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Size and Time-Resolved Automated Aerosol Sampling Field Campaign Report

S China
D Dexheimer
NN Lata
ASM Shawon
F Mei
Z Cheng
G Vandergrift

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S China, Pacific Northwest National Laboratory (PNNL)
Principal Investigator

F Mei, PNNL
D Dexheimer, Sandia National Laboratories
Z Cheng, PNNL
NN Lata, PNNL
G Vandergrift, PNNL
ASM Shawon, PNNL
Co-Investigators

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Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
CCSEM/EDX	computer-controlled scanning electron microscope coupled with energy-dispersive X-ray spectroscopy
CPC	condensation particle counter
DOE	U.S. Department of Energy
EMSL	Environmental Molecular Sciences Laboratory
FICUS	Facilities Integrating Collaborations for User Science
POPS	printed optical particle spectrometer
PNNL	Pacific Northwest National Laboratory
SGP	Southern Great Plains
STAC	size and time-resolved aerosol collector
TB	tethered balloons
TBI	TBS impactor
TBS	tethered balloon system
TEM	transmission electron microscopy

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1.0 Summary

Determining the size-resolved chemical composition of particles is crucial for understanding several atmospheric processes, including warm and cold cloud formation, but challenging to accomplish using currently available sampling devices. The main objective of this study was to deploy a size and time-resolved automated aerosol sampler via a tethered balloon system (TBS) to understand the vertical profile of atmospheric aerosol composition. The size and time-resolved aerosol collector (STAC) was used to collect aerosol for characterization of physical and chemical properties using multi-modal microscopy, spectroscopy, and mass spectrometry platforms. The automated sampler enabled the collection of samples with broad size range and high time resolution to fill the measurement gap in the current U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility observatory site. STAC was deployed via TBS. This enhanced capability enables the collection of atmospheric aerosol samples at multiple altitudes and improves our understanding of the vertical profile of aerosol composition.

Particle size distribution and chemical composition influence the optical properties of aerosol and their ability to form warm and cold cloud formations.[1, 2] The vertical distribution of different atmospheric aerosol species and their atmospheric processing can impact the atmospheric thermal structure, cloud dynamics, and regional-to-global circulation systems.[3, 4] The radiative forcing of the ambient aerosol is strongly influenced by the vertical distribution of aerosols throughout the entire atmospheric column and their chemical composition, which are not well constrained in climate models.[5, 6] Estimating the indirect effect of aerosol also strongly depends upon the aerosol properties as a function of height and has a significant uncertainty due to the spatial variation of the aerosol, especially at the cloud base.[7] Aerosol chemical composition as a function of height is critical to understand the interaction between aerosol and the atmospheric boundary layer.

Aerosol chemical composition is essential information in global aerosol models. Models found significant variances in vertical aerosol dispersion, which leads to large uncertainties in estimating aerosol lifetimes in the atmosphere.[8-10] Model comparison performs well at ground level due to numerous measurements. However, the vertical distribution of aerosol properties data is limited compared to ground measurements.[8, 11] Measurements of the vertical distribution of aerosol properties such as size distribution, shape, mixing state, and chemical composition at various altitudes are required to improve understanding of the climate effects of aerosols.[12] Unmanned systems have gained significant interest in aerosol properties measurement, which could minimize environmental research errors, risks, and costs. Tethered balloons (TB) have been deployed in several geographical locations to study aerosol properties [4, 11, 13-15], gaseous air pollution[16, 17], and microphysical parameters of the atmosphere.[18, 19]

Detailed measurements of single-particle aerosol composition as a function of altitude are limited but needed.[8, 11] To improve the understanding of the impacts of the vertical distribution of aerosol chemical composition on radiative forcing of aerosol, we performed a detailed physicochemical characterization of aerosol particles sampled during ARM deployment of the tethered balloon system at two ARM sites. We used multi-modal microscopy/spectroscopy techniques and advanced mass spectrometry platforms for the analysis of particles. We found variations in size-resolved aerosol composition at different altitudes. We also observed changes in aerosol composition during different

seasons. Some of our observations at ARM's Oliktok Point, Alaska site suggest cloud processing of aerosol while sampling in clouds, below clouds, and above clouds.

2.0 Results

The TBS was deployed at two ARM sites, the Southern Great Plains atmospheric observatory (SGP; ARM megasite), Oklahoma, and the ARM mobile facility at Oliktok Point, Alaska. Atmospheric aerosol samples were collected from different altitudes using TBS and the recently developed STAC at the Pacific Northwest National Laboratory (PNNL)'s Environmental Molecular Sciences Laboratory (EMSL). During the TBS deployment, several events were captured, such as biomass burning events and dust events. Figure 1 shows the deployment of STAC at the SGP and Oliktok Point sites.

Figure 1. Deployment of STAC via TBS at SGP (left) and Oliktok Point (right). Photos by Darielle Dexheimer. The white box on the right panel is the STAC system.

STAC is an automated sampler that collects aerosol particles in a size range of 0.1-5.0 µm and with a few seconds' time resolution. STAC consists of several cascade impactors (STAC impactors) with electronic valves, a circuit board, and a manifold and touch screen display. The user can set up the sampler with measurement delay (start time), number of samples, and sampling time at each point of sampling. The software automatically stores the data. The current version of STAC can sample up to 20 different altitudes per flight. The STAC impactor has four stages where size-resolved particles will be collected on multiple substrates for microscopy, spectroscopy, and substrate-bases mass spectrometry analysis. The STAC impactor is designed to collect particles in size range of 0.1-0.5 µm; 0.5-1.0 µm; 1-2.5 µm; and 2.5-5.0 µm. The STAC impactor has three substrate holders in each stage and can accommodate transmission electron microscopy (TEM) grids, silicon nitride substrates, or quartz substrates. Typically, two TEM grids (B-film and lacey film) and silicon nitride substrates or quartz substrates are placed on each stage.

During most of the flights, we deployed additional TBS impactor (TBI) packages, two printed optical particle spectrometers (POPS), and one condensation particle counter (CPC, model 3007, TSI). Two of three TBI are usually attached with the tether 1 foot away from the POPS. The third TBI is generally operated 10 feet above the ground. One POPS is operated just below the balloon to reach the maximum possible altitude, while the second POPS is generally operated lower on the tether (e.g., near cloud base).

We employed multi-modal micro-spectroscopy techniques to analyze atmospheric particles collected at different altitudes. We used a computer-controlled scanning electron microscope coupled with energy-dispersive X-ray spectroscopy (CCSEM/EDX). CCSEM/EDX enables automated particle detection and X-ray spectra acquisition of thousands of individual particles. Particles were classified into different categories based on their elemental compositions (atomic %) such as 1) Na-rich, 2) Na-rich sulfate, 3) Sulfate, 4) Carbonaceous, 5) Dust, and 6) Other.

In this report, we show some representative examples of size-resolved chemical composition data. Several samples will be analyzed in the following year. Figure 1 shows a typical flight pattern for STAC sampling via TBS at the SGP site. The bottom panel shows an example of the size-resolved chemical composition of particles and overall particle classes at different altitudes and different size ranges. Samples at lower altitudes are dominated by dust particles, as expected.

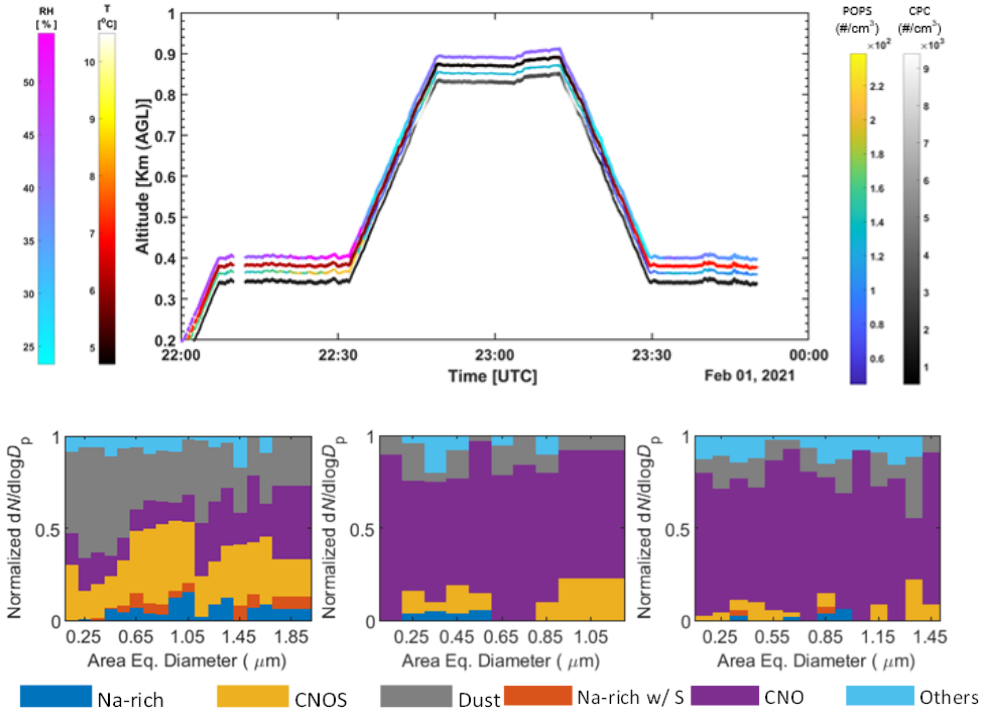

Figure 2. TBS profile during one of the flights in February 2021. STAC samples were collected at the ground level, 400m, and 900m. Color bars on the left represent relative humidity and temperature. Color bars on the right represent concentrations from POPS and CPC. The lower panel shows the size-resolved composition obtained from STAC samples.

Overall, our results show variations in the chemical composition of aerosol at different altitudes. Carbonaceous aerosols dominate at higher altitudes. The presence of dust particles is higher at ground level and typically decreases at higher altitude. However, during some dust events, we observed a high fraction of dust particles at higher altitudes. We also investigated the molecular composition of aerosol using high-resolution mass spectrometry platforms. Figure 3 shows representative high-resolution mass spectrometry data from samples collected during February 2021. Preliminary results suggest the abundance of nitrogen- and sulfur-containing species.

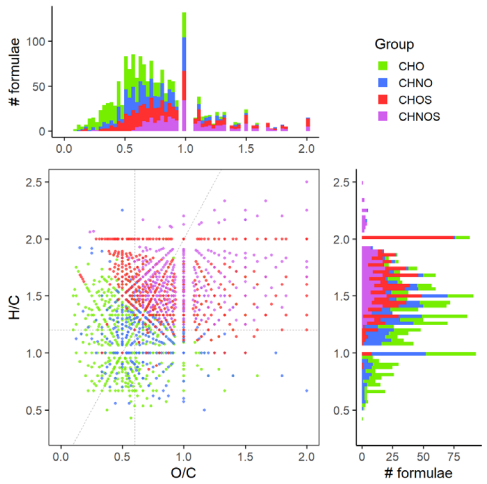

Figure 3. Representative example of high-resolution mass spectrometry data of the samples collected at the SGP site during February 2021.

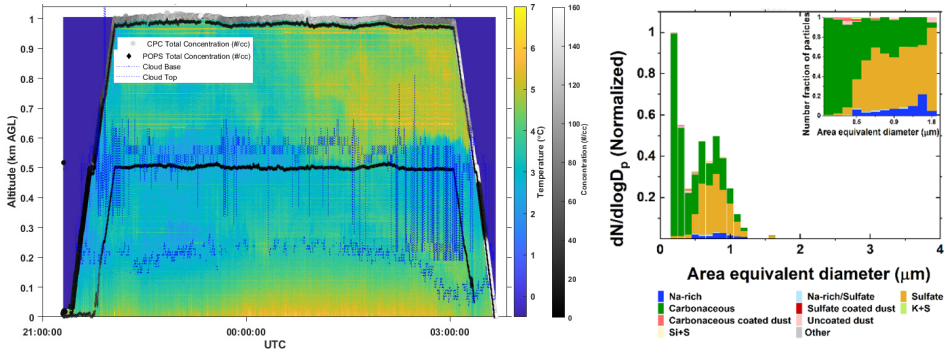

Figure 4. Left panel shows the TBS distributed temperature sensing, cloud thickness, CPC, and POPS total concentration on that day at different altitudes at Oliktok Point. The right panel shows size-resolved particle classes obtained from CCSEM/EDX at 500m.

Figure 4 shows a case study of the TBS flight that was conducted at Oliktok Point. Overall, our analysis shows a clear difference in aerosol chemical compositions at various altitudes on the same day. Broadening of the size distribution of the particles at high altitude was observed compared to low-altitude particles on the same sampling day. A relatively higher percent of sulfate and sulfate-coated dust aerosols was observed at higher altitudes, suggesting the possibility of cloud processing of aerosols.

3.0 Publications and References

Our team is currently preparing two manuscripts as an outcome of this field campaign. We collected more than 300 samples during different seasons and at different altitudes. Further analysis of samples will be performed later as part of the ARM/EMSL partnership and Facilities Integrating Collaborations for User Science (FICUS) projects.

3.1 Presentations

1. Cheng, Z, S Mathai, D Dexheimer, F Mei, G Vandergrift, A Ijaz, ASM Shawon, and S China. 2021. "Seasonal Variation in Phase State and composition of Ambient Particles Collected at the Southern Great Plains Site at Different Altitude." Poster presentation at 16th International Global Atmospheric Chemistry Scientific Conference, September 13-17, virtual.
2. Lata, NN, D Dexheimer, F Mei, Z Cheng, B Rhenton, and S China. 2021. "Size and time resolved Aerosol Sampling to Access the Physicochemical Properties of Vertically Resolved Aerosols." Poster presentation at ARM/ASR 2021 virtual joint meeting, June 21-24.
3. Cheng, Z, S Mathai, D Dexheimer, F Mei, G Vandergrift, A Ijaz, ASM Shawon, and S China. 2021. "Seasonal variation in phase state and composition of ambient particles collected at the Southern Great Plains site at different altitudes." Poster presentation at ARM/ASR 2021 virtual joint meeting, June 21-24.
4. Lata, NN, D Dexheimer, F Mei, D Tseng, and S China. 2020. "Physicochemical characterization of vertically resolved atmospheric particles via unmanned aerial system." Presented at the 31st Annual Symposium of the Pacific Northwest American Vacuum Society, September 24-25, virtual.

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