Harvard John A. Paulson School of Engineering and Applied Sciences

Mini-MOUDI analysis of Chemical and Morphological Properties of UT/LS particles from SABRE

SABRE Science Team Meeting, Spring 2024

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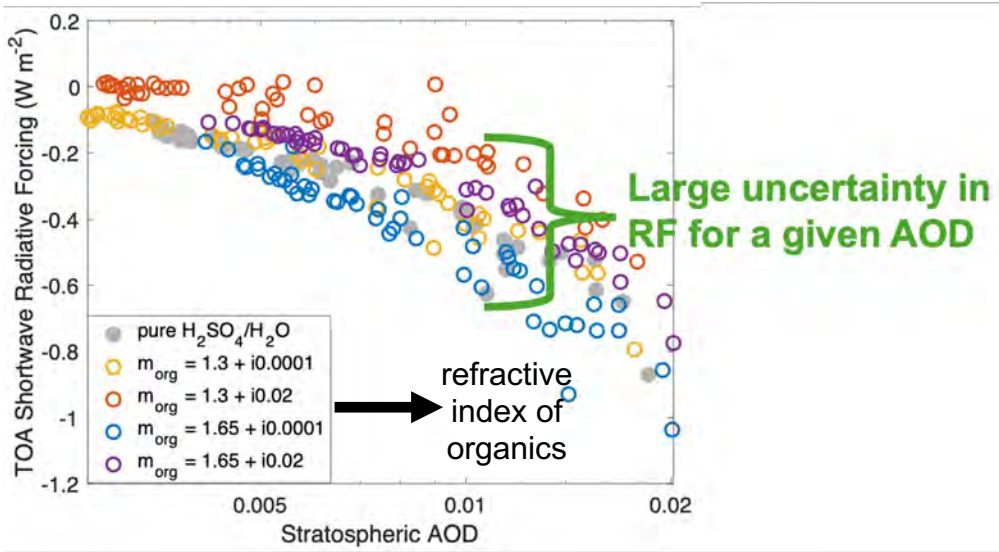
Yaowei Li, Frank Keutsch, Swarup China, Zezhen Cheng, Zhenli Lai, Gregory Vandergrift

Stratospheric aerosol radiative forcing is highly dependent on the particle composition and mixing states, not just sAOD

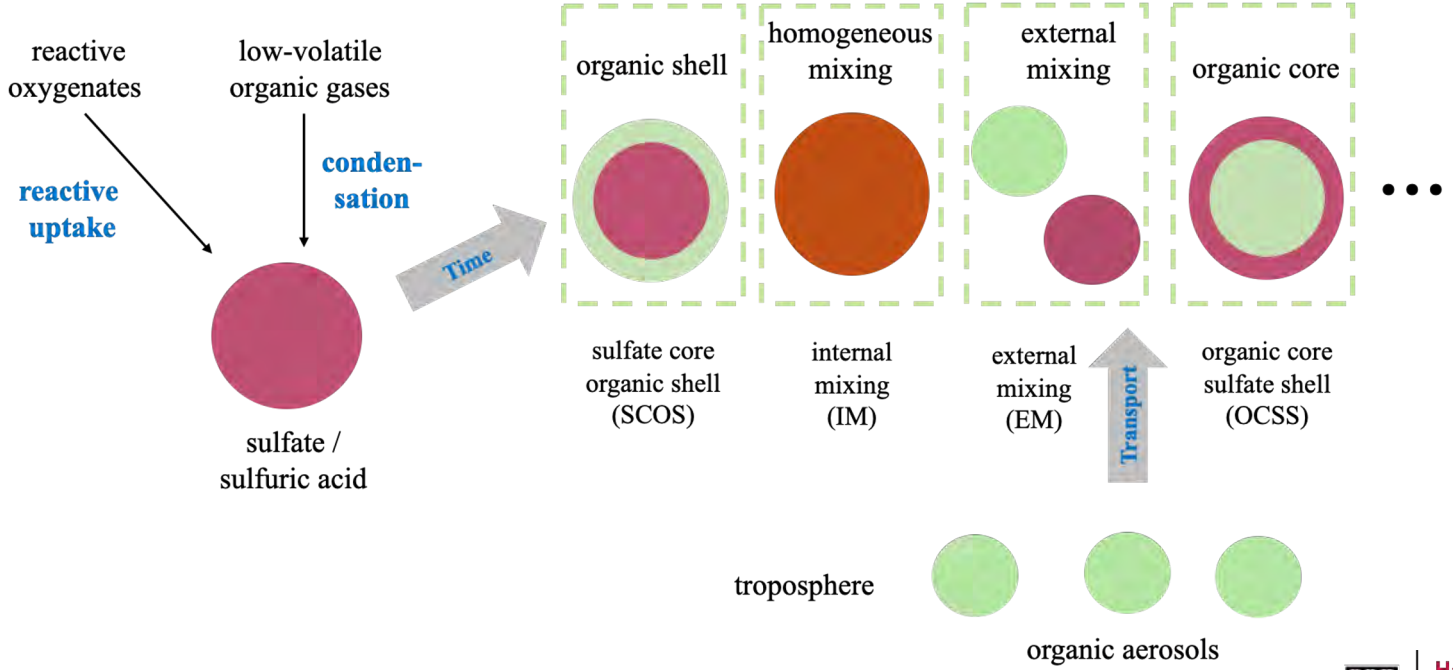


Sensitivity studies on U. Wyoming balloon-borne aerosol measurements

Courtesy of Yaowei Li

Li et al., GRL 2021

The mixing state of organics and sulfate in the stratosphere is largely unknown



Courtesy of Yaowei Li

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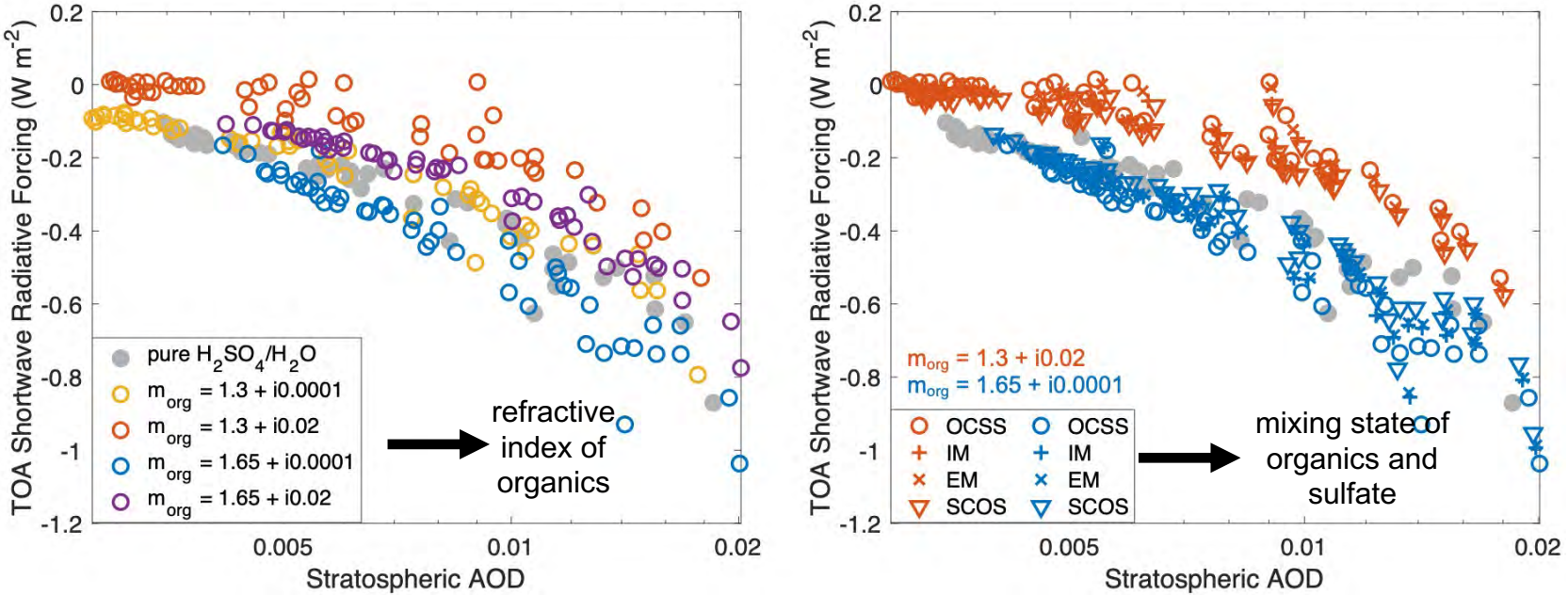

Compared to sulfate, organic-containing aerosol (50% organic volume fraction) may cause **±100% change in shortwave radiative forcing** depending on refractive index and mixing state. While there is very little data to constrain them.

Courtesy of Yaowei Li

Li et al., GRL 2021

Mini-MOUDI: miniature Micro-Orifice Uniform Deposit Impactor (Harvard University)

Size-fractionated aerosol collection for offline analysis

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Mini-MOUDI collection

Products: **chemical composition and morphology of individual particles**

Size stages: (F) 180-320 nm, (8) 320-560 nm, (7) 560-1000 nm, (6) 1000-1800 nm, (5) 1800-3200 nm

Each stage can collect multiple parallel samples of the same size from the same air mass

Chemical imaging analyses

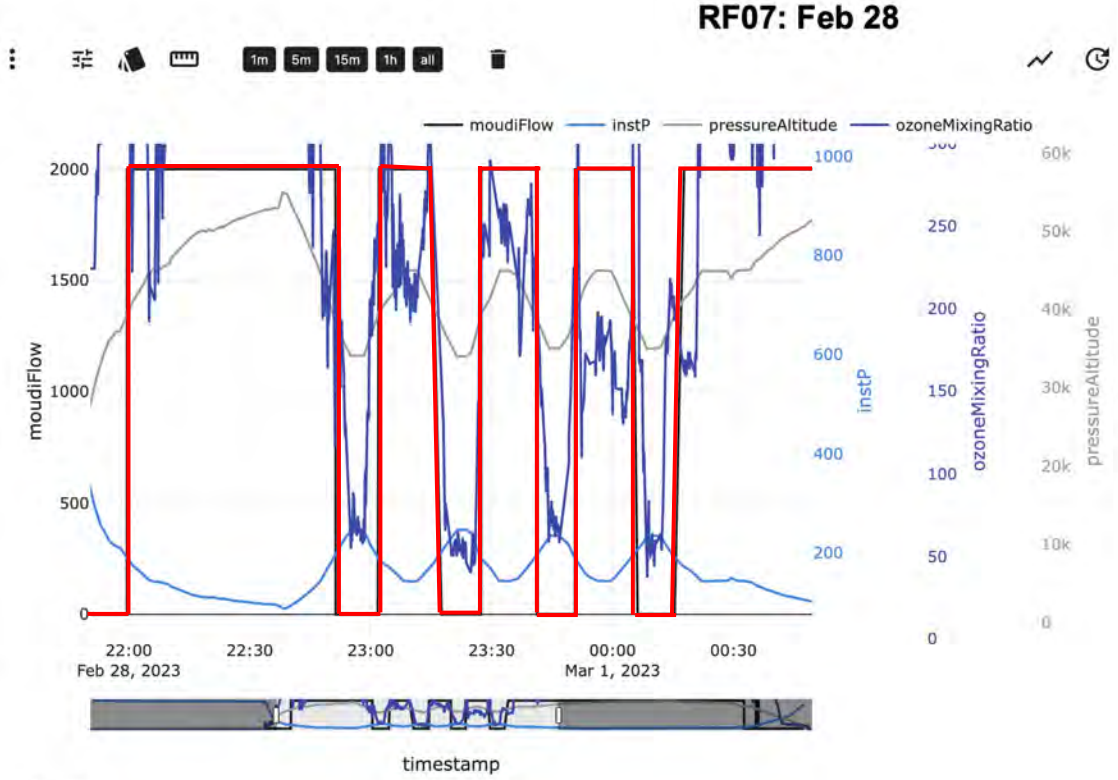
1. CC SEM-EDX
2. STXM-NEXAFS

Different substrates for different analytical instruments

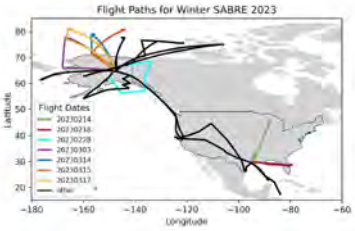
Electron microscope grid (round one): scanning electron microscope analysis (SEM)

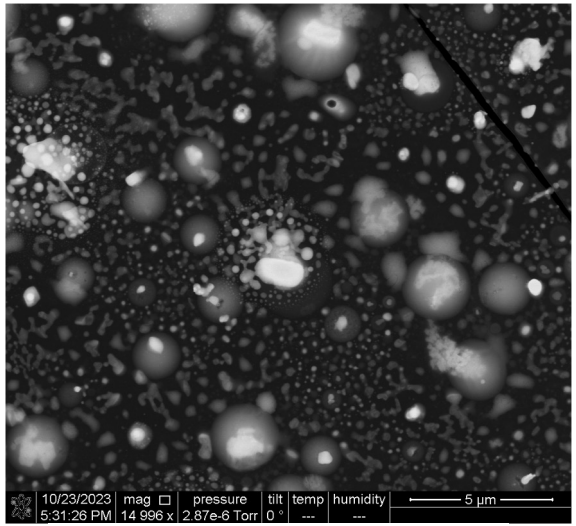
Silicon Nitride Window (square one): x-ray microscopy/chemical imaging (STXM)

Real-time control: sample specific targets (rocket plumes…) or avoid specific events (cirrus clouds…)

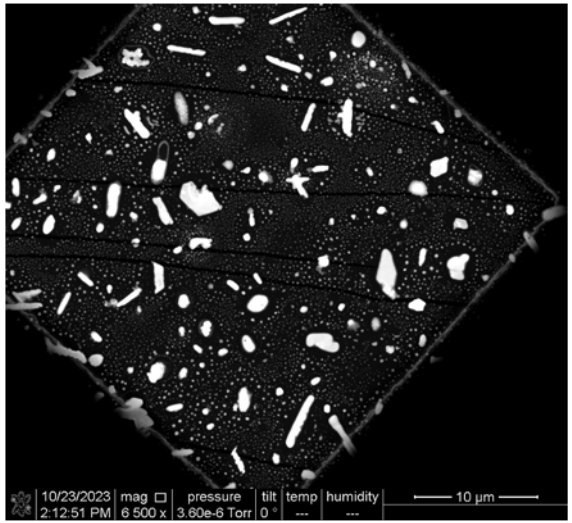


CCSEM-EDX Polar Vortex particles show range of morphologies

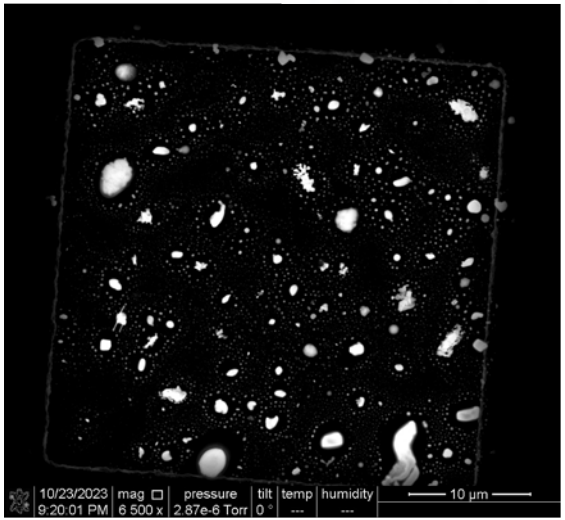




RF12: 03/14/2023



RF13: 03/15/2023

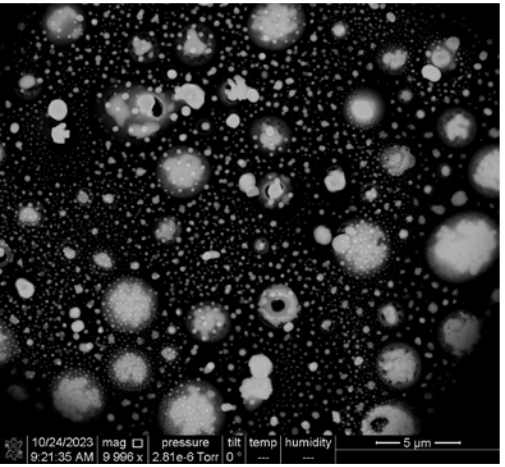


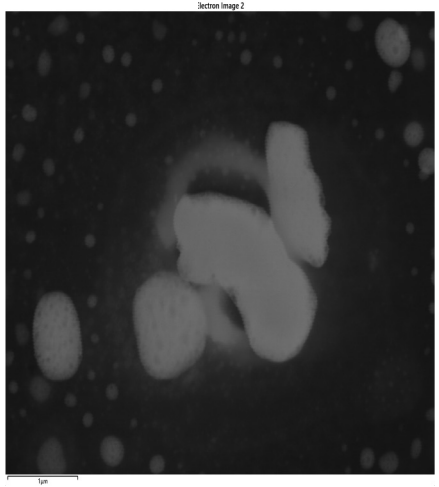
RF14: 03/17/2023

- Stratospheric polar vortex flights with low N_2O (99, 118 ppbv) and high O_3 levels (>1500 ppb) show abundant sulfate-rich and some carbonaceous particles, with Fe, Cu, Al signals likely from cosmic dust influence; carbon signals likely substrate interference

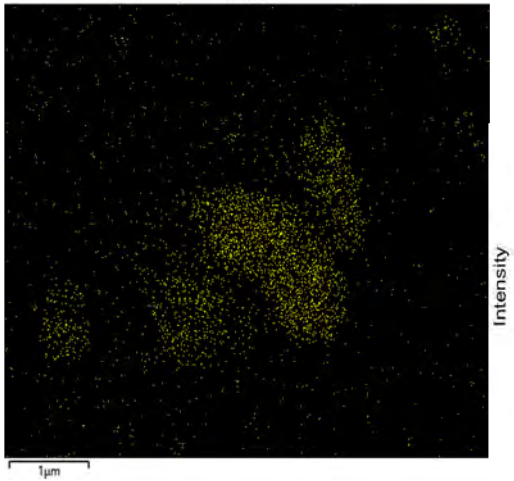
CCSEM-EDX Polar (non-vortex) particles also have range of morphologies

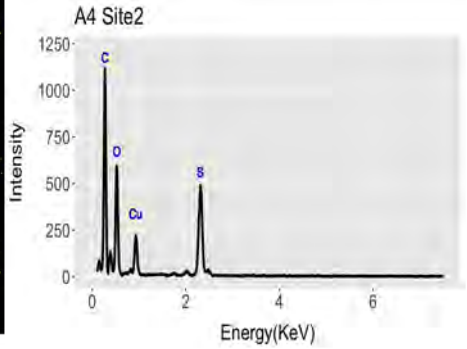
RF07: 03/03/2023





Sulfur Contributions

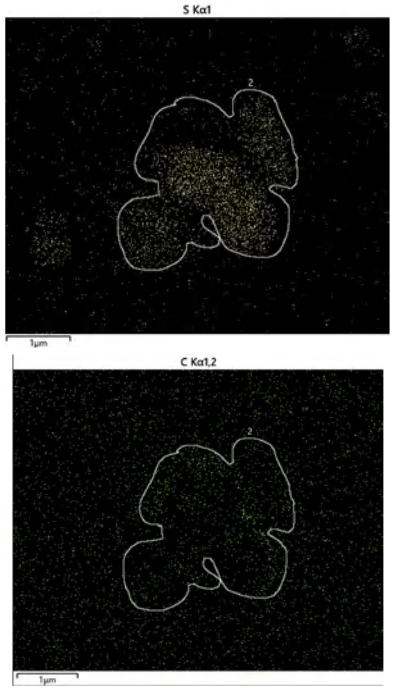


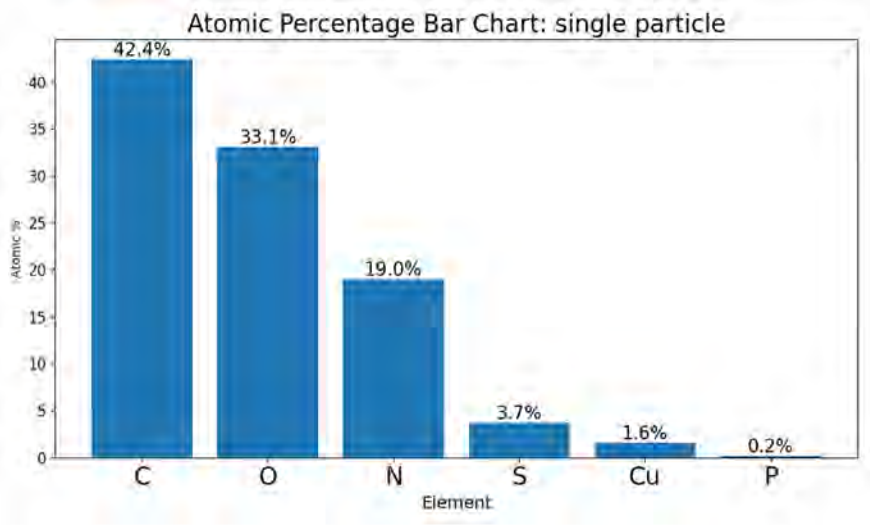


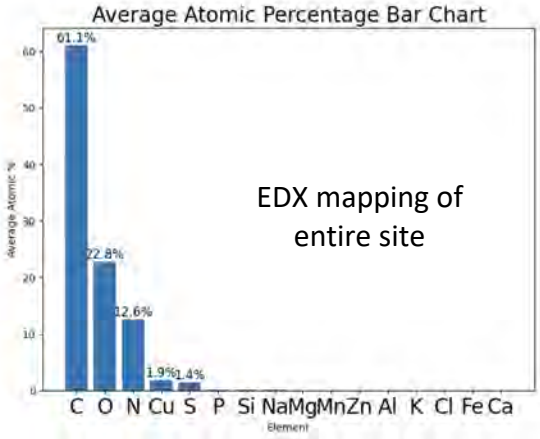
- >1200 ppb ozone, aged stratospheric air
- Stratospheric polar (non-vortex) flight show strong sulfate signal for many particles
- CCSEM-EDX can give us mixing state information

CCSEM-EDX allows for entire mapping and single particle tracing

RF07: 03/03/2023







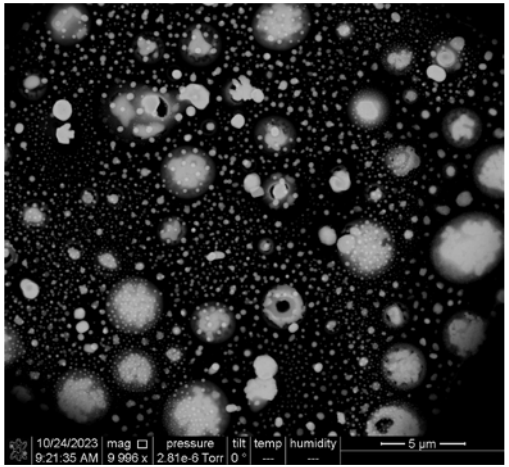
EDX mapping of entire site

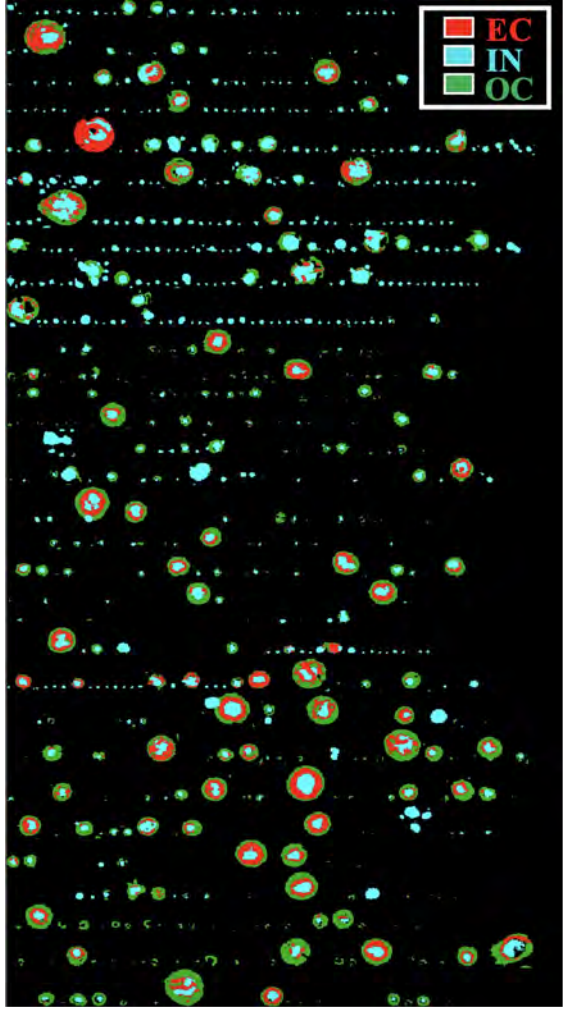
- Substrate interferences are likely carbon, oxygen, nitrogen, copper

In collaboration with Joy Li at PNNL

STXM analysis offers chemical composition information on the single particle basis for the entire substrate

RF07: 03/03/2023

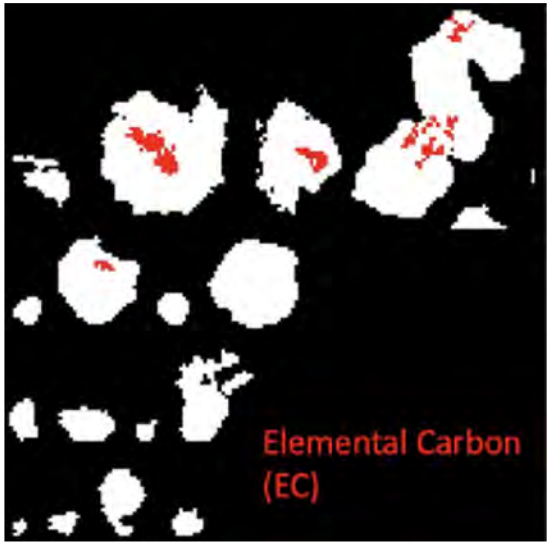


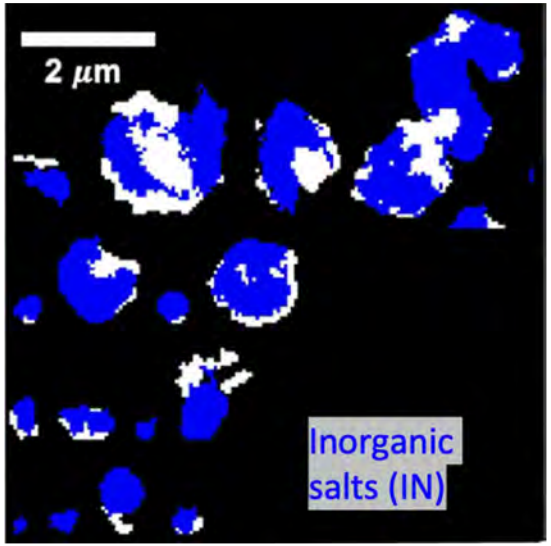


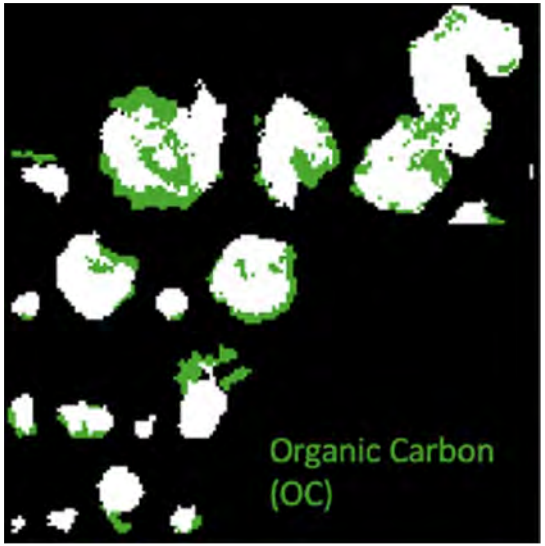
- STXM can characterize mixing states (EC, IN, or OC) which in turn will help us determine RF

In collaboration with Joy Li at PNNL

DCOTSS Mini-MOUDI characterization via STXM





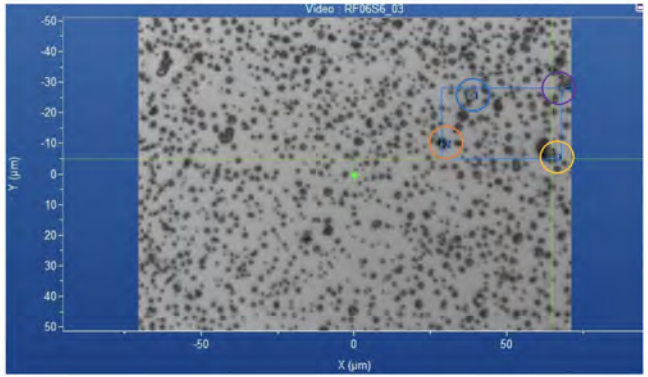


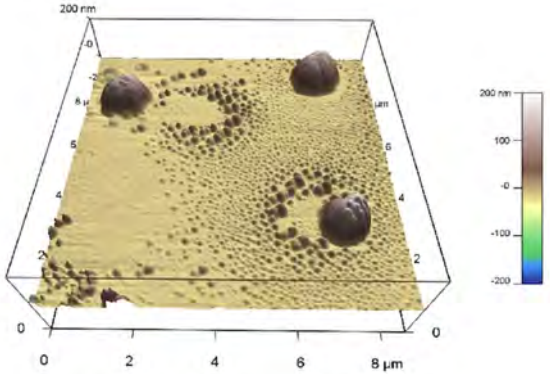
DCOTSS offline analysis of stratospheric aerosol samples: most of the particles were internally mixed with inorganics and organics, sometimes BC

Li et al., in prep. In collaboration with Alex Laskin, Steven Sharpe, and Felipe Rivera-Adorno at Purdue University

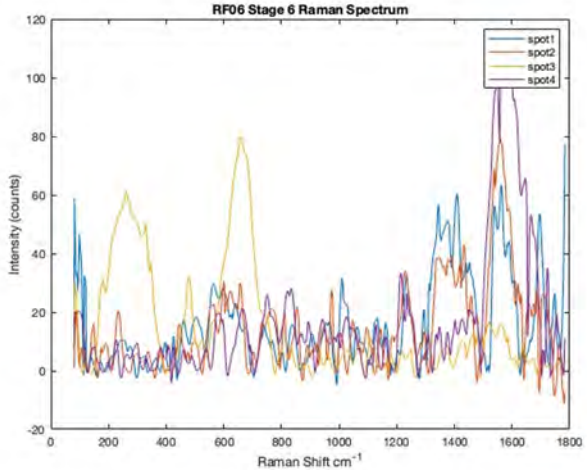
Courtesy of Yaowei Li

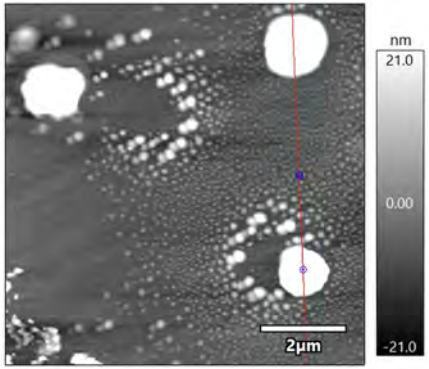
Raman Spectroscopy/Atomic Force Microscopy possibilities…

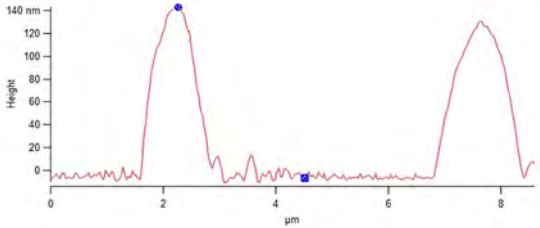




Take note of different axis units (nm vs um)

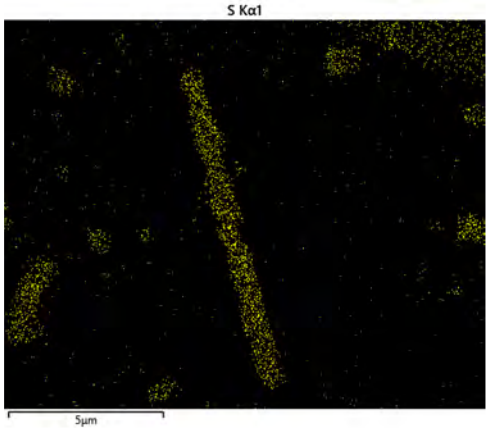






Near-future directions

- SEM-EDX analysis of polar vortex flights compared to Houston UTLS flights for single particle elemental composition and morphologies (i.e. the figure to the right)



- Metal inclusion particles on the single particle analysis basis, especially those with potential spacecraft influence (*Murphy et al. PNAS 2023*)

- STXM clustering analysis

Open to collaborations!

Thank you!

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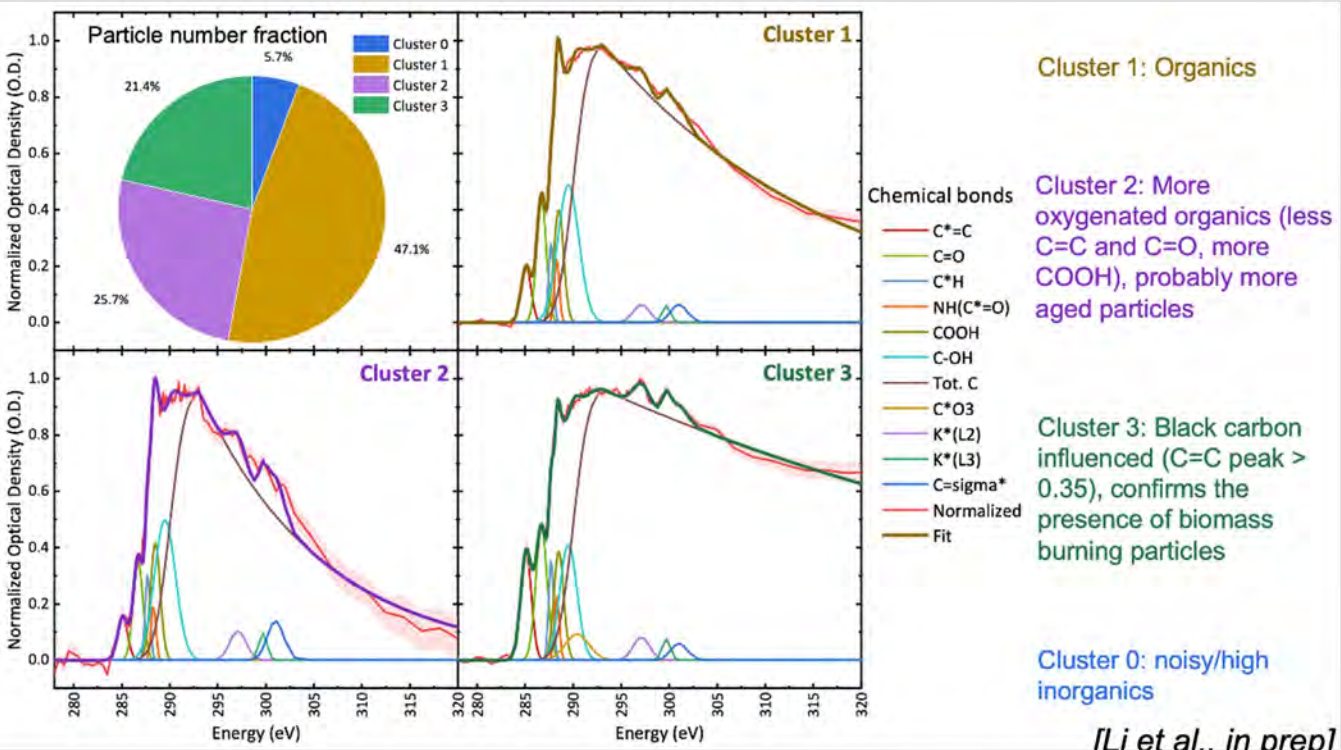
Supplemental Slides

DCOTSS Mini-MOUDI samples show substantial organics and biomass burning influence, as well as oxygenated organics

Near Edge X-ray Absorption Fine Structure (NEXAFS) spectra



Cluster 1: Organics

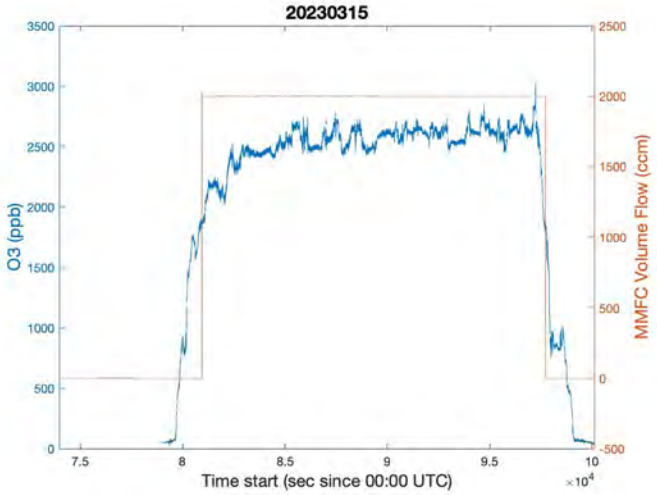
Cluster 2: More oxygenated organics (less C=C and C=O, more COOH), probably more aged particles

Cluster 3: Black carbon influenced (C=C peak > 0.35), confirms the presence of biomass burning particles

Cluster 0: noisy/high inorganics

[Li et al., in prep]

Harvard Flight Summary: A1 site RF13 S6 March 15, 2023



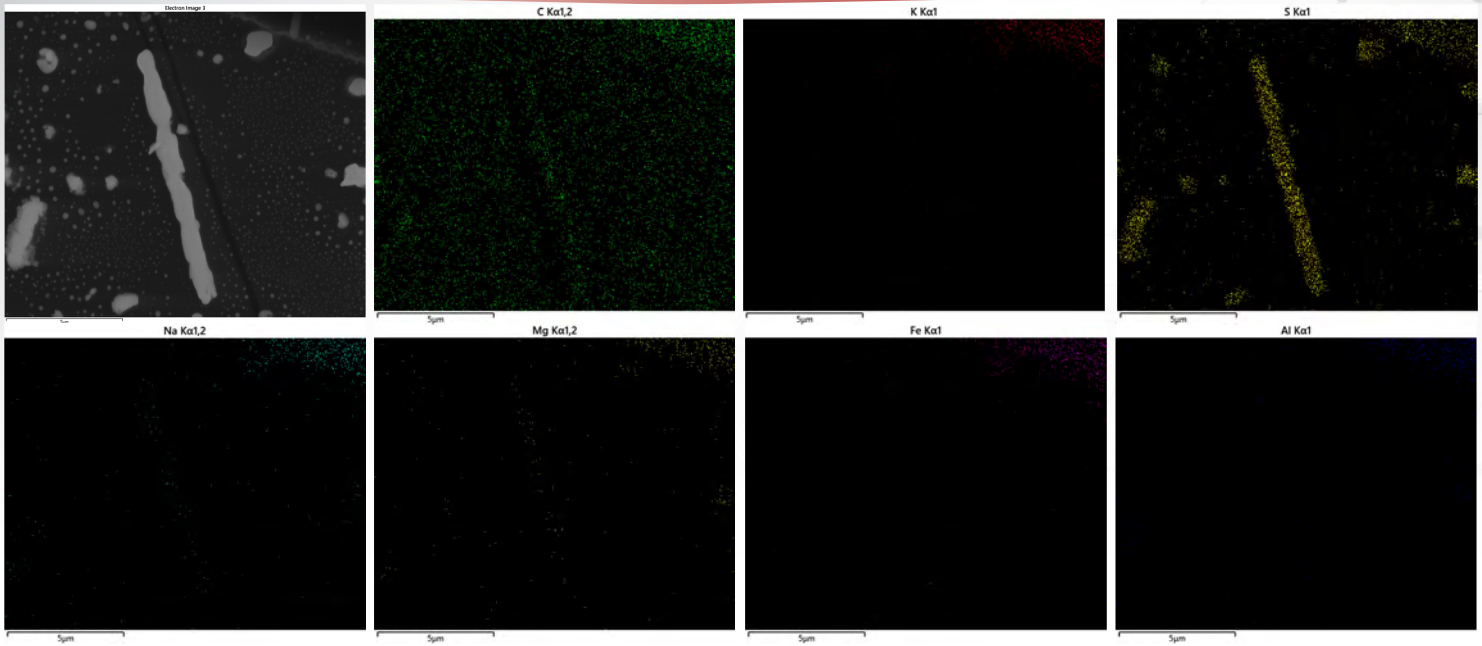
Polar Vortex air in stratosphere with N2O levels as low as 99ppbv

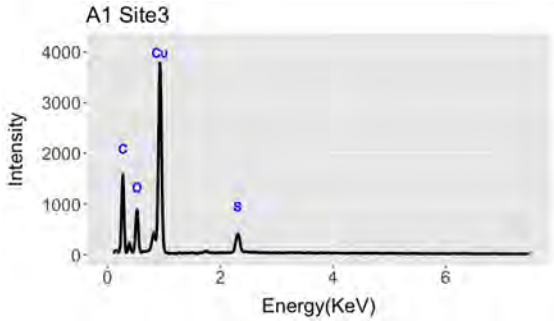
Mostly sulfate-rich carbonaceous particles

Some copper + k-rich particles -> could be cosmic dust(?)

Smaller particles surrounding center, larger, particles could be debris of droplet impaction during sample collection

A1 site3





Sulfate-rich carbonaceous particles. Sulfate-rich particles are abundant in this sample