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Ice Nucleation Measurements during TRACER Field Campaign Report

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Acronyms and Abbreviations

AMF	ARM Mobile Facility
ANC	ancillary site
ARM	Atmospheric Radiation Measurement
CCNC	cloud condensation nuclei counter
CPC	condensation particle counter
DRUM	Davis Rotating Uniform size-cut Monitor
GPS	Global Positioning System
INP	ice nucleating particle
LPM	liters per minute
MPL	micropulse lidar
PDF	probability density function
ROAM-V	Rapid Onsite Atmospheric Measurements Van
SBF	sea-breeze front
SMPS	scanning mobility particle sizer
TRACER	Tracking Aerosol Convection Interactions Experiment
UHPLC	ultra-high-performance liquid chromatography

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1.0 Summary

This campaign, Ice Nucleation Measurements during TRACER, was conducted during the TRacking Aerosol Convection Interactions ExpeRiment (TRACER) in Houston, Texas during June-September, 2022. Specifically, our ice nucleation campaign was designed to improve understanding of ice nucleating particles (INPs) and their role in deep convective clouds across the Houston region. Summertime deep convection in southeast Texas is regularly tied to the sea-breeze front (SBF) that pushes inland from the Gulf of Mexico across the region.

During TRACER, three Davis Rotating Uniform size-cut Monitors (DRUM; DRUMAir 4-DRUM) were used to collect aerosols for offline ice nucleation measurements in the Brooks laboratory at Texas A&M University. The three instruments were located at the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) first Mobile Facility (AMF1) in La Porte, at the ancillary site in Guy (ANC), and onboard the newly developed Texas A&M Rapid Onsite Atmospheric Measurements Van (ROAM-V). Aerosol at the main ARM TRACER site, ARM1, is expected to arise from local urban, industrial, and ship channel sources. In contrast, the ANC is isolated and rural, and can therefore provide a continental background aerosol population. On select days when convective development was forecast to be probable, ROAM-V commuted from College Station, Texas to Galveston, sampled in the morning/mid-day on the coast, and then transited to a second inland site for the afternoon/evening. Sampling from the ROAM-V allowed us to capture aerosol and meteorological differences between maritime and continental airmasses by strategically positioning the ROAM-V near the coast in the morning in the opposite airmass from the AMF1 site.

ROAM-V was deployed to capture airmasses behind (maritime) and ahead (continental) of the passage of the SBF through Houston. On select sampling days, ROAM-V sampled in the morning/mid-day on the coast and then transited to a second inland site for the afternoon/evening. The suite of instruments deployed on ROAM-V included a condensation particle counter (CPC; GRIMM Model 5.403 CPC), scanning mobility particle sizer (SMPS; TSI 3750 detector, TSI 3082 classifier, TSI 3088 neutralizer, TSI 3081A differential mobility analyzer), cloud condensation nuclei counter (Droplet Measurement Technologies CCN counter), micropulse lidar (Droplet Measurement Technologies micropulse lidar [miniMPL]), and one of the Davis Rotating Uniform size-cut Monitors (DRUM; DRUMAir 4-DRUM). Before sampling at each location, the latitude and longitude were recorded using the Global Positioning System (GPS) on the phone application "My Altitude".

Each DRUM sampler was operated at a flow rate of 23 LPM. Each DRUM has four stages in which the aerodynamic diameter size cuts are as follows: stage 1, larger than 3 µm; stage 2, 3 to 1.2 µm; stage 3, 1.2 to 0.34 µm; and stage 4, 0.34 to 0.15 µm. Pretreated aluminum foil was used as a substrate on all stages. The DRUM onboard ROAM-V was operated at a rotation rate of 150 mm per day to clearly separate the multiple deployment locations for ROAM-V, (out of practical need this is faster than the rotation selected for the weekly AMF1 and ANC DRUMS and thus the time resolution from the ROAM-V drum is substantially higher). For the ROAM-V DRUM, substrates were changed every deployment and transported to Texas A&M for storage in a -80 °C freezer until analysis.

The DRUMs at the AMF1 and ANC sites were delivered and installed at the start of the TRACER intensive operational period in June 2022 and sampled through September 2022. At the AMF1 and ANC

sites, the DRUMs were operated on the shared aerosol inlet, with generous support of the ARM site staff. These instruments rotated 24 mm per day and only contained aluminum foil substrates. Substrates were changed weekly and transported to Texas A&M for storage in a -80 °C freezer until analysis. Every week, with a few exceptions, during this period has substrates with aerosol sample to be analyzed.

At Texas A&M, ice nucleation experiments were conducted to measure the ice nucleation temperature of the ambient aerosol samples collected from the three DRUMs using our previously established procedures (Alsante et al. 2023, Fornea et al. 2009, Matthews et al. 2023). For ice nucleation, only samples collected on stage 3 were analyzed, given that these are the most relevant size (1.2 to 0.34 μm diameter) for potential ice nucleating particles. For the AMF1 and ANC sites, we cut and analyzed 2-mm (2-hour) samples. We analyzed the time periods of the AMF1 site instrument when the ROAM-V was deployed. A 72-hour period from July 11th at 23:49 through July 15th at 1:49 was analyzed from the ANC site instrument. For the ROAM-V instrument, we separated the daily samples by site location. Between 1- and 6-hour independent samples were analyzed at each location. We also cut the ROAM-V samples in half to allow for compositional analysis of the aerosol on the other half of the substrate. All viable samples from the ROAM-V were analyzed.

During the lab experiments, ice nucleation temperatures were determined with a custom experimental apparatus, recently updated to include an array of 16 individual samples (Matthews et al. 2023). In our experimental procedure, we used 100 μL of ultra-high-performance liquid chromatography (UHPLC) water (Sigma Aldrich, >99.9% purity) to wash off the aerosol from the DRUM substrate. Then, we micro-pipetted 2-μl droplet samples into each of the 16 wells of the array, using hydrophobically coated microscope slides. Experiments would cycle 28 times from 10 °C to -40 °C over a 20-hour period. During analysis of the TRACER samples, nine experiments were conducted with UHPLC water process blanks that followed an identical preparation procedure. The average freezing temperature and standard deviation for these process blanks is -27.7±2.4 °C.

2.0 Results

The ROAM-V started sampling in mid-July and was typically only deployed to the field when the TRACER team called for an enhanced sounding day. In total, the DRUM onboard the ROAM-V had an aerosol sample for 33 days from mid-July to the end of September 2022. The vast majority of ROAM-V samples were useable, all of which have been analyzed and reported in the ARM Data Center. Nevertheless, a small number of samples were not useable due to instrument malfunctions as well as user error (either in the field or during analysis). For example, the substrates would become wet, ruining the sample, in the event of a heavy rain event that lasted for more than about 10 minutes. In contrast, the instrument setup on the shared aerosol inlet at the AMF1 and ANC sites was more susceptible to condensation on the DRUM substrates, which made it difficult to use some of the substrates. Additionally, the AMF1 and ANC instruments (particularly the AMF1 DRUM) had issues throughout the campaign with its impactor rotation. The impactors stopped rotating during several of the weeks of the campaign. This issue makes time-resolved analysis of impacted aerosol impossible for each period after the impactors stopped rotating.

Samples were from the Al foil trips of the DRUM Impaction Stage 3 as described above. Next, deposited sample was washed from the substrate using 100 μL of UHPLC water (Sigma Aldrich, >99.9% purity). Sample mean nucleation temperature can be qualitative described as including weakly effective, moderately effective, and effective, which corresponds to mean freezing temperatures of -32 to -25 °C, -25 to -20 °C, and -20 to -10 °C, respectively (Alsante et al. 2023). With these definitions, the process blank is considered a weakly efficient INP, suggesting the processing technique introduces weakly efficient INPs to the UHPLC water. Each sample had a single ice nucleation experiment with either 27 or 28 freezing events.

In addition to the impacted aerosol samples, we also tested the ice nucleating ability of the UHPLC water samples, which were subjected to the same series of rinse and preparation steps as the DRUM samples. These data are considered a process blank for our experimental process employed here. This provided a baseline to compare our ice nucleation results for the aerosol samples. The average freezing temperature of the ultrapure water process blank is -27.7±2.35 °C from nine independent nucleation experiments with 27 freezing events per experiment. The theoretical nucleation temperature of pure water is ~ -38 °C (Heymsfield and Miloshevich 1993). The difference between our experimental freezing temperature and the theoretical value can be explained by minor contamination of the UHPLC water introduced during preparation of the process blank (processed the same as our samples) and the setup of the ice nucleation experiment.

Ice nucleation experiments, for the entire campaign, from the DRUM sampler mounted in the ROAM-V are shown in Figure 1 as a probability density function (PDF). It can be seen that the aerosol sampled by ROAM-V nucleates ice over a range of temperatures (-29 to -15 °C) in one of three distinct modes. The coldest mode has the highest proportion of freezing temperatures with a peak around -25 °C and a range from about -29 to -21 °C. First, we note that for any data with freezing observed at temperatures at or below the temperature of the blank 27.7±2.35 °C, it cannot be said for certain that the sample contains any INPs at all. A significant fraction of the samples were in this category and will not be considered in further assessment of the ice nucleation ability of aerosols collected in the field. Above the threshold of the blank, there remain three distinct modes of freezing temperatures, suggesting that three independent types of aerosols with independent ice-nucleating abilities were present in air masses throughout the campaign. The majority of aerosols lie in the mode of the coldest freezing temperatures. Theses did not freeze until the samples were cooled to -21 °C and below. The INP activating in the coldest mode can be described as either weakly efficient or moderately efficient, in line with many other INPs (Alsante et al. 2023).

In contrast, the INPs activating to form the middle and warmest freezing modes can be described as efficient. The middle freezing mode has a center at -18.5 °C with a range from -20.5 to -17.5 °C. The warmest freezing mode, with the smallest proportion of freezing events, ranges from -17.5 to -14.5 °C with a peak at -16 °C.

For the M1 site DRUM, the analyzed two-hour samples have a range of freezing temperatures similar to the ROAM-V DRUM samples. The range in freezing temperatures runs from -31.5 to -14 °C. Like the ROAM-V samples, the range in freezing temperatures suggests that weakly efficient, moderately efficient, and efficient INPs were all present at the AMF1 site while the ROAM-V was deployed for TRACER. Additionally, the coldest mode of freezing temperatures has the largest proportion of freezing events like the ROAM-V data. The three-day period analyzed for the ANC site showed that only one two-hour sample would be considered to have activated efficient INP, according to the mean freezing temperature. Next steps in this project include compositional analysis of the aerosols from Stage 3 drums to assess the types and sources of aerosols that froze in these modes.

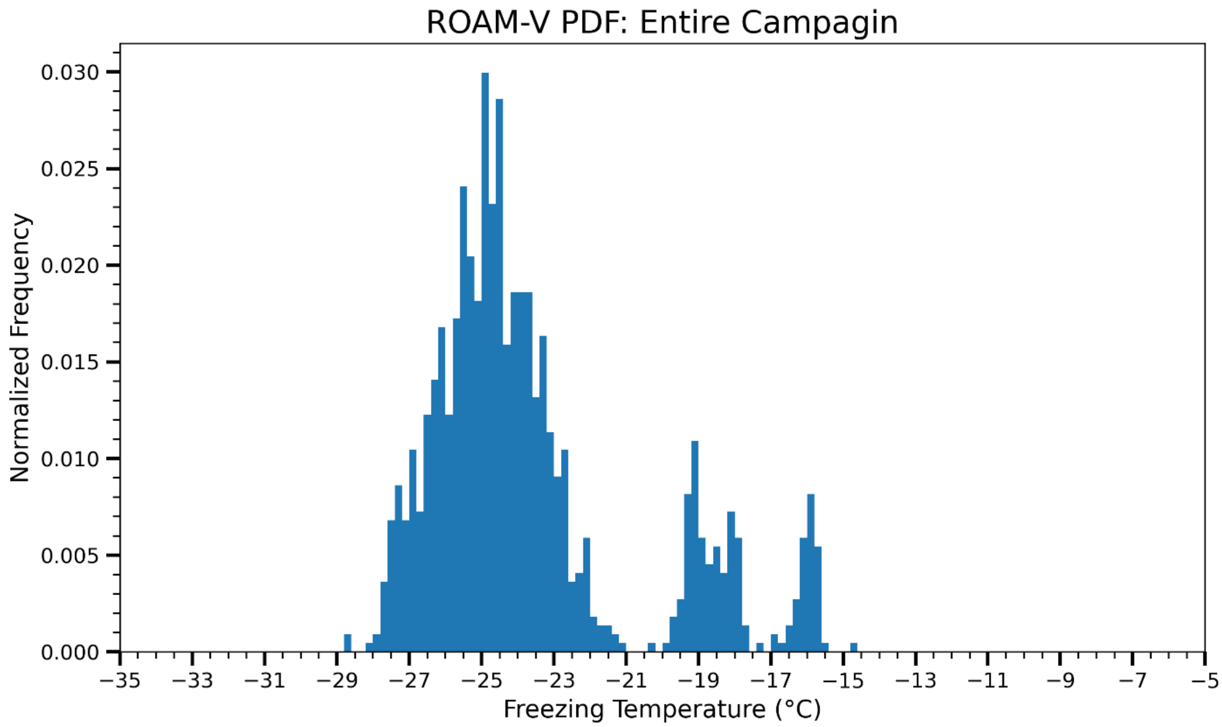

Figure 1. Probability density function (PDF) of measured freezing temperatures for analyzed impacted aerosol samples collected by the DRUM onboard the ROAM-V over the entire TRACER campaign.

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4.0 Lessons Learned

The operation of the DRUM samplers at AMF1 and ANC would not have been possible without the tremendous hard work and technical knowledge of the ARM staff supporting the operation on site.

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