U.S. DEPARTMENT OF ENERGY | Office of Science

DOE/SC-ARM-24-004

Cloud Processing of Aerosol during SAIL Field Campaign Report

S China
Z Cheng
G Vandergrift
NN Lata

March 2024

ARM

DISCLAIMER

This report was prepared as an account of work sponsored by the U.S. Government. Neither the United States nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof.

DOE/SC-ARM-24-004

Cloud Processing of Aerosol during SAIL Field Campaign Report

S China
Principal Investigator

Z Cheng
G Vandergrift
NN Lata
Co-Investigators

All at Pacific Northwest National Laboratory

March 2024

How to cite this document:

China, S, Z Cheng, G Vandergrift, and NN Lata. 2024. Cloud Processing of Aerosol during SAIL Field Campaign Report. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM-24-004.

Work supported by the U.S. Department of Energy,
Office of Science, Office of Biological and Environmental Research

Acronyms and Abbreviations

AED	area equivalent diameter
AOS	Aerosol Observing System
ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
CCSEM/EDX	computer-controlled scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy
CPC	condensation particle counter
DOE	U.S. Department of Energy
ELVOC	extremely low-volatility organic compound
EMSL	Environmental Molecular Sciences Laboratory
HRMS	high-resolution mass spectrometry
INP	ice nucleating particle
nano-DESI	nanospray desorption electrospray ionization
PTFE	polytetrafluoroethylene
RH	relative humidity
SAIL	Surface Atmosphere Integrated Field Laboratory
STAC	size- and time-resolved aerosol collector
STXM/NEXAFS	scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy
TBAC	total bulk aerosol collector
TEM	transmission electron microscopy

Contents

Figures

1.0 Summary

Globally, on average, the release rate of aerosols from clouds is estimated to be about 6,000 Tg/yr and an aerosol particle is estimated to experience about three cloud cycles. Cloud processing of aerosol produces aerosols that have dramatically different physicochemical properties compared to their original form. Cloud processing may transform particles to efficient cloud condensation nuclei (CCN) or ice nucleating particles (INP) that can further participate in cloud formation. We investigated cloud processing of aerosol during the U.S. Department of Energy (DOE)'s Atmospheric Radiation Measurement (ARM) Surface Atmosphere Integrated Field Laboratory (SAIL) field campaign using multi-modal offline analysis techniques available at DOE's Environmental Molecular Sciences Laboratory (EMSL). The main scientific aim of this project was to understand the processes that transform aerosols (chemically, and physically), and influence aerosol-cloud interactions. Aerosol particles undergo significant modifications due to warm (cloud droplets) and cold cloud (ice crystals) processing. Cloud processing of aerosol has important effects on the atmospheric aerosol number and mass concentration, size distribution, phase separation, and chemical composition.

Detailed size-resolved chemical composition of aerosol and their microphysical properties along with in situ measurements from ARM's mobile facilities provided opportunities to improve our understanding of the aerosol-cloud processes. EMSL's automated size- and time-resolved aerosol collector (STAC) and a total bulk aerosol collector (TBAC) were deployed at the SAIL Aerosol Observing System (AOS) site at Crested Butte Mountain, Colorado to collect impactor samples and polytetrafluoroethylene (PTFE) filter samples continuously from November 2021 to June 2023. The STAC and TBAC were deployed inside the AOS container, with an aerosol inlet approximately 5 m above ground (Figure 1).

Figure 1. STAC and TBAC deployed at the SAIL AOS site. A separate inlet was installed for particle sampling.

The STAC platform was loaded with an array of 20 customized four-stage STAC impactors. The TBAC system was loaded with five filter holders for aerosol collection on PTFE filters. Each impactor and filter collected for 24 hours. We loaded one transmission electron microscopy (TEM) carbon type B-film grid on stages A and B, two TEM carbon type B-film grids and one silicon nitride (Si_3N_4) substrate on stage

C, and a TEM carbon type B-film grid, a TEM carbon lacey grid, and a SiN substrate on stage D. Over 250 impactor samples were collected, covering more than 90% of the days during that period of the SAIL campaign. Determining size-resolved chemical composition of particles was crucial for understanding several atmospheric processes during different seasons. Our long-term sample collection during SAIL provided the opportunity to study these processes.

During our long-term measurements we observed several events such as dust transport and biomass burning. For example, we observed a series of events during September 2022 such as elevated coarse mode particles, high abundance of bioaerosols, and a biomass burning-influenced plume. It is an interesting month to focus on few case studies. We discussed a few events during December 2021.

2.0 Results

We investigated several samples for single-particle analysis and bulk molecular composition analysis. Figure 2 shows the daily variation of particle concentration from the condensation particle counter (CPC), temperature, and relative humidity. The highest mean particle concentration was observed on December 6th (~4100 #/cc) and the lowest mean concentration was observed on December 9th (~500 #/cc). Throughout the month, the lowest temperature was around -15°C (December 10th), and the highest mean temperature was around 2°C (December 4th). The RH was also fluctuating with a maximum mean value of ~ 94% (December 23rd) and a minimum mean of ~43% (December 5th). We used multi-modal chemical imaging techniques such as computer-controlled scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (CCSEM/EDX) to characterize the single-particle chemical compositions and their morphologies. Carbon features in the particles are probed with scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS) at the Advanced Light Source at Lawrence Berkeley National Laboratory to determine particle mixing state.

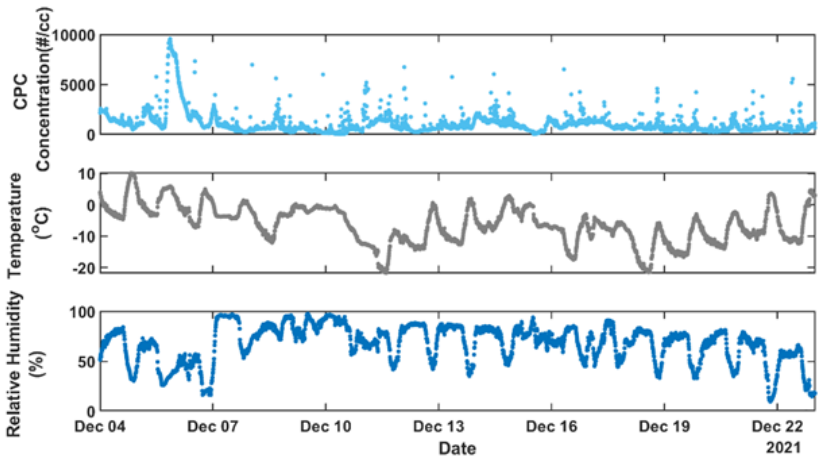

Figure 2. Ground-based measurement of condensation particle counter concentration, temperature, and relative humidity (RH) from December 4th, 2021, to December 24th, 2021.

Figure 3 shows different particle classes observed during December 2021. While most of the days were dominated by carbonaceous particles, a few days were influenced by dust and sulfate particles.

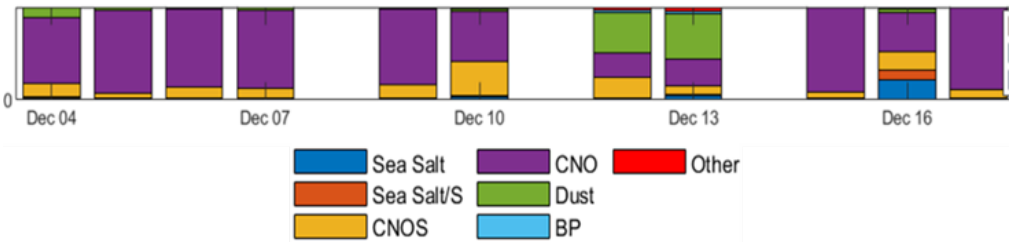

Figure 3. Number fraction of particle classes using CCSEM/EDX during December 2021.

Figure 4 shows the representative size-resolved chemical composition of STAC-collected particles with medium, high, and low concentration case observed during December 2021. Aerosol particles collected on stage D were analyzed with CCSEM-EDX at EMSL to probe morphology (size and shape) and relative element percentage of 15 elements (C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn, Fe, and Zn) of individual particles. Based on this information, we classified each particle as biological, sulfate, carbonaceous, carbonaceous+dust, sodium-rich (Na-rich), sodium-rich with sulfate (Na-rich-sulfate), dust, and others. The leftmost panel of Figure 4 shows that the major fraction of particles are carbonaceous and sulfate particles where the CPC concentration of the particle was moderate (Figure 2, top panel). Bi-modality in size distribution is observed and both modes are dominated by carbonaceous particles. Particles with AED>1µm are dominated by sulfate-rich particles. The middle panel shows the dominance of carbonaceous with the highest mean CPC concentration (Figure 2, top panel). The rightmost panel shows the dominance of sulfate-rich particles with AED > 0.5 µm and low CPC concentration (Figure 2 top panel).

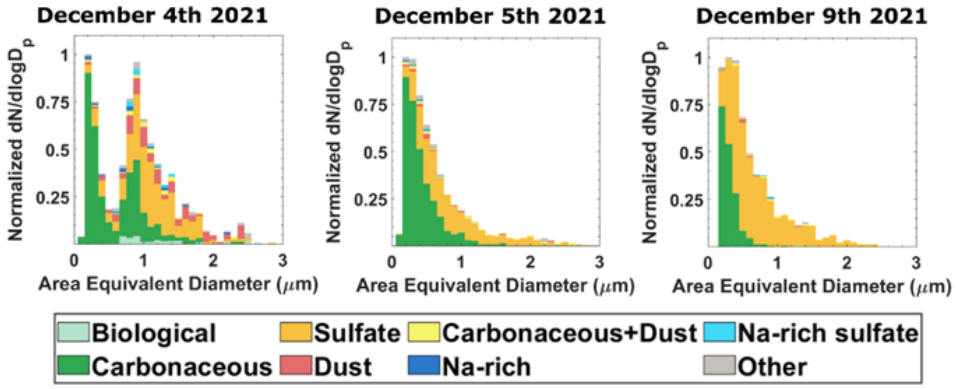

Figure 4. Representative size-resolved chemical composition of the particles from December 2021. December 4th (medium CPC concentration), December 5th (very high CPC concentration), and December 9th (low CPC concentration or background condition) are shown.

Aerosol samples were subsequently analyzed by a direct sampling high-resolution mass spectrometry (HRMS) modality to achieve molecular-level compositional sample information. Specifically, nanospray desorption electrospray ionization (nano-DESI) HRMS was used, which directly sampled the collected particles from the microscopy substrate via a ~50 µm solvent junction. Consecutive HRMS scans were obtained from even this minimal amount of material, which were then subjected to organic molecular formula analysis based upon the measured accurate masses. Figure 5 shows representative sample results, including a mass spectrum, distribution of detected molecular classes, and volatility plot. In general, hundreds to thousands of molecular-level features were observed for each sample, including a variety of

organosulfates (i.e., CHOS compounds) and organonitrates (i.e., CHNO compounds). Comparable results were obtained across the duration of the sampling period. Current efforts involve correlating the enhanced detail observed of the organic particle fraction via the nano-DESI HRMS approach with the microscopy results.

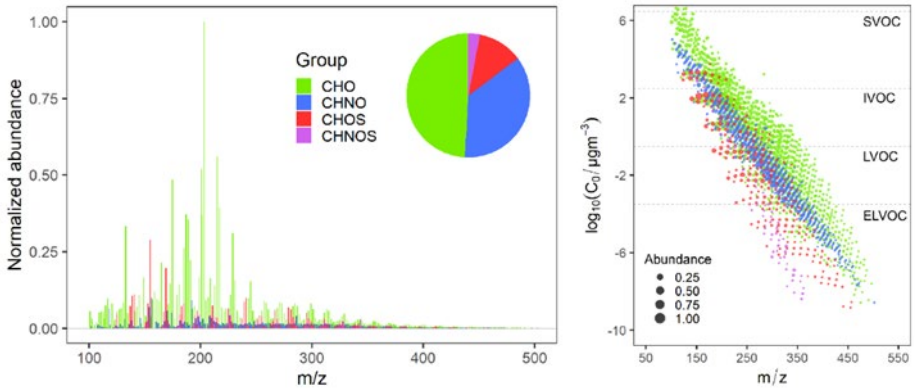

Figure 5. Representative nano-DESI HRMS results for samples collected at the AOS SAIL site. On the left, a representative mass spectrum is shown, with an inset pie chart that displays the proportion of molecular formulae that were detected by class. On the right, the detected molecular formulae were parametrized to represent volatility, allowing for the visualization of different volatility classes that were present in the ground particle samples (e.g., extremely low-volatility organic compounds; ELVOC).

3.0 Publications and References

We have presented or plan to present our preliminary results in scientific meetings and conferences. We have plans to publish several manuscripts as part of this ARM field campaign.

1. Roberts, A, N Lata, Z Cheng, and S China, S. 2022. "Investigating Source Impacts on Aerosol Concentrations and Composition." *American Geophysical Union Fall Meeting Abstracts*, A11L-01, https://agu.confex.com/agu/fm22/prelim.cgi/Paper/1096279

2. China, S, Z Cheng, G Vandergrift, N Lata, A Rahman, M Zawadowicz, A Aiken, P Demott, and D Feldman. 2023. "Insights into the single-particle composition and molecular composition of organic aerosol during SAIL." Presented at the Joint Atmospheric Radiation Measurement (ARM) User Facility/Atmospheric System Research (ASR) Principal Investigators (PI) Meeting.

3. Lata, N, J Creamean, T Hill, P DeMott, R Perkins, S Kreidenweis, Z Cheng, D Dexheimer, A Laskin, D Feldman, and S China. 2023. "Unraveling the Complexity of Aerosol Composition and Ice Nucleation Potential at Variable Altitudes: A Study from Crested Butte Mountain." Presented at the American Geophysical Union Annual Meeting.

4. Lata, N, J Creamean, T Hill, P DeMott, R Perkins, S Kreidenweis, Z Cheng, D Dexheimer, A Laskin, D Feldman, and S China. 2023. "Investigating the Vertical Variability of Aerosol Composition, Morphology and Mixing State over Crested Butte Mountain: Impacts on Ice Nucleation." Presented at the American Association for Aerosol Research Annual Conference, Portland, Oregon.

5. Lata, N, J Creamean, T Hill, P DeMott, R Perkins, S Kreidenweis, Z Cheng, D Dexheimer, A Laskin, D Feldman, and S China. 2023. “Assessing the Vertical Heterogeneity of Aerosol Composition and its Influence on Ice Nucleation over Crested Butte Mountain.” Virtual poster presented at the Joint Atmospheric Radiation Measurement (ARM) User Facility/Atmospheric System Research (ASR) Principal Investigators (PI) Meeting.

www.arm.gov

U.S. DEPARTMENT OF
ENERGY

Office of Science