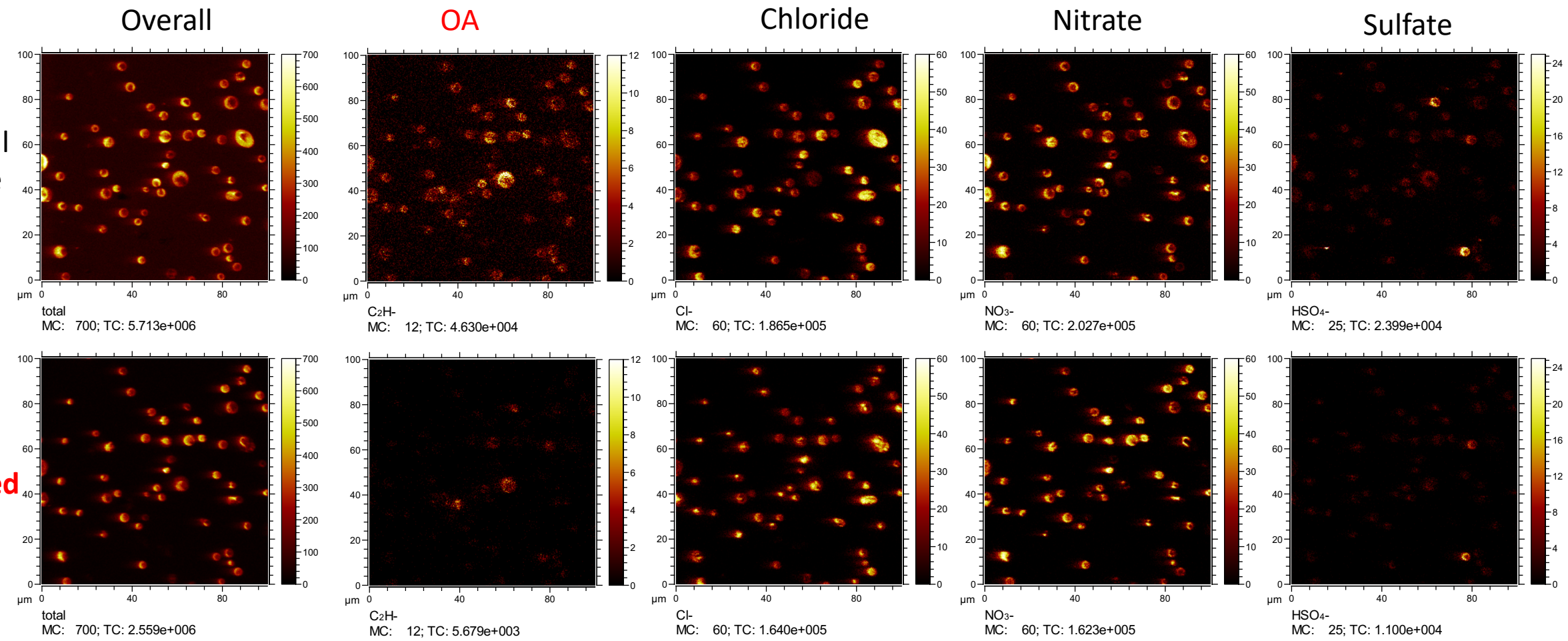
How does SIMS detect thin organic coating layer?

(Y. Li et al., 2023 EST)

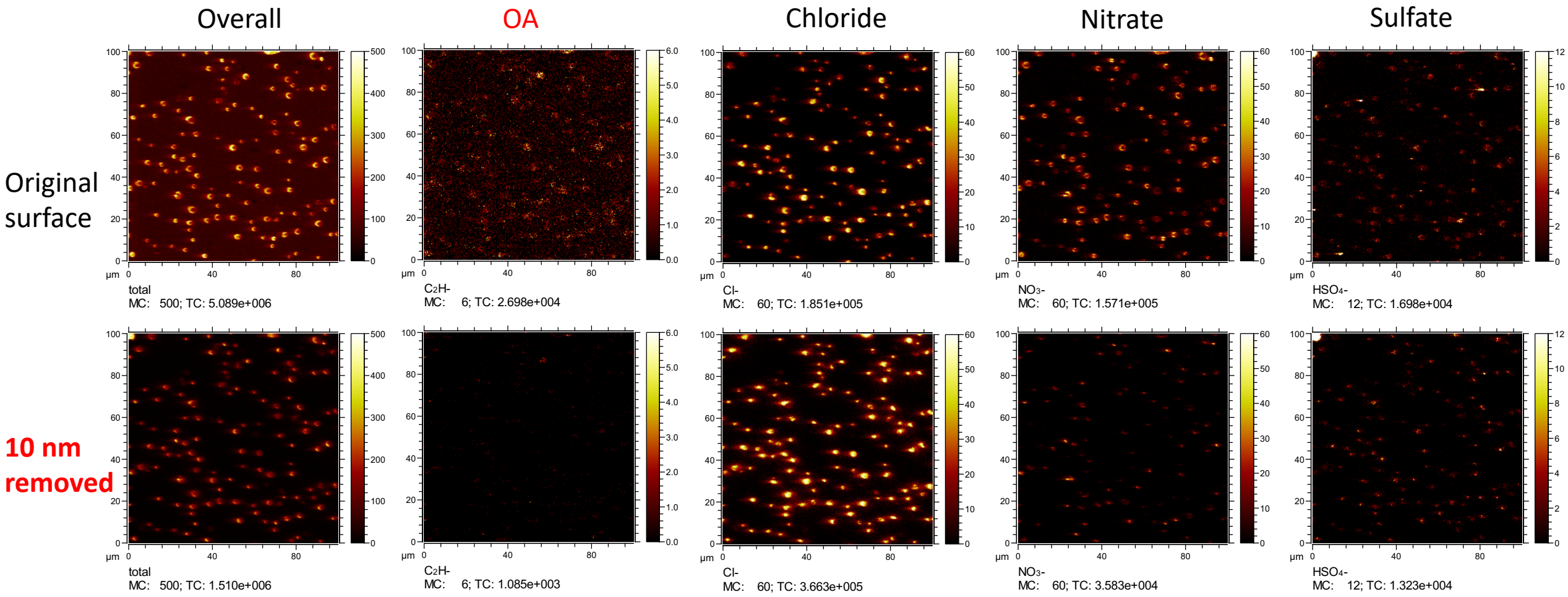
- SIMS is different from TEM (transmission electron microscopy) for particle imaging
- TEM beams pass through the thin coating layer
- SIMS has shallow information depth and controllable sputtering (removing 5-10 nm layer at a time)



ToF-SIMS surface imaging of RF156/157 sample

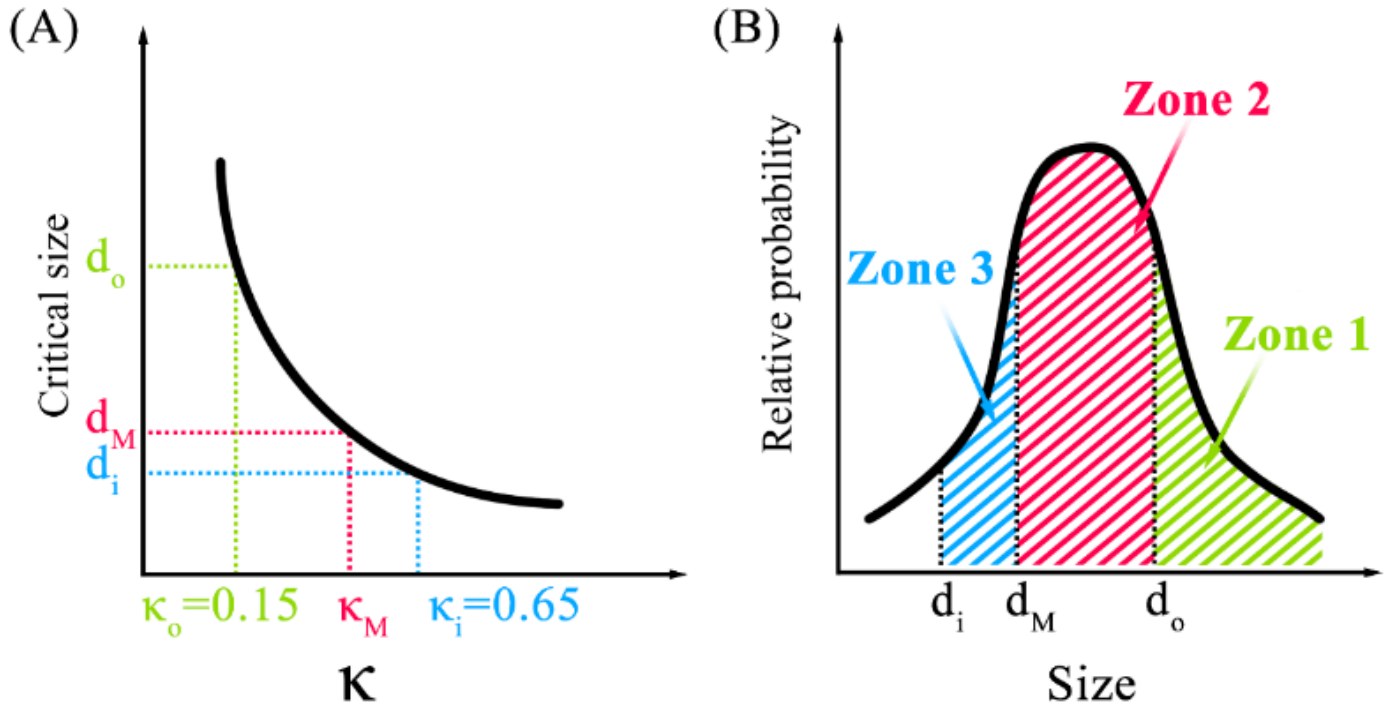


SIMS surface imaging of Bermuda (RF174) sample



Impact of surface chemistry on aerosol activation

- Inorganic aerosol has a larger kappa (k_i) and smaller critical size (d_i) than organic aerosols (k_o, d_o) for activation at a certain SS
- A mean k_m between k_i and k_o is used in ESMs with internal mixing assumption
- Particles in zone 1 and zone 2 are activated, but with organic-shell-inorganic-core structure only zone 1 is activated



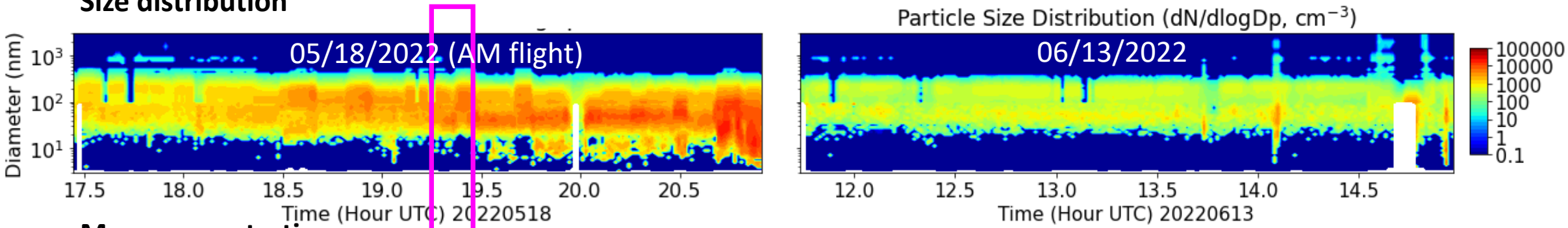
(Y. Li et al., 2023 EST)

CCN closure analysis based on ACTIVATE measurements

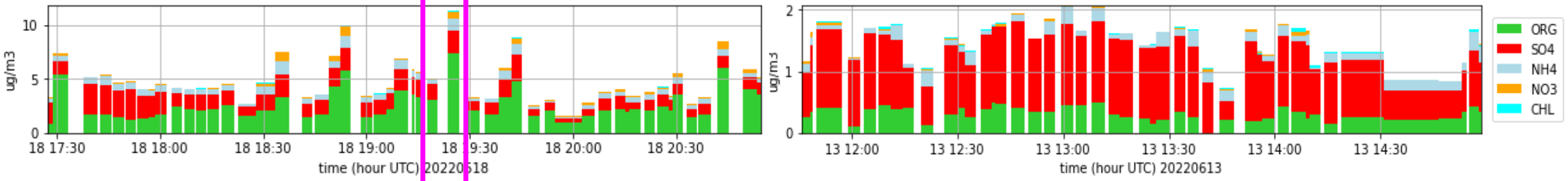
- **Hypothesis**: most aerosol particles are coated with organics, having smaller kappa
  - Kappa derived from chemical composition mass fractions is greater than measured
  - Analyzing **CCN closure** instead due to large uncertainties in estimating kappa from CN and CCN
- **ACTIVATE data**: mass of aerosol components (AMS, PILS), CN size distribution (LAS, SMPS) and CCN (@0.1-1%, 0.37% SS)
- **Method**: Calculating CCN number from CN size distribution and kappa derived from chemical composition and comparing with measured CCN at the same SS
- **Expectation**: Calculated CCN > Measured CCN

ACTIVATE measurements for the two cases

Size distribution

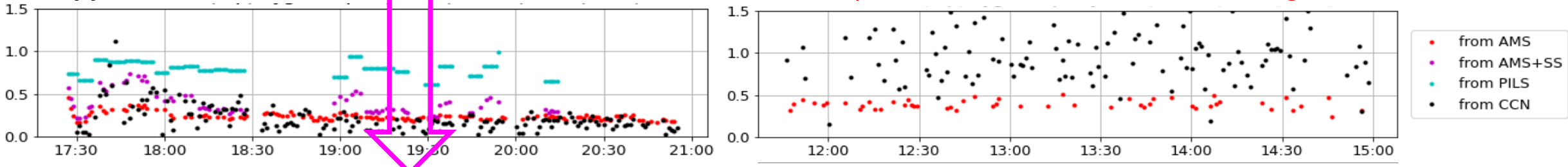


Mass concentration



Kappa

Fewer particles and smaller fraction of organics

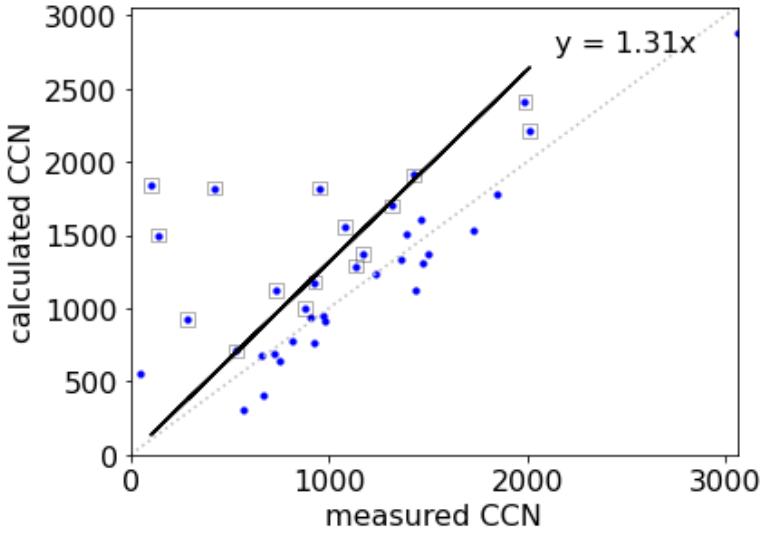


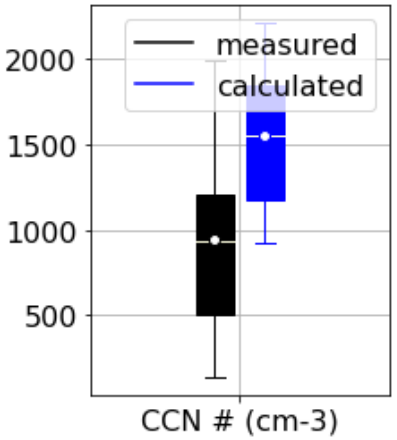
Kappa=0.108 (HSRL-2, 532nm)

0.195 (PM), 0.32 (AM), 0.15 (in situ)

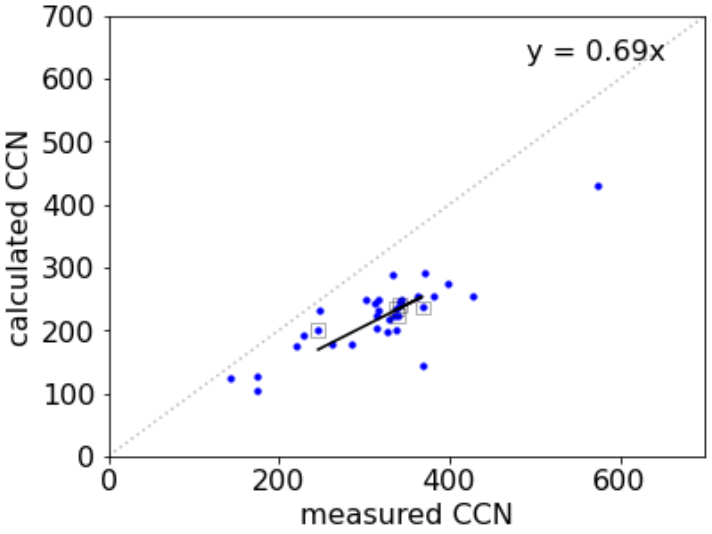
CCN closure for the two cases

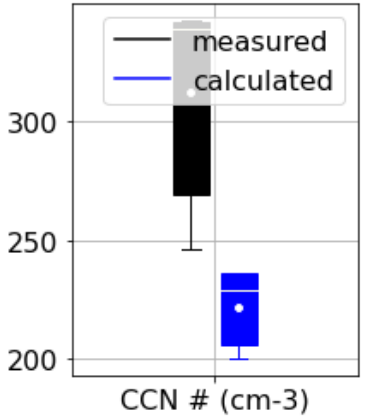
05/18/2022 (two RFs)

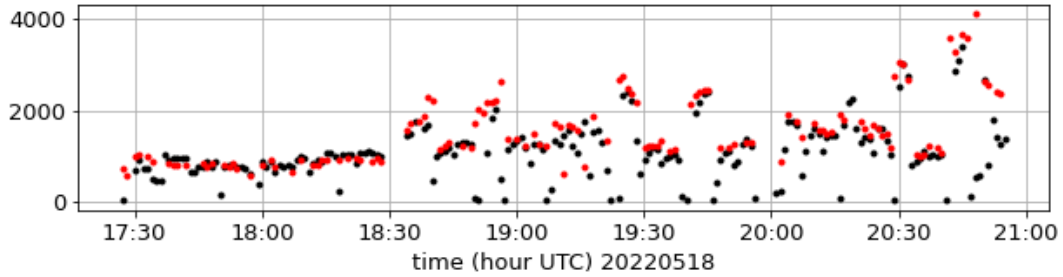


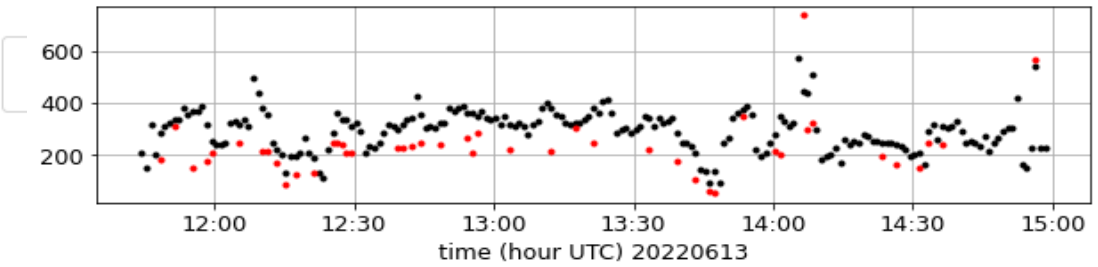


06/13/2022







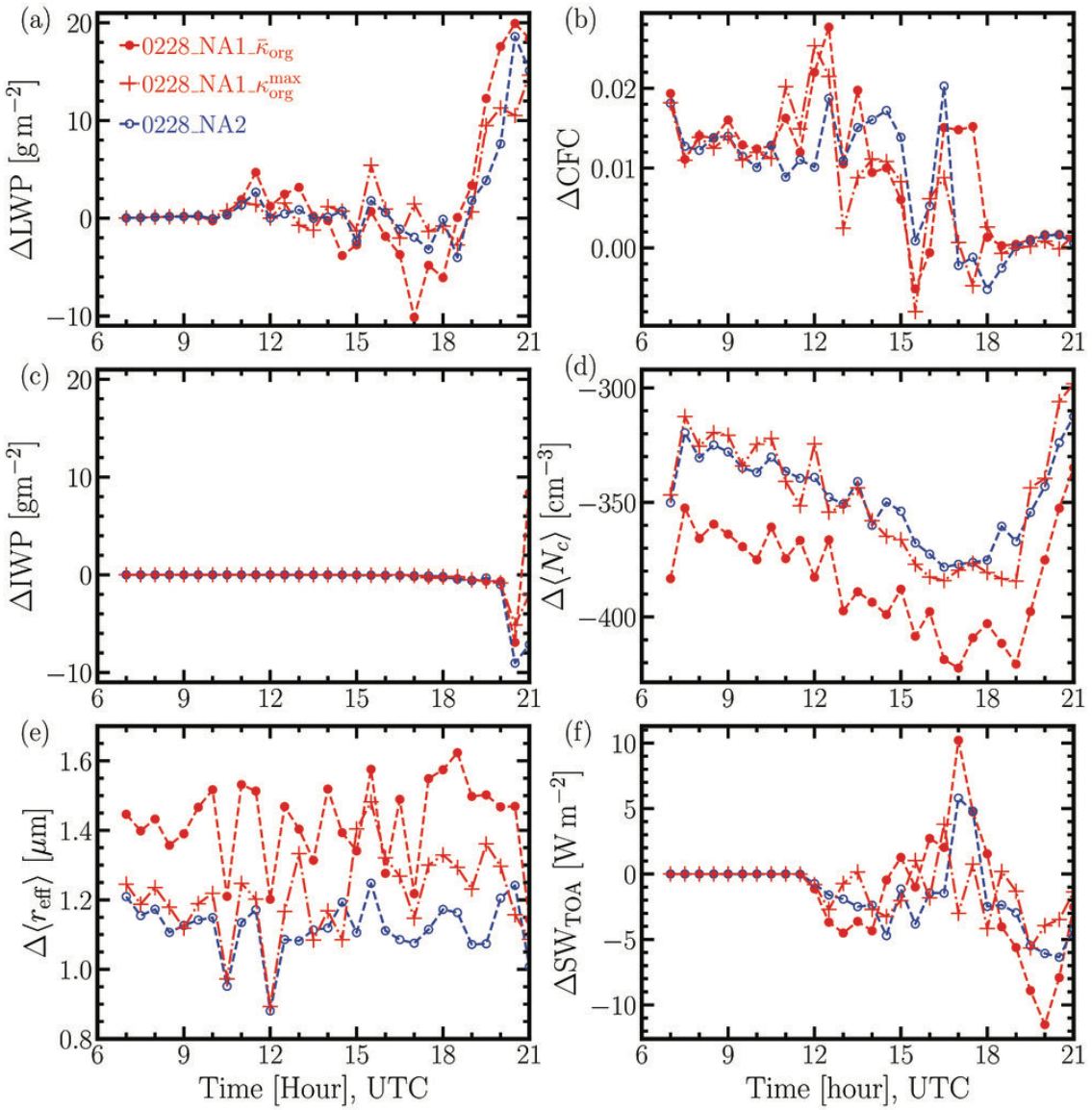


✓ calculated CCN > Measured CCN

Changes in k_o make a difference to LES simulated cloud properties

- About 50% of organic components in a CAO case (2/28/2020)
- k_o (mean=0.1, max=0.229), leading to k_m= 0.31 and k_{max}=0.39
- A significant impact on Nc, r_e , LWP and SW cloud forcing
- Even strong impact when $k = k_{min}$

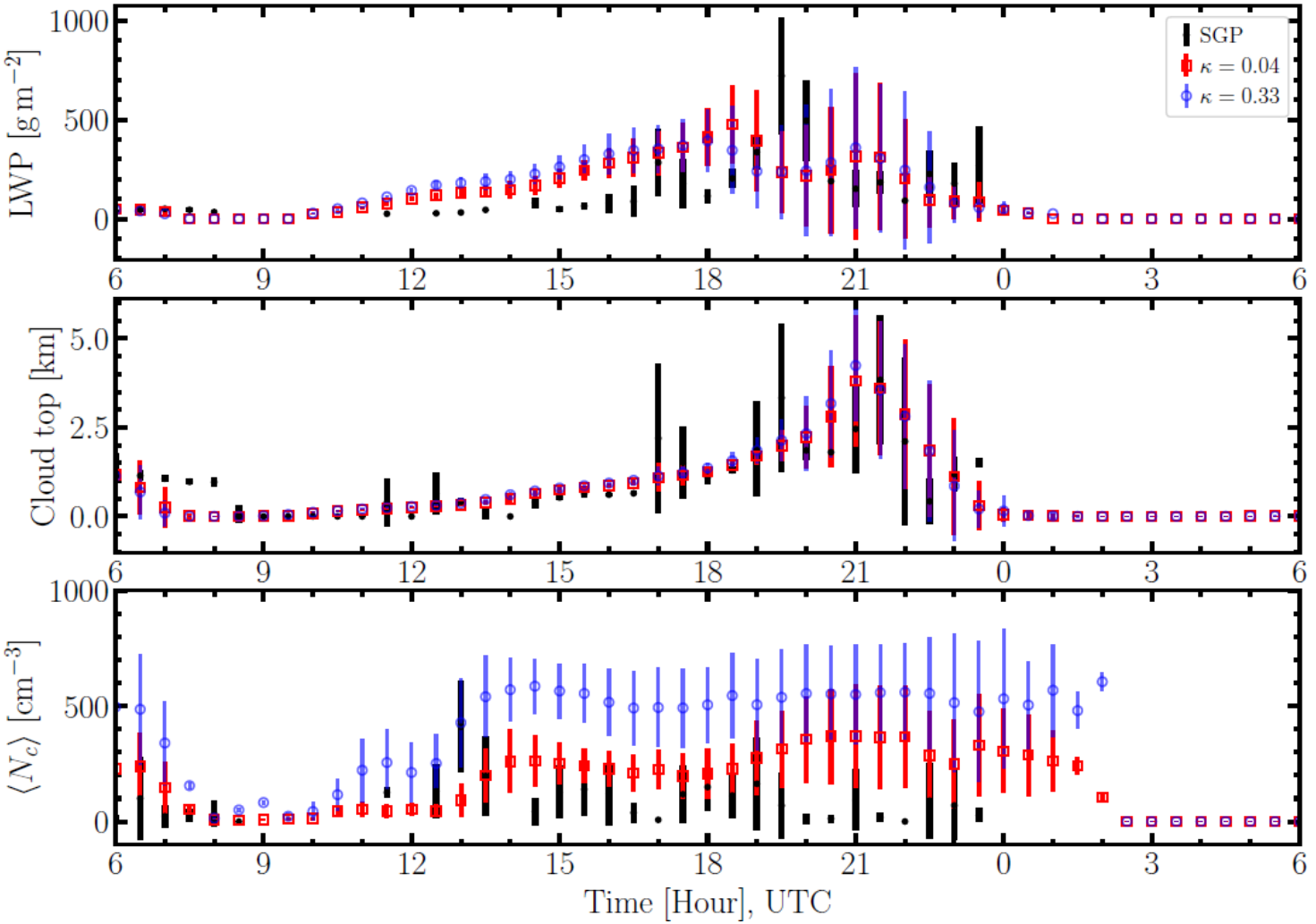
(X. Li et al., 2023 JAS)



Sensitivity of clouds to aerosol chemistry at ARM SGP site

- LWP and cloud top are not sensitive to kappa (k_m=0.33 versus k_{min} = 0.04), compared well with ARM measurements
- N_c (r_e) is much smaller (larger) when k = k_{min}, closer to ARM measurements
- 12-h mean SW cooling is 1.2 W m^{-2} weaker (instant as large as 10 W m^{-2})

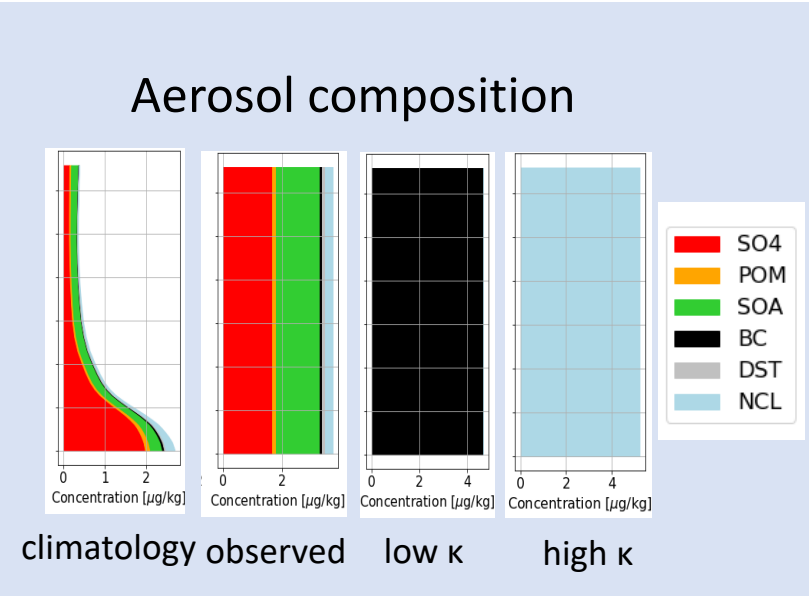
WRF-LES simulations

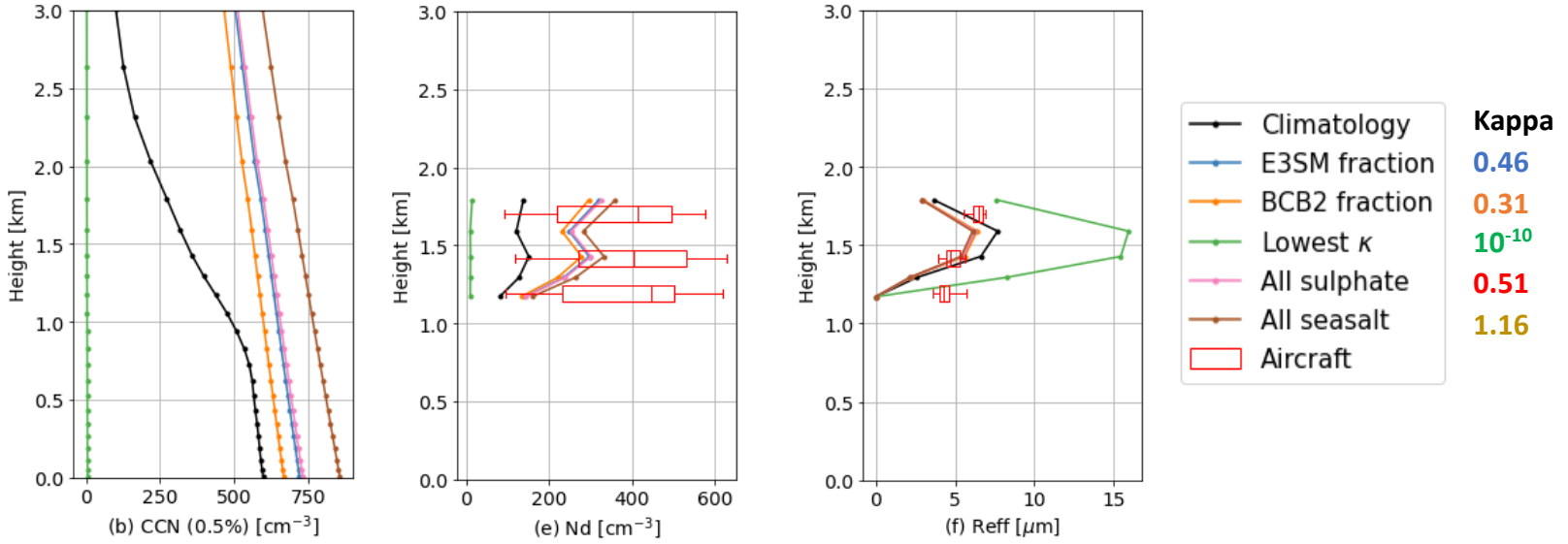


(Mei et al., in review for BAMS)

Single-column E3SM sensitivity tests on kappa

(Tang et al., To Be Submitted)





- Cold air outbreak case (March 1, 2020)
- CCN (0.5% ss), N_d and r_e are sensitive to the kappa values from assumed aerosol composition
- Much smaller impact on LWC, except for the lowest k
- The lowest k pushes N_d to an artificial lower bound (10 cm^{-3}), having an impact on precipitation

Summary

- Chemical analysis reveals organic coating structure of ACTIVATE aerosols
- Hygroscopicity (kappa) and CCN closure analysis shows an important impact of the organic coating
  - Kappa estimation is subject to large uncertainty and algorithm limitation
  - Estimated CCN from aerosol composition and size distribution measurements are compared to CCN measurements
  - The 5-18-2022 case shows clear overestimation of CCN, while the 6-13-2022 case shows the opposite, likely due to too small organic fraction and/or too clean condition
  - 2020-2022 flights overall show an overestimation of CCN during some legs (Takeoff/Landing, Ascent, ACT, ABL and BBL)
  - Results are sensitive to supersaturation (larger discrepancy in CCN at lower SS)
- LES, SCM and GCM show sensitivities of clouds to kappa changes