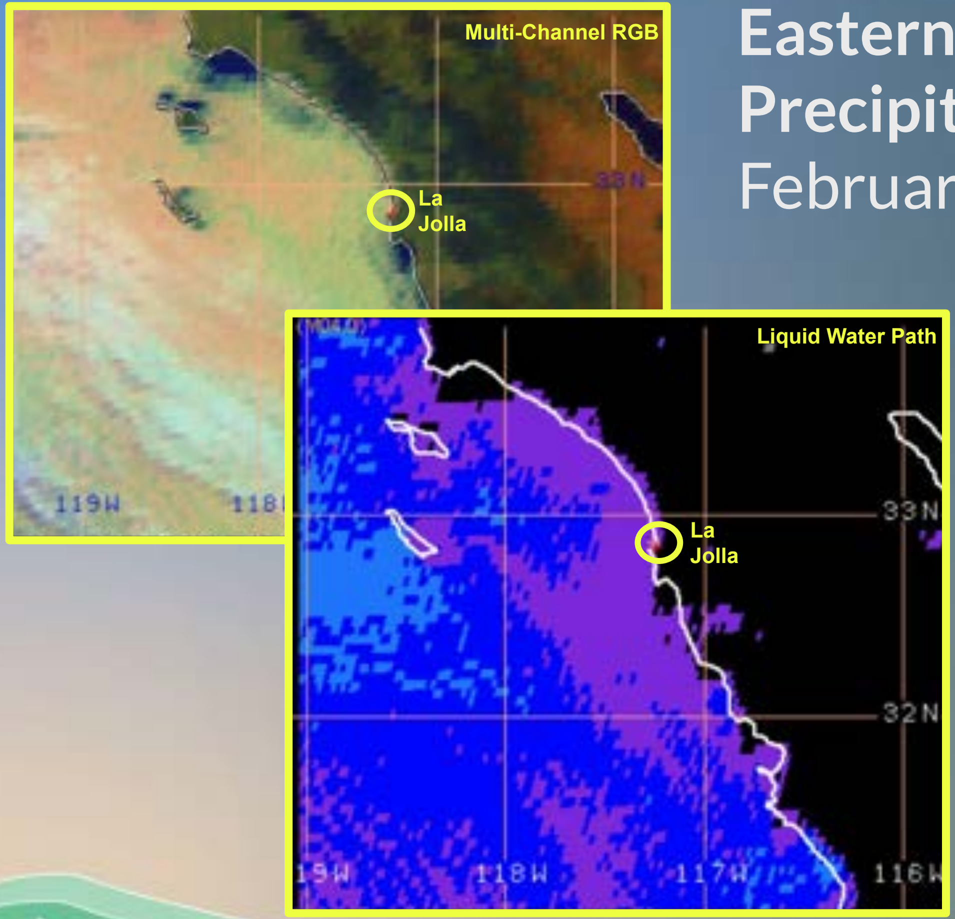
Factors Affecting Aerosol Activation to Cloud Droplets and Aerosol Growth by Cloud Processing at EPCAPE

Lynn Russell of Scripps Institution of Oceanography
with contributions from the **EPCAPE Science Team**



Eastern Pacific Cloud Aerosol Precipitation Experiment (EPCAPE)

February 2023 through February 2024

Scientific Objectives:

- Characterize aerosol and <u>cloud</u> climatology
- Quantify aerosol-<u>cloud</u> interactions
- Measure <u>cloud</u> radiative fluxes

Frequent Coastal Cloud Coverage

Prior Aerosol Studies at La Jolla:

- Hawkins and Russell, 2010 *Atmos. Env.*
- S. Liu et al., 2011 *Atmos. Chem. Phys.*
- Day et al., 2011 *Atmos. Env.*
- R. Zhao et al., 2014 *Geophys. Res. Lett.*
- Modini et al., 2015 *JGR Atmos.*
- Sanchez et al., 2016 *JGR Atmos.*

SATCorps.larc.nasa.gov by Painemal and Thieman

Simultaneous sampling of aerosol below cloud at Pier and in cloud (**800+ hr**) at Soledad shows aerosol activation and cloud processing.

EPCAPE Measurements

ARM AMF1 with SACR and AOS (mostly at Pier):
Proposal Team: Lynn Russell, Markus Petters, Mark Miller, Dan Lubin, Israel Silber, Ed Eloranta, Johannes Muelmenstaedt, Susannah Burrows, Allison Aiken, Die Wang, Andy Ackerman, Ann Fridlind, Mikael Witte, Matt Lebsock, David Painemal, Rachel Chang, John Liggio, Michael Wheeler.

Guest PI Instruments (mostly at Soledad):
Russell – HR-ToF-AMS w/ET, DMA, APS, SP2, FTIR.
Petters – Denuded and Undenuded, Size-Resolved CCN.
Paulson – Aqueous OH^-.
Wheeler/Liggio/Wentzell/Lee – CVI, CIMS, SP2, PAX.
Chang – Fog Drop Monitor.
Smith – TD-CIMS, Nano-TDMA.
Lubin – Integrated radiometers.
Galewsky – Water vapor isotopes.
Aiken – Intensive CVI, SP-AMS, SP2.
Farmer – Particle fluxes (at Pier).
Lebsock – G-band Doppler Radar (at Pier).

EPCAPE Manuscripts

- **Silber et al., 2024 *ESSD*: ARMTRAJ back-trajectories for EPCAPE.**
- Socuellamos, Lebsock, et al., 2024 *ESSD*, *AMT*: G-band Doppler radar for drizzle.
- Yurk, Lebsock, et al., in review: G-band radar for drop size distributions.
- Rybecky and Galewsky, in review: Water vapor deuterium excess in clouds.
- **Han, Russell, et al., in review: Semi-volatile and local aerosol contributions.**
- **Maneenoi, Russell, et al., in review: Sulfate sources and formation processes.**
- **Ravichandran, Petters, et al., in review: Hygroscopicity effects of denuding.**
- **Dedrick, Russell, et al., in preparation: Aerosol feedbacks on cloud supersaturation.**
- **Williams, Russell, et al., in preparation: Size and composition effects on activation.**
- Farley, Aiken, et al., in preparation: Organic aerosol source types.
- Chellappan, Painemal, et al., in preparation: Regimes of cloud variability.
- **Kapp, Smith, et al., in preparation: Ultrafine aerosol composition in and out of cloud.**
- **Berta, Russell, et al., in preparation: Constraining organic hygroscopicity with composition.**
- **Pelayo, Russell, et al., in preparation: Separating organic aerosol contributions.**
- Zawadowicz et al., in preparation: Multi-site composition comparison.
- Lubin et al., in preparation: Surface energy balance differences for coastal ACI.
- Witte et al., in preparation: Low cloud interactions with aerosol and land.
- Pujiastuti et al., in preparation: Emission fluxes of sea spray from Doppler LIDAR.
- Zhang et al., in review: Comparisons to SCREAM modeling of clouds and water.

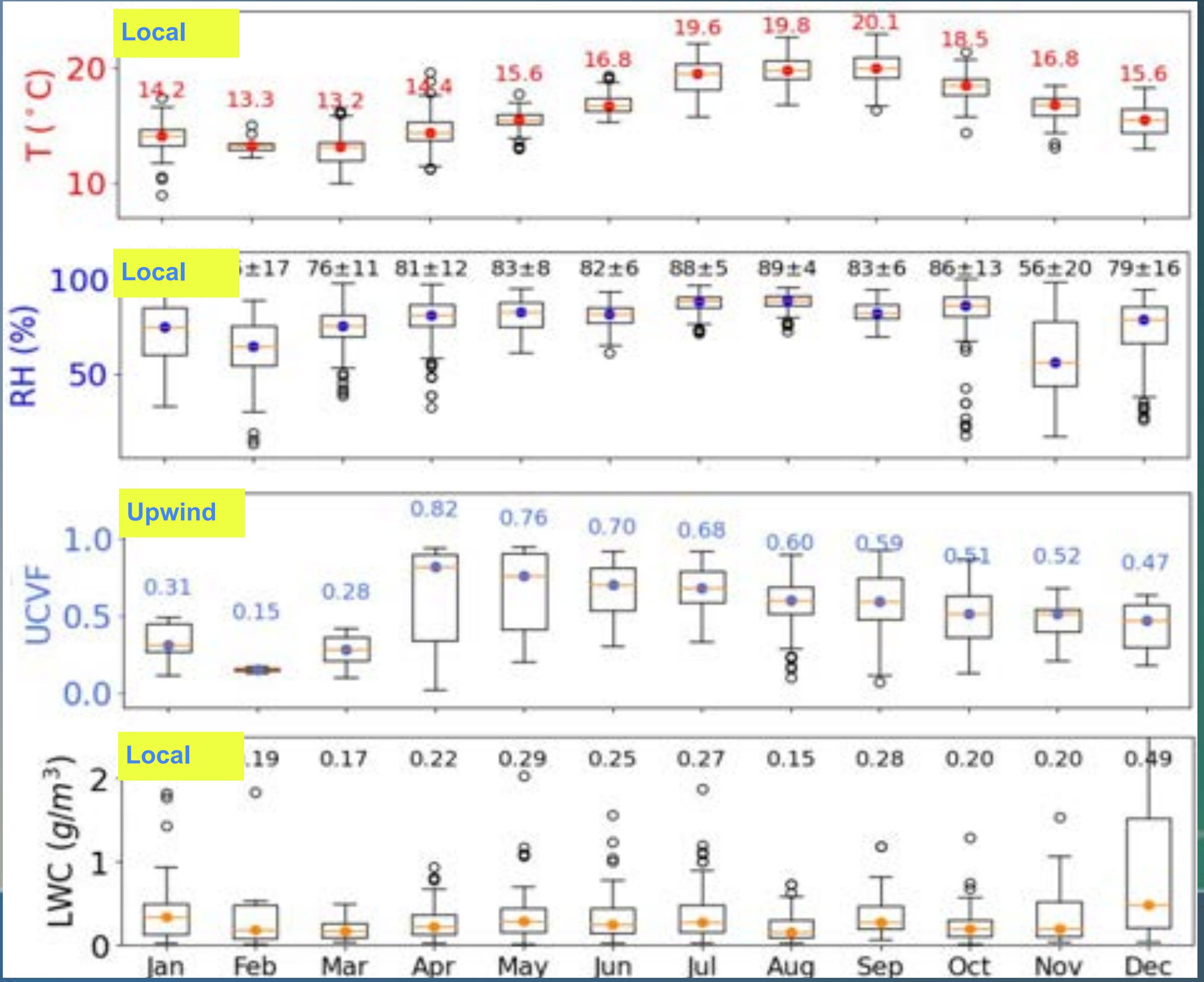
Papers in Progress!

Late Breaking Results!

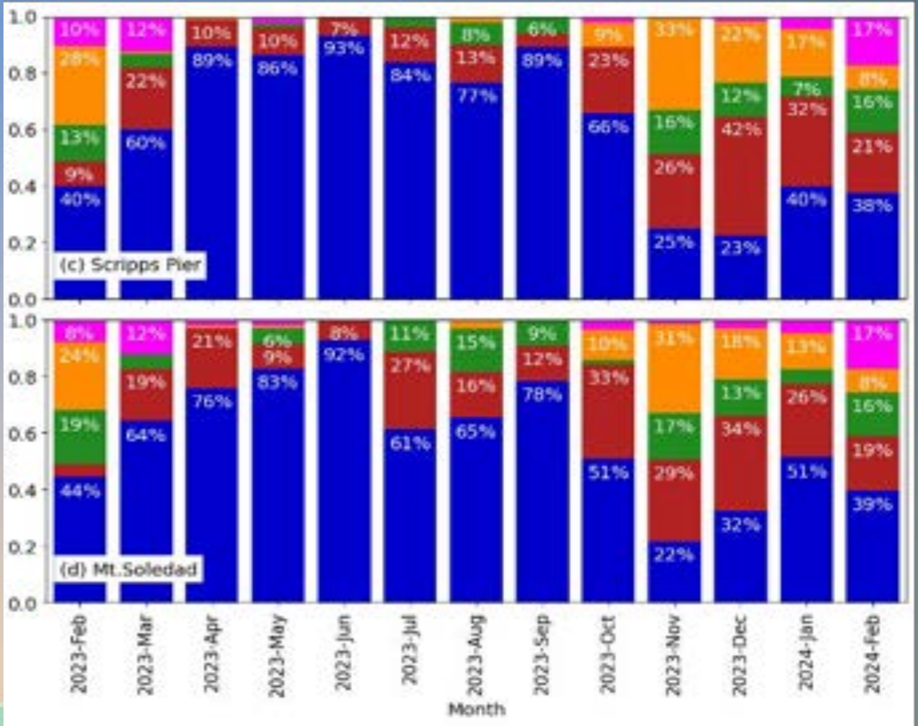
National Science Foundation | U.S. DEPARTMENT OF ENERGY | ASR Atmospheric System Research | ARM | Environment and Climate Change Canada

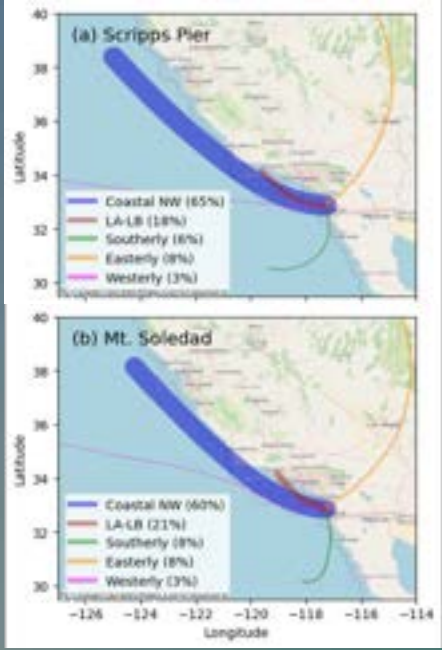
Climatology of Local and Upwind Meteorology and Clouds

UCVF=1-CBH/CTH
for 24h upwind



Climatology of ARMTRAJ Back-Trajectories





<u>Seasonal Differences</u>

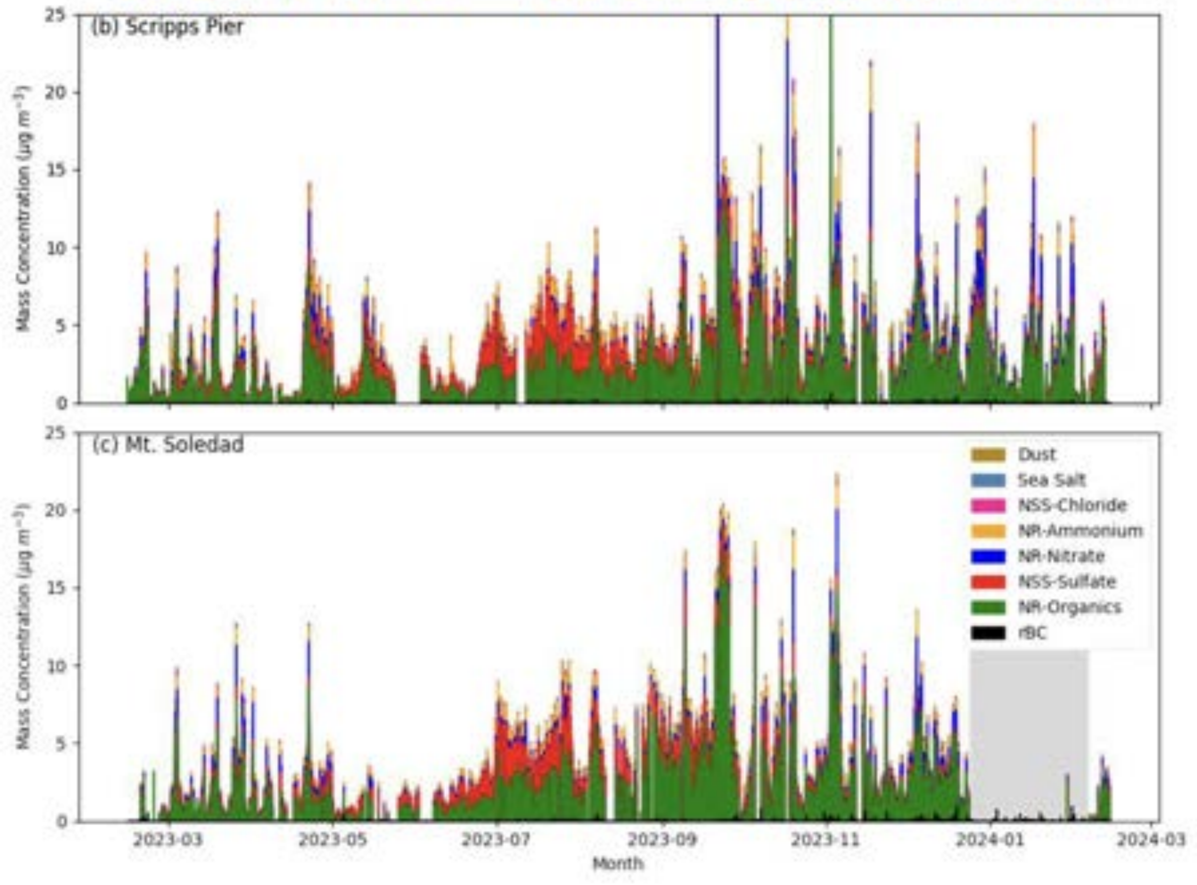
April-October:
>60% Coastal NW

November-March:
LA-LB, Easterlies,
<60% Coastal NW

See Silber et al. (ESSD, 2024)

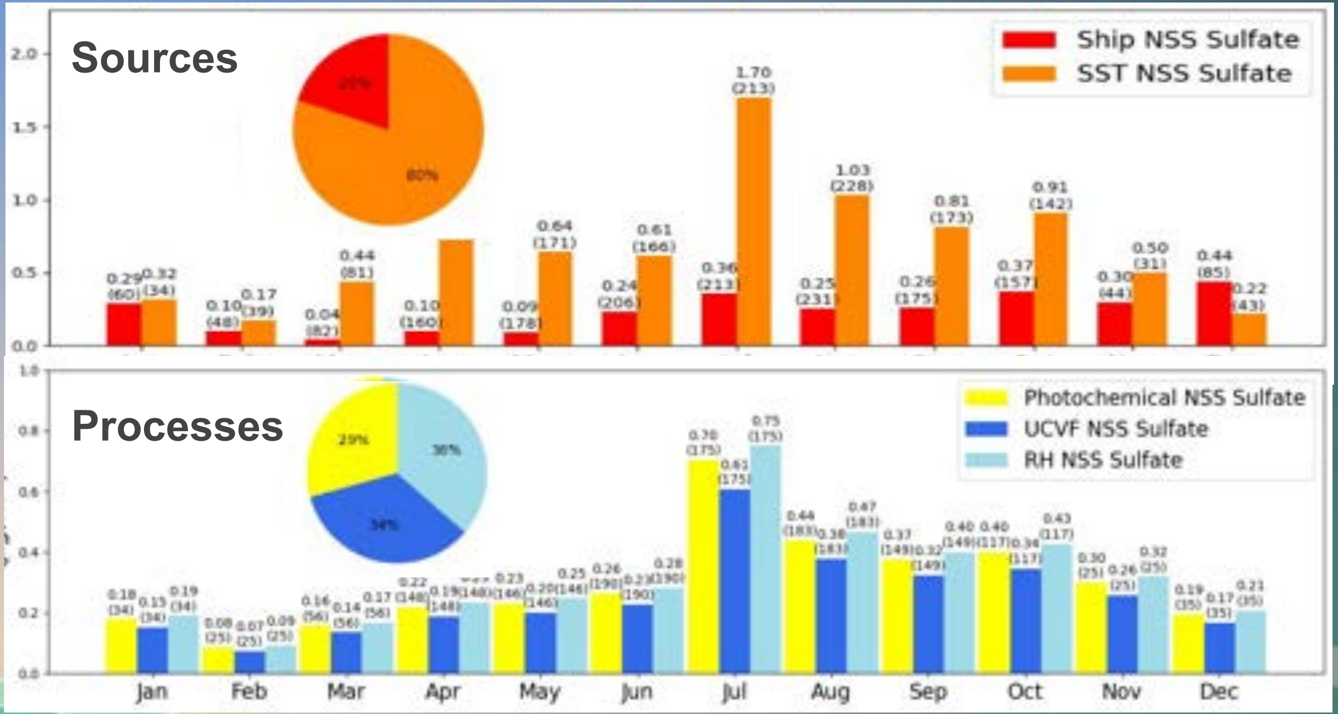
Aerosol Climatology of Composition: Large Contributions from Organics plus Summer Sulfate and Winter Nitrate

- LA-LB, Southerly, and Easterly trajectories had more than double Organic and Nitrate mass concentrations compared to Coastal NW.



See Han, Russell, et al. (In Review)

Aerosol Sulfate Sources and Cloud-Related Sulfate Oxidation Processes



Regionally-emitted sulfate sources

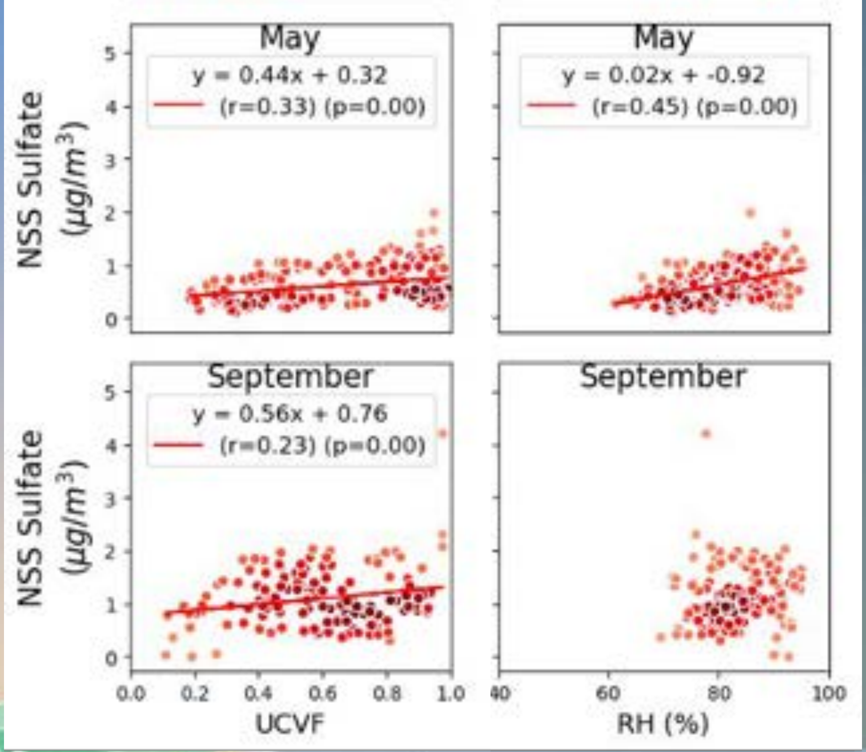
- 76-80% biogenic
- 20-24% shipping

Regional oxidation

- 29% photochemical
- 70% aqueous
  - 36% aerosol
  - 34% cloud

See Maneenoi, Russell, et al. (In Review)

Site-Specific Process Correlations Explain Variability for Regional Contributions



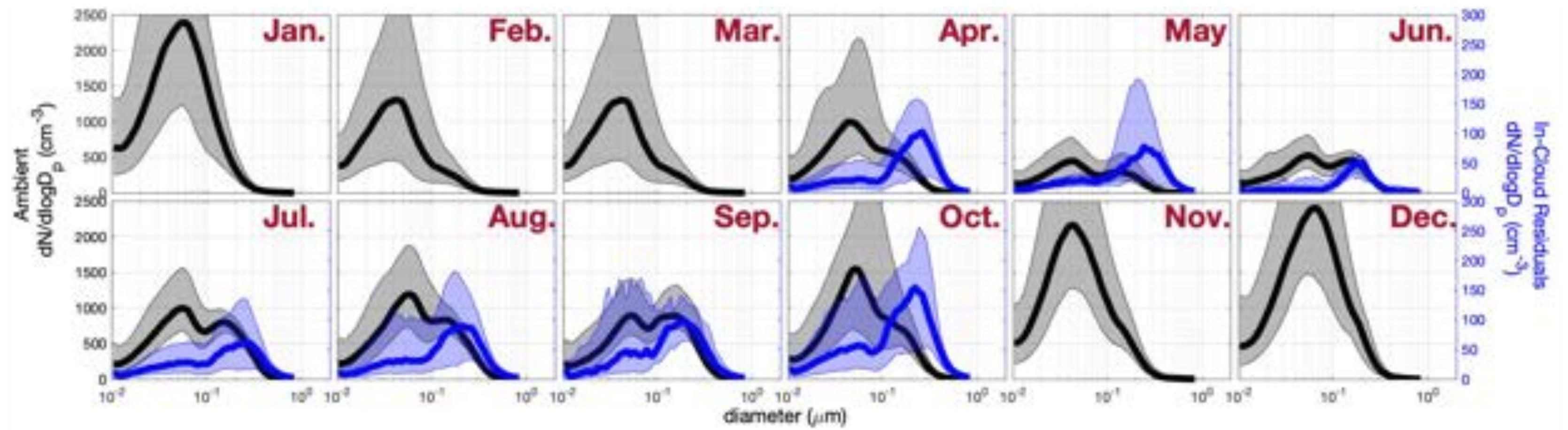
Co-located variables indicate regional process contributions, explaining some of the variability.

- Retrieving similar variables from global models should explain similar amount of variability.
- Future work with site-specific sampling of global models could be applied to other ARM sites.

Join us at EPCAPE Breakout Thursday 8:30am

In-Cloud Residual Size Distributions Show Clear and Consistent Hoppel Minimum

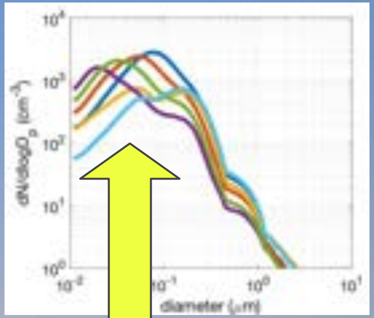
Most in-cloud residual particles collected by Counterflow Virtual Impactor (CVI) are accumulation mode but there is a tail into the Aitken mode.



Residual small mode (<100 nm) activation are attributed to higher LWC, lower cloud base, and variable turbulence.

See Dedrick, Russell, et al. (In Preparation)

Aerosol Climatology of Size Distributions: Frequent Large Aitken Mode Hides Hoppel Minimum



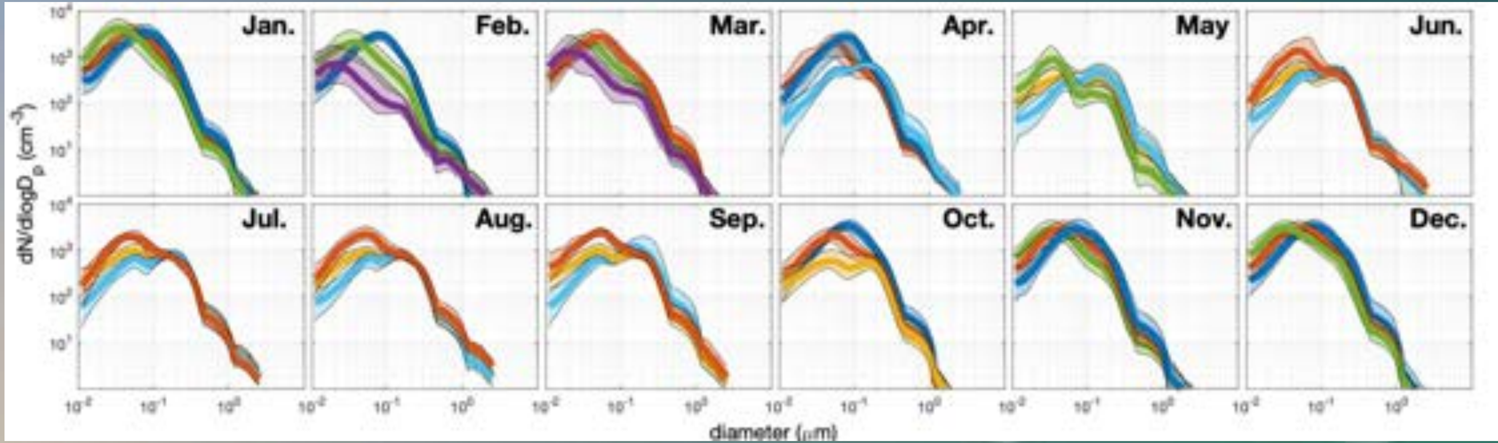
Unimodal 1: Larger (19%)
Unimodal 2: Smaller (20%)
Bimodal 1: Low Aitken (18%)
Bimodal 2: High Aitken 1(10%)
Bimodal 3: High Aitken 2 (17%)
Bimodal 4: High Accumulation (16%)

<u>Seasonal Trends</u>

April-October: >12-38% Bimodal 1

November-March: >40-63% Unimodal

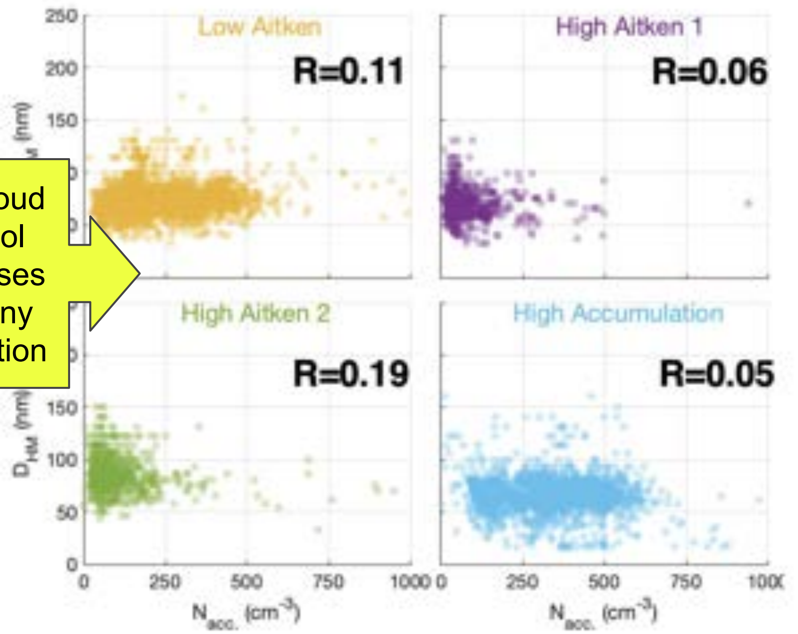
High Aitken modes show multiple recent (non-cloud) aerosol sources/ processes



See Dedrick, Russell, et al. (In Preparation)

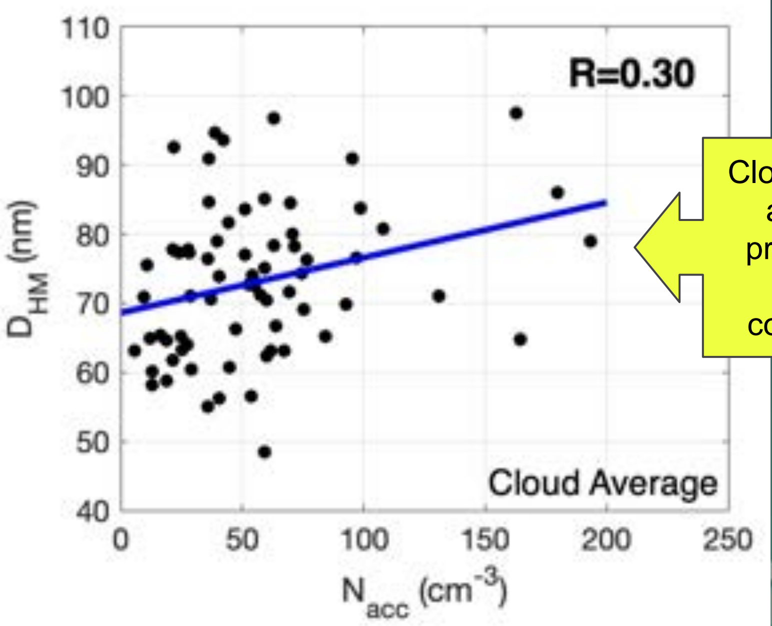
Hoppel Minimum Diameter (D_{HM}) Shows Cloud Feedback on Supersaturation for In-Cloud but Not Out-of-Cloud

Out-of-Cloud Distributions



Non-cloud aerosol processes hide any correlation

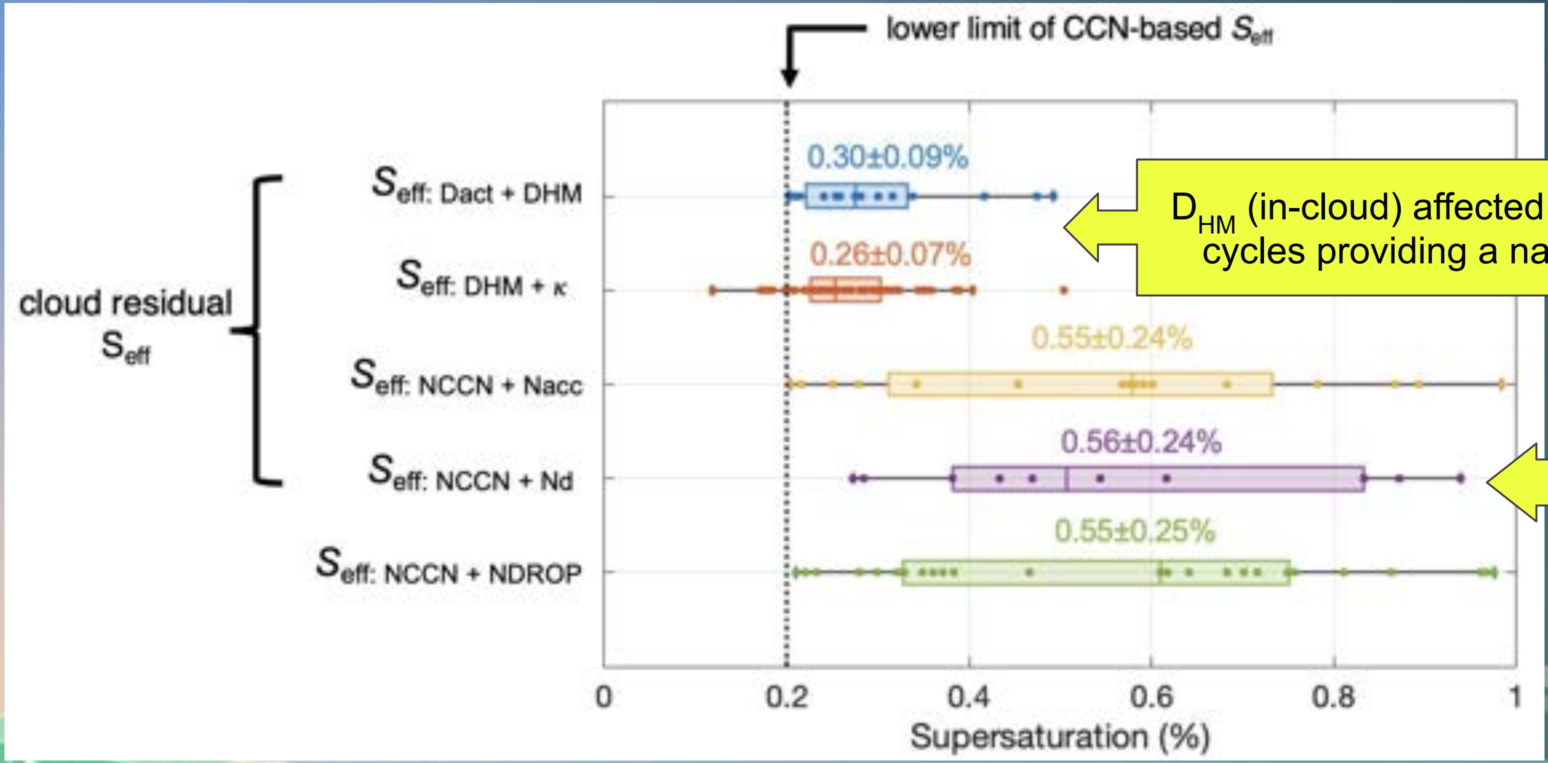
In-Cloud Residual Distributions



Cloud-driven aerosol processes are correlated

Compare to LASIC (Dedrick et al., 2024 GRL)

Comparisons of Effective Cloud Supersaturations Show Differences based on Instruments and Sampling

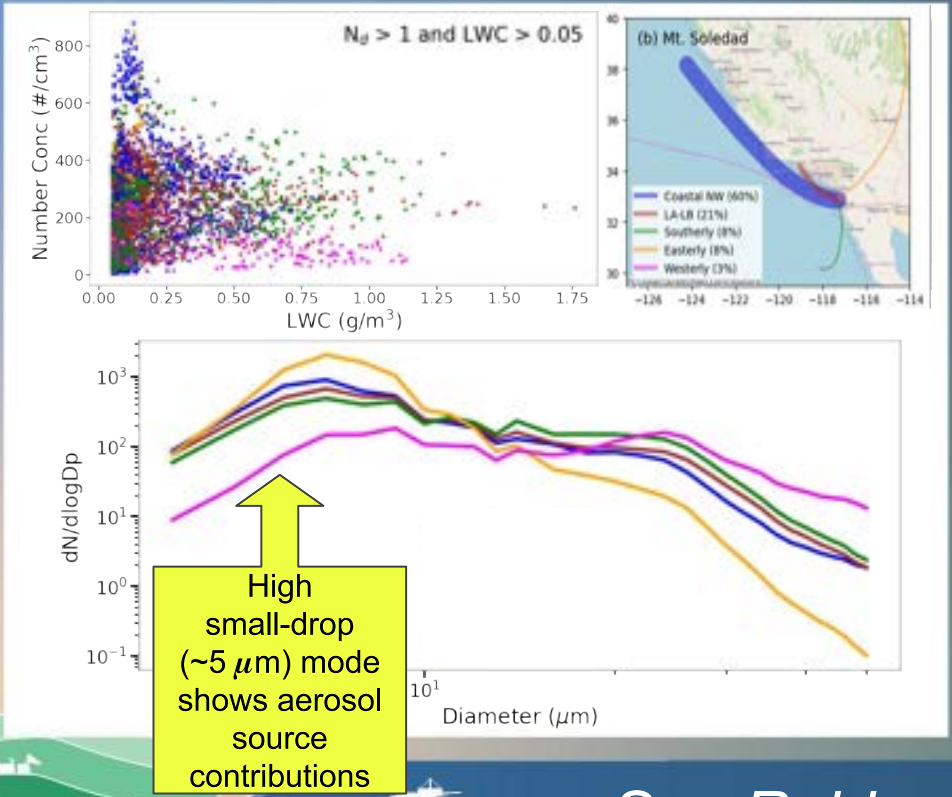


D_{HM} (in-cloud) affected by multiple cloud cycles providing a narrower average

CCN-S_c>0.2% excludes lower values and increases observed average

Compare to LASIC (Dedrick et al., 2025 PNAS)

Cloud Droplet Size Distributions Vary with Back-Trajectories

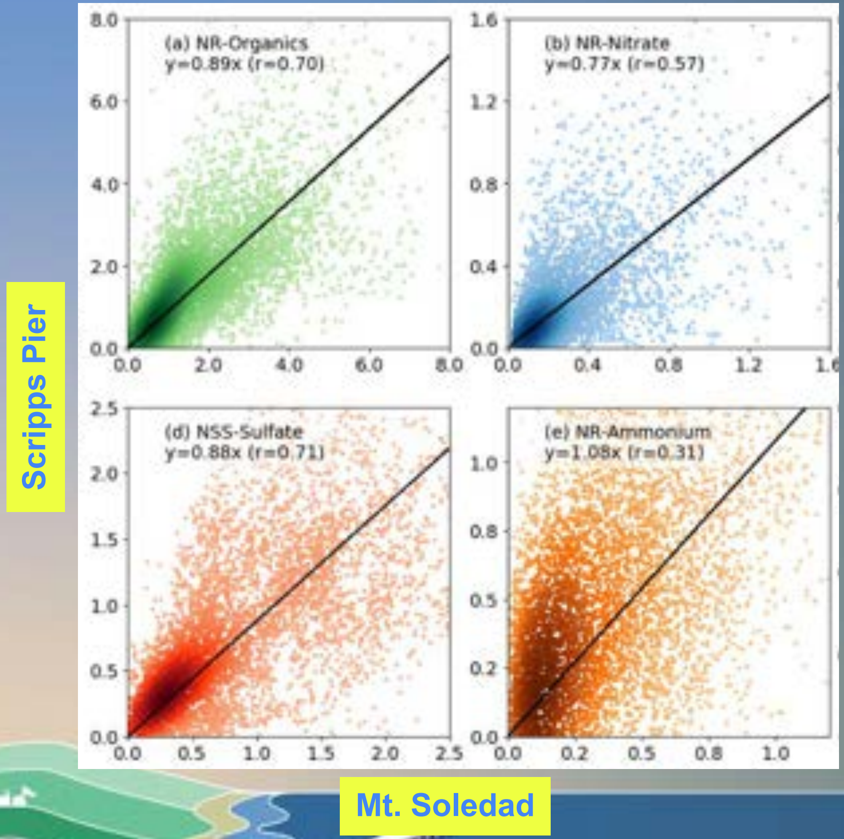


Coastal NW, LA-LB, and Southerly sources have similar distributions, which represent mean background conditions in the region.

Marine Westerly cases have lower N_d and a higher LWC, which indicate a cleaner source.

See Robinson, Chang, et al. (In Preparation)

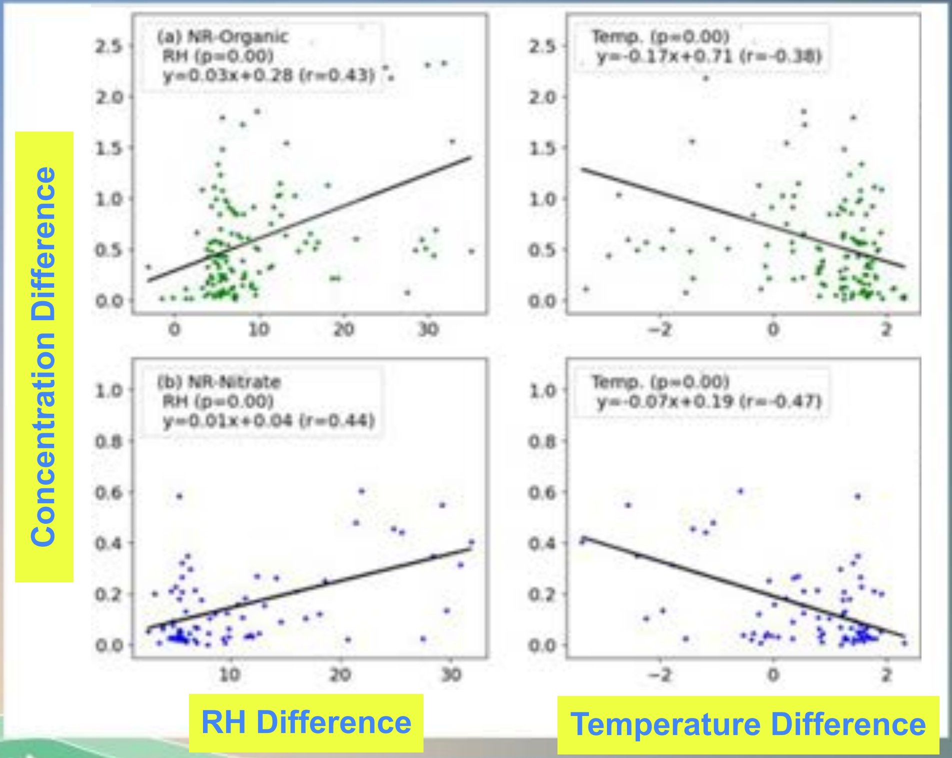
Composition at Scripps Pier and Mt. Soledad More Similar for Non-Volatile Sulfate and Less Similar for Semi-Volatile Nitrate



- Scripps Pier and Mt. Soledad NR components are similar and correlated.
  - Sulfate and Organics strongly correlated (non/low-volatile).
  - Ammonium moderately or weakly correlated (semi-volatile).
  - Nitrate has higher sources at Soledad but also semi-volatile.
- Local sources at Mt. Soledad contributed 38-52% of Organics and Nitrate plus brake-metal tracers.
- Land/Sea breezes caused daily cycles.

See Han, Russell, et al. (In Review)

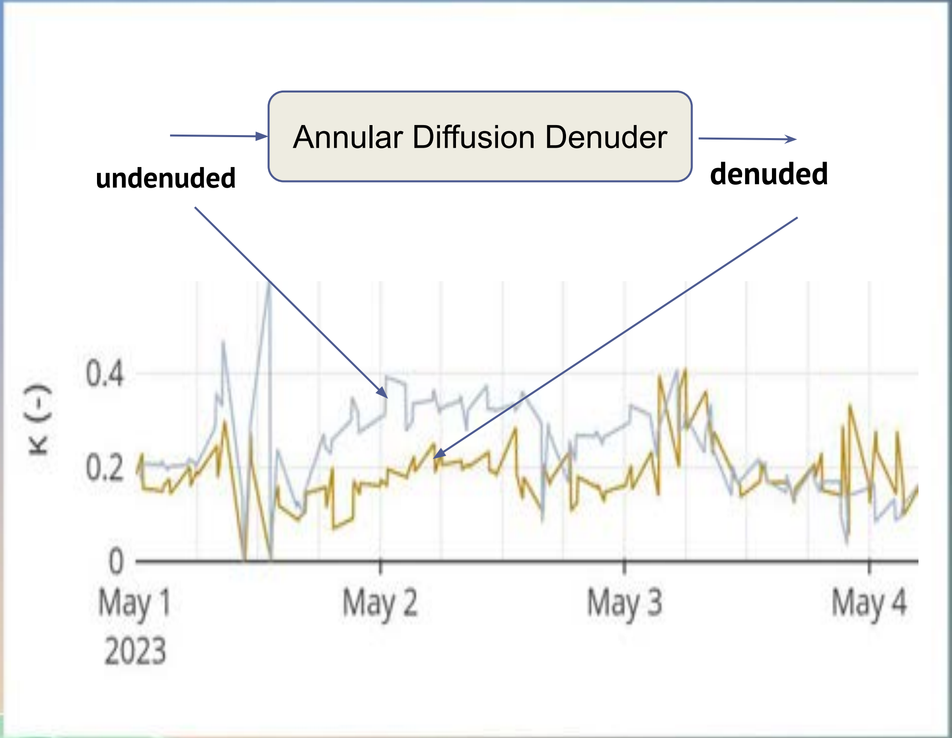
Semivolatile Nitrate and Organics Increase with RH and Decrease with Temperature



- Differences are partly explained by semivolatile partitioning.
- Organics and Nitrate are higher at
  - Higher Relative Humidity because of water uptake.
  - Lower Temperatures because of reduced evaporation.

See Han, Russell, et al. (In Review)

Denuded and Undenuded Cloud Condensation Nuclei Show Semivolatile Effects



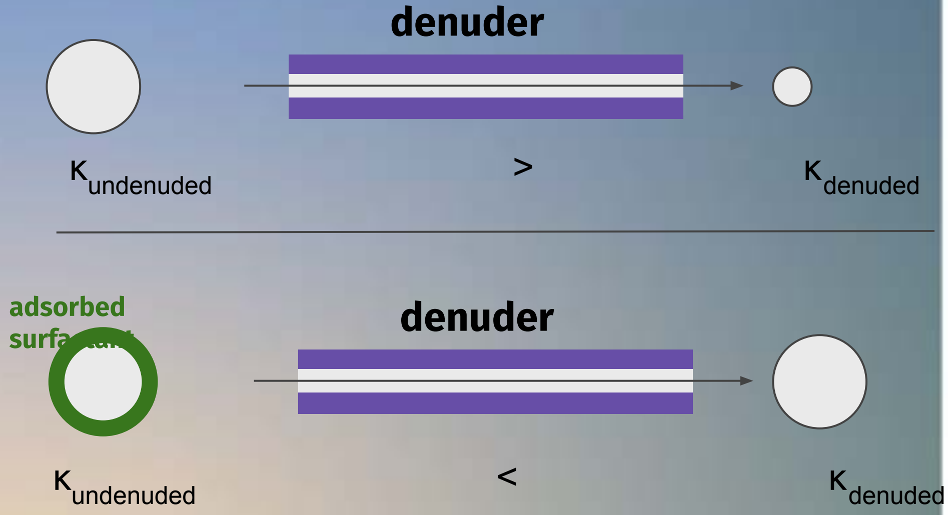
Hygroscopicity parameter varies between 0.1 and 0.4 for supersaturations >0.2 %.

Denuding sometimes, *but not always*, reduces hygroscopicity by up to 50%, activating at lower supersaturation in the atmosphere relative to predictions by standard CCN measurements.

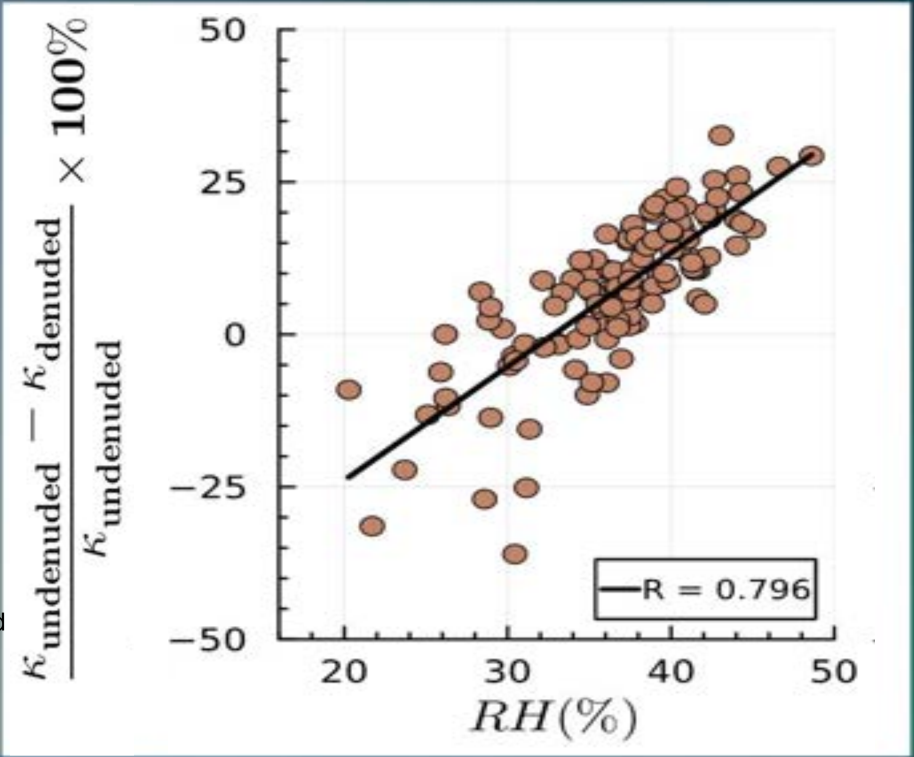
See Ravichandran, Petters, et al. (In Review)

Removing Semivolatiles by Drying Increases or Decreases CCN-derived K (Hygrosocopicity) by up to 50%

Solute Effect: Removing semivolatiles decreases the amount of dissolved solutes and decreases $\kappa_{denuded}$.

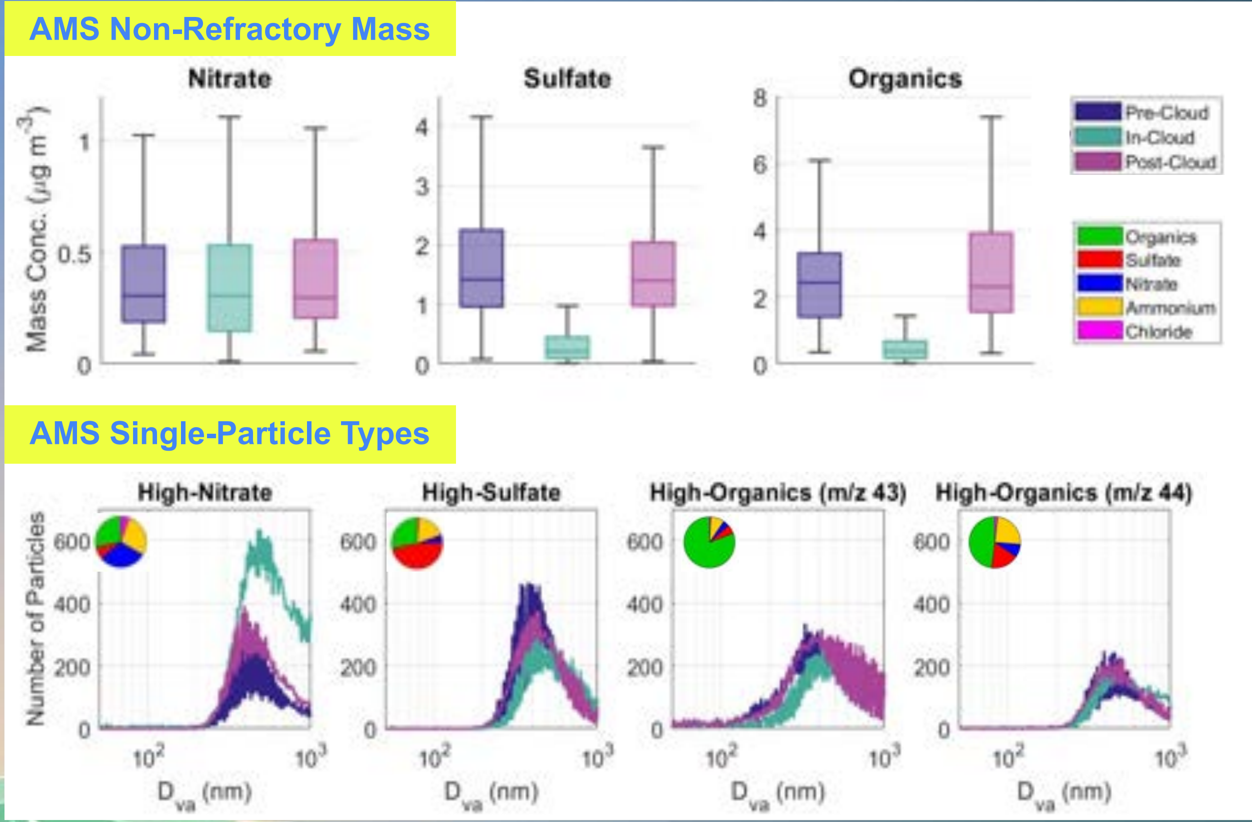


Surface Effect: Removing semivolatiles changes the surface tension and increases $\kappa_{denuded}$.



See Ravichandran, Petters, et al. (In Review)

Nitrate Enhanced in Cloud Drop Residual Particles



Compared to out-of-cloud, residuals droplets have:

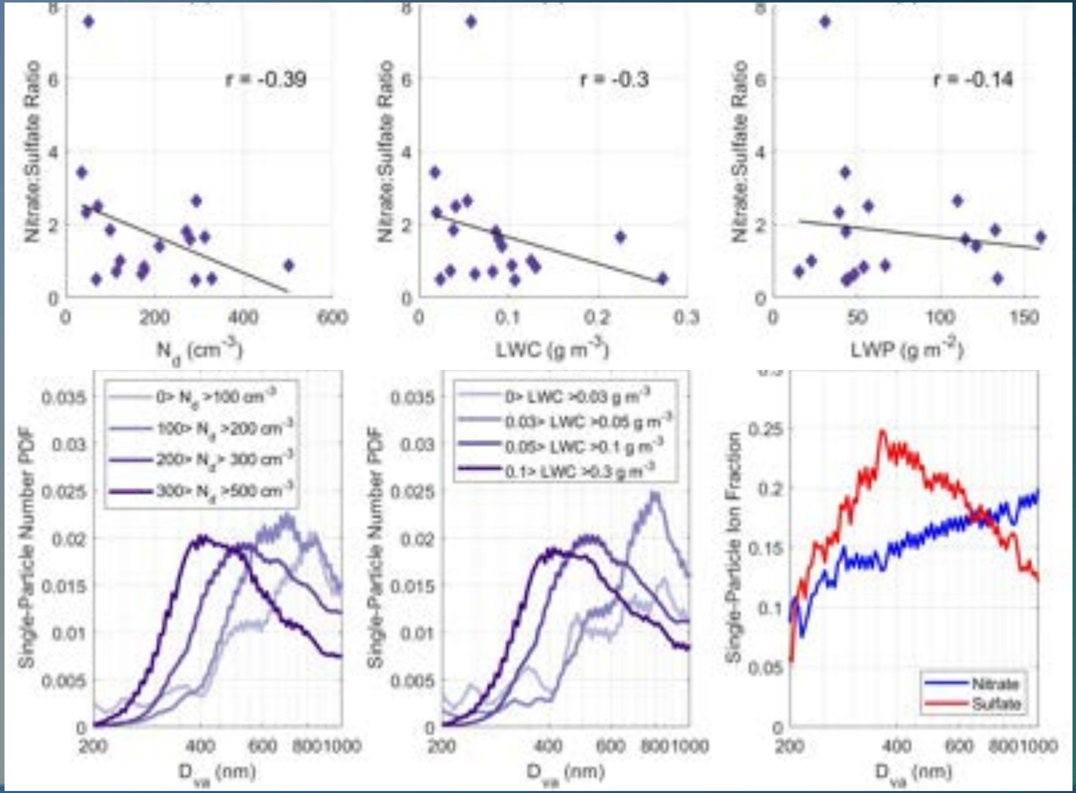
- **Similar nitrate mass concentration** but lower sulfate and organic.
- **More high-nitrate single particles** that are larger in size than other types.

See Williams, Russell, et al. (In Preparation)

Larger Nitrate Particles Activate in Clouds with Low N_d and LWC

For 19 single-layer cloud events at Mt. Soledad

- **Nitrate/Sulfate decreases with increasing cloud N_d, LWC, and LWP**
- **Smaller sizes activate with increasing N_d and LWC**
- **Single-particle Nitrate/Sulfate is highest at larger sizes**



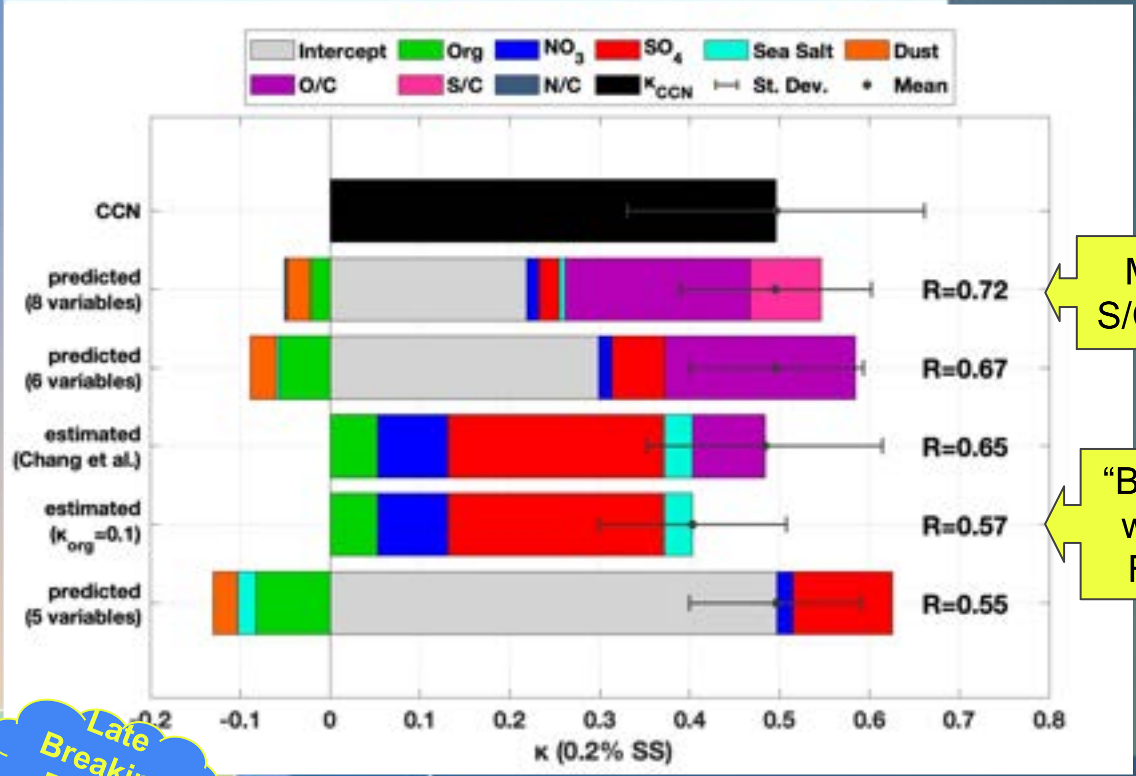
See Williams, Russell, et al. (In Preparation)

ACI Highlights from EPCAPE

Coastal NW back-trajectories bring clouds and a range of coastal emissions.

- Aerosol Activation to Cloud Droplets:
  - Semivolatile nitrate and organic partitioning driven by RH and T difference between Pier and Soledad.
  - Semivolatile components change aerosol activation.
  - Larger nitrate particles activate more at low LWC and smaller sulfate at high LWC.
- Aerosol Growth by Cloud Processing:
  - In-cloud size distributions show supersaturation feedback but ambient hidden by emissions.
  - Regional sulfate oxidation is 34% by cloud and 36% by aerosol aqueous reactions.

Hygroscopicity Representation is Improved by Incorporating Measured O/C and S/C



MLR with S/C and O/C

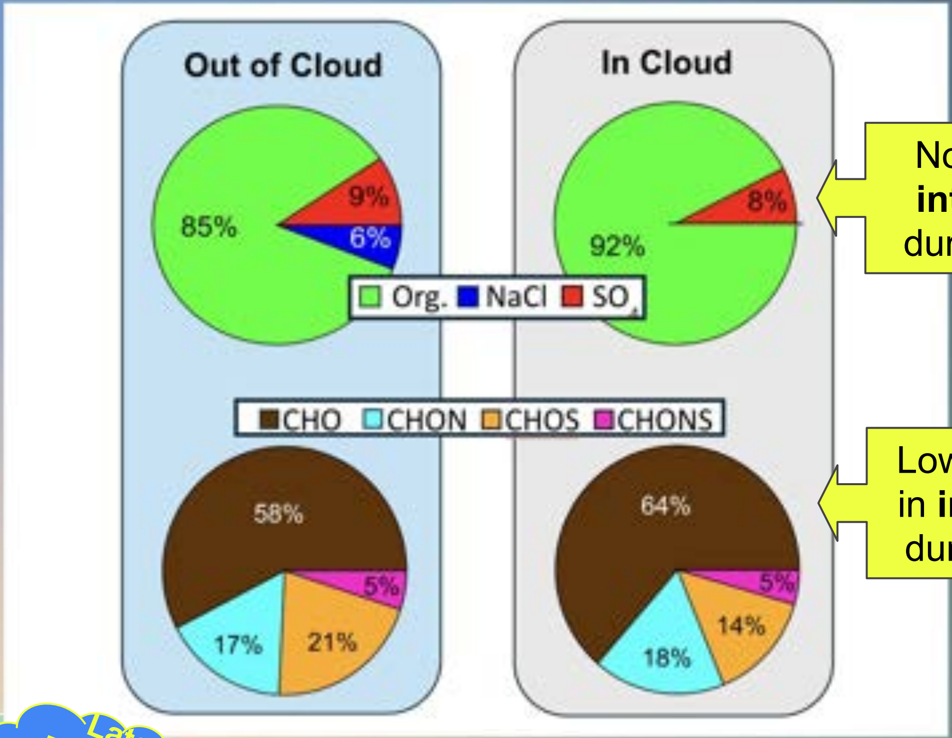
"Bottom-Up" with Mass Fractions

Highest correlation to CCN-measured hygroscopicity (K) is multi-linear regression with O/C and S/C, even though sulfate and nitrate account for most water uptake.

Late Breaking Result!

See Berta, Russell, et al. (In Preparation)

Ultrafine Aerosol Composition Measurements for April-June



No NaCl in **interstitial** during cloud

Lower CHOS in **interstitial** during cloud

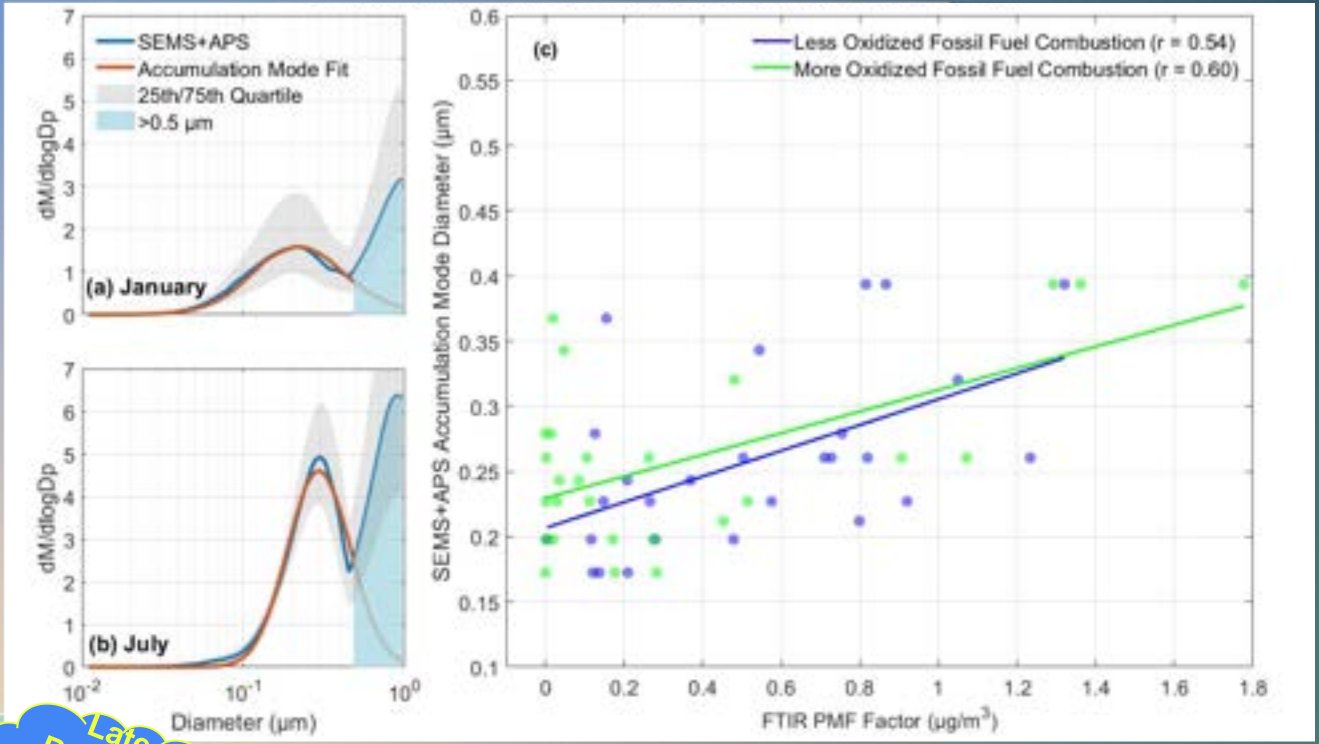
During clouds, ultrafine (UF) particles have

- No NaCl, suggesting UF with salt were activated.
- Reduced organosulfate but higher oxygenated organics.

Late Breaking Result!

See Kapp, Smith, et al. (In Preparation).

Combustion-Related Organics Contribute to Increasing Particle Size



For LA-LB back-trajectories, organics from fossil fuel combustion are associated with more mass 0.5-1 μm and larger accumulation mode.

Late Breaking Result!

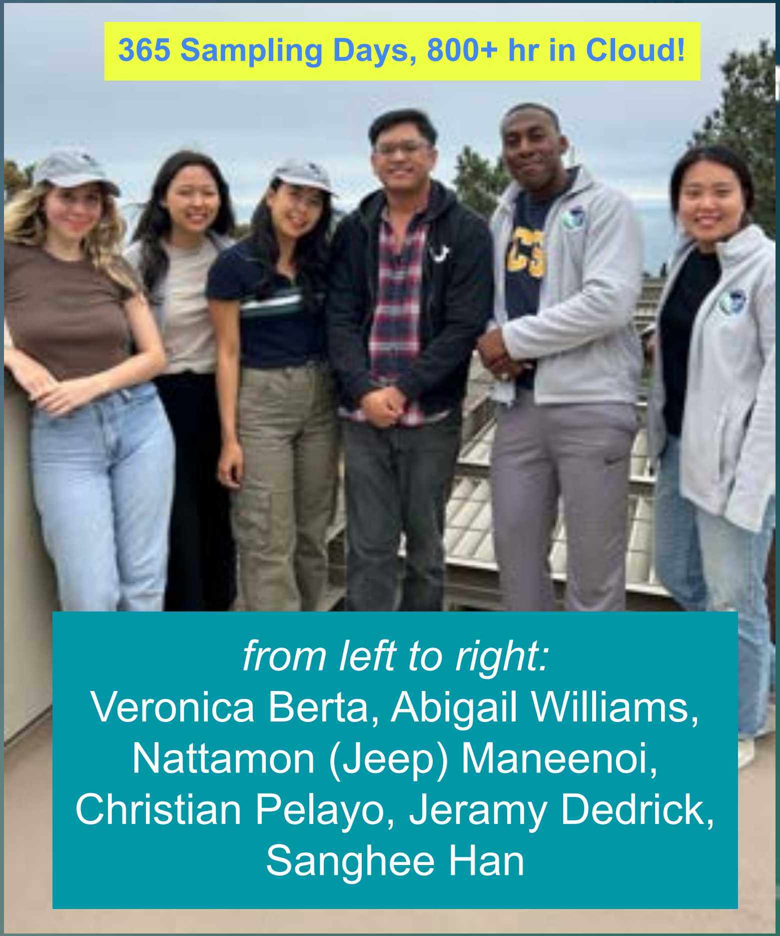
See Pelayo, Russell, et al. (In Preparation)

Acknowledgments

DOE ARM&ASR Funding, Techs, Mentors
EPCAPE Science Team for Measurements

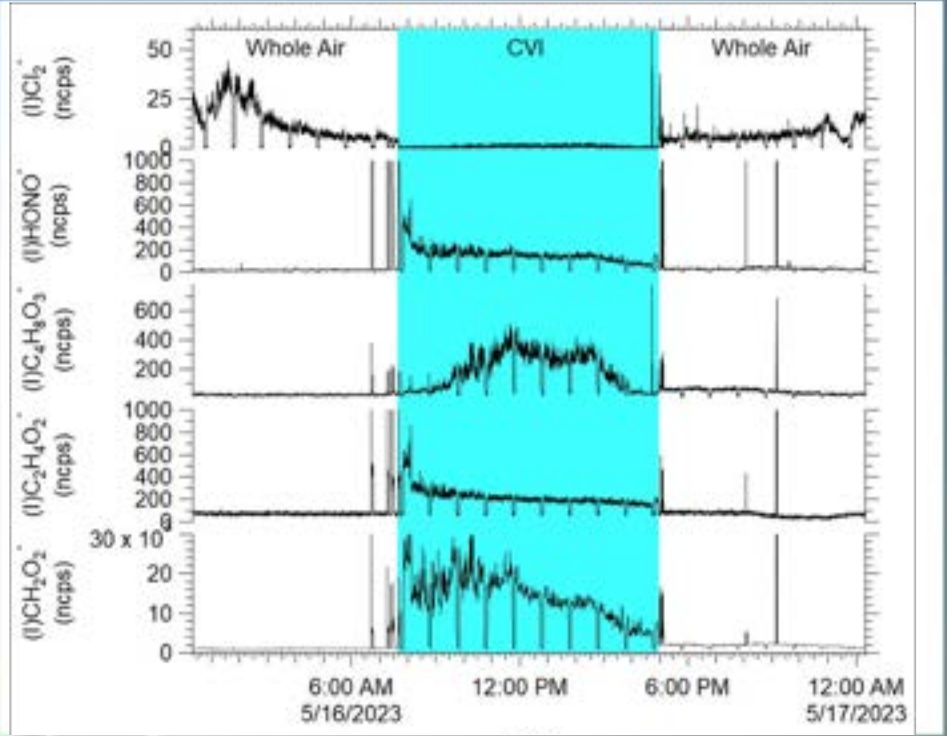
- Mt. Soledad Data
  https://library.ucsd.edu/dc/collection/bb0898306q
- EPCAPE Websites
  https://www.arm.gov/research/campaigns/amf2023epcape
  https://wordpress.cels.anl.gov/clouds/epcape-plots/
  https://dq.arm.gov/dq-plotbrowser/#

Thank you…Questions?



National Science Foundation | U.S. Department of Energy | ASR Atmospheric System Research | ARM | Environment and Climate Change Canada

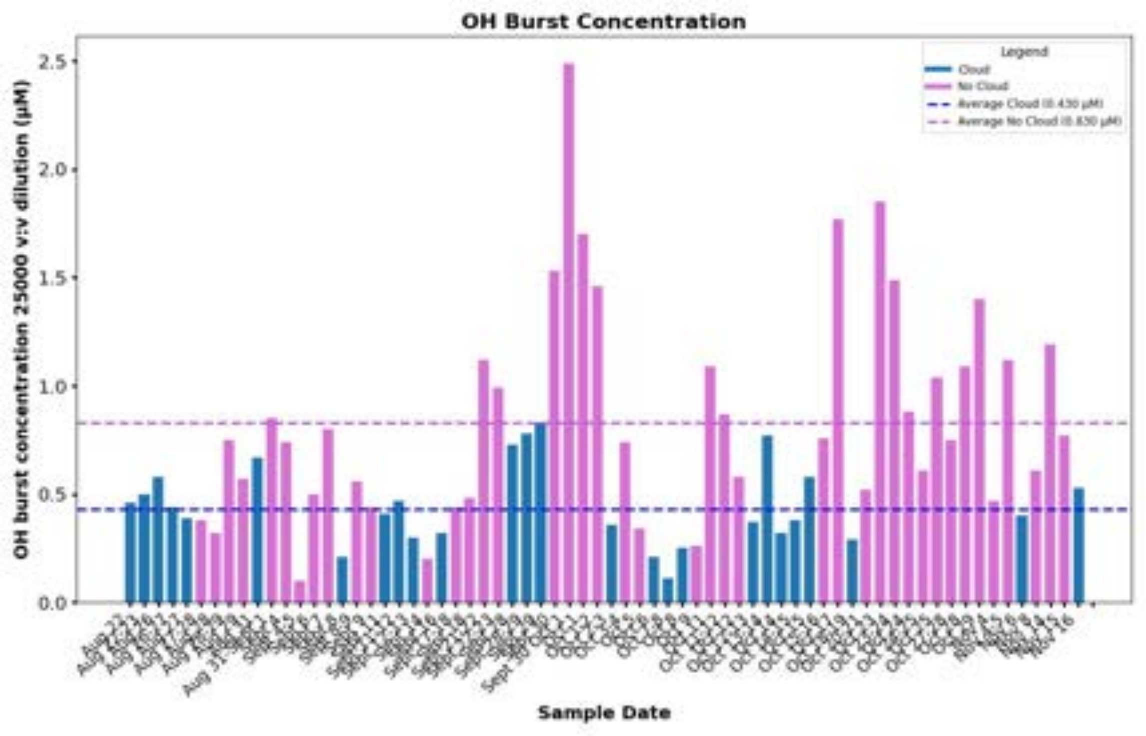
Organic Vapors Evaporated from Cloud Water for May 2023



Iodide-CIMS preliminary results indicate many organics are **enhanced in the evaporated cloud water** relative to ambient air, above what is expected via Henry's Law, suggesting rather rapid and active cloud organic chemistry.

See Wentzell, Liggio, Wheeler, et al. (in prep)

OH Burst in Aerosols for Clear and Cloudy Days during August-November 2023

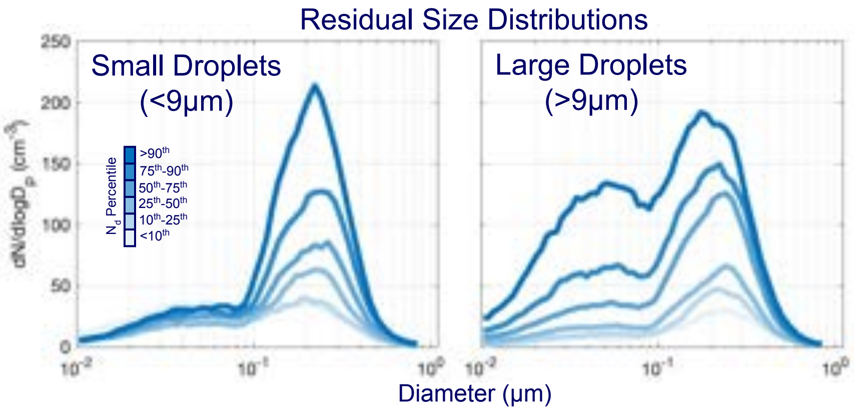


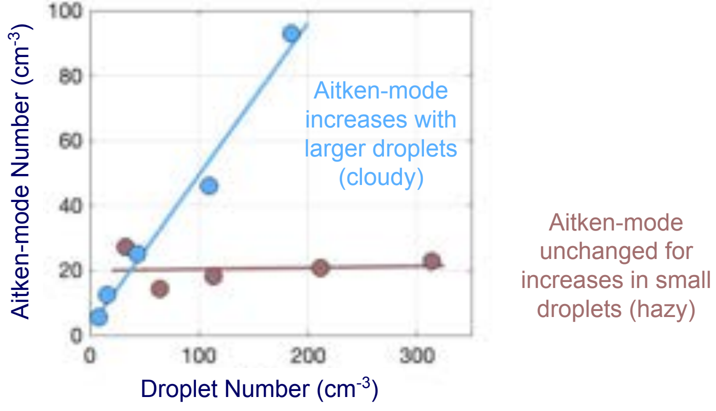
The first remotely controlled, automated Direct-to-Reagent OH burst measurements.

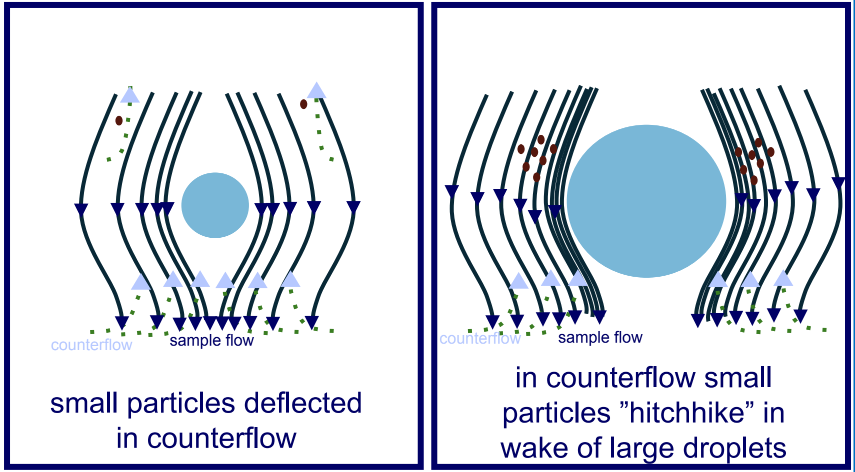
Samples collected during **cloudy periods are less active**, potentially because OH burst precursors are consumed when aerosols activate.

See Banach, Paulson, et al. (in prep)

CVI Operation and Efficiency





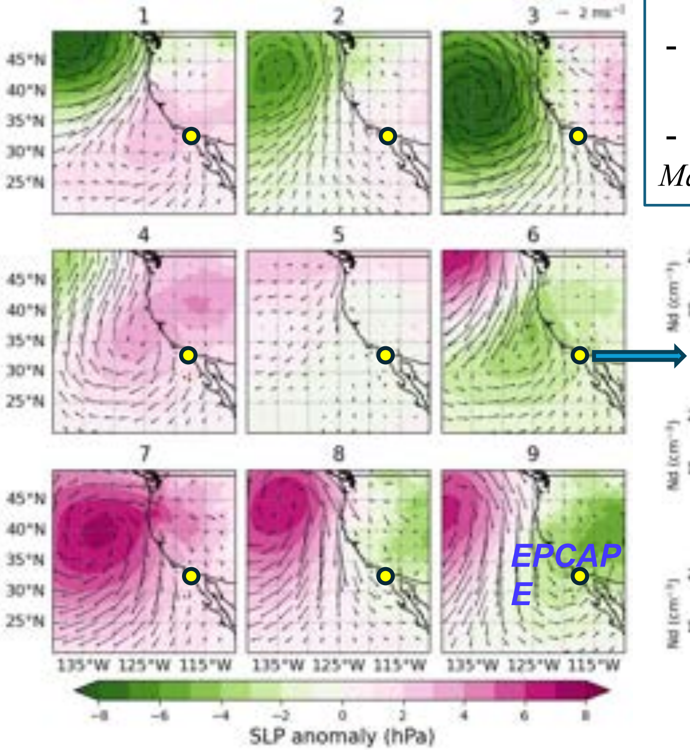


In cloudy conditions, Hoppel minimum represents critical size of residual aerosol activated in >9μm droplets.

Atmospheric regimes and drivers of cloud variability and aerosol-cloud-radiation interactions over the coastal northeast Pacific

Seethala Chellappan, David Painemal, Mandy Thieman, William Smith Jr. (NASA LaRC Team, DE-FOA-0002850)

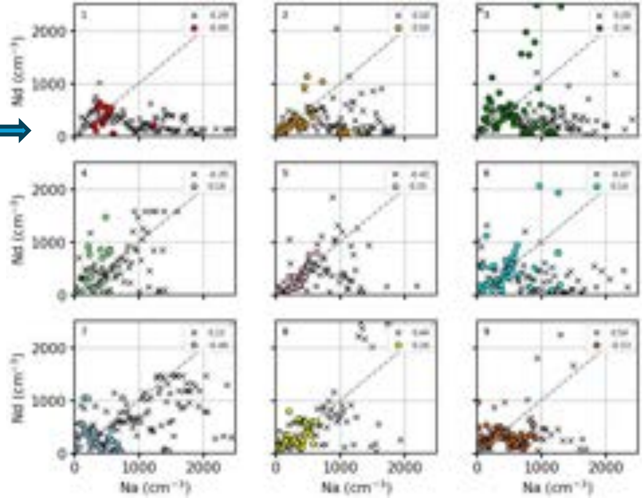
Synoptic regimes based on Self Organizing Maps (SLP and 975 hPa winds)



Objectives

- Understand the control of coastal meteorology over cloud variability and aerosol transport at the EPCAPE site.
- Isolate the cloud response to aerosol concentration from the control of environmental factors through the use of atmospheric regimes.
- Determine magnitudes of aerosol-cloud interactions and cloud adjustments.

Manuscript in preparation to be submitted to ACP in spring.

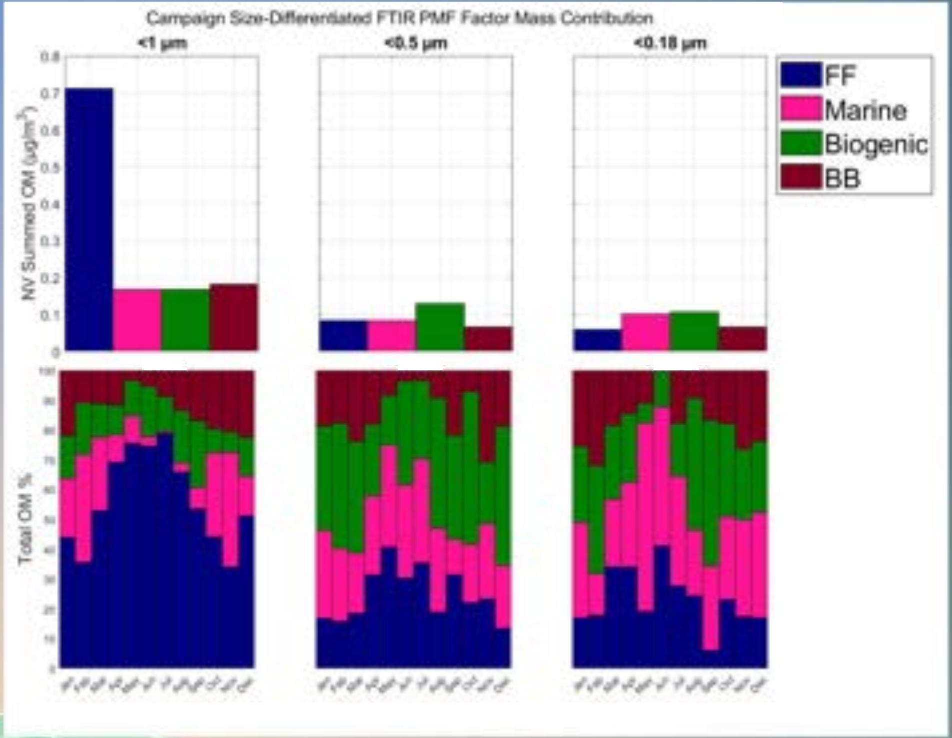


Relationship between ARM Aerosol Concentrations and Cloud Droplet Number Concentrations at Scripps Pier for different atmospheric regimes

VAPs request:

1. Cloud Optical Depth (SPHOTCOD) from Cimel Sunphotometer
2. Drizzle/Precipitation Rate from Ka band Zenith Radar (KAZR)

Sources of Size-Resolved Aerosol by PMF Analysis of FTIR for Organic Functional Groups

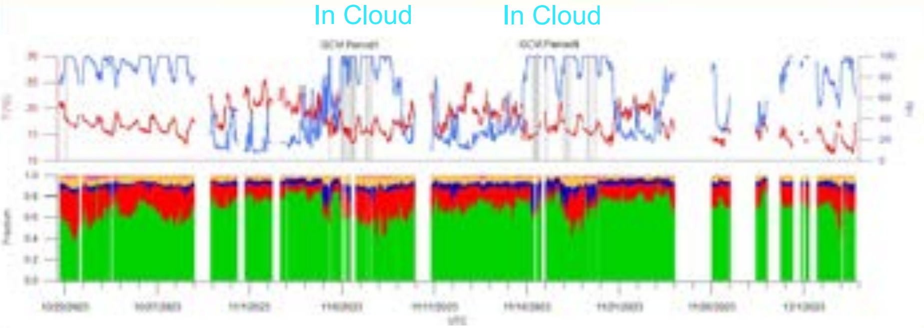


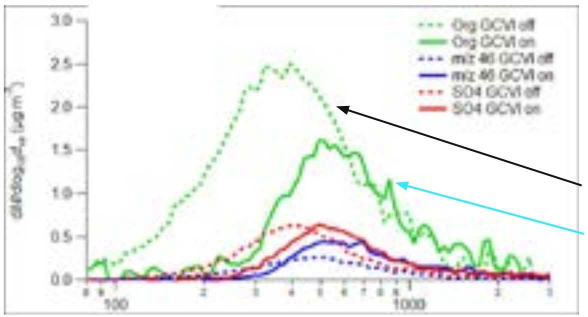
PMF analysis of FTIR spectra shows four sources of organic aerosol for most of the project.

Fossil fuel combustion contribution largest for 500-1000 nm particles.

See Pelayo et al. (in prep)

Cloud Processing of Carbonaceous Aerosol





Aerosol Mass Spectrometer (AMS)
Particle Time-of-Flight (PToF) Size Distributions

Out of cloud (dashed)

In Cloud (solid)

ASR Atmospheric System Research

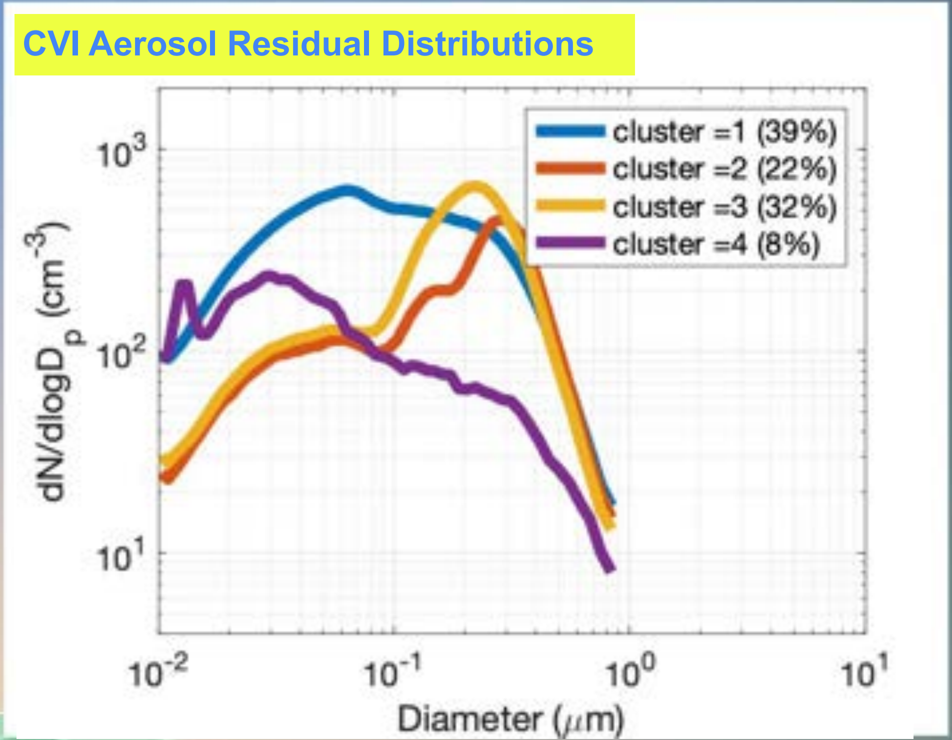
Cloud residuals in general had **larger mean particle size.**

Chemical composition, supermicron, bioaerosol, water uptake, optical properties and VOC analysis in progress

Allison C. Aiken, Ryan Farley, Kyle Gorkowski, Katherine Benedict, James Lee, Manvendra Dubey, Abu Sayeed Md Shawon

Contact: aikenac@lanl.gov

Aerosol Number and Updraft Variability Influence Particle Activation and Supersaturation



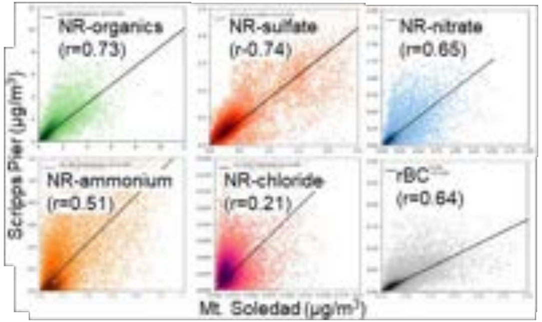
Most (60%) residual size distributions are bimodal, reflecting mean critical size of particles activated as CCN and supersaturation.

Mode <100 nm likely associated with updraft variability during cloud events.

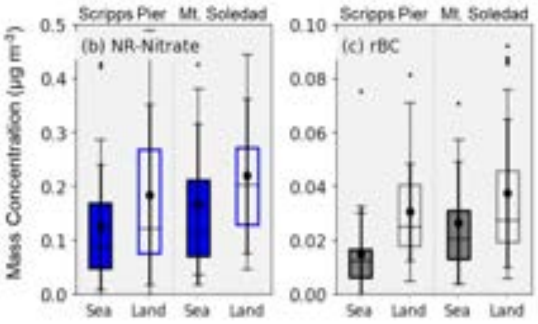
See Dedrick et al. (in prep)

Local and Upwind Effects on Aerosol Composition at Scripps Pier and Mt. Soledad during EPCAPE

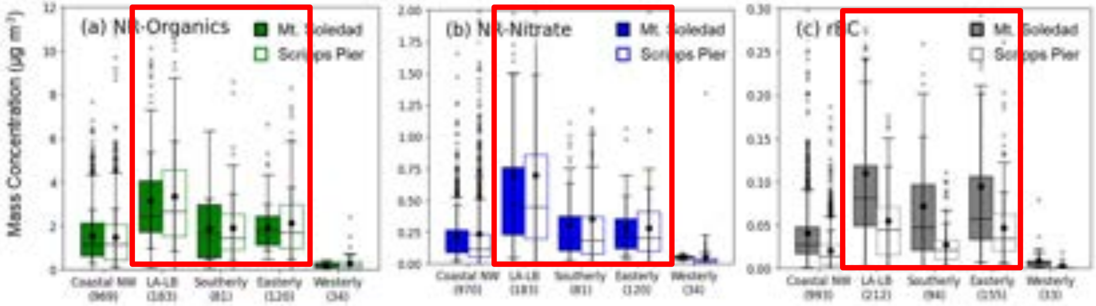
Site-to-site comparison



Land/sea breeze effects



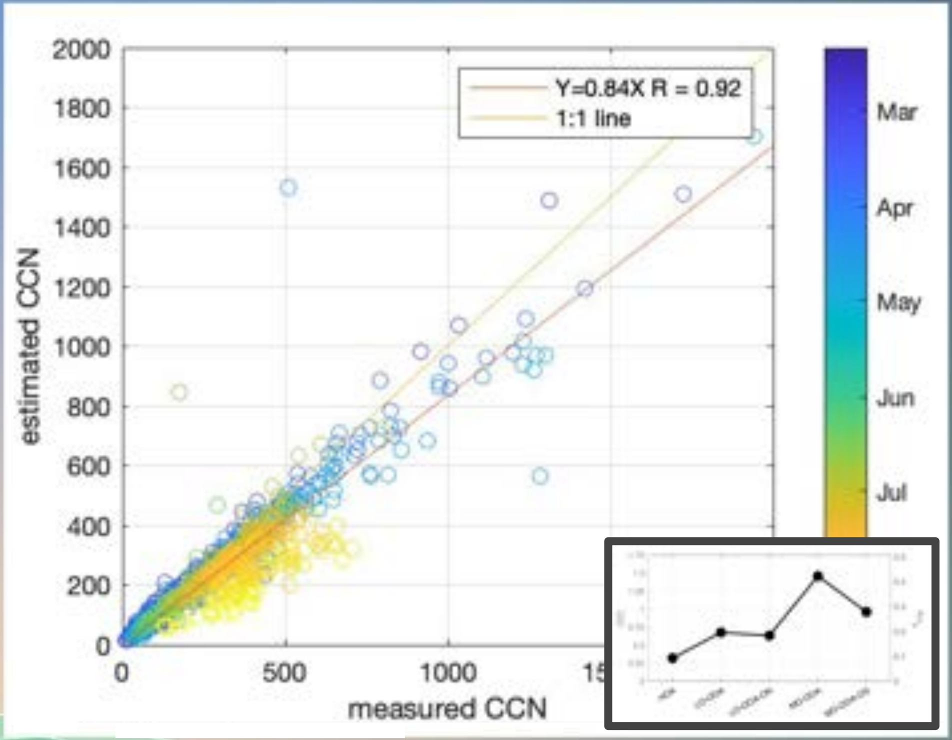
Dependence on upwind regions



- NR-sulfate and NR-organics at Scripps Pier are similar to Mt. Soledad.
- NR-organics, NR-nitrate and rBC were higher for LA-LB, Southerly, and Easterly trajectories.
- Local sources at Mt. Soledad accounted for about 38-52%.
- Land breeze increased Scripps Pier concentrations by approximately 65% of rBC, 40% of NR-nitrate, and 33% of NR-organics.

See Han et al. (In Review)

Organic Factors Improve Closure on Cloud Condensation Nuclei



Using organic hygroscopicity (κ_{org}) based on the atomic oxygen to carbon ratios (O/C) of **organic PMF factors improves closure on cloud condensation nuclei** by ~13%.

Retrieving κ_{org} from organic PMF factors that correlate with Aitken mode mass (HOA, LO-OOA) resulted in improved estimation of κ at higher %SS.

See Berta et al. (In Preparation)