ENVIRONMENT AND CLIMATE CHANGE CANADA (ECCC)

Air Quality Processes Research Section (ARQP)

Mt. Soledad Droplet and Residual Measurements

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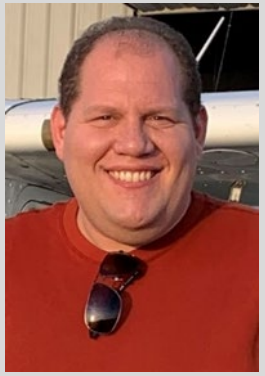

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FDM
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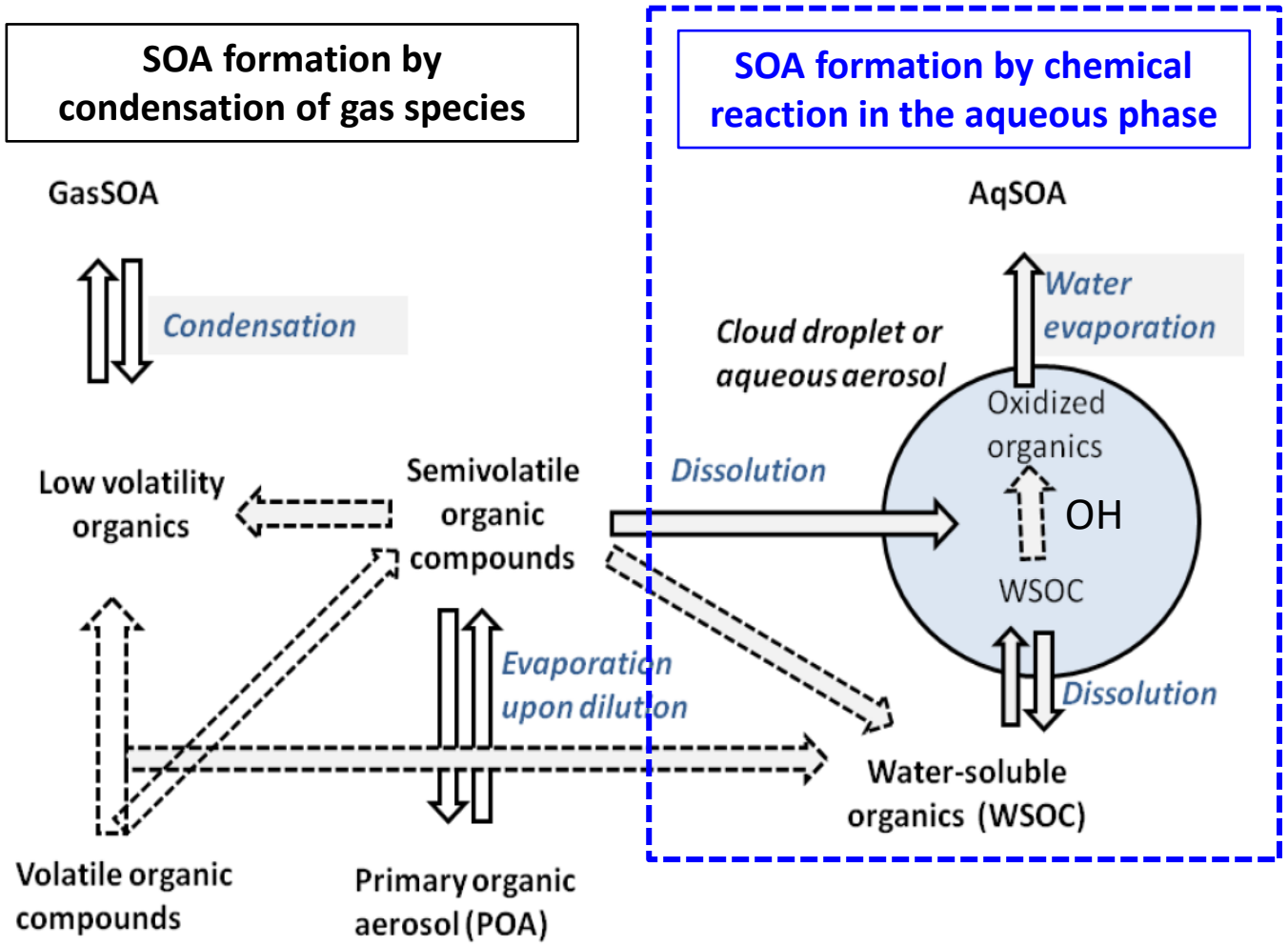
Some important aspects of cloud chemistry and organics

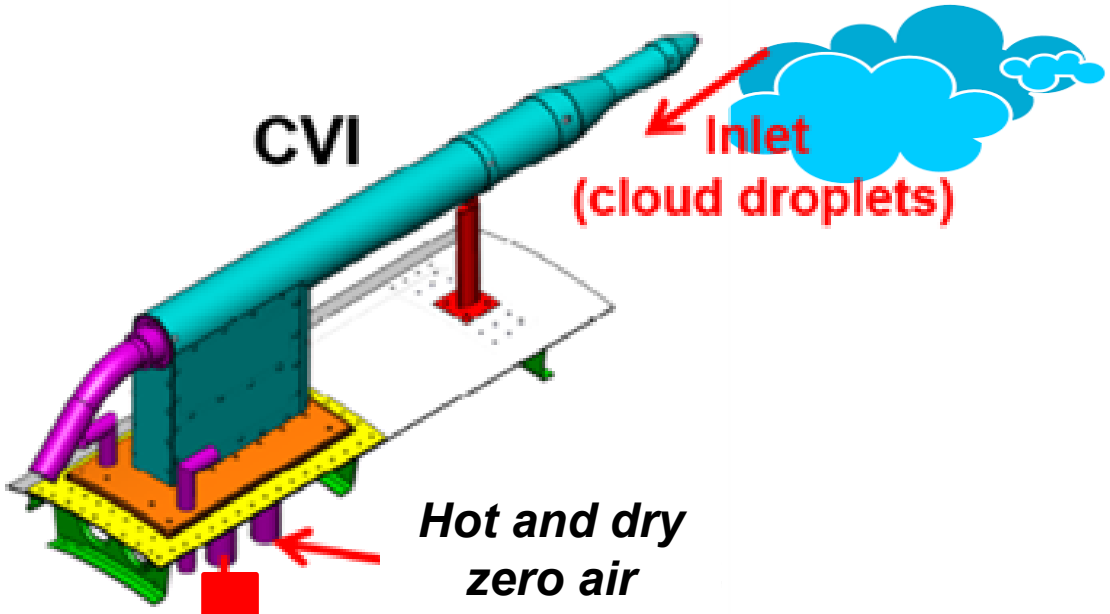


Ervens et al., ACP, 2011

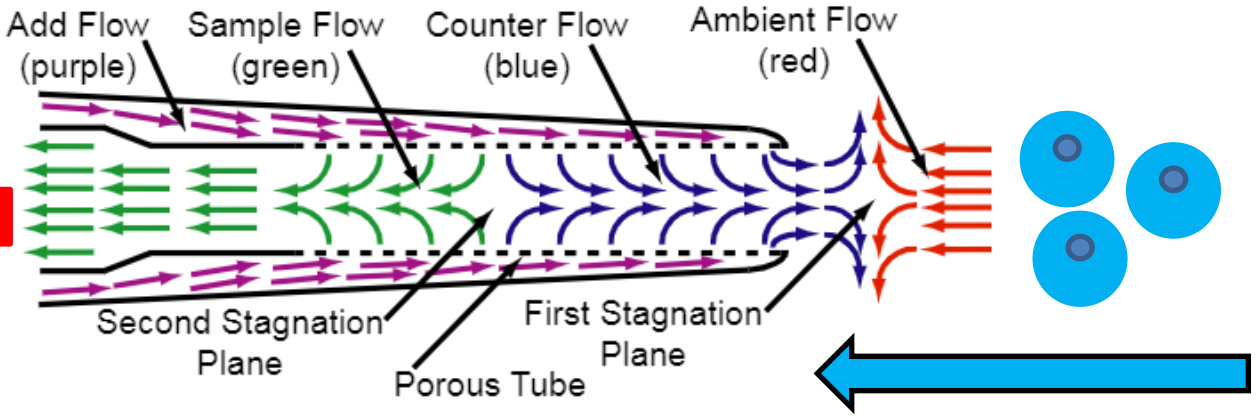
- Cloud droplets are an important source of secondary organic aerosol (SOA) and brown carbon
- An important loss process for water soluble organics
- Organic composition in particles/droplets impact oxidant formation (which go on to make more WSOC)
- Organic trace gases can alter particle surface tension and impact CCN activation
- **Understanding of these effects of organics are poorly understood from ambient studies**

Improving our cloud chemistry understanding via Mt. Soledad

- **Key instrument is a Counter-flow Virtual Impactor (CVI):** *(Shingler et al., AMT, 2012)*

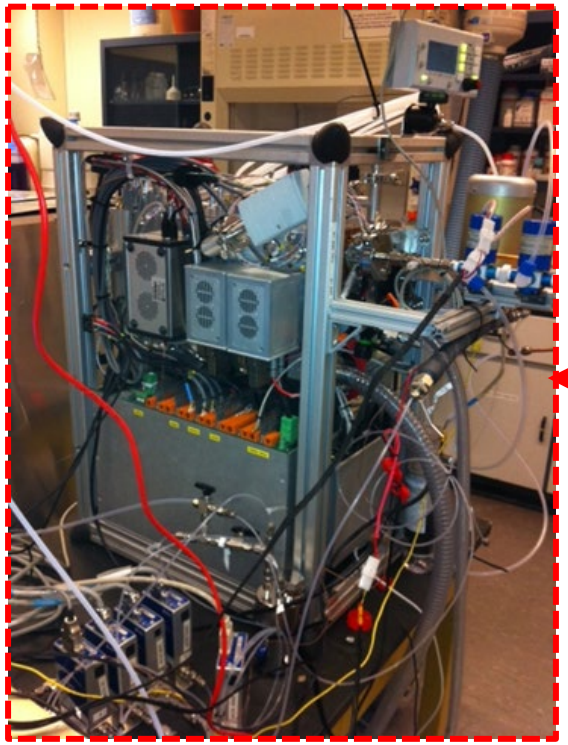


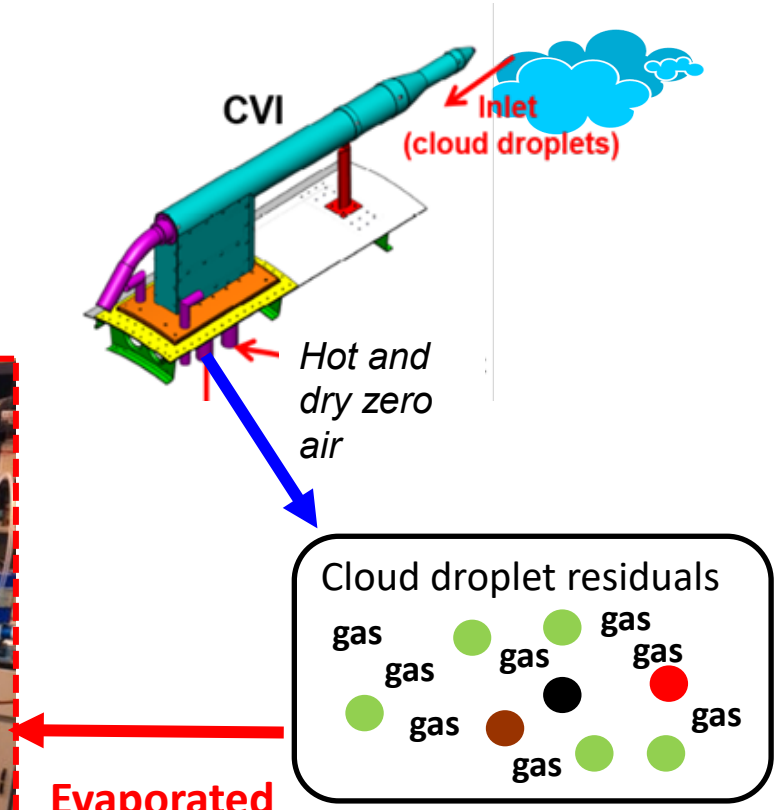
- Only droplets greater than a defined size enter CVI
- Cloud droplets evaporated with zero air
- Surfaces and transfer lines heated to reduce loses
- **Only droplet residuals (particles) and evaporated organic/inorganic gases remain**



Chemical Ionization Mass Spectrometry (CIMS) measurements

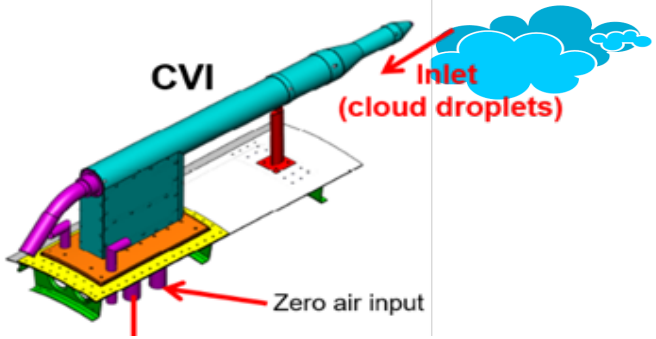
HR-ToF-CIMS

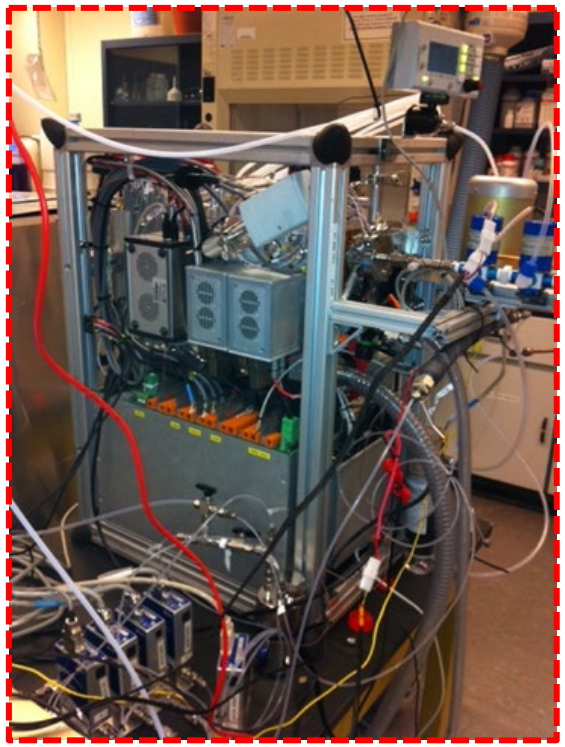




- Evaporated organic gases (ie: aqueous fraction) measured by CIMS in real-time
- Iodide ionization: $I^-\cdot(H_2O)_n + M \rightarrow I^-\cdot(M) + n\ (H_2O)$ where n=0,1,2……
- Oxidized organic gases (organic acids, org-nitrates, etc..)
- Inorganic Nitrogen (N_2O_5, HNCO, HONO)
- <10 ppt DL in 1 sec for many species
- Likely small volatile oxidized molecules will evaporate best (indicator of oxidation chemistry)

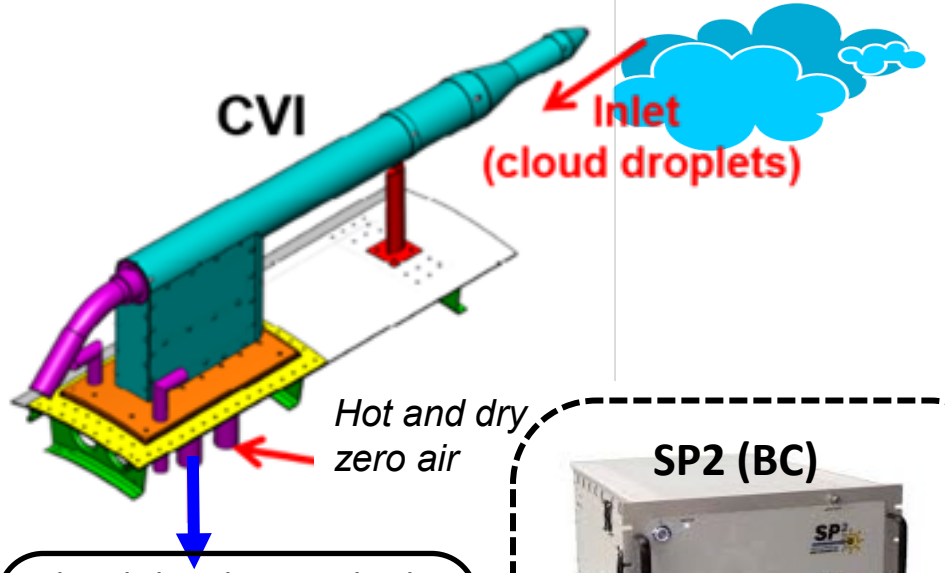
CIMS related objectives/questions





- In-cloud oxidation should form a variety of oxidized species in aqueous phase, that we can measure. How fast do they form? – relate measured species to photochemical/air mass age
- Improved understanding of composition of water soluble organics within cloud droplets
- Investigate organic gas-cloud droplet partitioning and/or chemistry (in cloud vs out vs interstitial air)
- Can we measure processing of organics at the same time that we measure “OH bursts”? What species are formed?
- Are some organic species associated with changes CCN activity? Which ones?

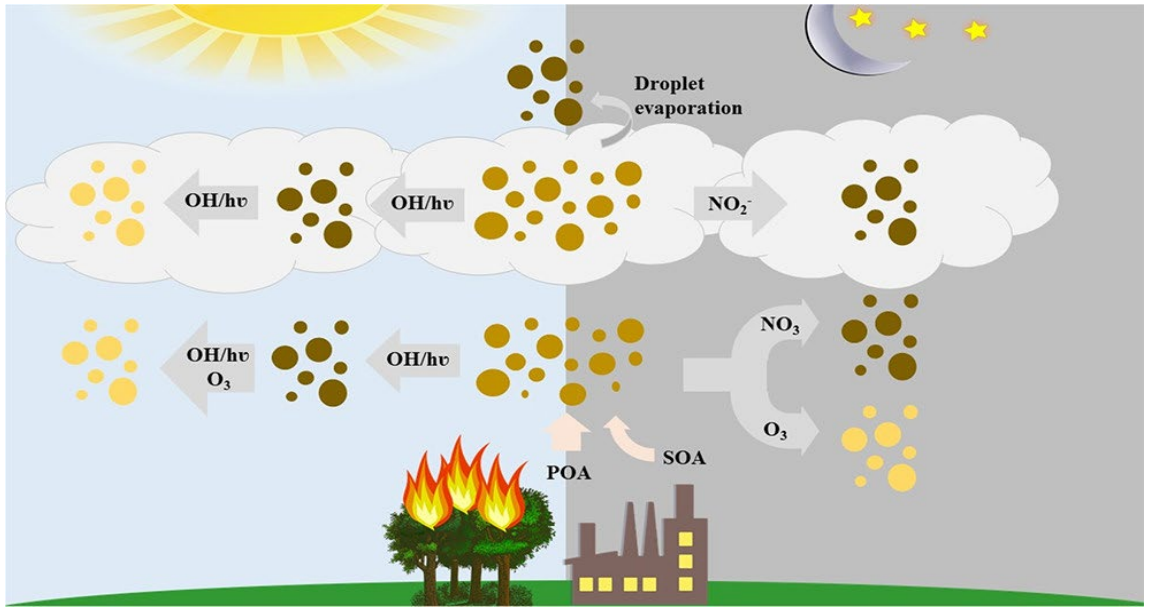
Measurements of PM residuals



- **Aerosol light scattering and absorption measured by Photoacoustic Extinctiometers (PAX, 405 and 870 nm)**

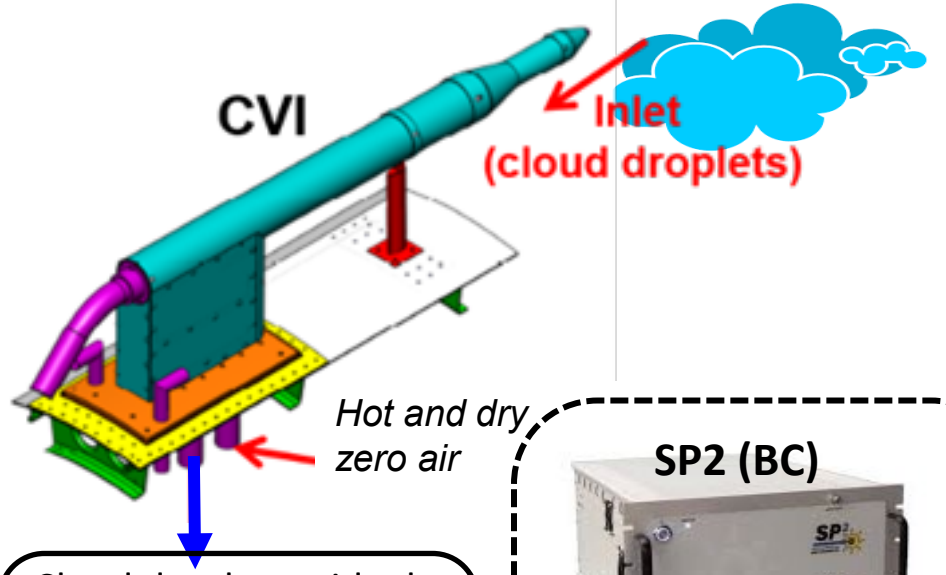
Objectives/questions:

- Investigate potential impacts of in-cloud processing on brown carbon (BrC) (in cloud vs out vs interstitial air)
- Improve understanding of secondary BrC formation chemistry (possible insights from AMS and CIMS?)



Hems et al., ACS Earth Space Chem., 2021

Measurements of PM residuals

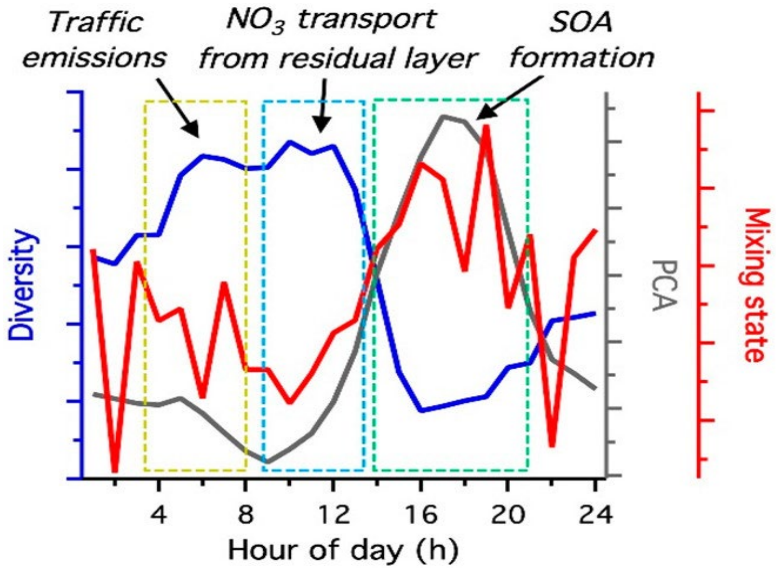


Other PM instruments (**HR-ToF-AMS, CPC, etc**…)

- **Non-refractory aerosol chemical composition measured by a high-resolution aerosol mass spectrometer**
- Event-trigger mode: Size-resolved chemical composition of single particle (Organic, SO_4^{2-}, NO_3^-, NH_4^+, Cl^-) → Clustering

Objectives/questions:

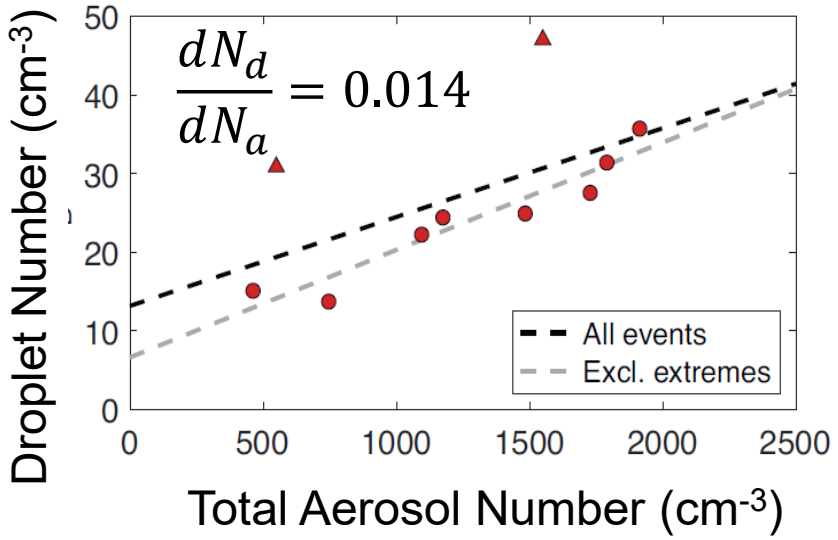
- Understand the size-resolved chemical composition and mixing state of ambient aerosols and cloud droplets
- Investigate the potential impacts of in-cloud processing on aerosol composition and mixing state

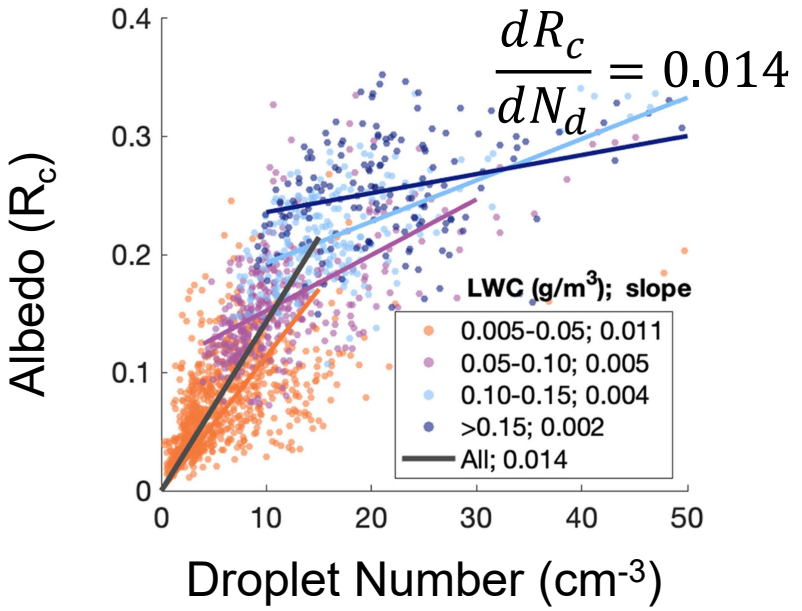


Fontana, CA (Summer 2015)

Lee et al., ES&T, 2020

Exploring aerosol-droplet interactions in fog





Aerosol-cloud albedo effect can be constrained by measuring:

- Total aerosol concentrations (residuals + interstitial)
- Droplet concentrations
- Cloud extinction, which can be used to approximate albedo
- Results from Nova Scotia showed that albedo increased 0.55 – 3.8 x 10^{-4} per additional particle cm^{-3} (Duplessis et al., *Atmospheric Research*, 2021)

$$\frac{dR_c}{dN_a} = \frac{dN_d}{dN_a} \times \frac{dR_c}{dN_d}$$