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Multimodal Chemical Characterization of Brown Carbon in Atmosphere and Snowpack during SAIL Field Campaign Report

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Acronyms and Abbreviations

3D	three-dimensional
AAA	absorption Ångström exponent
AMF2	second ARM Mobile Facility
ARM	Atmospheric Radiation Measurement
BC	black carbon
BrC	brown carbon
CCN	cloud condensation nuclei counter
CCSEM/EDX	computer-controlled scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
LAP	light-absorbing particles
LC	liquid chromatography
MD	mineral dust
NOAA	National Oceanic and Atmospheric Administration
OVF	organic volume fraction
PAH	polycyclic aromatic hydrocarbons
PSD	particle size distribution
PU	Purdue University
RMBL	Rocky Mountain Biological Laboratory
SAIL	Surface Atmosphere Integrated Field Laboratory
SMPS	scanning mobility particle sizer
STI	Sonoma Technology, Inc.
STXM/NEXAFS	synchrotron-based scanning transmission X-ray microscopy coupled with near-edge X-ray absorption fine structure spectroscopy
TBS	tethered balloon system
TRAC	time-resolved aerosol collector
UV	ultraviolet
Vis	visible spectrum
WSOC	water-soluble organic carbon

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1.0 Summary

The main goal of this field project was to characterize the chemical and physical properties of light-absorbing particles (LAP) present in the atmosphere and snowpack. The specific field efforts included: 1) online measurement and sampling of light-absorbing particles using a Magee Scientific model AE33 aethalometer (AE33) and a time-resolved aerosol collector (TRAC); and 2) sampling of snowpack during observed events of LAP deposition on snow, confirmed by onsite U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility measurements and meteorological data. The ongoing research objectives of our follow-up sample and data analysis include: 1) Characterization of aerosol regimes and sources, identifying particle-type populations, mixing states, and atmospheric transformations based on correlative analysis of our detailed chemical imaging and chemical characterization measurements and real-time records from the second ARM Mobile Facility (AMF2) and other instruments available from the Surface Atmosphere Integrated Field Laboratory (SAIL) experiment; 2) Assessment of the optical and chemical properties of snowpack deposits to investigate how aerosol deposition influences snowpack lifetime.

The AE33 and TRAC were deployed at the Rocky Mountain Biological Laboratory (RMBL), located 200 m north of the main AMF2 site. Because of various restrictions on travel, field work, and user facility operations during COVID-19 pandemic outbreak, our field project started later than originally planned. Specifically, we started our field operation at SAIL only in April 2022, after travel restrictions by Purdue University to its personnel were lifted. The AE33 was deployed from 04/05/2022 until 10/09/2023, providing real-time measurements of the atmospheric mass loadings of LAP along with the collecting of time-tagged bulk samples of aerosols deposited on the filter tape. The AE33 was down from 10/03/2022 until 12/08/2022 due to lost communication and remote troubleshooting of the underlying problems. The TRAC was deployed from 06/17/2023 until 07/31/2023 to collect samples of individual particles for chemical imaging. In collaboration with RMBL staff, we collected samples of snow polluted by long-range transported atmospheric aerosol from pollution events in February 2022 and April 2023, These were two major LAP haze episodes that darkened the surface of snowpack in the area of study, as illustrated in Figure 1.

Figure 1. LAP deposits on the snowpack at the SAIL field site. Photo courtesy of Dr. C. Cox, NOAA.

A combination of molecular characterization and chemical imaging techniques are used by our group to characterize the aerosol and snowpack samples. Assessment of the analyses results will contribute essential data to improve predictive understanding of aerosol effects on the climate.[1]

LAP are primarily composed of organic brown carbon (BrC), mineral dust (MD), and black carbon (BC). The concentration of BrC and BC has increased due to increases in the population density in the Rocky Mountains and more intense wildfire events.[2,3] Additionally, changes in climate and land use has increased the concentration of mineral dust.[4] While BrC and MD are less absorbing than BC, their regional mass loadings are high, and therefore their contribution to changes in the snow surface albedo is substantial.[5,6] Snow albedo is directly decreased by the presence of LAP, and indirectly decreased by enhanced snow grain growth.[7] The net effect of light-absorbing particles on alpine climates is uncertain due to the effects of snow albedo feedback.[8,9]

Atmospheric particles have complex composition of organic compounds, inorganic salts, mineral dust, and black carbon (soot), exhibiting a wide range of molecular structures, morphologies, physical properties, and chemical reactivity.[10–14] The key aerosol metrics are particle external and internal 'mixing states'.[15,16] External mixing denotes particle ensembles when different chemical species exist in distinct particles, while internal mixing occurs when different chemical components are homogeneously distributed throughout the individual particles.[17] The mixing states of aerosol populations are complex and vary as a function of particle size, altitude, and age.[18–21] Fundamental understanding of the physical properties and chemical composition of aerosols requires advanced analytical approaches for imaging of particle components and quantitative assessment of their mixing states.[15,22] In our project, we use synchrotron-based scanning transmission X-ray microscopy coupled with near-edge X-ray absorption fine structure (STXM/NEXAFS) spectroscopy and computer-controlled scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (CCSEM/EDX) for chemical imaging of individual particles.[18,23,24] Additionally, we investigate molecular composition and optical properties of BrC chromophores in bulk aerosol and snowpack samples using multimodal high-resolution mass spectrometry techniques developed and advanced by our team.[27-29]

2.0 Results

The field deployment tasks of our project were completed on 10/09/2023. The AE33 was operated to monitor the concentrations of LAP. The instrument inlet was equipped with a cyclone with a particle cutoff of 2.5 μm and placed 5 m above ground level. Hour-resolved LAP mass loadings at seven wavelengths were reported by the instrument's built-in algorithm, which calculates mass loading from the rate of change of the attenuation of light transmitted through the aethalometer filter tape. The absorbance of the filter-deposited bulk aerosol samples measured at 880 nm is an operationally defined reference value for calculation of the BC atmospheric mass loading (ng/m^3). Hourly resolved measurements from AE 33 have been uploaded to the ARM data server as an ARM0816 data product. The data set includes: 1) absorbances (Mm^{-1}) at seven wavelengths of 370, 470, 520 590, 660, 880, and 950 nm, 2) estimated mass loadings ($ng\ m^{-3}$) at each of the wavelengths assuming mass absorption coefficients of BC, 3) absorption Ångström exponent (*AAE*) of BrC calculated as the slope of a linear fit of $\log(\lambda)$ versus $\log(Abs_\lambda)$, based on 370, 470, and 520 nm datapoints, 4) *AAE* of BC calculated as the slope of a linear fit of $\log(\lambda)$ versus $\log(Abs_\lambda)$, based on 590, 660, 880, and 950 nm datapoints, 5) CO_2 atmospheric mixing (ppm) at the site monitored with a Vaisala CARBOCAP carbon dioxide probe

GMP343. A summary data file with the time tags of particle samples collected on microscopy substrates using TRAC has been uploaded as an ARM0822 data product.

The sample analyses tasks are still ongoing at the time of this report; they will be followed with data analysis and interpretation. The chemical imaging experiments require use of Environmental Molecular Sciences Laboratory/Pacific Northwest National Laboratory and Advanced Light Source/Lawrence Berkeley National Laboratory user facilities, which are scheduled for our group later in 2024 and in 2025. Chemical composition, volatility, and optical properties of organic carbon in aerosol and snow samples are now being investigated following methodologies developed in our recent laboratory studies[28,29] of snowpack samples collected in China.[6,27] The latter was conducted withing the framework of our project as a case study to establish analytical protocols to streamline and facilitate analysis of the expected snowpack samples from SAIL.

Below we highlight our selected ongoing efforts on the chemical characterization of samples collected during SAIL.

AE33 measurements were used to select samples for detailed chemical analysis. Figure 2 exemplifies two different cases of BrC events inferred from field records. The first case shown in panel (a) was attributed to long-range transport of biomass burning aerosol from Canadian wildfires that reached the SAIL site on May 20, 2023, and effected the area until May 25, 2023. Filter samples caught the start, middle, and end of the plume. The second case shown in panel (b) is a diurnal series of biomass burning events from local campfires observed in mid-July of 2023. Each of these events started at 18:00 local time and ended by midnight. The time records and high AAE is consistent with fresh BrC emissions from local sources. Tan bars in Figure 2b represent individual TRAC samples. Within the period of local campfire episodes, *k*-means a machine learning clustering algorithm was used to process AE33 records and identify samples with representative and statistically significant differences in particle optical properties. Three types of light-absorbing particles were identified: 1) strongly and 2) moderately absorbing BrC from biomass burning events, 3) background BrC-influenced various unspecific local sources. Samples from each of these distinct clusters are selected for the planned chemical imaging studies at U.S. Department of Energy user facilities. These samples are shown as green overlays in Figure 2b.

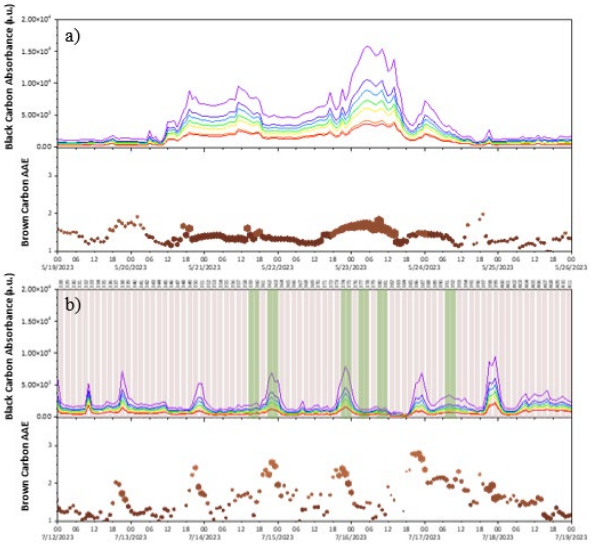

Figure 2. Two events identified during deployment of the AE33. a) Canadian wildfire plume that effected the region in late May 2023 shows characteristics of aged BrC. b) Local campfires north of AMF2 show characteristics of fresh BrC. Tan bars represent individual TRAC samples; green overlay represent samples selected for chemical imaging.

In a separate task, atmospheric particles from different altitudes were collected on tethered balloon system (TBS) flights on May 16, 2022, which coincided with a BC/BrC plume recorded by the AE33. Particle samples are analyzed with STXM/NEXAFS and CCSEM/EDX to characterize particle components and mixing states. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) backwards trajectories of the airmass show it moved over several populated areas in western Colorado. Sampled particles likely originated when the air mass stagnated over Delta, Colorado. Figure 3 shows STXM particle size distributions (PSD) at separate sampling altitudes. Altitudes below 250 m show higher particle diversity with increased organic volume fraction that suggest freshly emitted biomass burning samples while altitudes above 250 m have aerosols typical of internally mixed aged aerosol.

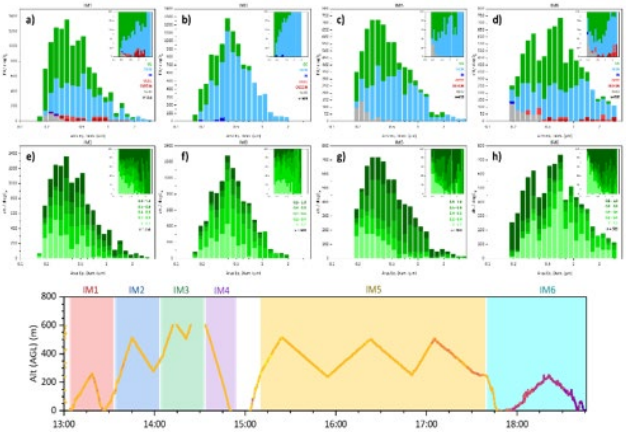

Figure 3. STXM/NEXAFS particle information. (a-d) Chemical mixing PSD of experimentally defined chemical components. (e-h) Organic volume fraction (OVF) measurements of the same PSD. (i) IM1 ascended from 0-250 m, IM3 ascended from 300-750 m, IM5 loitered between 300 and 500 m, and IM6 ascended from 0-250 m during the top of the BC event.

Multimodal chemical analysis of bulk snow samples provides detailed chemical information on the composition of MD, particle size distributions of BC, and chemical speciation of BrC components. The exact timing of the haze events was identified with scanning mobility particle sizer (SMPS), cloud condensation nuclei counter (CCN), and nephelometer records from real-time measurements at AMF2. HYPSLIT backwards trajectories reveal the source of the particle haze events to be from central Arizona and likely influenced by the Phoenix metro area. Analysis of the BrC components shows that most water-soluble BrC chromophores are monoaromatic compounds with minor influence from substituted polyaromatic hydrocarbons (Figure 4).

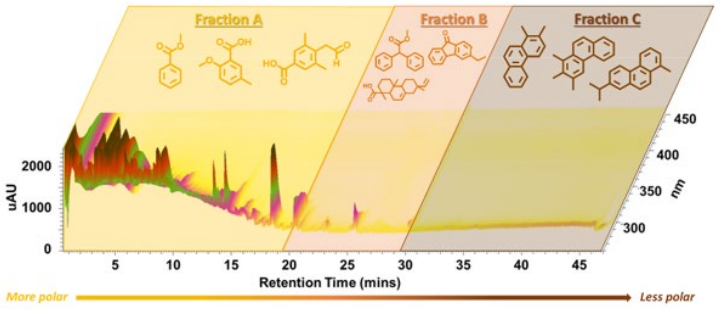

Figure 4. 3D LC-UV/Vis chromatogram of water-soluble organic compounds (WSOC) present in February snow samples. Fraction A is characterized as monoaromatic compounds, Fraction B is lignin depolymerization products associated with biomass burning, and Fraction C is substituted polycyclic aromatic hydrocarbons (PAH) associated with high-temperature combustion.

High amounts of monoaromatic compounds without significant contribution from lignin depolymerization products are typical of urban emissions. CCSEM/EDX characterization of MD is focused on the Fe-containing particles as they tend to be the most absorbing in the visible range. [25,26] Identified MD are Fe-aluminosilicates that ranged from 1-3 μm that were consistent with Arizona dust standards.[25] Insoluble BC particles are imaged with a spatial resolution of 35 nm and they are identified based on their distinct NEXAFS absorption features at 285.4 eV. Sizes of the detected BC particles range from 30 to 100 nm, and they exhibit NEXAFS spectra consistent with biomass burning particles.

Additional work involves chemical analysis of the aerosol samples collected on the AE33 tape using temperature-programed desorption direct analysis in real-time, high-resolution mass spectrometry,[29] quantifying the gas-particle partitioning and viscosity of deposited organic aerosol and quantifying its BrC components. Samples collected during the haze event apportioned to the Canadian wildfire in 2023 and the local campfires events (Figure 1) will be the primary focus of this analysis. Additionally, chemical imaging of particle samples collected on the TRAC corresponding to the same campfire events will be performed to evaluate particle morphology and mixing states.

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Li, W, N Riemer, L Xu, Y Wang, K Adachi, Z Shi, D Zhang, Z Zheng, and A Laskin. 2024. "Microphysical properties of atmospheric soot and organic particles: Measurements, modeling, and impacts." *NPJ Climate and Atmospheric Science* 7: 65, https://doi.org/10. 1038/s41612-024-00610-8

Rivera-Adorno, FA, JM Tomlin, M Fraund, E Morgan, M Laskin, R Moffet, and A Laskin. 2023. "Estimating Viscosity of Individual Substrate-Deposited Particles from Measurement of their Height-to-Width Ratios." *Aerosol Science and Technology*, published online, https://doi.org/10.1080/02786826.2023.2270503

Zhou, Y, CP West, D Calderon–Arrieta, M Misovich, APS Hettiyadura, T Shi, J Cui, H Wen, W Pu, X Wang, and A Laskin. 2024. "Photolytic Degradation of Water–Soluble Organic Carbon from Ambient Snow: Changes in Molecular Characteristics, Composition of Brown Carbon Chromophores, and Radiative Effects." Submitted to *Journal of Geophysical Research – Atmospheres*

3.2 Conference Presentations

Sharpe, S, F Rivera-Adorno, J Tomlin, K Siemens, NN Lata, Z Cheng, E Hulm, R Moffet, S China, and A Laskin. 2024. "Multimodal Chemical Characterization of Brown Carbon in Atmosphere and Snowpack from the Colorado Rockies." American Meteorology Society 104th Annual Meeting, Baltimore, Maryland, oral 13A.1.

Sharpe, S, F Rivera-Adorno, J Tomlin, K Siemens, NN Lata, Z Cheng, E Hulm, R Moffet, S China, and A Laskin. 2023. "Multimodal Chemical Characterization of Brown Carbon in Atmosphere and Snowpack from the Colorado Rockies." The American Association for Aerosol Research 41st Annual Conference, Portland, Oregon, poster RA.9.

Sharpe, S, F Rivera-Adorno, J Tomlin, K Siemens, NN Lata, Z Cheng, E Hulm, R Moffet, S China, and A Laskin. 2023. "Multimodal Chemical Characterization of Brown Carbon in Atmosphere and Snowpack from the Colorado Rockies." American Chemical Society Spring Meeting, Indianapolis, Indiana, oral Room 132.

Sharpe, S, F Rivera-Adorno, J Tomlin, NN Lata, Z Cheng, E Hulm, R Moffet, S China, and A Laskin. 2022. "Chemical Characterization of Brown Carbon in Atmosphere and Snowpack from the Colorado Rockies." The American Geophysical Union Fall Meeting, Chicago, Illinois, poster H25M-1260.

Sharpe, S, F Rivera-Adorno, J Tomlin, NN Lata, Z Cheng, E Hulm, R Moffet, S China, and A Laskin. 2022. "Multimodal Chemical Characterization of Brown Carbon in Atmosphere and Snowpack from the Colorado Rockies." The American Association for Aerosol Research 40th Annual Conference, Raleigh, North Carolina, poster RA.10.

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