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Ice Nucleation Spectrometer (INS) Instrument Handbook

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ARM

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Acronyms and Abbreviations

3D	three-dimensional
AMF	ARM Mobile Facility
AOS	Aerosol Observing System
ARM	Atmospheric Radiation Measurement
CACTI	Cloud, Aerosol, and Complex Terrain Interactions
CCD	charge-coupled device
CCN	cloud condensation nuclei
CI	confidence interval
CSU	Colorado State University
DI	deionized
HEPA	high-efficiency particulate air
IN	ice nuclei
INP	ice nucleating particle
INS	ice nucleation spectrometer
IS	Ice Spectrometer
PCR	polymerase chain reaction
QA	quality assurance
QC	quality control
SAIL	Surface Atmosphere Integrated Field Laboratory
SGP	Southern Great Plains
STP	standard temperature and pressure
TBS	tethered balloon system
TRAPS	Time-Resolved Aerosol Particle Sampler
UV-B	ultraviolet light with wavelengths of 290-320 nanometers

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2.0 Instrument Technical Specifications

The Ice nucleation spectrometer (INS) is an offline analytical measurement system used to process filter samples for freezing temperature spectra of immersion-mode ice nucleating particle (INP) number concentrations. It is almost identical to the Colorado State University (CSU) Ice Spectrometer (IS) design. Filter samples are collected at U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facilities and transferred to CSU for offline analysis with the INS, as described in detail below.

2.1 Filter Preparation

The single-use Nalgene™ Sterile Analytical Filter Units are prepared by replacing their cellulose nitrate filters with 0.2-µm polycarbonate filters backed with 10-µm polycarbonate filters (each are 47-mm

diameter Whatman® Nuclepore™ Track-Etched Membranes), both pre-cleaned in-house (Barry et al. 2021). Filter units are dis- and re-assembled under ultraclean conditions in a laminar flow cabinet with near-zero ambient particle concentrations. The modified filter units are then capped and individually stored in clean airtight bags until use.

2.2 Filter Collection

Filter samples are collected routinely at select ARM facilities. Each filter unit sampling apparatus consists of sterile single-use filter units prepared at CSU, a totalizing mass flow meter (TSI Mass Flow Meter 5200-1, TSI, Inc.), vacuum pump (2688CE44 Oil-less Piston Compressor/Vacuum Pump, Thomas), tubing, and precipitation shields (Figure 1). Two identical apparatuses are operated in tandem to collect primary filters for INP analyses and duplicate filters that serve as backup or as archival samples for users to obtain for their own analytical ventures. The filter units are open-faced, secured outside to the ARM Aerosol Observing System (AOS) railing, and shielded from precipitation. Vacuum line tubing connects each filter unit to the flow meter followed by vacuum pump, both of which are housed inside the AOS container.

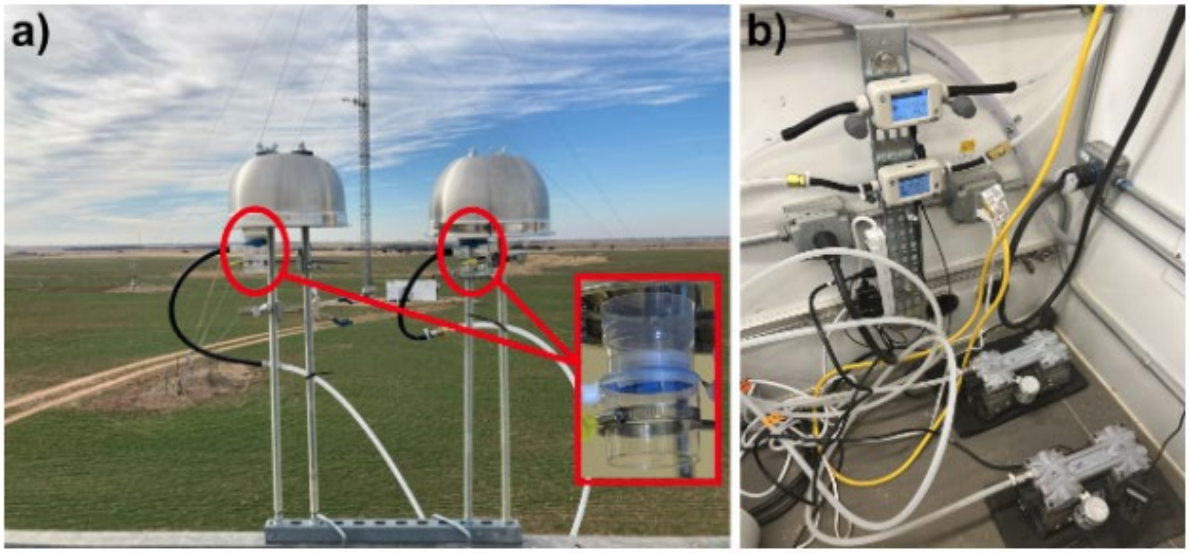

Figure 1. Filter unit sampling apparatuses, including a) single-use filter units under precipitation shields that are connected via tubing to b) the mass flow meters and vacuum pumps. The inset in a) shows a magnified photo of a filter unit. Photos in a) and b) are from ARM's Southern Great Plains (SGP) observatory in Oklahoma and the Surface Atmosphere Integrated Field Laboratory (SAIL) site in Colorado, respectively.

2.3 Filter Use and Preservation

Once sampling is complete, typically after 24 hours (option to collect between 2 to 72 hours, depending on total aerosol loadings at each site), the 0.2-µm filters containing the collected aerosol loadings are removed from the single-use filter unit and preserved frozen at approximately –20 °C on site in individual sterile Petri dishes. Batches of samples preserved in Petri dishes are transported frozen to CSU where they are further preserved frozen until processing.

2.4 INS Sample Processing

For resuspension of particles prior to measurement of INPs in the INS, filters are placed in sterile 50-mL polypropylene tubes, 7-10 mL (smaller amounts for filters from "cleaner" environments, to increase sensitivity) of 0.1 µm-filtered deionized (DI) water added, and particles resuspended by tumbling end over end on a rotator for 20 minutes. Each INS is constructed using two 96-well aluminum incubation blocks, designed for incubating polymerase chain reaction (PCR) plates, placed end to end and encased on their sides and base by cold plates (Figure 2a). Two INS instruments are placed side by side to double sample processing capacity (Figure 2b).

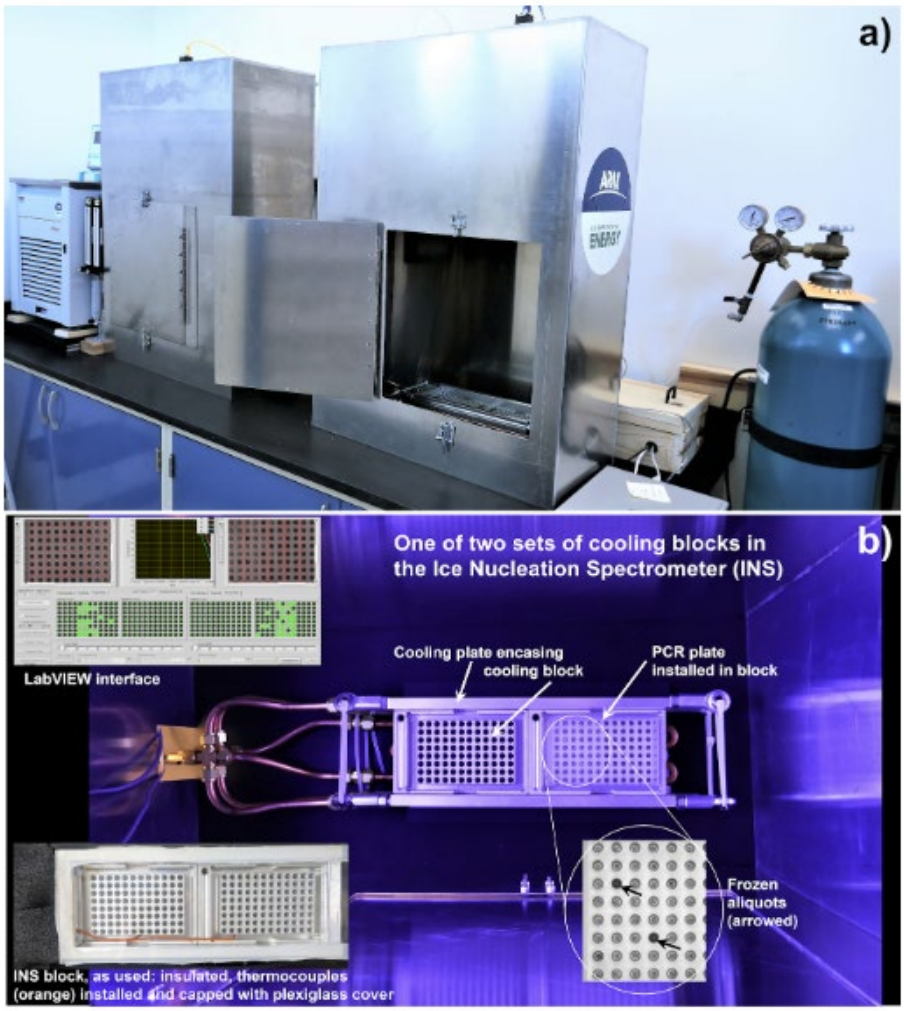

Figure 2. The INS, including: a) a photo of the exterior showing both cooling systems, one with the door open exposing the cooling blocks for scale and b) interior view of one system. PCR trays are loaded into the cooling blocks, the headspace purged with cooled and high-efficiency particulate air (HEPA)-filtered air, blocks cooled at 0.33 °C min^{-1}, and freezing events logged automatically through a LabVIEW interface with a camera.

Immersion freezing temperature spectra are obtained by dispensing 50-µL aliquots of aerosol suspensions into four sterile, 96-well PCR trays in a laminar flow cabinet. One INS run, therefore, processes 384 × 50 µL aliquots of suspension. There are typically 32 aliquots per level of dilution, and we use up to

five, 11- to 15-fold serial dilutions to cover the full temperature range/INP concentration. PCR plates are then placed into the blocks of the INS, the device covered with a plexiglass window, and the headspace purged with 750 mL min^{-1} of cooled and HEPA-filtered N_2. The device is cooled at 0.33°C min^{-1} using a recirculating low temperature bath, and the freezing of wells is recorded every 0.5 °C automatically through an interface with a charge-coupled device (CCD) camera system. Limit of measurement is between –27 and –29 °C, depending on INPs in the DI water used for resuspension (this baseline is subtracted before subsequent calculations).

2.5 INP Number Concentration Calculation

From the fraction of drops frozen and the known total volume of air filtered at each temperature interval, we can calculate INP concentration with a universally used equation (Vali 1971):

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

where f is the proportion of droplets frozen, V_{drop} is the volume of each drop, $V_{suspension}$ is the volume of the suspension, and V_{air} is the volume of air per sample (liters at standard temperature and pressure (STP): 0°C and 101.32 kPa).

2.6 Sample Treatments

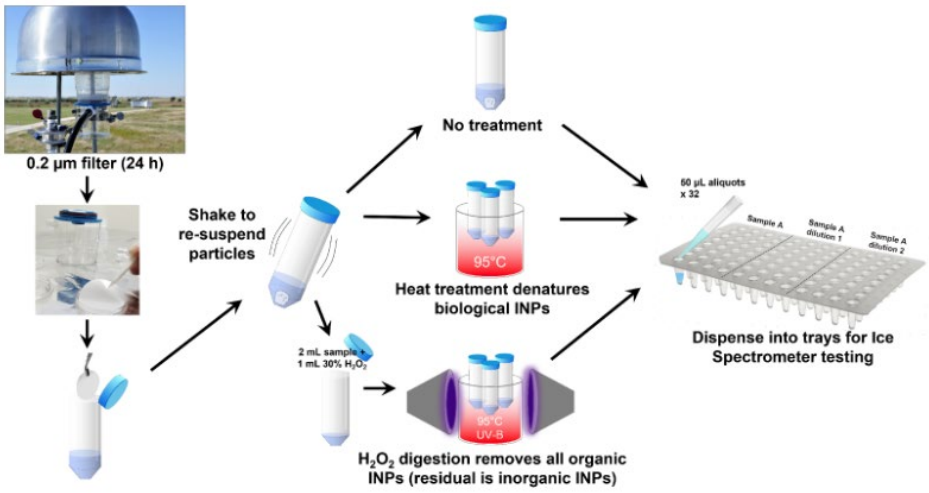

Figure 3. Schematic of workflow used to prepare samples for INP measurement in the INS.

Thermal treatments and peroxide digestions provide valuable insights into INP composition (see Figure 3). Heat treatments are performed by heating 2.5 mL of sample suspension to 95 °C for 20 min to denature heat-labile INPs, such as proteins. Peroxide digestions are performed on a further 2 mL of suspension in a solution containing 10% H_2O_2 (by adding 1 mL of 30% H_2O_2 in water, Sigma-Aldrich®) and heating to 95 °C for 20 min while illuminating with UV-B fluorescent bulbs to generate hydroxyl radicals. Following digestion, residual H_2O_2 is removed using catalase (MP Biomedicals™ catalase from bovine liver). This digestion removes all bio-organic INPs as described in detail in McCluskey et al. 2018, Suski et al. 2018, and Testa et al. 2021. The difference in the INP temperature

spectra after both treatments determines the influence of that INP type in the original sample, and the residual spectrum gives the inorganic/mineral INP component. Thus, this processing provides four key measures from each sample: total, heat-labile (i.e., biological), bio-organic, and inorganic (i.e., mineral) INP concentration. Treatments are typically applied to 1/3 of the total samples collected at any given location or during a given intensive operational period.

The temperature measurement range of the INS is between 0 and approximately –27 to –29 °C. The range of INP concentrations measurable depends mainly on total volume of air filtered. For a sampling period of 24 hours, and a total volume filtered of 27 m^{-3} (current average at SGP), the detection limit is 0.0002 INPs L^{-1}. There is no upper limit, since we use serial dilutions (typically 11-fold) to bring suspensions within range. INP concentrations may exceed 100 L^{-1} at -28 °C (e.g., at SGP).

2.7 Tethered Balloon System Sampling

INP sampling is also occasionally conducted on the ARM tethered balloon system (TBS) at select sites, based on user/principal investigator requests. The filter preparation, sampling, and pre-processing handling are slightly different as described here, while the sample and data processing remain the same as the standard filters collected at the sites.

The filter collection is conducted using a prototype miniaturized time-resolved sampler called the IcePuck (Handix Scientific, Inc.; Figure 4). The IcePuck has eight separate sampling channels controlled by individual mass flow controllers. It is modeled based on the National Oceanic and Atmospheric Administration's Continuous Light Absorption Photometer (Ogren et al. 2017), which was then modified into the home-built Time-Resolved Aerosol Particle Sampler (TRAPS) for balloon-borne filter collections of aerosols and INPs (Creamean et al. 2018). The IcePuck is an improved version of the TRAPS that is lighter and more user friendly. Its case is 3D printed, and it has an internally integrated miniature vacuum pump system. It is powered by an external 12-V battery used to supply power to other instruments on the TBS system.

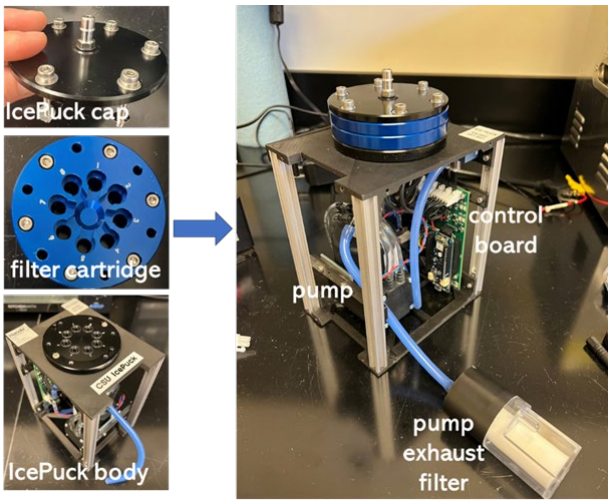

Figure 4. Images of some of the components of IcePuck and of the IcePuck assembled.

The removable and reusable aluminum filter cartridges have eight separate channels and sample spots to collect air sequentially at any time resolution defined by the user within the IcePuck programming code.

For TBS flights, ideally, new samples are collected at different altitude ranges. Thus, up to eight separate samples can be collected around the circumference of the same 47-mm filter. On average, TBS sampling ranges from 30 minutes to 2 hours per sample, depending on flight duration, typically affording 2-3 samples at different altitude ranges per flight.

Filter cartridges are prepared and loaded with pre-cleaned filters prior to deployment in a similar fashion to sterile filter units. The cartridges contain the same 0.2-µm polycarbonate filters backed with 10-µm polycarbonate filters (47-mm diameter Whatman® Nuclepore™ Track-Etched Membranes). The cartridges are sonicated in Windex® then in DI water to remove potential background contaminants. Sample and backing filters are carefully loaded into the base of the cartridge; then the cartridge lid is secured on top of the filters, only exposing the eight sample spots. Once loaded, cartridges are carefully wrapped in foil and stored in sterile bags until use.

Immediately prior to TBS flights, a pre-cleaned and pre-loaded filter cartridge is secured to the inlet manifold of the IcePuck. The programming is pre-set and IcePuck is started once airborne. The first sample is typically the ascent (< 30 minutes) and is not used for analysis. Flow rates are typically 0.5-1 Slpm through the sample and backing filters at any given altitude. Colder temperatures tend to decrease the flow rate to the lower end of this range. After collection during flights, the entire cartridge is removed, recovered with fresh foil, and stored frozen in a new bag until sample processing. Prior to processing on the IS, the filter sample spots are cut out of the main 47-mm filter, in addition to 1-3 sample blanks from non-sample spot locations on the filter. Each sample spot is then processed in the same manner as the main site filters.

3.0 Data

3.1 Data Description

The primary measurement output of the INS is freezing temperature spectra of cumulative immersion-mode INP number concentrations in aerosols resuspended from individual filter samples, calculated in the units of # per L of air. Note that INPs are also interchangeably called ice nuclei (IN).

Output data from the INS include freezing temperature (°C), INP number concentration (L^{-1} STP, with STP being 0 °C and 101.32 kPa), upper and lower 95% CI values, and treatment flag. The treatment flag indicates if the suspension was untreated to obtain total INP number concentrations, heat-treated to deactivate biological (e.g., proteinaceous) INPs, or H_2O_2-treated to remove all organic INPs. These are measured and/or calculated from preliminary output data files that contain date and time of processing, freezing temperature, and number of wells frozen (typically out of an array of thirty-two, each containing a 50-uL aliquot of resuspended aerosol) per 0.5 °C interval. Images of the unfrozen and frozen wells acquired throughout INS runs are obtained for visual quality control of the output data files.

INP number concentration data from the INS sample processing can be accessed and ordered on the ARM website. Data can be found by searching for the measurement “ice nuclei” or data source of “ice nucleation spectrometer”. “Ice spectrometer” is also commonly used as an alternative to “ice nucleation spectrometer” for the data product description. INS data available on the ARM website include intensive operational period data from previous campaigns in addition to routine INP mentor data.

3.2 Data Quality and Uncertainty

A flow diagram of our quality assurance/quality control (QA/QC) protocols is illustrated in Figure 5. These are described in detail in the follow sections.

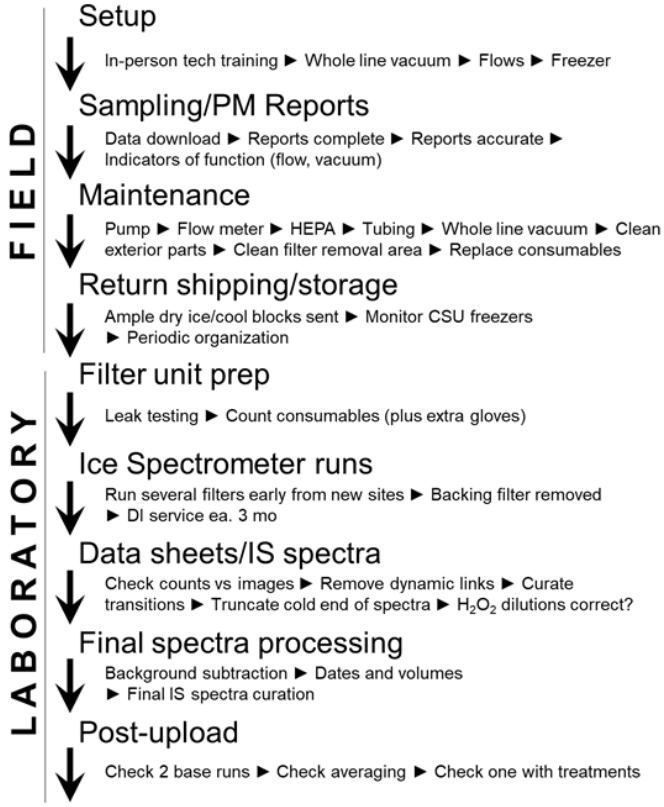

Figure 5. Flow diagram of QA/QC protocols designed for INPs. Quality assurance ensures quality requirements are fulfilled for both ARM management and end users. Quality control maintains quality requirements via inspection and testing to ensure performance characteristics conform to pre-specified requirements.

3.2.1 Filter Sample Collections

Collection of filter samples for offline INS processing are quality-controlled through monitoring the in-line pressure (kPa) and flow rate (Slpm) at the start and end times of filter collection. These values are used to determine if sampling errors occurred during collection (e.g., a significant change in pressure and/or flow rate may indicate a leak in the filter unit, tubing, or other connections throughout the system). To ensure accurate measurement of total volumes filtered, because flow rates decrease over the sampling period, we use a totalizing mass flow meter (TSI 5200-1, accuracy ±2%) updating every second.

TSI 5200 mass flow meters are checked against a TSI 5230 meter, which has enhanced 1.7% accuracy, and is reserved for this purpose. If TSI 5200 units are more than 5% adrift they are returned to TSI for servicing and calibration.

3.2.2 INS Processing

To minimize contamination of the filters with INPs on laboratory surfaces or in consumables (e.g., tubes used for filter processing, pipet tips, PCR plates), and to ensure that the DI water used for re-suspension of particles from filters is as INP-free as possible, we apply a comprehensive protocol during preparation of the samples for measurement of INPs in the INS (Barry et al. 2021). Pipets are calibrated annually. A 0.1-µm filtered DI water blank is included in each INS run, to correct for INPs present in PCR trays and the DI water used for re-suspension. Blanks (using DI water in place of the sample re-suspension) are also run for H_2O_2 digestions to check for any INP contamination in the hydrogen peroxide and catalase used in the procedure.

For accurate temperature measurement in the INS, thermocouples are inserted just below the wells, via a horizontally drilled hole plugged with thermal grease. There is one thermocouple in each of the four PCR blocks. For each set of double blocks, readings from the two thermocouples are averaged. The HEPA-filtered N_2 that purges the headspace above the PCR trays is precooled to a few degrees above the block temperature to prevent it warming the aliquots dispensed into the trays.

In addition to automated detection of well freezing, camera images are taken every 20 seconds, or approximately every 0.1 °C, to check for program errors. Checking images against automated detection output is performed on every IS run.

The temperature uncertainty in the INS technique is ± 0.2 °C (a combination of thermocouple uncertainty and temperature variation across the blocks due to gradients in cooling). Accuracy is also controlled by the inherent uncertainty in using count data of frozen wells. Binomial sampling confidence intervals (CI, 95%) are derived following Agresti and Coull (1998) to estimate the uncertainty in INP number concentrations, and their ranges vary according to the proportion of wells frozen. For a single well frozen (out of 32), the 95% CI ranges from ~0.2 to ~5.5 times the estimated INP concentration, while for 16 of the 32 wells frozen it ranges from ~0.6 to ~1.6 times.

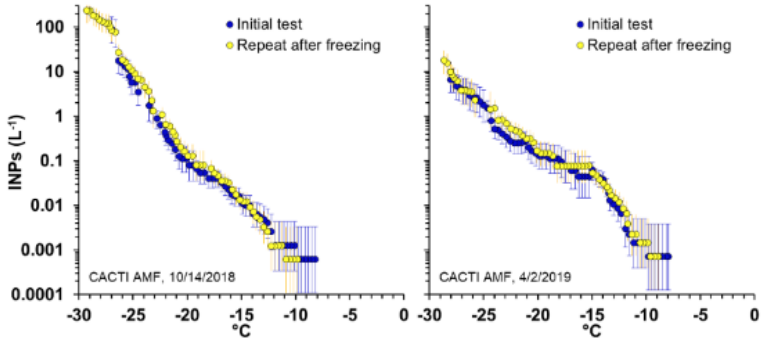

Figure 6. Repeatability of INP tests using suspensions that were tested immediately after re-suspension of particles form filers, then re-tested after frozen storage of the suspension (tubes were briefly pulse-vortexed after thawing). Vertical bars are 95% confidence intervals.

On several occasions, in previous campaigns, we have confirmed the repeatability of measures within the range inherent to the method (see section 7.3). Two examples are given in Figure 6, from filter samples

taken in ARM's Cloud, Aerosol, and Complex Terrain Interactions (CACTI) field campaign in Argentina. We have also tested replicate filters to confirm their comparability (Figure 7).

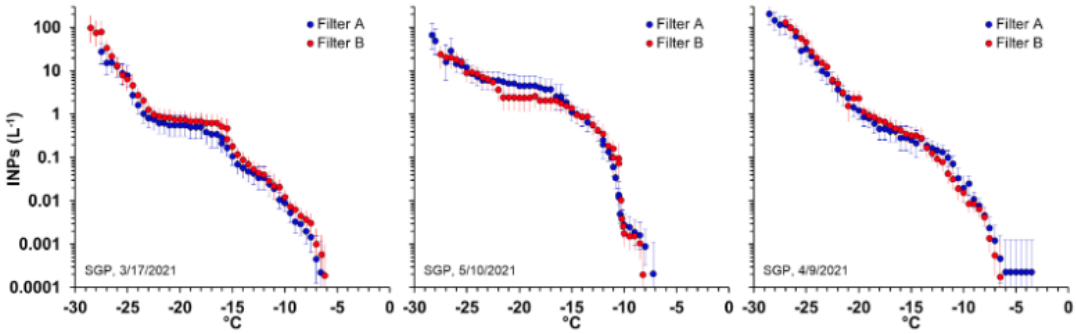

Figure 7. Comparability of INP spectra obtained from replicate filters taken at SGP. Vertical bars are 95% confidence intervals.

3.2.3 Control Blank Sample Collection and Processing

Field filter unit blanks are prepared identically to the sampling filters and exposed briefly to air at the sampling position to monitor possible contamination during filter sample preparation and handling. These samples are preserved and processed in the same manner as the collected aerosol filter samples to obtain a mean background INP spectrum. Filter unit and DI water control blank tests are conducted routinely to obtain background INP spectra for calibration of the measured INP spectra from the aerosol filter samples (Figure 8).

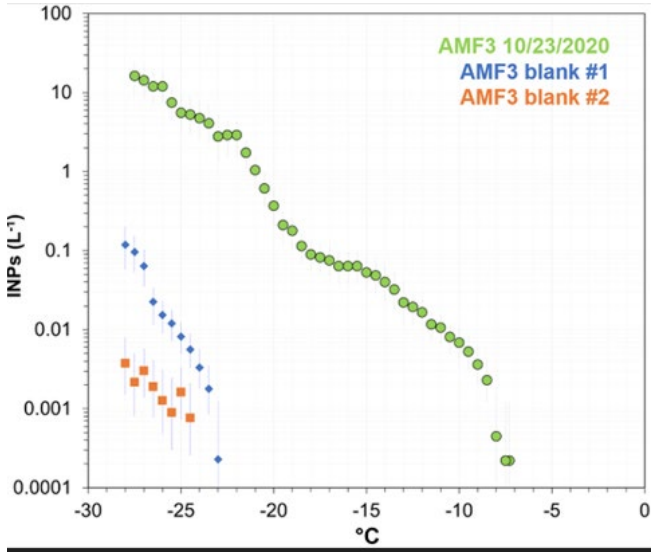

Figure 8. Example of a 24-hour sample from the third ARM Mobile Facility (AMF3; Oliktok Point, Alaska) on 10/23/2020 and two blank filters collected at the same site.

3.3 Examples of Data

A plot containing representative INS output data is shown in Figure 9. These data include cumulative freezing spectra of untreated (total) INP number concentrations, heat-treated (e.g., proteinaceous) INP number concentrations, and organic INP number concentrations from a peroxide treatment. INPs left after peroxide digestion are inorganic (e.g., mineral) INPs. Data are from the CACTI campaign.

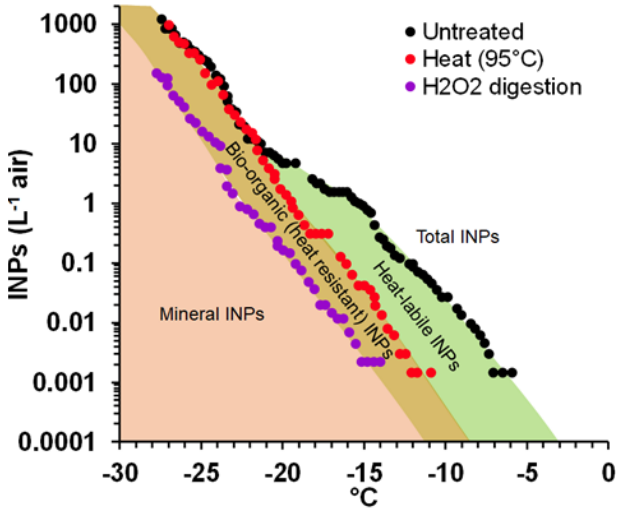

Figure 9. Cumulative INP spectra from a filter collected during the ARM CACTI campaign. Spectra include total INPs in addition to unique treatments that differential heat-labile (proteinaceous), heat-resistant (bio-organic), and mineral INP concentrations.

4.0 Historical Background

The formation and microphysical modulation of cloud droplets and ice crystals are highly dependent upon aerosols that serve as cloud condensation nuclei (CCN) and INPs. In general, INP observations are limited relative to other aerosol properties, yet central to elucidating the role of aerosols in cloud formation, and subsequent cloud microphysical and radiative properties. Immersion freezing, whereby an INP first serves as a CCN, then freezes at temperatures above homogeneous freezing (–38 ˚C) (Kanji et al. 2017, Murray et al. 2012), is a particularly important glaciation process for the formation and impacts of mixed-phase clouds. The INS mimics immersion freezing of cloud ice through ambient aerosols serving as INPs by way of heterogeneous ice nucleation. This technique provides quantitative information on the population of ambient aerosols that can facilitate cloud ice formation at a wide range of subzero temperatures.

The efficacy of an aerosol to serve as an INP largely depends on temperature and vapor saturation with respect to water and ice, in addition to its composition (chemical, mineral, or biological makeup), morphology, and size, and thus, its source (Hoose and Möhler 2012). Aerosols such as mineral dust, soil dust, sea spray, volcanic ash, black carbon, and biologically derived particles (e.g., intact or fragmented bacteria, pollen, fungal spores, lichens, algae, diatoms, soil organic matter, fatty acids, proteins, and other macromolecules) have been shown to serve as INPs (e.g., Conen et al. 2011, Creamean et al. 2013, 2019, 2020, Cziczo et al. 2017, DeMott et al. 1999, 2016, 2017, 2018a, 2018b, Hill et al. 2016, Huang et al. 2021, Kaufmann et al. 2016, Levin et al. 2010, McCluskey et al. 2017,

O'Sullivan et al. 2014, 2016). Among the natural sources, mineral dust and biologically derived particles are arguably the most crucial INPs found in the atmosphere. Mineral dust is an abundant INP, forming ice primarily at temperatures < −15 °C, while classes of biological particles such as certain bacteria are capable of initiating freezing up to −1.5 °C (Després et al. 2012, Fröhlich-Nowoisky et al. 2016, Huang et al. 2021, Vali et al. 1976). Measuring total INPs, in addition to their biological and mineral components, yields critical information on INP abundances and sources.

While basic immersion freezing methods have been applied for decades, recent intercomparison studies (DeMott et al. 2017, 2018b) with other methods for sampling ambient INPs support the method's utility. Use of the INS with filters produces spectra spanning wide dynamic ranges of temperature and, hence, INP concentration (e.g., six orders of magnitude). The INS is supported with well-established experimental protocols and has been applied in many diverse scenarios (e.g., Beall et al. 2017, DeMott et al. 2017, Hill et al. 2016, Hiranuma et al. 2015, McCluskey et al. 2017, 2018, Suski et al. 2018).

5.0 Maintenance Plan

The following maintenance procedures are required for the filter unit apparatuses:

- Check in-line temperature, pressure, and flow rate at the start of sample collection.
- Check in-line temperature, pressure, and flow rate at the end of sample collection.
- Clean and clean precipitation shields, as needed.
- Check for leaks in single-use filter units before use.
- Check for leaks/obstructions in vacuum tubing and connection points, as needed.
- Check TSI 5200 mass flow meters (against TSI 5230, which has enhanced 1.7% accuracy, and is reserved for this purpose) annually, and return for servicing if required.
- Check performance of Thomas 2688CE44 pumps using TSI flow meters. When operating efficiently these pumps will be capable of producing a 0.5 kPa vacuum.

These maintenance procedures are required for the INS:

- Clean plexiglass lids with Windex and DI water every two weeks.
- Deep-clean laboratory space with Windex and Kimwipes once per month.
- Check copper piping for SYLTHERM™ XLT heat transfer fluid leaks.
- Watch rate of N_2 tank depletion for leaks.

6.0 User Notes and Known Issues

Chemicals including 30% H_2O_2 and catalase for the peroxide treatment processing, methanol for preparing sterile filters, and SYLTHERM™ XLT heat transfer fluid for the INS coolant are all used during filter collection and processing. These chemicals can be flammable and/or toxic.

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