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Arctic Ice Nucleation Sampling during MOSAiC (INPMOSAIC2) Field Campaign Report

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Acronyms and Abbreviations

AMF2	second ARM Mobile Facility
AMPC	arctic mixed-phase cloud
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
AWI	Alfred Wegener Institute
CCD	charge-coupled device
COMBLE	Cold Air Outbreaks in the Marine Boundary Layer Experiment
CP	Cold Plate
CSU	Colorado State University
DNA	deoxyribonucleic acid
DOE	U.S. Department of Energy
DRUM	Davis Rotating drum Unit for Monitoring
INP	ice nucleating particle
IS	Ice Spectrometer
MARCUS	Measurements of Aerosols, Radiation, and Clouds over the Southern Ocean
MICRE	Macquarie Island Cloud and Radiation Experiment
MOSAiC	Multidisciplinary drifting Observatory for Study of Arctic Climate
NGS	Next-Generation Sequencing
PCR	polymerase chain reaction
PI	principle investigator
RNA	ribonucleic acid
rpm	revolutions per minute
SHEBA	Surface Heat Budget of the Arctic Ocean
UV	ultraviolet

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1.0 Summary

The accelerated rate of warming in the Arctic is of great concern due to potential impacts that include release of greenhouse gases from permafrost, melting glacial ice contributing to sea level rise, and declining sea ice cover exposing the darker ocean surface. These processes induce positive feedbacks and contribute to further warming that affects climate globally. Clouds play a crucial role in regulating the energy reaching the sea ice and snow surfaces, but the magnitude of their effects on surface temperature is not well constrained in the Arctic. Aerosols are an important contributor by serving as seeds for cloud particle formation, but even less is known about their overall impact and origin. In particular, aerosols that serve as ice nucleating particles (INPs) are vastly understudied, especially above the central Arctic Ocean. However, INPs likely play a significant role in arctic mixed-phase cloud (AMPC) microphysics and the resulting impacts of such clouds on the surface energy budget over the sea ice. Previous studies have improved understanding of arctic INPs but were extremely limited in terms of the aerosol properties measured and their temporal/spatial coverage. There have been no INP data reported over the central Arctic throughout winter, where INPs could potentially have a great impact directly over the sea ice surface when ice coverage is at its maximum and the surface is more susceptible to warming by clouds.[2,3] It has also been reported that clouds act to warm the Arctic much of the year and briefly cool it during the summer, emphasizing the need to evaluate aerosol-cloud interactions during an entire year.[4] The Surface Heat Budget of the Arctic Ocean (SHEBA)[5] experiment was a combined icebreaker/ice camp campaign in which measurements were obtained over the central Arctic during the course of a year (1997–1998); however, only very select aerosol properties were measured via aircraft during the spring (including INPs, but only at –30 °C)[7] and ice nucleation measurements were not conducted at all on the icebreaker.

Until now, a full year's worth of INP measurements have not been conducted ***anywhere*** *in the Arctic and no INP data exist from the central Arctic throughout the winter and spring*, creating a significant gap in understanding AMPC microphysical processes. The year-long transpolar drift experiment, Multidisciplinary drifting Observatory for Study of Arctic Climate (MOSAiC), took place in the central Arctic sea ice pack from September 2019 to October 2020 (Figure 1) and provided the unique and exclusive opportunity to execute novel INP measurements. The INP measurements for MOSAiC forego the limitations of previous studies by: 1) obtaining aerosol/INP observations in the central Arctic, 2) doing so during a full annual cycle, and 3) using a multidisciplinary approach to collect a comprehensive set of INP characterization measurements to help delineate their identities and define their prospective sources.

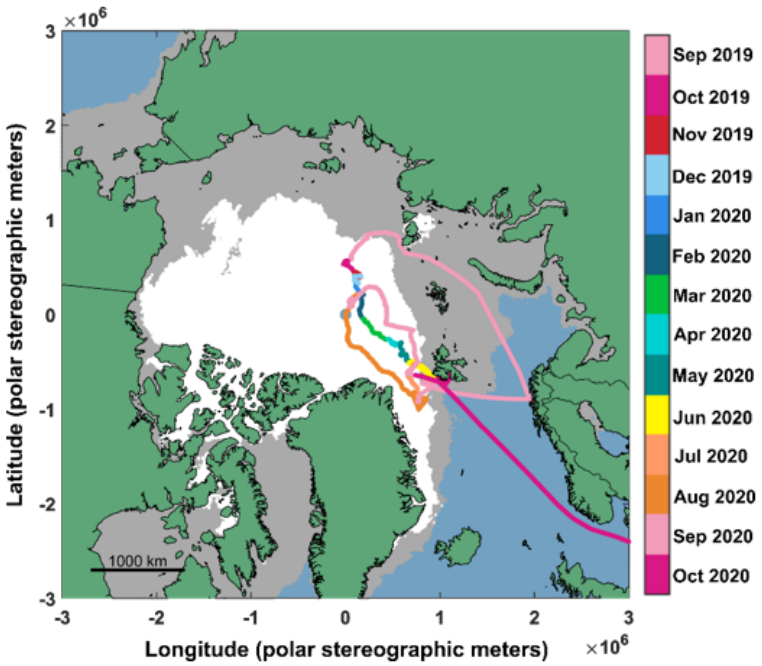

Figure 1. Map of drift during the MOSAiC expedition. The ship track is colored by date. Gray and white shaded areas represent the maximum and minimum sea ice extents in March and September 2020, respectively. Sea ice data were obtained from the National Snow and Ice Data Center.[1] Arctic Mapping Tools for Matlab were used for plotting the data.[6]

The overarching goal of this project is to achieve an unprecedented characterization of INP abundance and sources (including biological) to evaluate their capacity to modulate cloud ice formation over the central Arctic. Specifically, we focused our measurements to address a set of targeted scientific research questions based on current gaps in the understanding of INPs in the central Arctic:

1. How do seasonal changes in sea ice and air mass transport influence INP abundance and sources in the central Arctic?
2. Are marine and sea ice biological processes a significant source of atmospheric INPs as compared to terrestrial sources?
3. Are open-water environments such as leads and melt ponds viable sources of INPs over the sea ice and do such environments exchange INPs with the atmosphere?

To achieve the overarching goal, funding from both the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility and Atmospheric Systems Research (ASR) program was granted. ARM funded continuous collection of total and size-resolved aerosol samples at the second ARM Mobile Facility (AMF2) on the Alfred Wegener Institute's (AWI's) *Polarstern* icebreaker over the entire MOSAiC expedition and ASR-funded offline sample processing for INPs and deoxyribonucleic acid (DNA), in addition to collection of seawater, sea ice, and snow samples for local INP source characterization. Principal Investigator (PI) Creamean was present during the first leg of the expedition (September 2019–January 2020) to establish the sampling protocols (berthing fees were funded by the National Science Foundation). Collaboration with several of the MOSAiC teams, including the Atmosphere, Ecosystems, Biogeochemistry, Ocean, and Sea Ice teams, provided synergistic opportunities to evaluating aerosol-cloud interactions through coordination of sample collection.

In general, these INP data fill a crucial gap in MOSAiC and, more generally, arctic science; they could also help to correct biases in current models from large-eddy simulations to climate models, yielding a better understanding and projection of arctic climate. The cross-disciplinary nature of integrated measurements of INPs and other collocated aerosol, atmospheric state, cloud, ocean, sea ice, and ecological properties has the potential for transformative outcomes and collaborative opportunities with respect to improving understanding of the arctic system.

2.0 Results

The activities funded under ARM involved deployment of: 1) a size-resolved Davis Rotating-drum Unit for Monitoring (DRUM) for immersion INP measurements and 2) two disposable sterile filter unit samplers for immersion INP measurements and DNA sequencing to characterize the airborne microbial community. Both sampling methods were deployed for continuous collection of time-resolved aerosol during MOSAiC as a part of this ARM project. Offline INP processing and DNA sequencing analyses were conducted under the ASR-funded portion of this project.

The DRUM-collected aerosol particles at four size ranges (i.e., from 0.15 to >12 μm in diameter with size cuts at 2.96, 1.21, and 0.34 μm) and sampled at 20–35 lpm. This size range covers a wide array of aerosols—particularly those that serve as INPs.[8-10] Aerosols were impacted on sterile perfluoroalkoxy substrate strips coated with petrolatum that were adhered to discs in each size chamber. The discs moved slowly over time, such that aerosol loading was streaked onto the strips. Every 24 hours, a blank was created on the PFA to separate daily samples. Discs rotated for approximately 24 days before the sampling per strip was complete and changed to a new disc. Sampled discs were stored frozen on *Polarstern* in cleaned, custom holders. The DRUM has been successfully used for size-resolved INP measurements in other arctic and high-alpine locations.[11-13]

The filter units for multi-day integration (72-hour samples) of aerosols were the same as those used for aerosol collections in the recent DOE MARCUS (Measurements of Aerosols, Radiation, and CloUds over the Southern Ocean), MICRE (Macquarie Island Cloud and Radiation Experiment), and COMBLE (Cold air Outbreaks in the Marine Boundary Layer Experiment) studies, and other AMF2 campaigns prior to those.[14-16] The filter system comprised a vacuum pump pulling through Nuclepore polycarbonate filters (0.2 and 0.4 μm backed with 10-μm filters) at 15–35 lpm covered by precipitation shields. Collected filters were removed from the disposable filter units and stored frozen in sterile Petri dishes on *Polarstern.*

All collected samples were transported frozen back to Colorado State University (CSU) for immersion freezing processing. DOE ASR funding supported processing of a subset of the size-resolved and total aerosol samples. For the DRUM samples, one daily sample set every 3–4 days of the study for all four stages was processed using the CSU Cold Plate (CP).[11-13,17,18] Daily samples were cut up and stored frozen in sterile centrifuge tubes prior to processing. Immediately after removal from the freezer, 2 mL of 0.02-μm filtered deionized water was added to the tubes, then mixed using a vortex mixer for at least 30 minutes at 500 rpm to resuspend particles from the strips into the deionized water. Following sample preparation, another single-use syringe was used to create 100 aliquots on a 3-inch-diameter copper plate placed on a cold stage, then covered with a transparent plastic dome. The drops were approximately the same volume (2.5 μL each). The plate was cooled at approximately 3–10 K min^{-1} from room temperature until all drops froze on the plate or until approximately –30 °C. Freezing was visually detected but

recorded through monitoring software to provide the time frozen, temperature, and cooling rate for each drop. Each sample was tested three times. For the total aerosol filter samples, about 3/4 of the 72-hour integrated INP samples were processed using the CSU Ice Spectrometer (IS).[19-26] Filters were placed in sterile centrifuge tubes, 7 mL of 0.02 µm-filtered deionized water added, and particles re-suspended by tumbling end-over-end on a rotator for 20 min. Thirty-two 50-µL aliquots of aerosol suspensions were distributed into sterile, 96-well polymerase chain reaction (PCR) trays in a laminar flow cabinet. Serial dilutions were applied to cover the full temperature range/INP concentration. Plates were then placed into the blocks of the IS, the device covered with a plexiglass window and the headspace purged with filtered N_2. The device was cooled at 0.33 °C min^{-1} until approximately –30 °C using a recirculating low temperature bath, and the freezing of wells recorded every 0.5 °C via a charge-coupled device (CCD) camera system. For both the IS and CP, INP concentrations were calculated per liter of air using the equation from Vali (1971):[27] [INPs] = $-(\ln(f)/V_{aliquot})*(V_{suspension}/V_{air})$ where f is the proportion of droplets not frozen, $V_{aliquot}$ is the volume of each aliquot, $V_{suspension}$ is the volume of the suspension, and V_{air} is the volume of air per sample.

Further, thermal treatments and peroxide digestions provide valuable insights into INP composition. Heat treatments were performed on approximately 1/3 of the IS filter samples by heating 2 mL of suspension to 95 °C for 20 min to denature heat-labile INPs, such as proteins. Hydrogen peroxide (H_2O_2) digestions were performed on a further 2 mL of suspension to remove all bio-organic material. The difference in the INP-temperature spectra after treatment determines the influence of that INP type in the original sample, and the residual spectrum gives the mineral INP component. Thus, this processing provides four key measures from each sample: total, heat-labile (i.e., biological), bio-organic, and mineral INP concentrations.

A subset of the second filters from the total aerosol samplers are undergoing extraction for DNA sequencing under ASR funding. The composition of the microbial community will be profiled using Next-Generation Sequencing (NGS) of the bacteria and archaea (16S rRNA), and eukarya (18S rRNA), after amplification with taxon-specific primers.[28] Prior to DNA extraction, samples of aerosol (in water) are concentrated using 0.22-µm Sterivex cartridge filters. We then use magnetic beads to further increase DNA concentration in the extract. DNA extraction is performed in a UV-decontaminated laminar flow cabinet using pipettes, racks, and consumables reserved exclusively for low copy number samples. For initial PCR we use purified enzyme to minimize contamination, and optimized PCR cycling conditions developed by our group. PCR will target the V4-V5 region of the bacterial 16S and eukaryote 18S genes, using primer pairs 515yF/926pfR[29] and TAReukF/TAReukR, respectively.[30] PCR, amplicon analysis, and amplicon cleanup will be performed at CSU's NGS Core Facility. PCR products will be sequenced using Illumina MiSeq paired-end sequencing after preparation using MiSeq V3 reagents kits. The ability to conduct NGS for all sample types provides the opportunity to perform source tracking analyses to estimate contributions from potential sources (i.e., melted ice, seawater) to the aerosol samples.[31] Further, we can search for bacteria known to contain ice nucleating species/strains across all sample types.[32]

Preliminary results demonstrate unique INP features based on annual cycle variability and differences in the sizes of INPs. Figure 2 shows an example of the seasonal variability in the total INP concentrations measured by the IS. Fall and winter typically exhibited low INP concentrations, especially at the warmer freezing tempearutres. However, enhanced INP concentrations, especially > −10 °C, were observed during spring and especially summer. These higher concentrations coincide with the onset and peak of sea ice melt—exposing higher fractions of open water in the form of open leads and melt ponds—and with the peak of sun exposure and biological productivity. Interpretation of the INP results with

complementary data sets, thermal and peroxide treatment results, and DNA results is ongoing to determine if local marine biological sources are responsible for the elevated INPs during the warmer months.

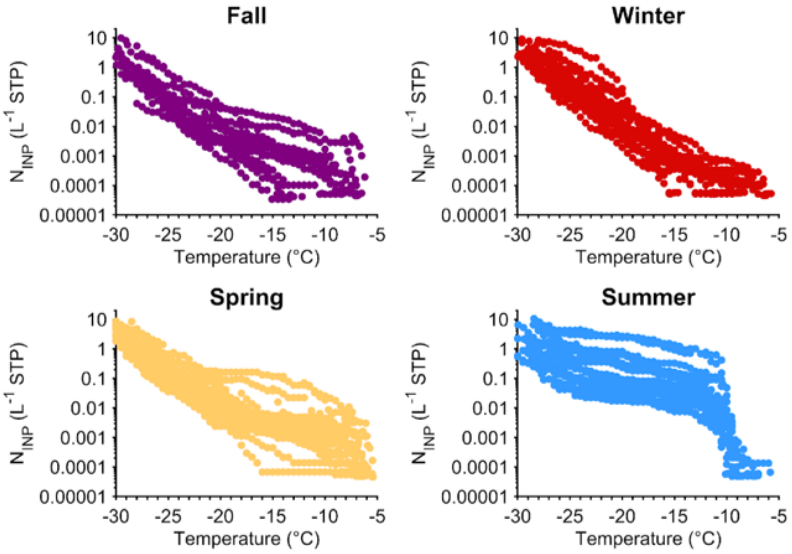

Figure 2. Cumulative INP spectra from the subset of processed total aerosol samples using the CSU IS. Spectra are separated by calendar season.

Size-resolved INP data also exhibit interesting preliminary results. Figure 3 shows the size-resolved INPs from the DRUM-CP subset of samples. Previous size-resolved INP studies have indicated that INP efficiency and concentrations increase with increasing particle size. However, this is not the case for the MOSAiC data set, whereby the 1.21–2.96-µm particles were more efficient than the largest (2.96–12 µm), namely during the summer months. The smallest particle sizes (0.34–1.21 and 0.15–0.34 µm particles) exhibited very small to no INP concentrations, especially during the fall and winter months. Examining these unique size-resolved INP relationships with supporting aerosol data sets (i.e., total aerosol number concentrations and size distributions) in addition to source analyses using air mass backward trajectory analyses is ongoing to explain these observed trends.

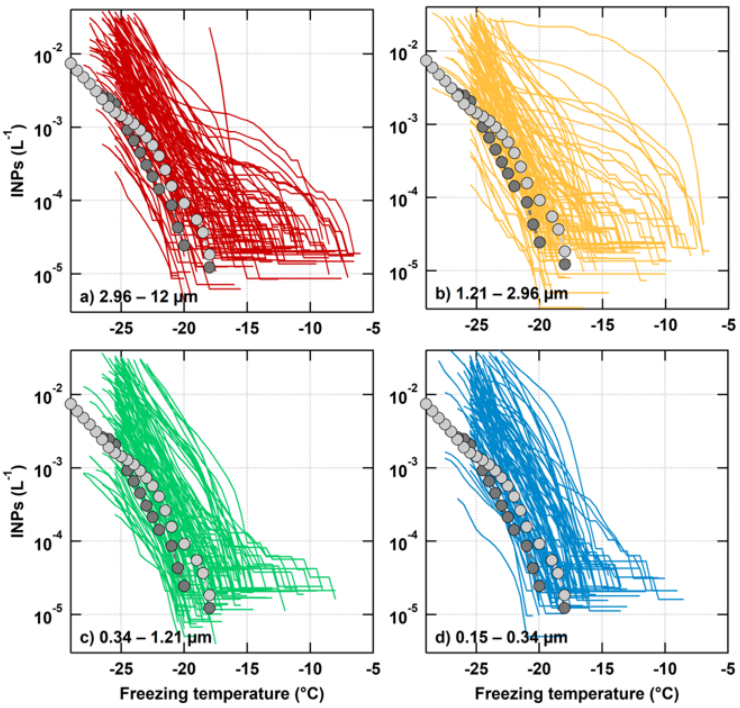

Figure 3. Size-resolved cumulative INP spectra from the full MOSAiC year. The grey spectra represent the deionized water and DRUM substrate blanks.

3.0 Publications and References

3.1 Conference Presentations

Creamean, J, T Hill, K Barry, P DeMott, and the MOSAiC field team. 2020. Evaluation of ice nucleating particles and their sources in the central Arctic during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) experiment. American Geophysical Union Fall Meeting, virtual online, 1−17 December.

Creamean, J, T Hill, K Barry, P DeMott, and the MOSAiC field team. 2021. Evaluation of ice nucleating particles and their sources in the central Arctic during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) experiment. American Meteorological Society annual meeting, virtual online, 11−14 January.

Creamean, J, T Hill, K Barry, P DeMott, and the MOSAiC field team. 2021. Evaluation of ice nucleating particles and their sources in the central Arctic during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) experiment. Arctic Frontiers meeting, virtual online, 1−4 February.

Barry, W, T Hill, J Creamean, P DeMott, and the MOSAiC field team. 2021. Tracking ice nucleating particles in the central Arctic during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) experiment. Arctic Science Summit Week, virtual online, 24−26 March.

Creamean, J, K Barry, T Hill, C Hume, P DeMott, and the MOSAiC field team. 2021. The seasonal variability of ice nucleating particles and their sources in the central Arctic during the MOSAiC experiment. DOE ARM-ASR Joint PI meeting, virtual online, 21−24 June.

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Creamean, J, K Barry, T Hill, C Hume, P DeMott, M Shupe, S Dahlke, S Willmes, J Schmale, I Beck, C Hoppe, A Fong, E Chamberlain, R Scharien, and O Persson. 2021. "The seasonal contrast of aerosols that can seed ice formation in central Arctic clouds." *Nature Geoscience*, in preparation.

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4.0 Lessons Learned

The sample collection went relatively very smoothly, especially considering the challenging central Arctic environment and duration of the campaign. The AMF crew did a tremendous job not only on maintaining our sample collections but also successfully operating all of the instruments deployed for MOSAiC. The ARM MOSAiC data set is an unprecedented resource that will be used by the scientific international community for years to come.

The lessons were learned as a result of missing data. A small amount of missing data (i.e., the latter half of February) was a result of personnel shifts not being able to change out sample filters and discs. Additionally, a small number of the stage D samples were lost due to sample substrate contamination during transportation. In the future, more care will be taken when handling and packing the samples for transportation.

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