Working with SAIL Aerosol Measurements

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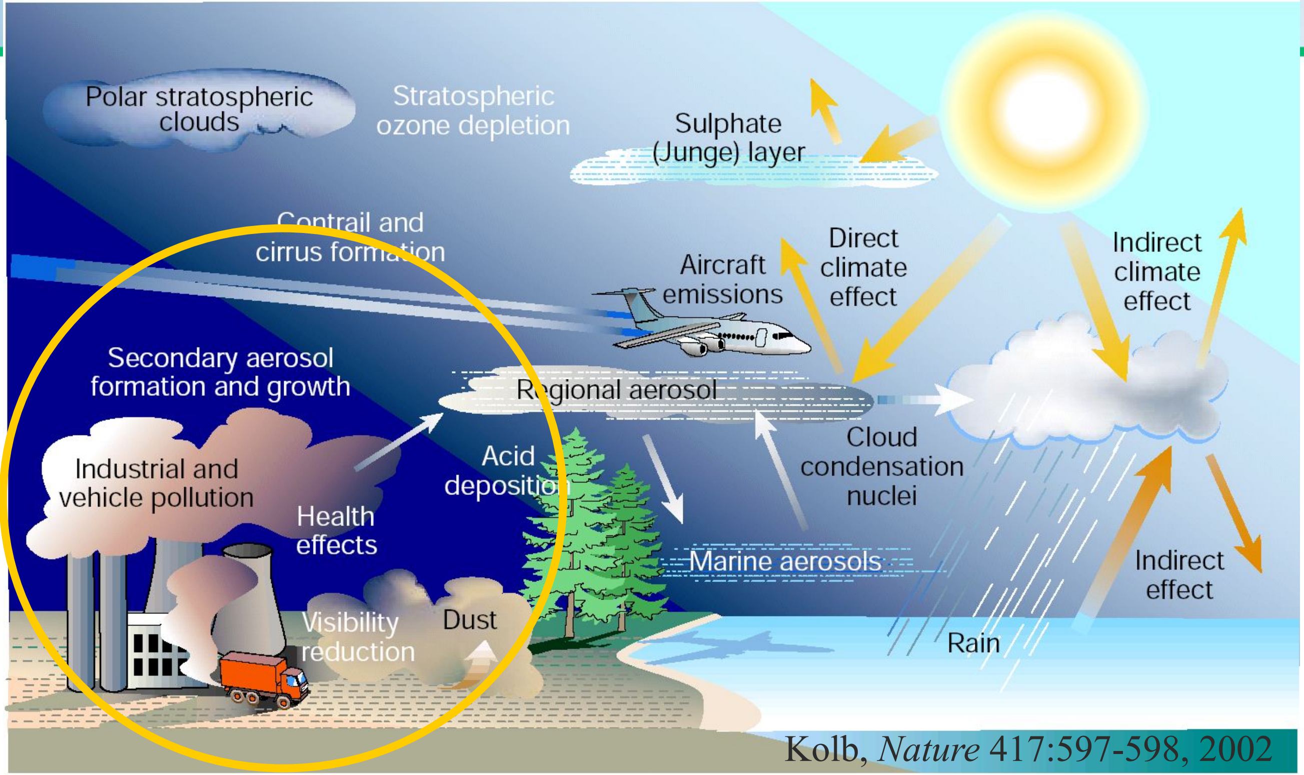
AMS Annual Meeting
SAIL Short Course
January 8, 2023

LA-UR-22-XXXXX

Why Study Atmospheric Aerosols?

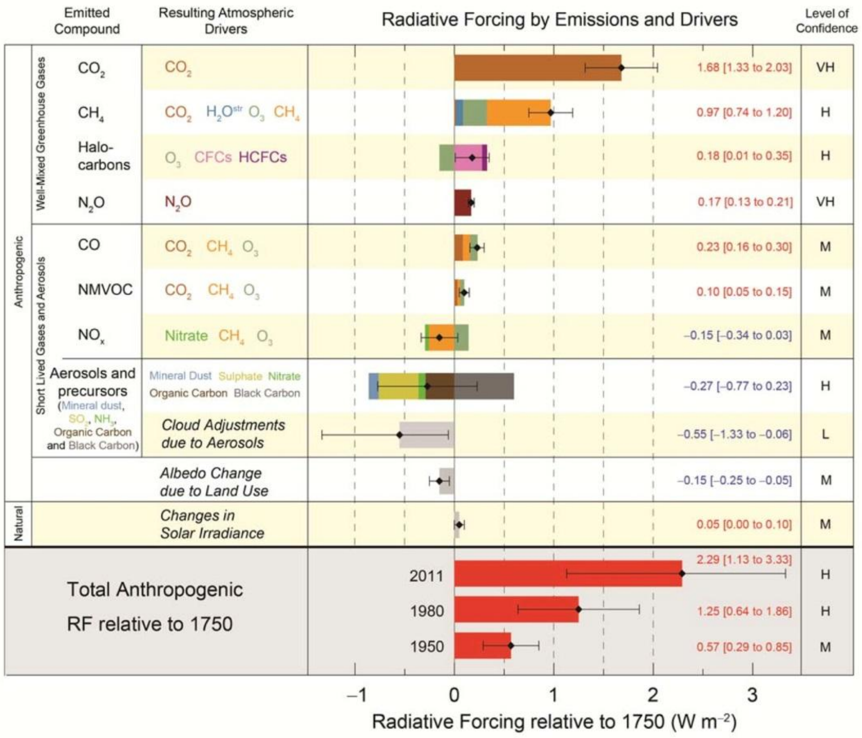


Goal: Decrease uncertainties with direct measurements (size, composition, optical properties, and concentration)

U.S. DEPARTMENT OF ENERGY

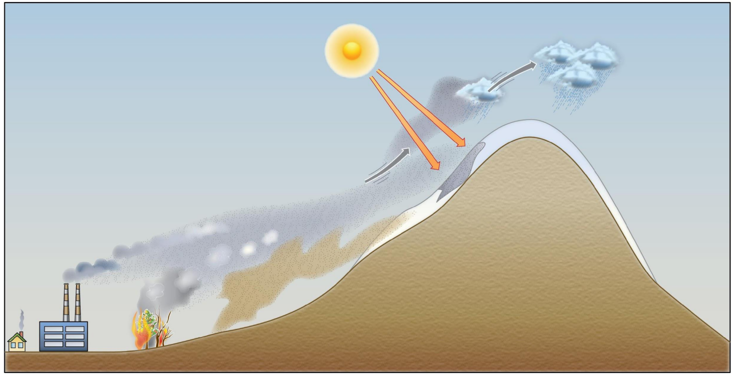
Ambient Aerosols: Impacts on Climate

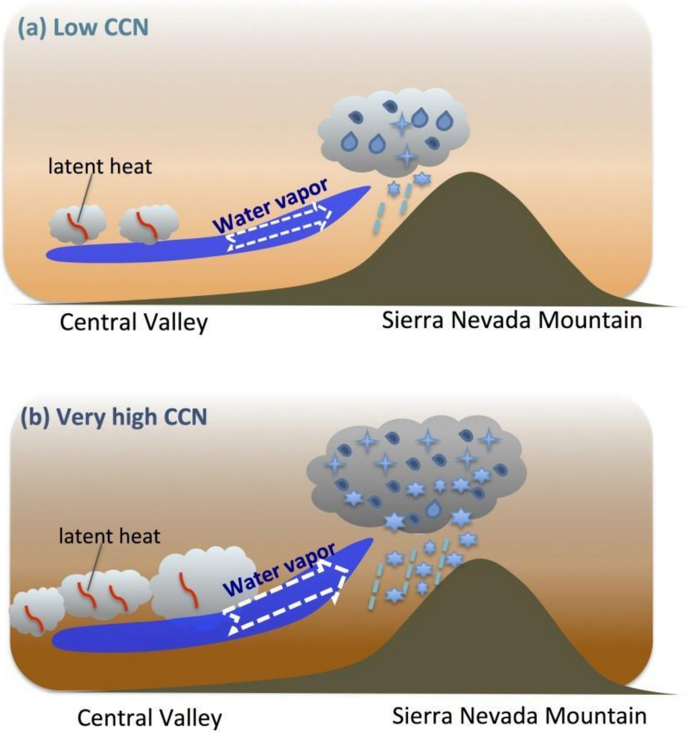
- **Greenhouse Gases – Terrestrial Warming from CO_2**
- **IPCC Aerosol Radiative Forcing**
  - First thought to be cooling and dominated by sulfates
  - 2007: Largest uncertainty is still in the aerosols
  - 2013 (AR5): BC absorption slightly increased (0.6 W m^{-2})
- **Chemical Information**
  - Most Aerosols absorb while others cool the atmosphere
  - **Black Carbon:** 2nd most important factor in global warming (behind CO_2) & most uncertain *(Bond et al., 2013)*
  - Sulfate and Organics – Cooling



IPCC, AR5, 2013.

Aerosol Regimes and Radiation





SCIENCE QUESTIONS

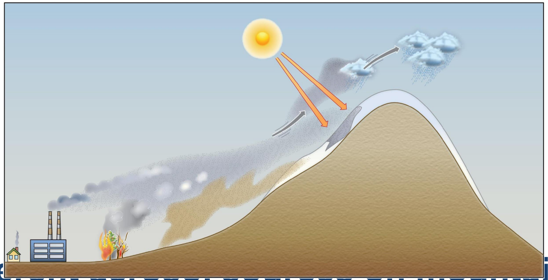
1) What are the dominant regimes of seasonal **aerosol transport, formation, growth, and removal processes** in the region?

2) Within these regimes, how do aerosol particles redistribute radiant energy, including **warming the atmosphere** and/or the **surface radiative forcing**?

3) How strongly do aerosols affect the surface energy and water balance by **altering and clouds and precipitation**?

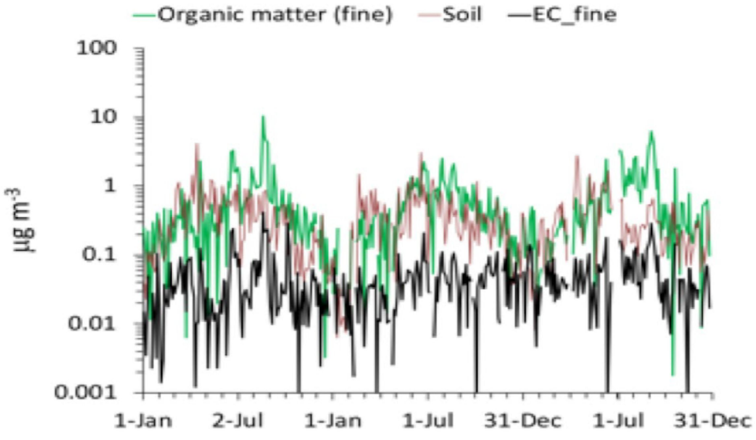
Aerosol Processes and Regimes

- **Aerosol processes impact the atmospheric and surface radiative environments**
  - Lifecycle (formation, growth, removal) and aerosol sources
  - Local and long-range transported aerosol
  - Atmospheric particles and deposition to the surface



- **Role of different aerosol sources on mountain hydrology**
  - Evidence of new particle and secondary organic aerosol formation and growth events in mountainous terrain
  - Spring and summer dust events and wildfires

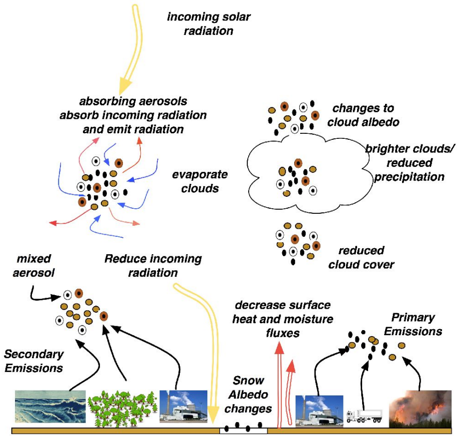
 Radiative impacts have been poorly constrained by observations – most studies rely on models



Annual cycles of aerosol mass concentrations of fine organic and elemental carbon, and soil dust at White River IMPROVE network [*Malm, et al.*, 1994] site, located close to the proposed SAIL site, at 3413 m MSL, 39.1536 latitude, -106.8209 longitude. The time period shown is from January 1, 2015 to December 31, 2017.

Absorbing Aerosols Alter Atmospheric and Surface Radiation

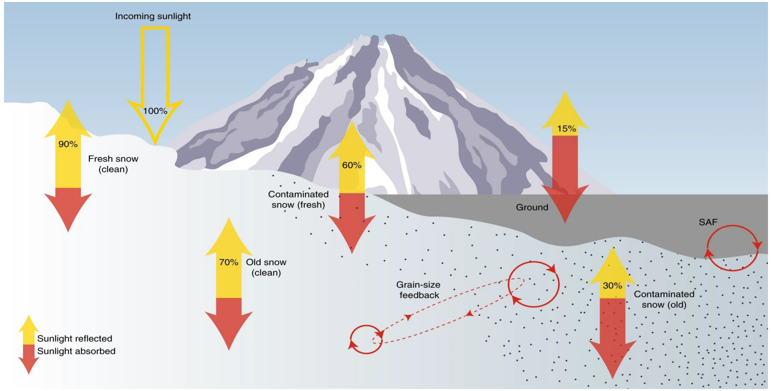
- **Atmospheric presence reduces solar radiation at the surface, increases atmospheric stability and decreases turbulent fluxes**
  - Absorbing dust species
  - Black carbon (BC, aka "soot")
  - Brown carbon and secondary organics
- **Brown carbon, implicated but understudied in high-altitude melt**
- **Transported particles vs. surface deposition**
- **Snow pit observation and sampling (Skiles/Univ. Utah)**
  - Snow pit albedo, depth, density, temp profiles, etc.
  - *In situ* Black Carbon (SP2) deposition
  - Offline dust and BC in snow



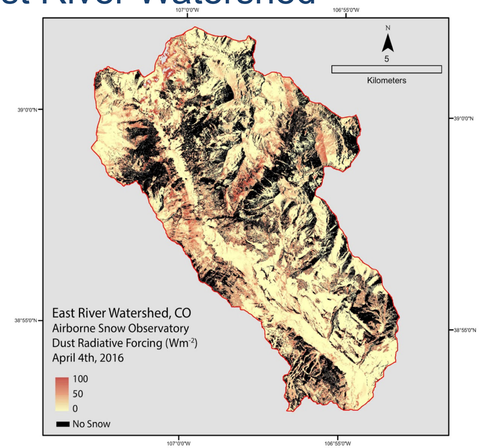
U.S. DOE BER Absorbing Aerosols Workshop Report (2016): Absorbing aerosols in the atmosphere and the various ways they interact with incoming solar radiation, clouds, and the dynamic and thermodynamic state of the atmosphere.

Absorbing Aerosols Alter Atmospheric and Surface Radiation

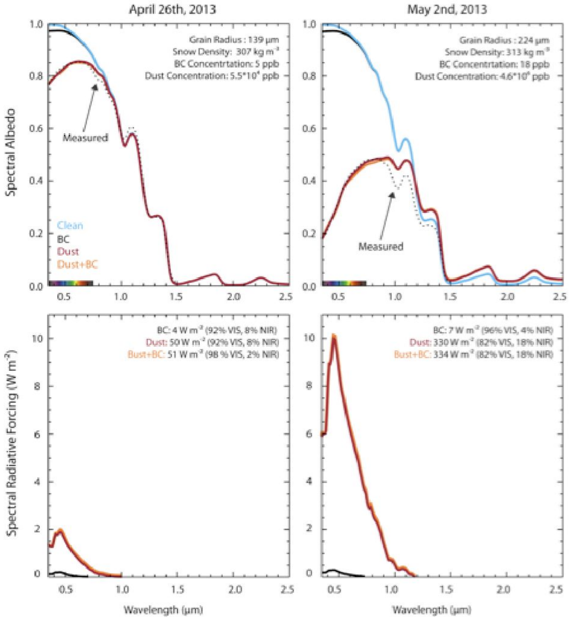
- **Absorbing aerosols deposit in/on snow**
  - Absorbed solar radiation increases at the surface
  - Snowmelt enhanced by lower surface albedo
  - Surface hydrology and watershed
- **Observations to constrain modelled radiative impacts in complex mountainous terrain**, e.g. Colorado East River Watershed



Skiles et al., Nature Climate Change, 2018.



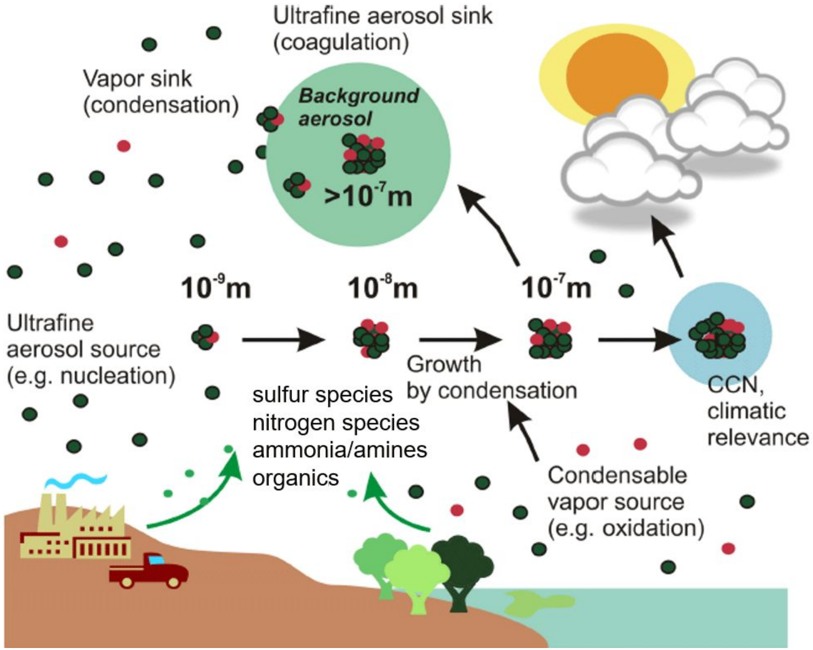
Radiative forcing by deposited aerosols in snow. NASA Airborne Snow Observatory (ASO), East River Watershed in April, 2016.



San Juan Mountain spectral albedo of snow. Lower plots associated SNICAR radiative forcing calculations.

Aerosol Lifecycle in Colorado

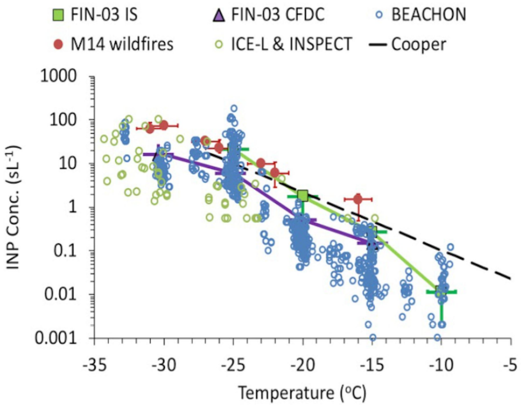
- **Aerosol lifecycles and the role of chemistry are key to understanding aerosol radiative impacts**
  - New Particle Formation and Growth
  - Removal (wet and dry deposition)
- **Are new particle events** on Colorado mountain terrain **observed when intrusions from the troposphere** mix with boundary layer trace gases?
- Do upslope valley winds transport **reduced nitrogen species to form secondary organic aerosol and Brown carbon**?
- What is the impact of **regional versus long-range transported dust, pollution and wildfire events**?
  - Black and Brown carbon
  - Secondary organic aerosols



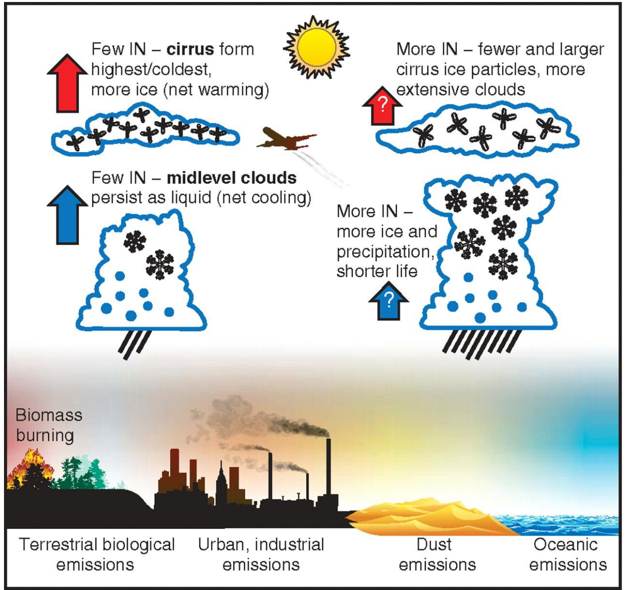
Riipinen, et al., ACP 2011.

Ice Nucleating Particles (INP)

- **Ice and mixed-phase clouds impact cloud radiative forcing, precipitation and cloud lifetime**
  - Biological INP often more important at higher temperatures (> -20 C)
  - Organic often dominate at lower temperatures
- Can we **observe/verify increased INP during summer convective storm and wildfire events**?
- What are the **dominant INP components for Colorado**?
- **Ice Nucleating Particle Sampler (Creamean + Hill/CSU)**
  - Immersion freezing measurements
  - Distinguish biogenic, inorganic and organic contributions



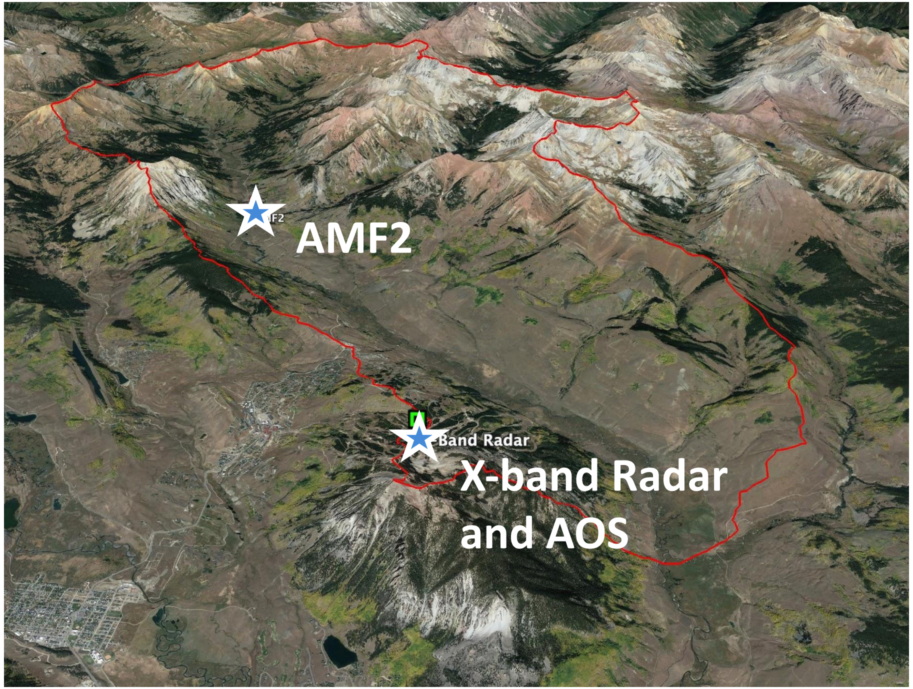
INP concentrations via immersion freezing from several campaigns since 2000 in the Colorado Mountains.



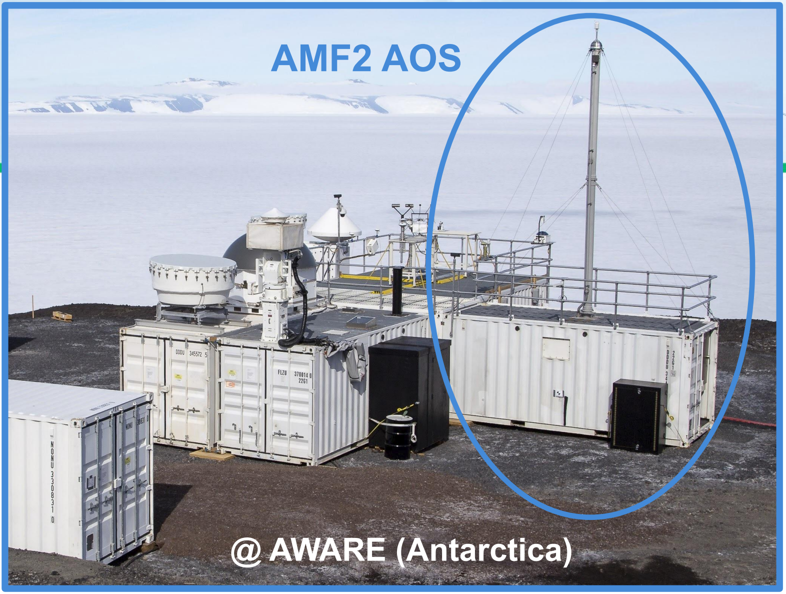
DeMott et al., PNAS, 2010.

ARM Aerosol Measurements

- **ARM Mobile Facility 2 (AMF2)**
  - Located in the town of Gothic
  - Rocky Mountain Biological Lab (RMBL)
  - EPA CASTNET Site
- **Aerosol Observing System (AOS)**
  - Co-located with the Radar on Crested Butte Mountain
  - *In situ* aerosol measurements
  - Regional aerosol processes
  - Ability to capture long-range transport
  - Minimal local sources
- **Tethered Balloon System (TBS)**
  - Aerosol configuration (CPC and POPS)



AMF2 *In Situ* Aerosol Measurements: Aerosol Observing System (AOS)



- **Chemistry**
  - Aerosol Chemical Speciation Monitor (ACSM)*
  - Single Particle Soot Photometer (SP2)*
- **Cloud Formation - Water Uptake and Ice Nucleation**
  - Cloud Condensation Nuclei Counter (CCN)
  - Humidified Tandem Differential Mobility Analyzer (HTDMA)
  - Ice Nucleating Particle (INP) Sampler
- **Optical Properties**
  - Nephelometer (NEPHDRY)
  - Particle Soot Absorption Photometer (PSAP)
- **Trace Gases and Meteorology**
  - CO Monitor (CO)
  - Ozone Monitor (O3)
  - Met Sensor (MET)
- **Physical Properties**
  - Optical Particle Counter (OPC)*
  - Condensation Particle Counter – fine (CPCF)
  - Condensation Particle Counter – ultrafine (CPCUF)
  - Scanning Mobility Particle Sizer (SMPS)*
  - Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)

LA-UR-21-25951

Uin, J., Aiken, A.C. et al. Atmospheric Radiation Measurement (ARM) Aerosol Observing Systems (AOS) for Surface-Based In Situ Atmospheric Aerosol and Trace Gas Measurements. *JAOT*, 2019. https://doi.org/10.1175/JTECH-D-19-0077.1

ARM Website information on the AOS

CAPABILITIES > INSTRUMENTS

AOS

HANDBOOK | CITE DATA | BROWSE DATA

AOS > AEROSOL OBSERVING SYSTEM

INSTRUMENT TYPE(S) > BASELINE • EXTERNAL • GUEST

The aerosol observing system (AOS) is the primary platform for in situ aerosol measurements at Earth's surface. Each AOS has a different complement of instruments, which are selected to ensure the best measurements at each deployment site.

The AOS measures aerosol optical properties to better understand how particles interact with solar radiation and influence the Earth's radiation balance. These measurements are useful for calculating parameters used in radiative forcing calculations, such as the aerosol single scattering albedo, asymmetry parameter, mass scattering efficiency, and hygroscopic growth. Measurements made with the AOS form a long-term record at the ARM fixed sites. Shorter measurement records are available from mobile deployments in a wide variety of geographical regions. These measurements are valuable for:

- identifying long-term changes in aerosol properties

RELATED DATA ANNOUNCEMENTS

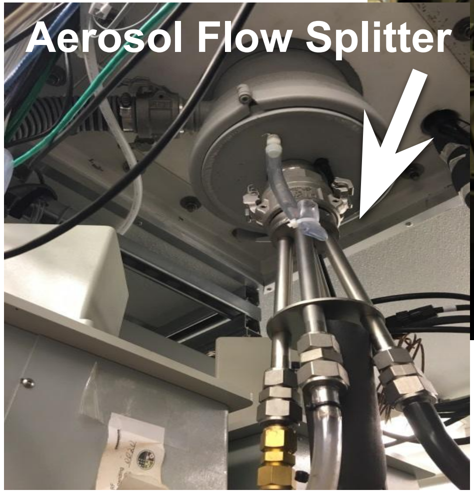
Boon of New Aerosol and Trace Gas Data for ARM Users

16 February 2021

REFERENCES

Uin et al. Southern Great Plains (SGP) Aerosol Observing System (AOS) Instrument Handbook. 2021. 10.2172/1756406. *View Citation*

In Situ Aerosol and Trace Gas Measurements



CAPABILITIES > INSTRUMENTS

NEPHELOMETER

HANDBOOK | CITE DATA | BROWSE DATA

NEPHELOMETER > NEPHELOMETER

INSTRUMENT TYPE(S) > BASELINE • EXTERNAL • GUEST

The nephelometer measures the total scattering and hemispheric backscattering of aerosol. Nephelometers are deployed in pairs, with one measuring the ambient conditions and the other measuring the scattering as a function of slowly increasing or decreasing relative humidity (RH). The combination can then be used to derive the hygroscopic growth factor as

RELATED DATA ANNOUNCEMENTS

New Best-Estimate Cloud Condensation Nuclei Data Product Available

20 April 2022

CAPABILITIES > INSTRUMENTS

SP2

HANDBOOK CITE DATA BROWSE DATA

SP2 > SINGLE PARTICLE SOOT PHOTOMETER

INSTRUMENT TYPE(S) > BASELINE • GUEST

The single-particle soot photometer (SP2) measures the soot (black carbon) mass of individual aerosol particles by laser-induced incandescence down to concentrations as low as ng/m^3.

The SP2 is part of the aerosol observing system (AOS).

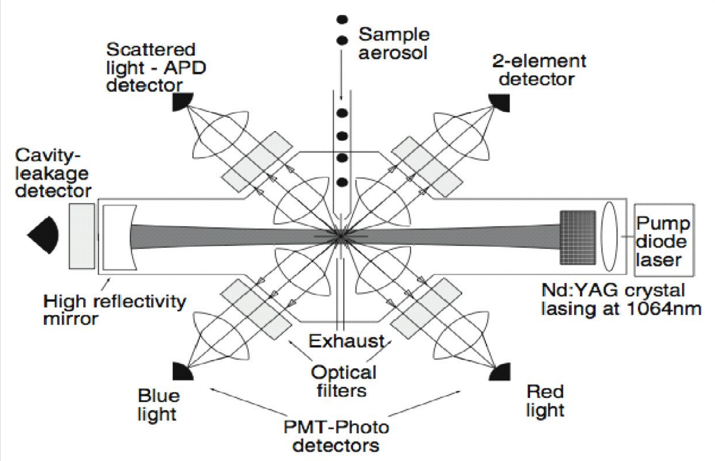
REFERENCES

Sedlacek et al. Single-Particle Soot Photometer (SP2) Instrument Handbook. 2017. 10.2172/1344179. ***View Citation***

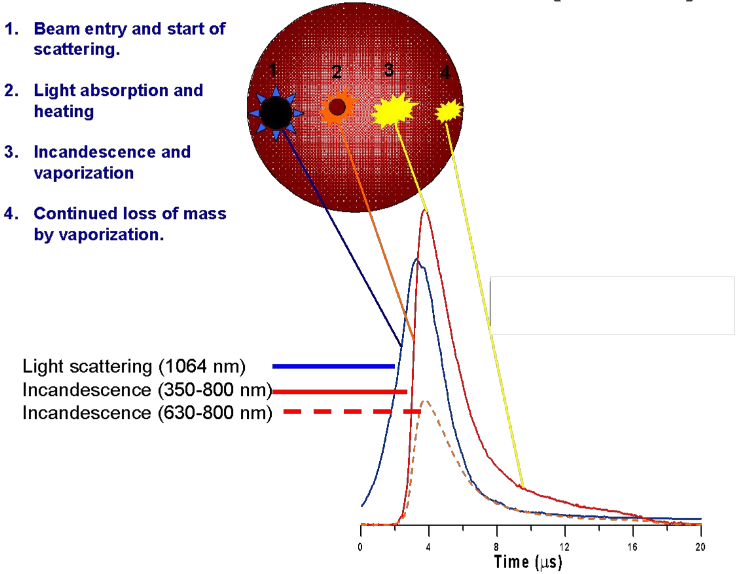
PRIMARY MEASUREMENTS

- Aerosol particle size distribution
- Black carbon concentration

Real-Time Direct Aerosol Instrumentation: CW LII in the Single Particle Soot Photometer (SP2)



Schwarz, J.P., et al. JGR-A, 2006.

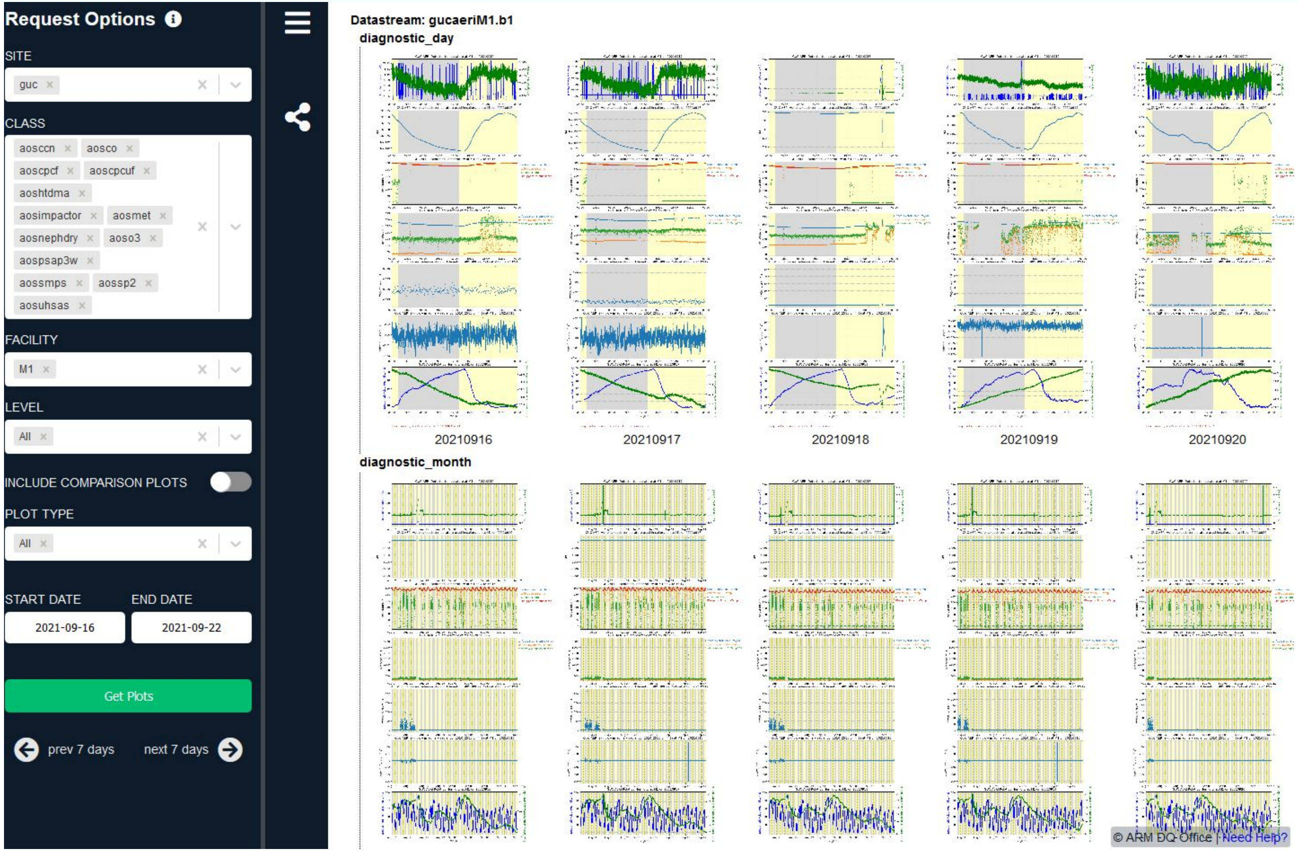


- **Refractory Black Carbon (rBC) Mass**
  - Direct, on-line, high time resolution measurement
  - Single particle incandescence and scattering
  - Highly sensitive: LOD ≤ 10 ng/m^3 (< 0.4/cm^3)
  - rBC size (derived from mass: approx. 50-700 nm *d)*

AOS Data is Online

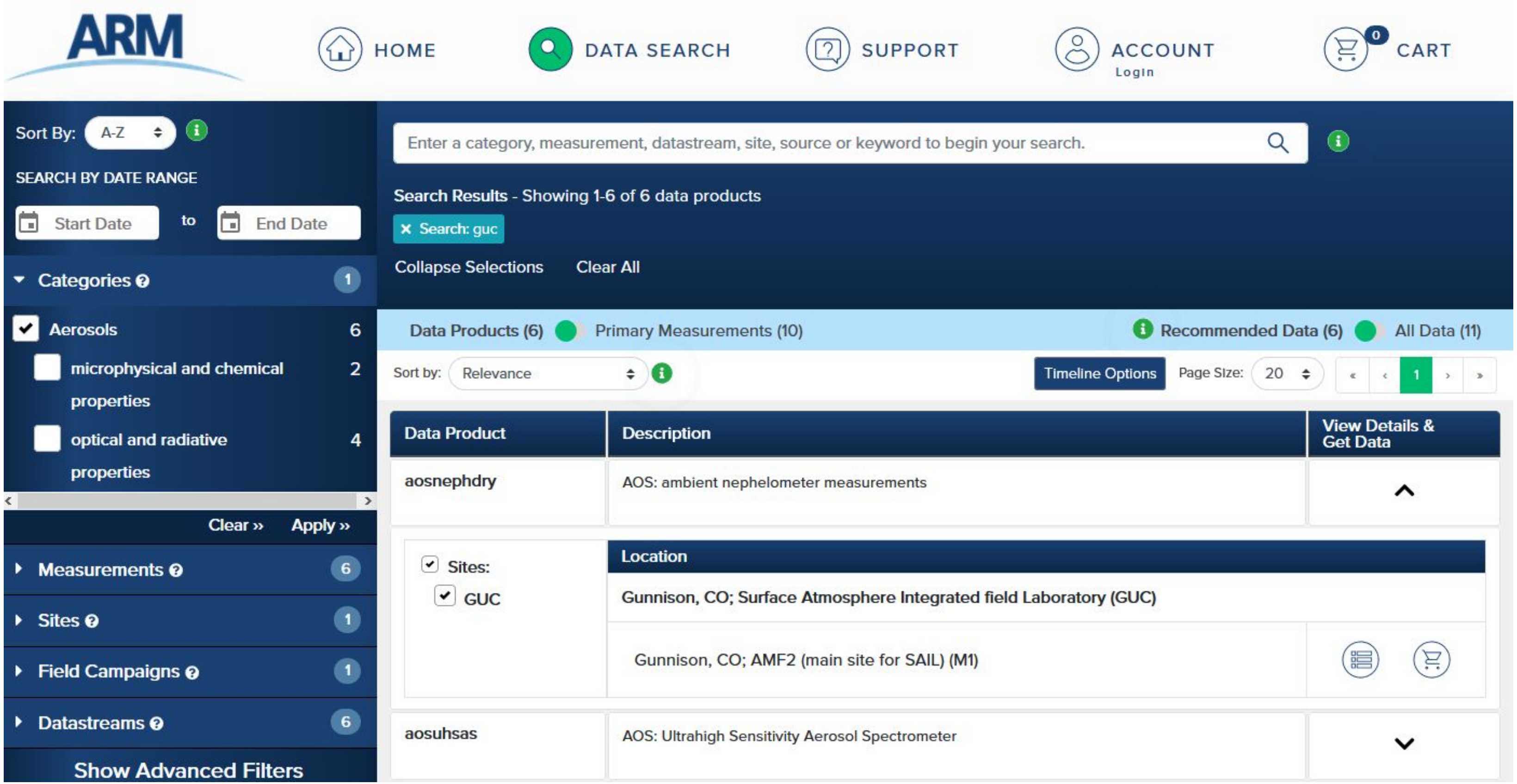
https://dq.arm.gov/dq-plotbrowser

ARM



ARM Data Discovery

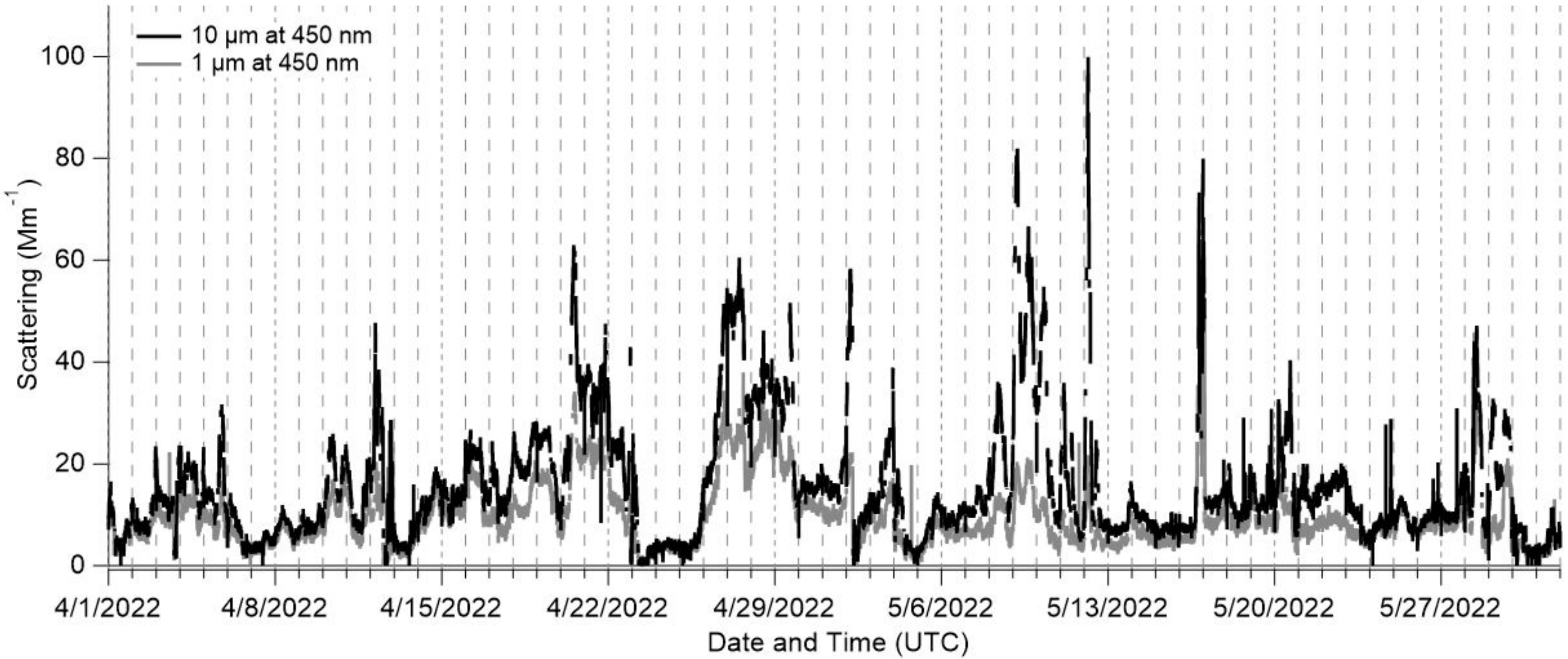
https://adc.arm.gov/discovery



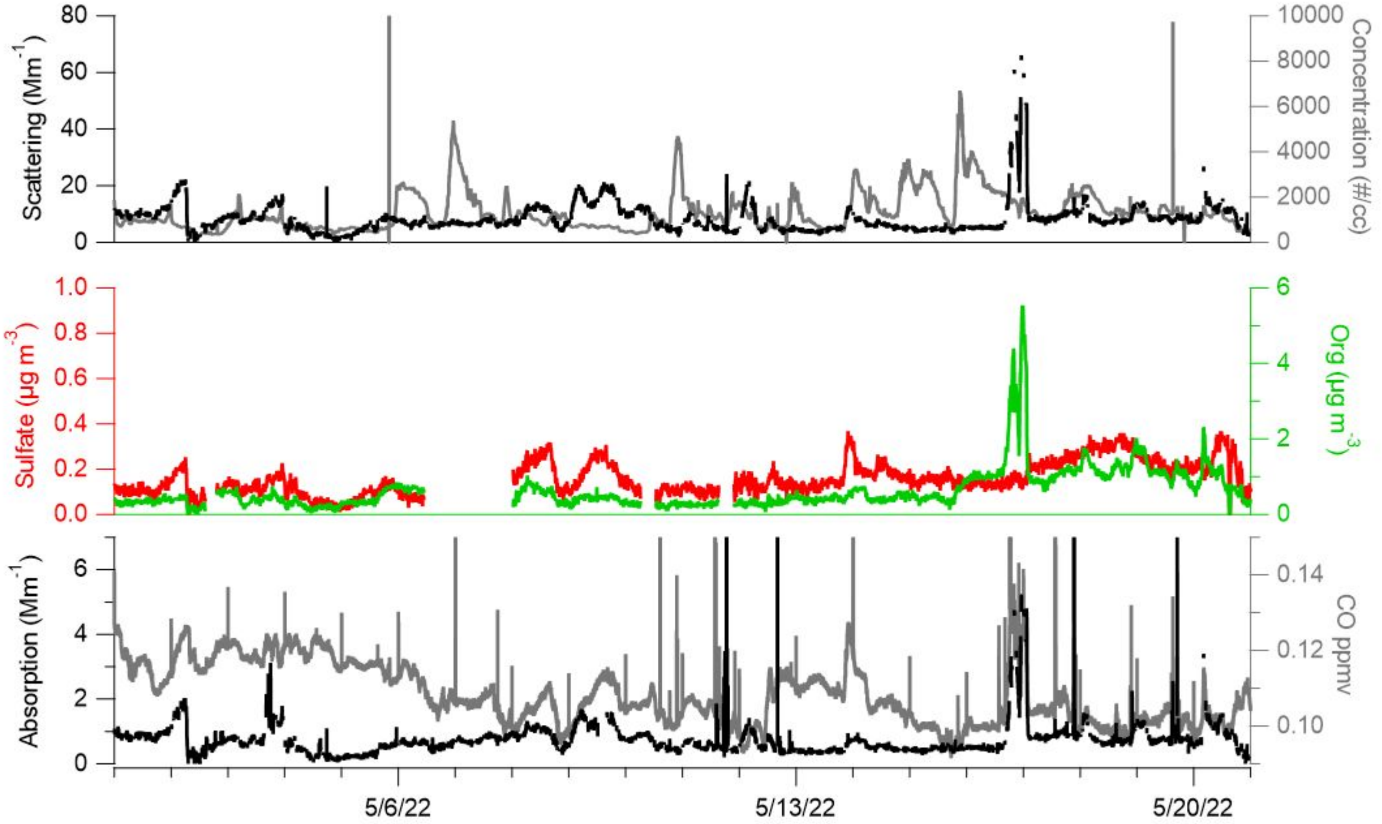
Supermicron Aerosol Sensors

Gothic AMF2 Site

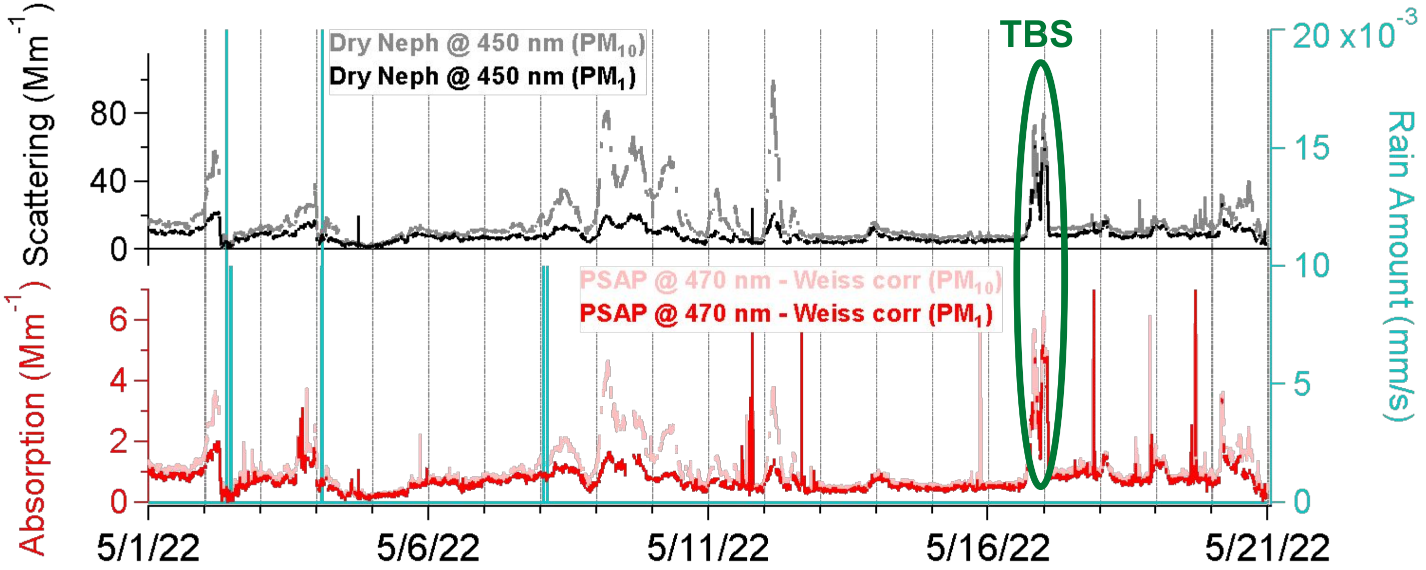
Aerosol Light Scattering



Aerosol Observing System (AOS) example data



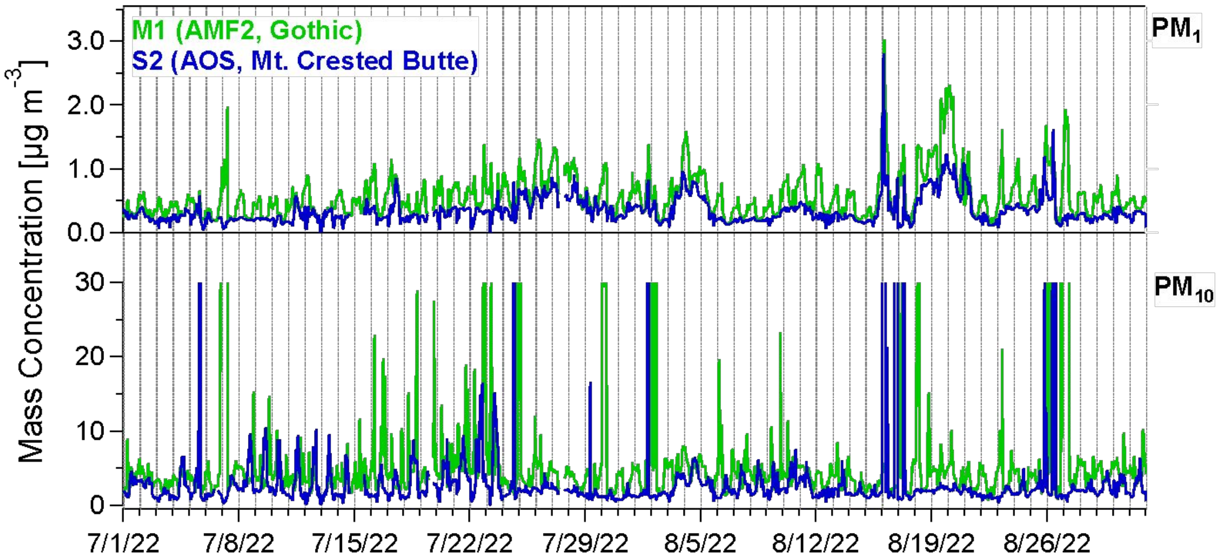
SAIL Aerosol Regimes and Processes: Results

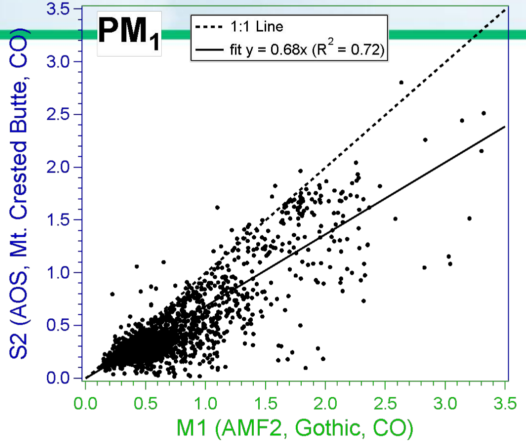


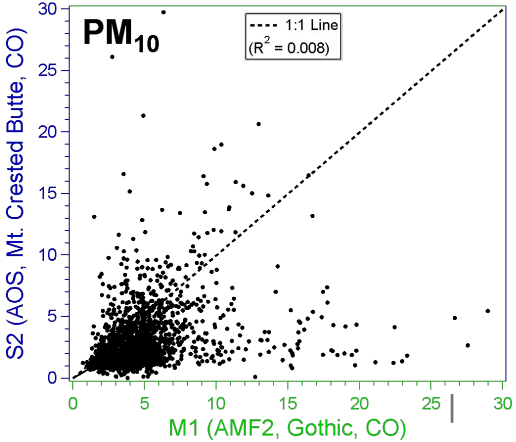
- AOS: Elevated supermicron events observed in May
- AOS and TBS: Submicron biomass burning event

SAIL Aerosol Regimes and Processes: Results

Summer 2022







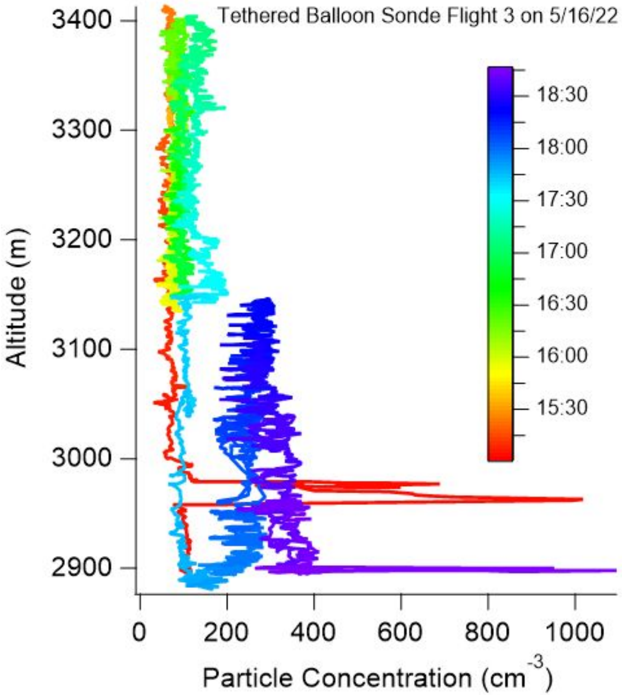
- PM_1 is well-correlated at the two sites and has a diurnal cycle at M1 that is not observed at S2 (AOS)
- PM_{10} is not well-correlated between the sites

SAIL Tethered Balloon Sonde (TBS)

Aerosol Properties

Instrument	Property Measured	Type
Printed Optical Particle Spectrometer (POPS) (6 units)	Aerosol size distribution from 140 nm to 3 µm	Baseline
Condensation Particle Counter (CPC) Model 3007 (4 units)	Total aerosol concentration from 0.01 µm to 1 µm	Baseline
Size- and Time-Resolved Aerosol Collector (STAC)	Size- and time-resolved chemical composition from 0.1 µm to 5.0 µm	Baseline
Cascade impactors (6 units)	Size-resolved chemical composition at four cut-off sizes (0.25, 0.5, 1.0, 2.5 µm)	Available upon request
MicroAeth AE-51	Black carbon concentration measured at 880 nm	Available upon request

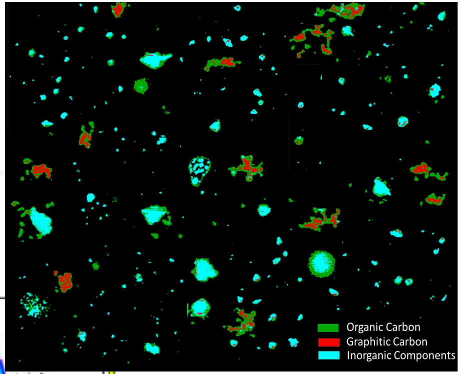
SAIL Tethered Balloon Sonde (TBS)

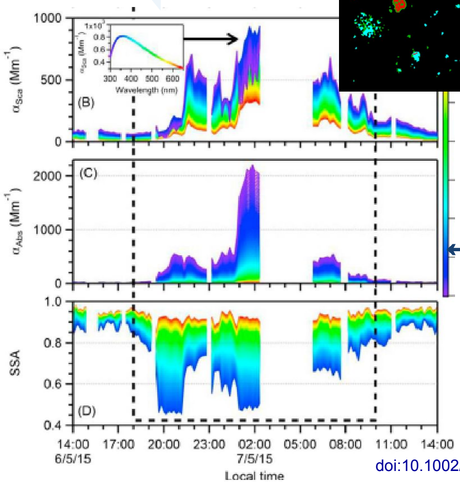


Off-line Studies of Aerosol Composition and Properties

- ## Chemical Imaging of Individual Particles

doi: 10.1021/acs.accounts.9b00396



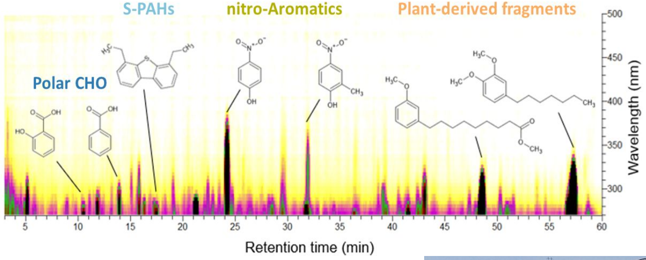


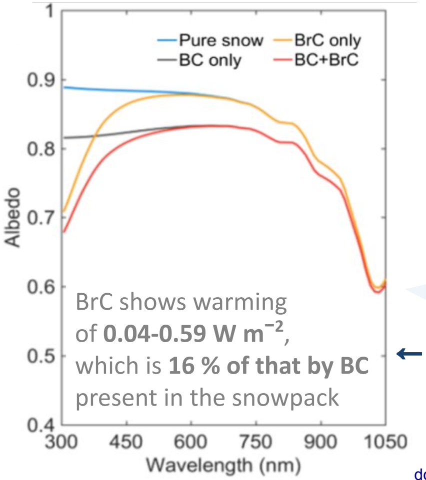
← **Informed interpretation of the *on-line* optical measurements**

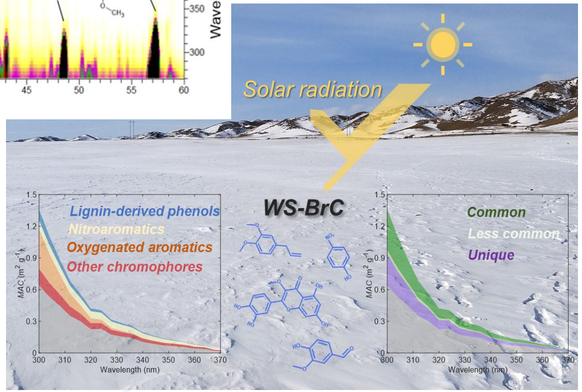
doi:10.1002/2016JD026230

- ## Composition-specific Optical Properties of Aerosol and Snow BrC

doi:10.1021/acs.analchem.8b02177



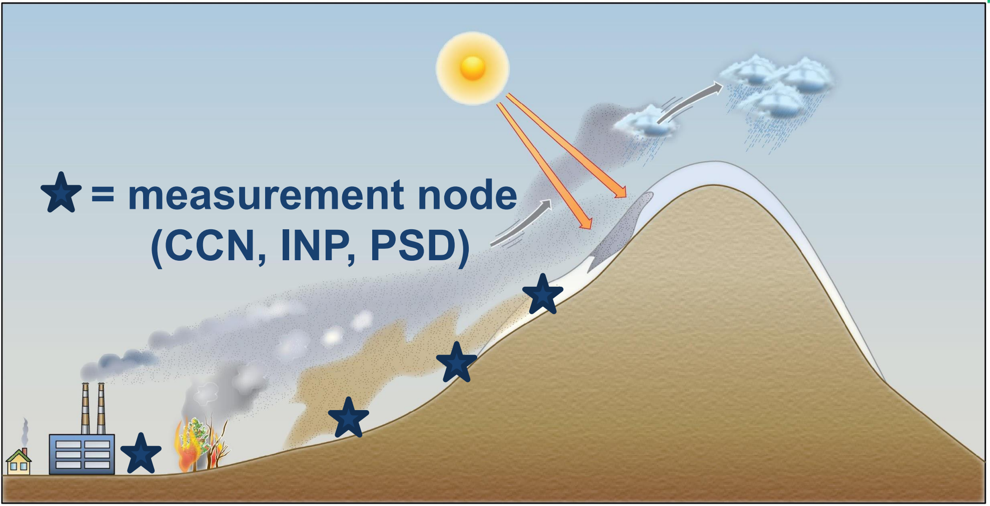


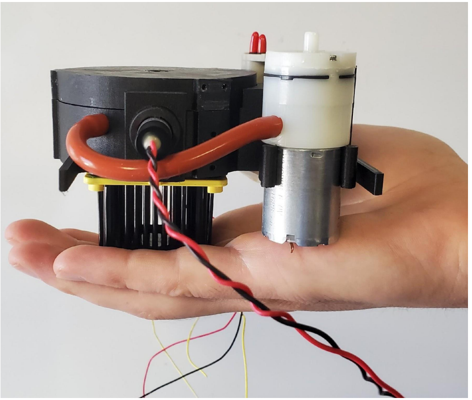


← **Modeled Effects of Deposited BrC on the Snowpack albedo**

doi:10.5194/acp-21-8531-2021

Aerosol Spatial Mapping





CloudPuck

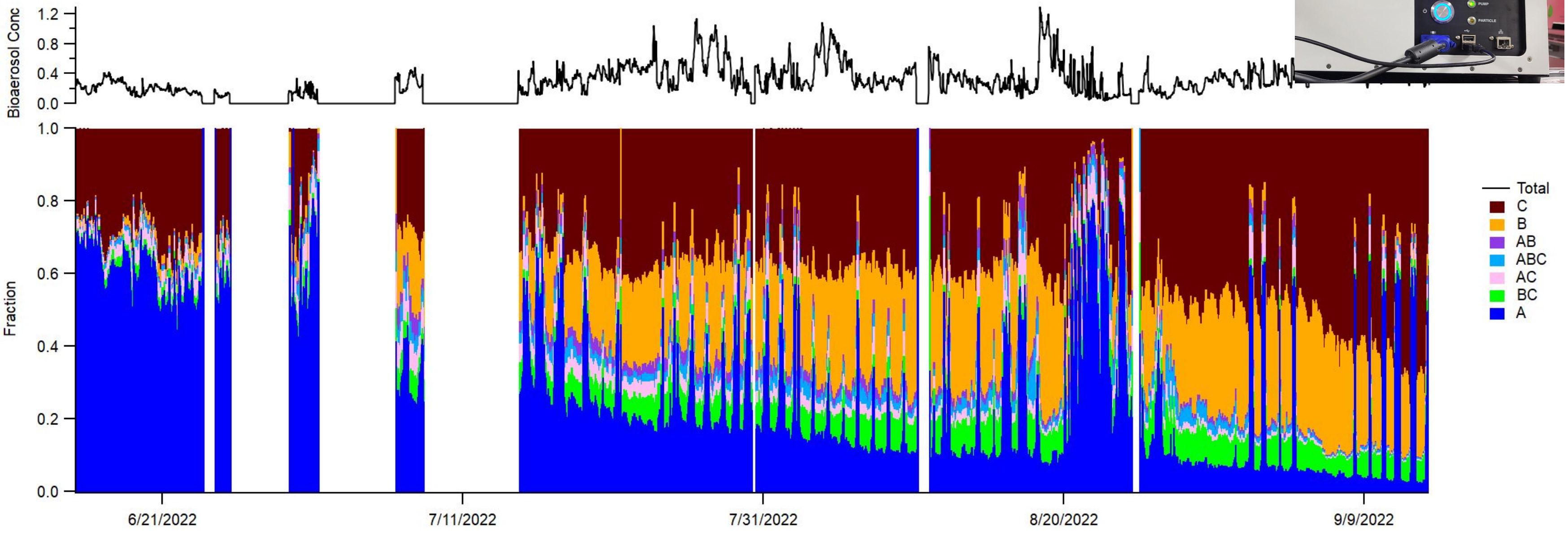
Solar-powered microphysics node

- Add **6+ instrument nodes** to SAIL domain
  - (new CloudPuck* CCN counter, INP filter sampler, POPS)
- Goal: **measure annual horizontal and vertical variability**
- Sites are TBD; happy to discuss locations with people!
  - anna@handixscientific.com

SAIL Aerosol Regimes and Processes: Results

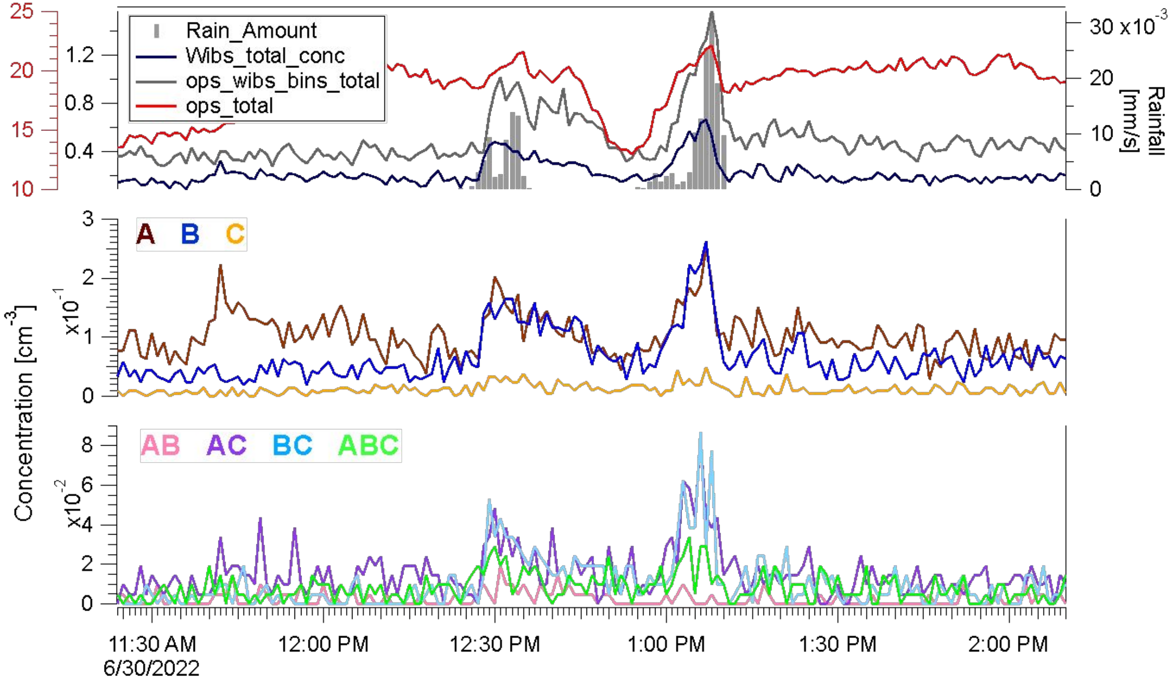
- Timeseries of different types of fluorescent particles changed during the 3 month initial deployment
- Offline filter samples of bioaerosol also were collected and are being analyzed

Wideband Integrated Bioaerosol Sensor (WIBS)



SAIL Aerosol Regimes and Processes: Results

- Initial WIBS deployment sampled 54 rain events from June 15 – Sept 13, 2022
- Example below of one event showing the fluorescent particle type time series



SAIL Aerosol Team at LANL

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Thank you for your attention!

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Backup Slides

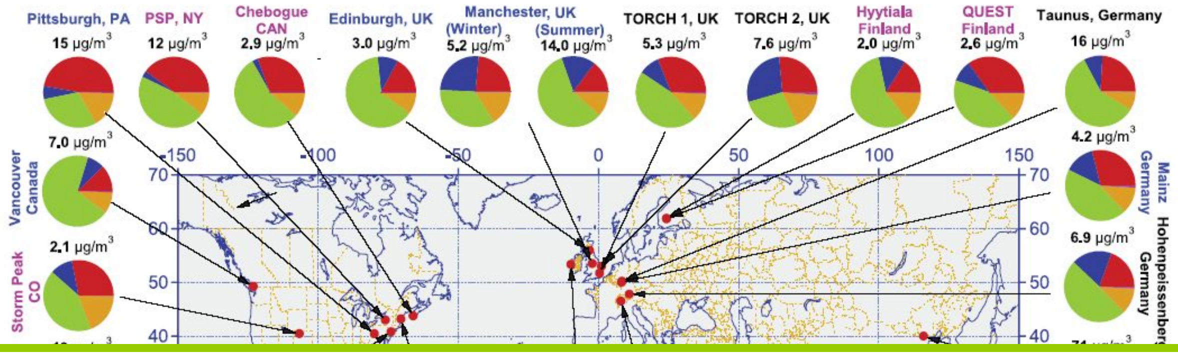
Color Code:
Organics
NH_4
NO_3
SO_4

Why Study Aerosol Chemistry?

20-90% of the particle mass from organics

Effects on climate, human health, etc…

Global sampling of particles < 1 micron diameter



- What are these OA?
  - Thousands of species
  - Extreme range of properties: MW, vapor pressure, functionality, polarity, hydrophilicity:
    - Hydrocarbons, aldehydes, ketones, peroxides, polyacids, oligomers, humic-like substances…
  - No technique or combination can analyze all OA at the compound level
    - Typical GC-MS: off-line, 6-24 hrs averages, 10% of the organic mass
  - Traditionally poorly characterized

Carbonaceous Aerosol Optical Properties + Direct Effects

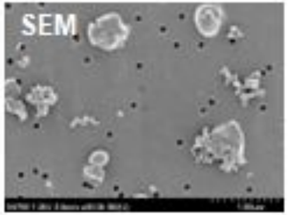
ARM

- **"Model" Soot: Fresh fractal, uncoated/denuded** *Cross et al., ACP, 2010*

Absorption Angstrom Exponents (AAE)

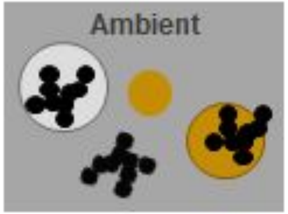
$$\frac{\beta_{\lambda}}{\beta_{\lambda'}} = \left(\frac{\lambda}{\lambda_0}\right)^{-AE}$$

- **Ambient Mixtures are heterogeneous – internal and external mixtures**



Internal mixtures (clear coatings)

External mixtures (Brown Carbon)



- **Coatings and Mixing with Brown Carbon (BrC)** *Cappa et al., Science, 2012; Liu, Aiken et al., Nature Comm., 2015*
  - Enhances Absorption → "How much?"
  - Changes the optical properties, e.g. Absorption Angstrom Exponent (AAE)
  - How is hygroscopicity (and the ability to form cloud droplets) affected?