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Exploratory Ice Nucleation Measurements at Oliktok Field Campaign Report

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Acronyms and Abbreviations

AMF3	third ARM Mobile Facility
AOS	Aerosol Observing System
ARM	Atmospheric Radiation Measurement (Climate Research Facility)
CPC	condensation particle counter
DFA	drop freezing assay
DOE	U.S. Department of Energy
DRUM	Davis Rotation-drum Unit for Monitoring
ICARUS	Inaugural Campaigns for ARM Research using Unmanned Systems
INP	ice nucleating particle
INPOP	Ice Nucleation Particles at Oliktok Point
LLC	limited-liability company
NPF	new particle formation
NSA	North Slope of Alaska
PBAP	primary biological aerosol particle
PFA	perfluoroalkoxy
PI	Principal Investigator
POPS	printed optical particle spectrometer
UHSAS	ultra-high-sensitivity aerosol spectrometer

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1.0 Summary

Ice nucleating particles (INPs)—such as mineral dust and bacteria (1)—are a crucial source of seeds for cloud ice crystal formation, and can subsequently impact cloud radiative properties, lifetime, and precipitation formation processes (2-4). The presence of such particles in the atmosphere can be rare, but large concentrations are not required to have substantial impacts on cloud microphysics (3). However, knowledge of INPs, particularly in the Arctic, is limited due to the dearth of measurements in time and space. It is especially important to evaluate the sources and efficiency of INPs in the Arctic due to the role mixed-phase clouds play in facilitating the surface energy budget and thus energy reaching the sea ice and snow surfaces (4, 5).

The purpose of this campaign, called INPOP (Ice Nucleating Particles at Oliktok Point), is to address the issue of limited information of Arctic INPs by providing time- and size-resolved INP concentrations over the course of a 3-month springtime campaign. This period is subject to Arctic haze, whereby pollutants from mid-latitudes are frequently transported to the Arctic and influence the relatively clean Arctic atmosphere (6-9). One unique aspect of this study is that the measurements were conducted at the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility third ARM Mobile Facility (AMF3) in Oliktok Point, North Slope of Alaska (NSA), which is located in the northwest edge of the Prudhoe Bay oilfield. Typically, oil and natural gas extraction emissions are not thought to serve as a significant source of INPs, since naturally produced primary aerosols are the most efficient and abundant INPs (e.g., 1). However, the goal of INPOP was to evaluate INPs in the context of supporting aerosol and meteorological observations to assess: (1) if local industrial activities due to the oilfield operations can serve as a source of INPs (e.g., such as road dust from routine vehicular operations) and (2) if other regional or long-range transported sources impact a polluted Arctic location by introducing higher concentrations of more efficient INPs (e.g., transport of primary biological aerosol particles [PBAPs] from open ocean water or Arctic haze INPs). Unlike other Arctic coastal locations in the spring, Oliktok Point has a distinct combination of aerosol sources including long-range transported haze, regional/local marine emissions, and the local oilfield activities.

Ground-based, immersion-mode, ice nucleation measurements were conducted on samples collected at the AMF3 in Oliktok Point from 1 March to 31 May, 2017. Samples were collected using a Davis Rotating-drum Unit for Monitoring (DRUM; DRUMAir, LLC) cascading impactor in 4 stages (i.e., size bins). Instrument information on the model used, the DA400, can be found at the DRUMAir website (http://www.drumair.com/). Stages A, B, C, and D had particle size cuts at 2.96, 1.21, 0.34, and 0.15 µm particle diameter. Strips of sample substrate (Mylar™; DuPont®) were adhered to each disc in each stage and coated with petrolatum to enable particles to stick on impact. The discs move slowly over time, such that aerosol loading is streaked onto the Mylar. Every 24 hours, a blank spot is created on the Mylar to separate daily samples. The pump typically pulled 27 lpm at the inlet with all the discs in place in each stage. Daily samples were collected for each of the four stages, equaling 38,880 total L of air per sample. Discs rotated for approximately 24 days before the sampling per strip was complete and a new disc was installed by the ARM operators. Discs were stored frozen after collection until the end of the study.

Following collection and transport from the AMF3 to Principal Investigator (PI) Creamean's laboratory in Boulder, Colorado, strips were removed from the discs and cut up to separate daily samples. Each daily portion of the strip was placed in a separate sterile bag and stored in a standard commercial freezer until

analysis (approximately 4-5 months after collection). Drop freezing assays (DFAs), a common technique used for immersion-mode ice nucleation measurements (e.g., 10, 11, 12), were conducted on one daily sample per week of the study for all four stages, with daily samples analyzed from 22 to 29 May, 2017. Immediately after removal from the freezer, 2 mL of ultrapure water were added with a single-use sterile syringe directly to the bags, then mixed using a vortex mixer for 2 hours at 500 rpm to re-suspend particles from the Mylar in the ultrapure water. Following sample preparation, another single-use syringe was used to create 100 drop aliquots (from 0.25 mL total per test) on a 3-inch-diameter copper plate, then covered with a transparent plastic dome. The drops were approximately the same volume (2.5 µL each), verified through careful inspection. The plate was cooled at approximately 3-10 °C min^{-1} from room temperature until all drops froze on the plate. Temperature was measured by an Omega thermocouple meter with the probe inside the copper plate. Drop freezing was visually detected, but recorded through monitoring software to provide the time frozen, channel of the meter used, temperature, and cooling rate for each drop. Not all 100 drops were always detected; the total number of rows in each data file equals the number of drops recorded, which typically was > 80%. Each sample was tested three times (i.e., a new set of 100 drops was created for each test). From the freezing temperatures recorded, the fraction frozen can be calculated as can the estimated INP concentration per L of air using the equation from Vali (1971) (13):

$$K(\theta)\ (L^{-1}) = \frac{\ln(1-f)}{V_{drop}} \times \frac{V_{suspension}}{V_{air}}$$

where f is the proportion of droplets frozen, V_{drop} is the volume of each drop (i.e., 2.5E-6 L), $V_{suspension}$ is the volume of the suspension (i.e., 0.002 L), and V_{air} is the volume of air per sample (i.e., 38,880 L).

The 22-29 May period was a unique event in which the aerosol observing system (AOS) measurements showed several processes occurring in this span of seven days. Figure 1 shows particle counts from the ultrafine condensation particle counter (CPC) (3 nm-3 µm), fine CPC (10 nm-3 µm), ultra-high-sensitivity aerosol spectrometer (UHSAS) (60 nm-1 µm), and wind speed and direction from the AOS. Above each "process" is the classification for that period based on particle counts, in the larger context of AOS measurements at Oliktok Point. The beginning of the period was relatively clean, then transitioned to a mixture of polluted conditions and a possible nucleation event followed by particle growth (i.e., new particle formation or NPF). Additionally, this time period overlaps with an interesting case study from Inaugural Campaigns for ARM Research using Unmanned Systems (ICARUS). As a result of this combination of processes in one period, daily INP sample analysis was conducted on all size bins from the DRUM.

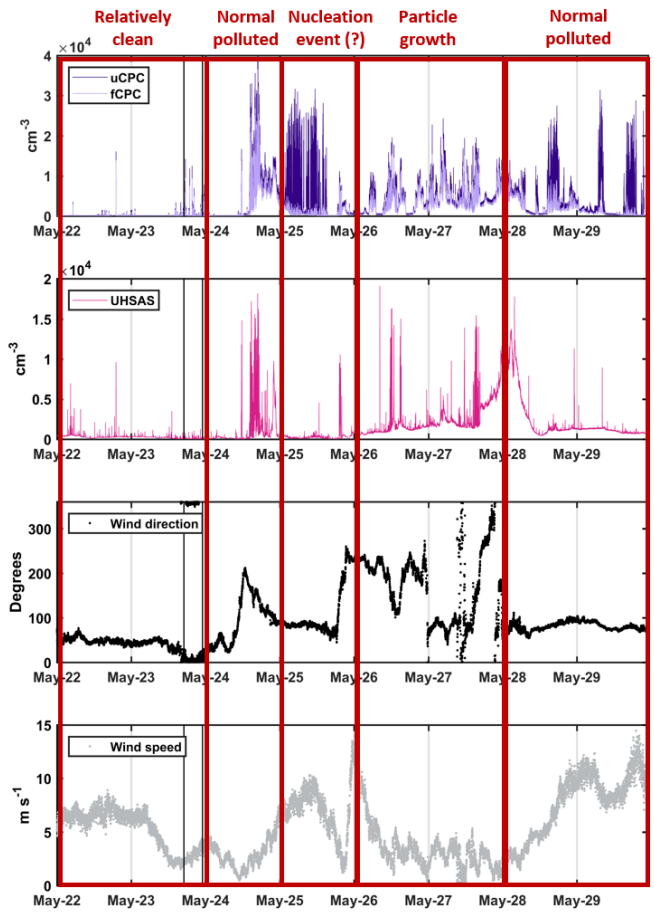

Figure 1. AOS observations during the unique period of interest. The vertical black lines in each panel are the balloon flight from the ARM ICARUS campaign at Oliktok Point, whereby a printed optical particle spectrometer (POPS) was deployed.

2.0 Results

Each triplicate test was plotted and checked for consistency between the three tests. Figure 2 shows cumulative freezing spectra from 26 May, 2017 for all stages. The fraction frozen represents the total number of drops frozen per 0.1 °C. The cooling rate (i.e., temperature slope) was typically held between −3 and −8 °C min^{-1}. Note that there are no significant differences in fraction frozen when the cooling rate varies within this range.

From the triplicate testing, an average cumulative estimated INP spectrum per sample can be created based on the Vali equation (13) and binned by 1 °C increments. Figure 3 shows such spectra for samples analyzed from the entire study. The black spectra for each stage represent the ultrapure water blank in the sterile bag and the colored spectra represent the daily samples from the case study of interest (22-29 May). Three interesting features are elucidated from the spectra: (1) the larger the particles, the higher the estimated concentrations of warm-temperature INPs (i.e., > −10 °C, which is indicative of biological INPs as compared to mineral or soot (1, 14-16)), (2) the smallest sizes (stages C and D) had INP concentrations below the sample blank, and (3) the case study time period had the highest concentrations of warm-temperature INPs in the largest sizes (stages A and B).

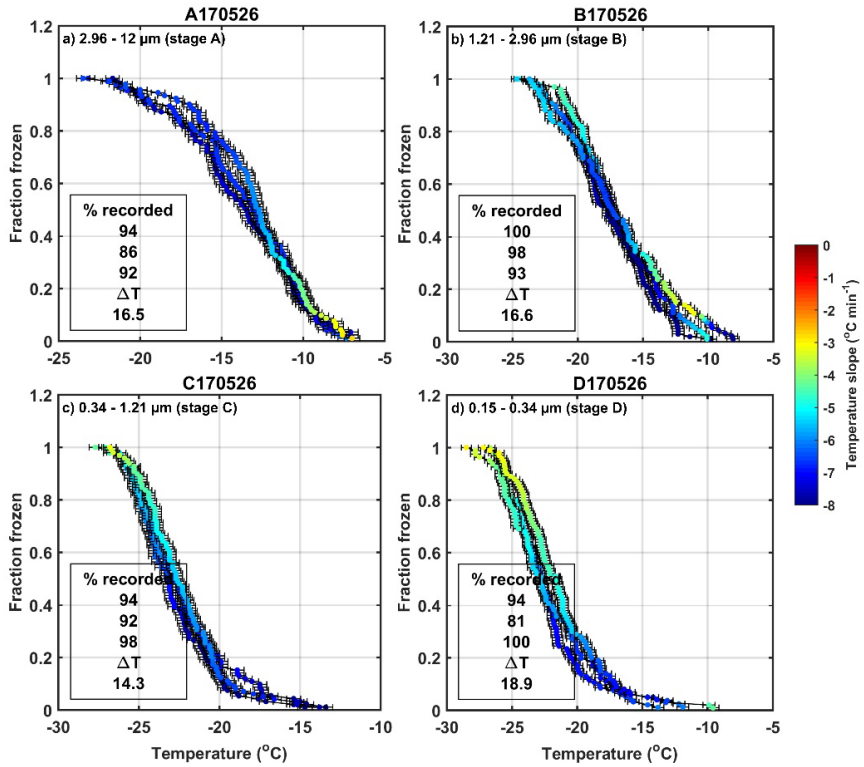

Figure 2. Example of freezing spectra from triplicate tests for all stages from 26 May, 2017. The spectra are colored by cold plate cooling rate (i.e., temperature slope). "%recorded" correspond to how many of the 100 drops were detected as frozen. "ΔT" is the difference between the warmest onset freezing temperature and the coldest temperature in which the last drop froze. The error bars represent uncertainty associated with the Omega probe.

The largest particles serving as the most efficient INPs (i.e., they form ice at relatively warm temperatures) and decreasing efficiency with decreasing size indicates that the size of the INP is important with regard to their ice nucleating ability (17). However, particle size is typically unique to its composition. For example, bacteria, algae, fungi, phytoplankton, and mineral dust tend to fall within this size range, and are some of the most efficient and abundant INPs. Nano-sized ice nucleating fragments have previously been reported, but these are rare and are typically attached to larger, host particles in the atmosphere (18).

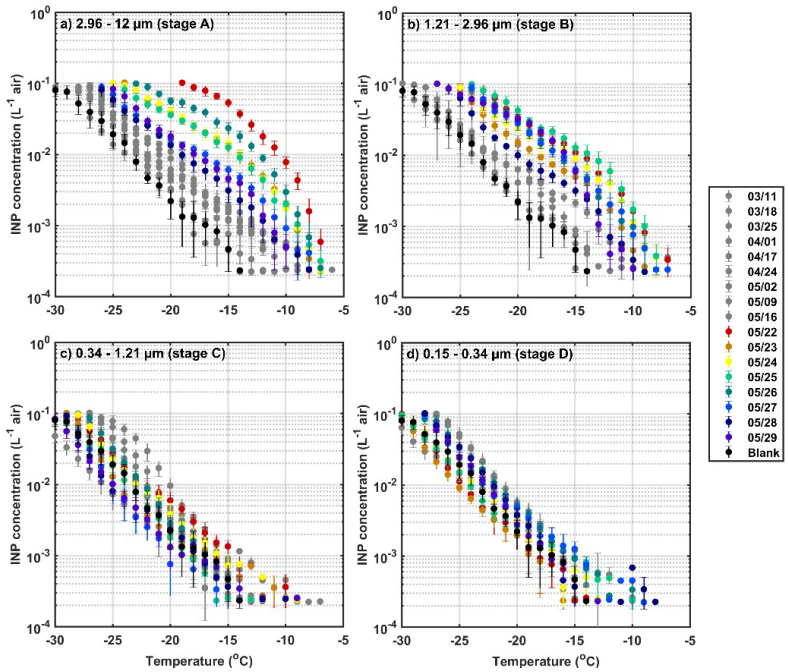

Figure 3. Cumulative INP spectra for all stages for the entire INPOP study.

In contrast, the smallest particles from INPOP (stages C and D) had less efficient INPs, especially for the case study time period. A possible explanation is that these stages were rich in organic aerosol, based on preliminary chemistry results by a collaborative ARM proposal from Kerri Pratt. High concentrations of organic aerosol could potentially suppress the ice nucleation abilities not only of the ambient particles, but also the artifacts in the ultrapure water (19). However, this effect needs to be further investigated.

The highest, warm-temperature INPs were observed during the case study in the largest sizes. This time period was influenced by winds from over the sea ice and open ocean water north and west of Alaska—the sea ice started retreating in this region mid-May (Figure 4). One explanation could be the influence of marine biological INPs (20-23), but this needs to be further investigated using the AMF3 meteorological measurements and air mass trajectory analysis.

These preliminary results inspire future research opportunities with regard to data analysis and interpretation of the three features highlighted above. J. Creamean will have one summer student focused on conducting DFAs on remaining INPOP samples. The student will also reanalyze samples presented here to check for reproducibility after extended frozen storage. The motivation for analyzing all samples is to investigate possible increases in INPs as the ocean water and vegetation surfaces (i.e., sources of PBAPs) are exposed. Ideally, future parallel studies could be conducted at AMF3 or the NSA site at Utqiaġvik, but for the entire duration of the spring and summer to evaluate seasonal variability in INP sources, since the Arctic Ocean water is a host of biological productivity in the summer (24) and the extent to which tundra emis INPs to the atmosphere is unknown.

Figure 4. Sea ice extent from the national Snow and Ice Data Center (http://inside.org/arcticseaicenews/sea-ice-comparison-tool/).

In general, INPOP sheds light into the possibility of long-term INP measurements in an Arctic environment. Although the measurements are more labor intensive due to the offline nature of the analysis, such a measurement could be implemented in the future if there are sufficient resources (e.g., specially trained personnel).

3.0 Publications and References

3.1 Presentations

The following are presentations highlighting results from INPOP, including near-future presentations:

Creamean, JM, J Cross, R Pickart, L McRaven, D Schmale, R Hanlon, E Collins, K Pratt, and R Kirpes. 2017. Understanding the sources of Arctic biological ice nucleating particles, Third Microbes in the Land-Atmosphere Feedback Workshop, Gothenburg, Sweden.

Creamean, JM, N Spada, R Kirpes, and K Pratt. 2017. Characterization of ice nucleating particles during continuous springtime measurements in Prudhoe Bay: an Arctic oilfield location, American Geophysical Union Fall Meeting, New Orleans, Louisiana.

Creamean, JM, A Solomon, R Hanlon, D Schmale, R Kirpes, K Pratt, N Spada, J Cross, R. Pickart, and V Phillips.2018. Understanding the sources of Arctic biological ice nucleating particles, POLAR 2018, Davos, Switzerland.

Currently, research is ongoing by evaluating supporting ARM measurements for INPOP. Two publication outlines have been prepared, but more work is needed before submission of the manuscripts. Plans for publications include:

Creamean, JM, R Kirpes, K Pratt, N Spada, and G de Boer. 2017. "Marine influence on aerosol-cloud interactions in an Arctic oilfield location." *Atmospheric Chemistry and Physics*, in preparation.

Creamean, JM, R Kirpes, K Pratt, N Spada, and G de Boer. 2017. "Increase in Arctic warm temperature ice nucleating particle concentrations during the transition from spring to summer." *Geophysical Research Letters*, in preparation.

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4.0 Lessons Learned

A couple of lessons were learned as a result of sample collection, storage, transport, and analysis. First, some missing data were a result of power outages, but stages C and D were lost during the first part of the study due to sample substrate contamination (i.e., one strip fell on the ground during disc replacement and one was contaminated during sample handling). In the future, more care will be taken to properly train the technicians and when handling the samples.

Another lesson could be eliminating possible uncertainties and errors in the results by conducting the DFAs in the field. J. Creamean has since streamlined the DFA technique, and has made it field-portable. This would eliminate possible sources of contamination or alteration of the sample particles from transport and extended storage, although the effects of these is unknown. However, this would require a PI in the field or a full-time technician trained to conduct DFA.

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