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Arctic Aerosol Sources and Mixing States Field Campaign Report

KA Pratt

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Acronyms and Abbreviations

AMF	ARM Mobile Facility
APS	aerodynamic particle sizer
APUN	Aerosols in the Polar Utqiaġvik Night
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
ATOFMS	aerosol time-of-flight mass spectrometer
CCN	cloud condensation nuclei
CCSEM-EDX	computer-controlled scanning electron microscopy with energy-dispersive X-ray
DOE	U.S. Department of Energy
DRUM	Davis Rotating-drum Universal-size-cut Monitoring
INP	ice nucleating particle
MOSAiC	Multidisciplinary drifting Observatory for the Study of Arctic Climate
MOUDI	micro-orifice uniform deposition impactor
NASA	National Aeronautics and Space Administration
NSA	North Slope of Alaska
SEM-EDX	scanning electron microscopy with energy-dispersive X-ray
SMPS	scanning mobility particle sizer
SSA	sea spray aerosol
TEM	transmission electron microscopy

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1.0 Summary

The Arctic is warming at a faster rate than anywhere else on Earth,[1] with rapidly shrinking sea ice extent[2] transforming the region. Depending on chemical and physical properties,[3] atmospheric aerosols directly scatter and/or absorb radiation, serve as cloud droplet and/or ice crystal nuclei, and/or reduce the reflectiveness of the snow surface, thereby altering the atmospheric energy budget.[4] There is a wide spread in the magnitude of simulated arctic aerosol radiative forcing, and significant differences in aerosol concentration levels and seasonal cycles often exist between models and observations.[5] Increasing local natural and anthropogenic emissions are significant, with uncertain climate impacts due to complex feedbacks.[6] Model evaluations, however, are limited by the dearth of arctic aerosol observations available and an inadequate understanding of arctic aerosol processes.[5] The majority of previous arctic aerosol observations have been made through intensive spring/summer field campaigns,[7-10] with few intensive measurement studies focused on the fall-winter transition,[11-13] a period when freeze-up is occurring later and thinning sea ice[14, 15] is resulting in wintertime ice fracturing.[16] Aerosol monitoring at arctic coastal stations has provided knowledge of long-term seasonal trends in aerosol concentrations.[17, 18] The completed ARM field campaign addresses observational and knowledge gaps through detailed aerosol size and chemical composition measurements during the fall-winter transition in the coastal Arctic and through the entire annual cycle in the central Arctic. The observations are improving our understanding of the sources and processes controlling the aerosol population in the rapidly changing Arctic.

Global models typically consider ~5-8 aerosol 'types' (sulfate, black carbon, organic aerosol, secondary organic aerosol, nitrate, biomass burning, dust, and sea salt),[5] yet these categories often do not represent observed single-particle mixing states.[19] Aerosol mixing state, the distribution of chemical species within and between individual aerosol particles, defines aerosol optical (absorption/scattering) properties,[20] hygroscopicity (cloud condensation nuclei (CCN) ability),[21, 22] and ice nucleating particle (INP) efficiency.[23] Estimated errors in simulations comparing assumptions of fully internally mixed aerosol (bulk aerosol assumption) versus size- and type-resolved aerosol (single-particle) show increased biases for freshly emitted (unaged aerosol) compared to aged aerosol, particularly for aerosol absorption simulations.[24] Yet, despite significant impacts on aerosol radiative forcing, few measurements of arctic aerosol mixing state exist, particularly outside of Svalbard.[12]

The overall goal of this ARM field campaign was to determine aerosol chemical composition, sources, mixing states, and aging processes across the entire annual cycle in the high Arctic, and in the Alaskan Arctic during fall-winter, to address the most significant gaps in aerosol observational data in the Arctic. Therefore, two deployments were conducted:

1) November-December 2018 near Utqiaġvik, Alaska (also known as North Slope of Alaska, NSA) during the Aerosols in the Polar Utqiaġvik Night (APUN) field campaign

2) October 2019-September 2020 on the icebreaker *Polarstern* during the year-long Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) campaign.

The Pratt Laboratory deployed guest instruments for aerosol measurements and sampling during both ARM field campaign deployments, and these intensive measurements were completed by routine aerosol and meteorological measurements by ARM. The sampling and measurements focused on online aerosol

size distribution measurements and individual atmospheric particle analyses. These measurements of single-particle size, chemical composition, and morphology are facilitating identification of arctic aerosol mixing states, sources, and aging mechanisms. This ARM field campaign addresses the key Atmospheric System Research (ASR) research areas of 'Aerosol Processes' and 'High-Latitude Processes'. The overarching impacts of the ARM field campaign are the generation of arctic aerosol observational data and improved understanding of arctic aerosol processes to inform and evaluate future simulations of arctic atmospheric composition and climate.

The APUN field campaign was conducted from November 8 to December 18, 2018 on the Barrow Environmental Observatory near Utqiaġvik, Alaska and the ARM NSA site. This focused on the rapidly changing and understudied transitional period of sea ice freeze-up of November-December 2018 using real-time aerosol time-of-flight mass spectrometry (ATOFMS), online aerosol sizing, and impactor sampling of atmospheric particles onto transmission electron microscopy (TEM) grids, silicon, and quartz substrates for offline individual particle analyses. Field measurements and sampling were conducted by former Pratt Laboratory postdoctoral researcher Dr. Jun Liu (now at the Bay Area Air Quality Management District) and Pratt Laboratory PhD graduate Dr. Jamy Lee (now at MilliPore Sigma). Laboratory ATOFMS calibration experiments and data analyses are being conducted by current PhD student Ms. Judy Wu, with assistance from additional PhD students. Computer-controlled scanning electron microscopy with energy-dispersive X-ray (CCSEM-EDX) and (originally unplanned) Raman microspectroscopy analyses are being conducted by postdoctoral researcher Dr. Jessica Mirrielees and PhD student Ms. Emily Costa.

The MOSAiC field campaign was conducted from October 2019 to September 2020 aboard the icebreaker *Polarstern* during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) field campaign. This was part of the second ARM Mobile Facility (AMF2) deployment and focused on atmospheric particle sampling onto aluminum foil and copper/nickel foil for offline CCSEM-EDX analysis. This is providing an unparalleled opportunity to study seasonal changes in aerosol processes in the high Arctic. Deployment of the aerosol impactor was completed in collaboration with Dr. Jessie Creamean (Colorado State University) and ARM technicians. Unfortunately, delays in the ARM technician crew change on Leg 2 led to the impactor not being run from mid-January to early March 2020, resulting in no samples collected during this period. CCSEM-EDX analyses and Raman microscopy analyses are being conducted by postdoctoral researcher Dr. Jessica Mirrielees and several undergraduate researchers. Prior to analysis, Dr. Mirrielees developed a protocol to isolate and analyze the samples collected on the Davis Rotating-drum Universal-size-cut Monitoring (DRUM) foil substrates.

The list of online instruments, corresponding parameters measured, sampling for offline analysis, and sampling dates and locations are provided in Table 1. Sample and data gaps primarily correspond to periods of instrument errors or downtime and/or weather conditions that prevented sampling or site access.

Table 1. Measurements and sampling during the University of Michigan APUN (NSA) and MOSAiC (icebreaker *Polarstern*) field deployments.

Instrument/sample	Parameter	Sampling dates and location
TSI 10-stage micro-orifice uniform deposition impactor (MOUDI) – daily, size-resolved collection of atmospheric particle samples	Offline individual particle morphology and elemental composition using CCSEM-EDX and offline individual particle functional group composition using Raman microspectroscopy for select samples	Nov. 8–Dec. 17, 2018 (NSA)
3-stage rotating DRUM impactor – daily, size-resolved collection of atmospheric particle samples	Offline individual particle morphology and elemental composition using CCSEM-EDX and offline individual particle functional group composition using Raman microspectroscopy for select samples	Oct. 24, 2019–Jan. 14, 2020; Mar. 7-29, Apr. 2–Jul. 12, Jul. 14–Sep. 19, 2020 (MOSAiC)
TSI scanning mobility particle sizer spectrometer (SMPS)	Size-resolved particle number concentrations from 12–638 nm	Nov. 8–14, Nov. 23–Dec. 18, 2018 (NSA)
TSI aerodynamic particle sizer (APS)	Size-resolved particle number concentrations from 0.54–19.8 µm	Nov. 7–Dec. 17, 2018 (NSA)
Aerosol time-of-flight mass spectrometer (ATOFMS)	Real-time size and chemical composition of individual aerosol particles	Nov. 8–Dec. 17, 2018 (NSA)
Snow sampling	Offline inorganic ion concentrations using ion chromatography	Nov. 4–Dec. 17, 2018 (NSA)
Routine ARM measurements	Meteorological and aerosol parameters	NSA and AMF2

2.0 Results

This ARM field campaign was associated with a U.S. Department of Energy (DOE) Early Career grant (DE-SC0019172) and DOE ASR grant (DE-SC0022046), as well as Pacific Northwest National Laboratory user facility grants (51363 and 60327). Sample and data analysis and interpretation are ongoing, with initial results presented here.

Figures 1 and 2 show the time series of total number concentrations and size-resolved number concentrations measured by the scanning mobility particle sizer spectrometer (SMPS) and aerodynamic particle sizer (APS) deployed during the APUN field campaign. SMPS number concentration spikes correspond to short local vehicle emission and town (Utqiaġvik) influence.

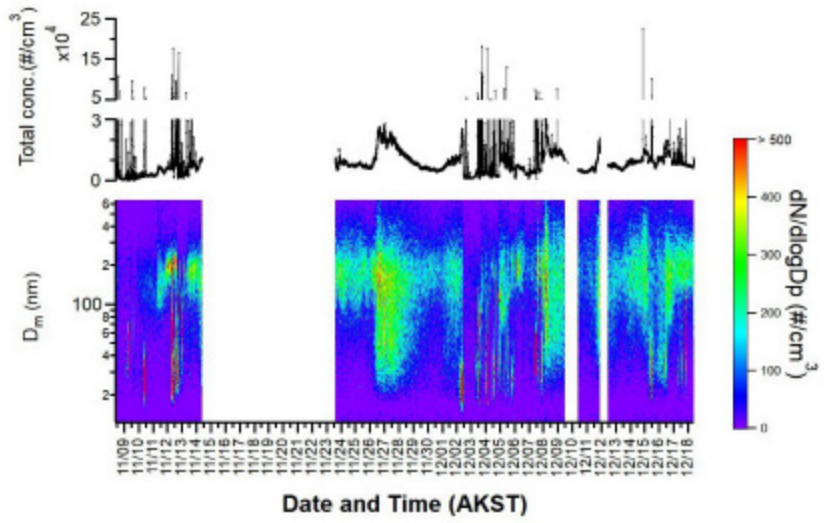

Figure 1. Total number concentrations (top) and aerosol size distributions (bottom) from 12–638 nm electrical mobility diameter (Dm) measured by the University of Michigan SMPS during the APUN field campaign at Utqiaġvik, Alaska.

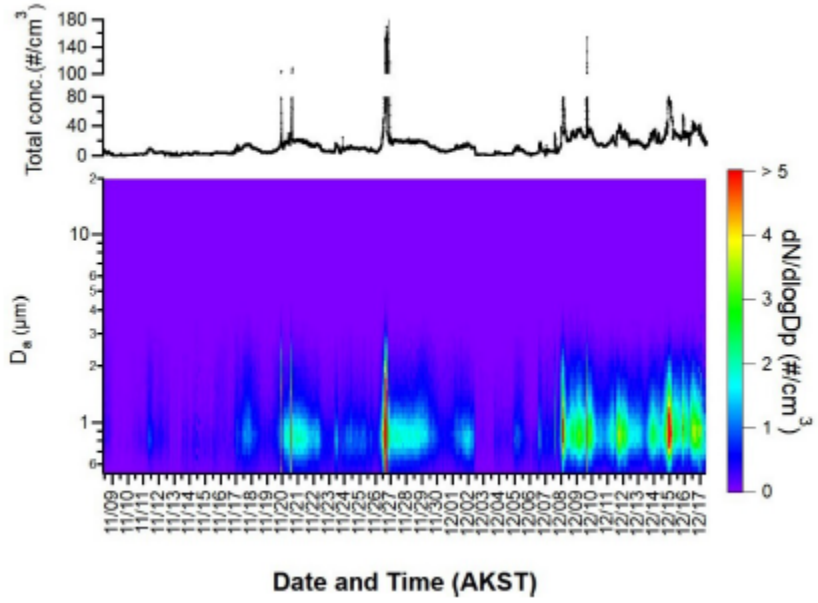

Figure 2. Total number concentrations (top) and aerosol size distributions (bottom) from 0.54–19.8 µm aerodynamic diameter (Da) measured by the University of Michigan APS during the APUN field campaign at Utqiaġvik, Alaska.

During the APUN campaign, 761,355 individual particles were chemically analyzed by the ATOFMS. Figure 3 shows the average mass spectra corresponding to the three individual particle types measured by the ATOFMS during the APUN field campaign. These correspond to dust, sea spray aerosol, and aged biomass burning particles. Laboratory calibration experiments are being used to produce number and mass concentrations of these submicron particle types. Thus far, 58,495 individual particles collected on TEM grids and silicon substrates have been chemically analyzed by CCSEM-EDX from seven select days of sampling. TEM grids provide superior imaging, while silicon substrates allow quantitation of carbon,

for example within organic coatings around sea salt aerosol. Example SEM images and EDX spectra of fresh sea spray aerosol (SSA), aged (chloride-depleted, sulfur-enriched) SSA, and organic aerosol particles are shown in Figure 4. Raman microspectroscopy analyses (not originally planned) are being conducted to identify the organic functional groups present in the organic coatings of the sea spray aerosol; these are being matched to marine organic compound Raman spectra (e.g., saccharides, fatty acids). We are also collaborating with the National Aeronautics and Space Administration (NASA) to connect the sea spray aerosol measurements to observations of leads through satellite sea ice imaging.

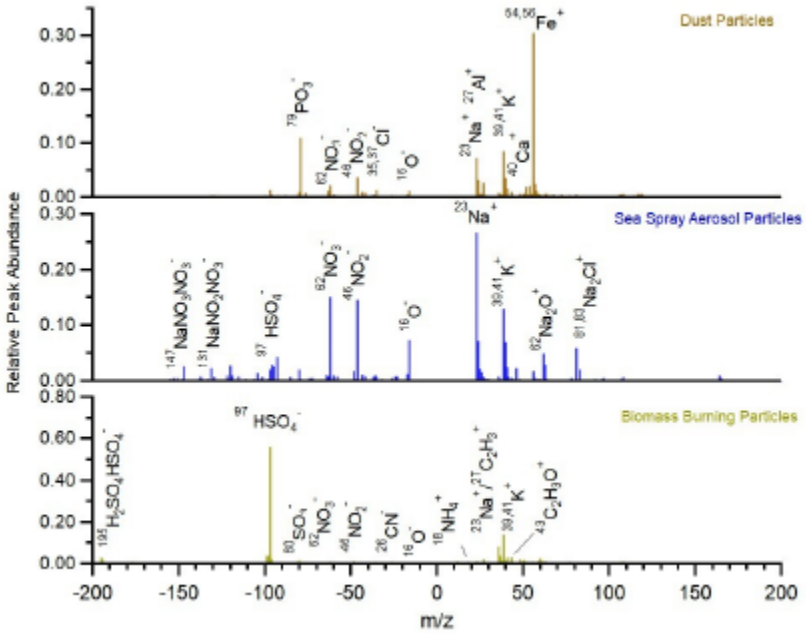

Figure 3. Average individual particle ATOFMS dual-polarity mass spectra during the 2018 APUN field campaign corresponding to three particle types: dust, sea spray aerosol, and aged biomass burning particles.

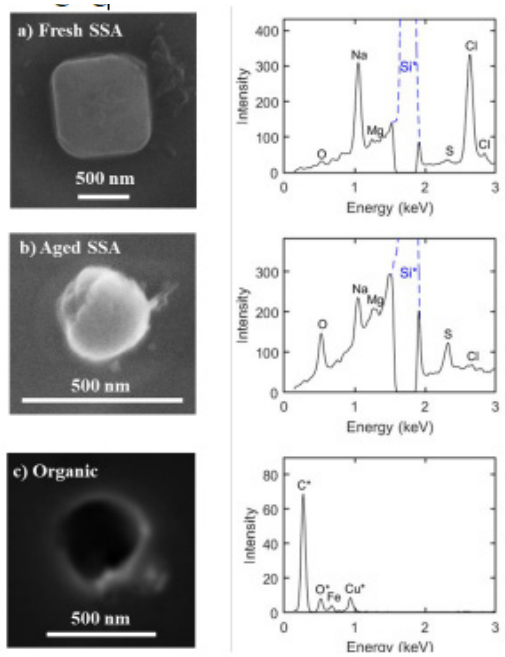

Figure 4. Representative individual particle SEM images (left) and EDX spectra (right) for (a) fresh sea spray aerosol (SSA), (b) aged SSA, and (c) organic aerosol collected during the 2018 APUN field campaign. The asterisk(*) indicates substrate EDX signal contribution.

For the MOSAiC expedition, CCSEM-EDX analyses have been conducted on samples collected on 11 days in November 2019, December 2019, March 2020, April 2020, and June 2020, with manual SEMEDX conducted on samples collected on an additional four days in November 2019, December 2019, May 2020, and June 2020. Sea spray aerosol particles, produced locally from bubble bursting in leads, were commonly observed, with an example particle shown in Figure 5. Manual image analyses are being conducted to examine morphology of the sea salt aerosol particles, to determine whether blowing snow sublimation may have contributed to the presence of these salt particles, in addition to bubble bursting in leads (which has a known morphology from many previous studies). Manual analyses are also being used to identify organic-rich particles that appear dark on the metal substrate and can be undercounted by the computer-controlled method.

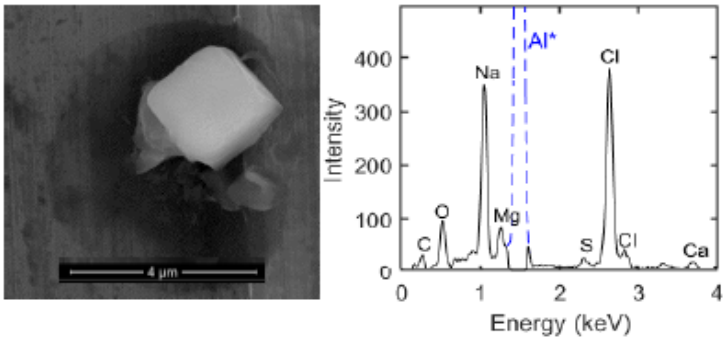

Figure 5. Example SEM image (left) and EDX spectrum (right) for an individual sea spray aerosol particle collected on December 8, 2019 during the MOSAiC expedition. The SEM image shows the bright NaCl cube surrounded by a dark organic coating on the aluminum substrate. The asterisk(*) indicates substrate EDX signal contribution.

3.0 Publications and References

The calibration, analysis, and interpretation of the real-time ATOFMS data are ongoing, as are offline CCSEM-EDX and Raman microspectroscopy measurements, analysis, and interpretation of collected atmospheric particle samples. These tasks have been significantly slowed due to COVID-19, which severely limited instrument and user facility access and led to required instrument maintenance and additional calibrations. Most notably, the Pratt Laboratory did not have microscopy facility access from March 2020 to July 2021. COVID-19 also impacted available personnel resources, leading to further delays. However, as of 2022, we are making significant research progress again. Additional presentations and many publications are expected in the future.

3.1 Presentations

Thus far, initial results have been presented to the scientific community through the following presentations:

Mirrielees, JA, JM Creamean, EJ Costa, N Bergner, J Schmale, M Frey, RM Kirpes, S China, AP Ault, and KA Pratt. 2023. “Identification and chemical composition of sea spray aerosol particles during MOSAiC.” Presented at the 2nd MOSAiC Science Conference, Boulder, Colorado.

Mirrielees, JA, EJ Costa, K Kolozsvari, S China, J Creamean, AP Ault, and KA Pratt. 2023. “Characterization of Arctic sea spray aerosol particles from the year-long MOSAiC expedition.” Presented at the American Meteorological Society Annual Meeting, Denver, Colorado.

Mirrielees, J, RM Kirpes, AM Grannas, V Boschi, N Nahar Lata, S China, P Matrai, AP Ault, and KA Pratt. 2022. “Chemical characterization of individual sea spray aerosol particles from the Arctic and North Atlantic regions.” Presented at the American Association for Aerosol Research Annual Conference, Raleigh, North Carolina.

Wu, J, J Liu, JY Lee, and KA. Pratt. 2021. “Analysis of wintertime Alaskan sea spray aerosol using single-particle mass spectrometry.” Poster presented at the American Association for Aerosol Research Annual Conference.

Wu, J, J Liu, J Lee, L Upchurch, P Quinn, and KA Pratt. 2020. “Mixing state of secondary species in Alaskan Arctic aerosol using single-particle mass spectrometry.” Poster presented at the American Association for Aerosol Research Annual Conference.

Pratt, KA. 2020. “Sea spray and biomass burning aerosol observed during fall-winter on the North Slope of Alaska.” Presented at the 2020 U.S. Department of Energy ARM/ASR Principle Investigator Meeting.

Liu, J, J Lee, L Upchurch, PK Quinn, and KA Pratt. 2019. “Arctic Sea Spray Aerosol (SSA) Production during Polar Night.” Poster presented at the American Geophysical Union Fall Meeting, San Francisco, California.

Liu, J, R Kirpes, AP Ault, and KA Pratt. 2019. “Arctic atmospheric particle sources, morphology, and composition." Presented at the 2019 Environmental Molecular Sciences Laboratory Integration Meeting – Plants, Soil and Aerosols, Richland, Washington.

Pratt, KA, J Liu, and J Lee. 2019. "Arctic aerosol chemical composition, sources, and mixing states on the North Slope of Alaska." Presented at the 2019 U.S. Department of Energy ARM/ASR Principal Investigators Meeting, Rockville, Maryland.

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