

SPICULE GV Ice Spectrometer measurements

Paul J. DeMott
Senior Research Scientist
Colorado State University
Department of Atmospheric Science
1371 Campus Delivery, Fort Collins, CO 80526
Paul.Demott@colostate.edu
<http://chem.atmos.colostate.edu/PJDeMott/>
<http://orcid.org/0000-0002-3719-1889>

Co-author: Thomas C. J. Hill
Research Scientist II
Colorado State University
Department of Atmospheric Science
1371 Campus Delivery, Fort Collins, CO 80526
Thomas.Hill@colostate.edu
<https://orcid.org/0000-0002-5293-3959>

Co-author: Ryan Patnaude
Graduate Research Assistant
Colorado State University
Department of Atmospheric Science
1371 Campus Delivery, Fort Collins, CO 80526
ryan.patnaude@colostate.edu
<https://orcid.org/0000-0003-3129-8279>

1.0 Data Set Overview

These measurements were part of the Secondary Production of Ice in Cumulus Experiment (SPICULE). The goal of the SPICULE campaign is to use in situ and remote sensing techniques to better understand the microphysical and dynamical parameters supportive of suspected Hallet-Mossop (HM) secondary ice process (SIP) in cumulus clouds. PI DeMott's group is focused on the question of interaction between clouds and aerosols in developing cumulus clouds and deployed instrumentation for measuring ice nucleating particles (INPs). This archive relates to the Colorado State University Ice Spectrometer (IS) instrument that utilized filters collected onboard the NSF/NCAR GV during SPICULE to measure the immersion freezing temperature spectra of ambient particles. Collections were from the HIAPER Modular Inlet (HIMIL), mounted on the front port side of the GV, adjacent to the rack that held filter holders. The IS was used to measure INP number concentrations from filters collected during approximately level-leg transects in the boundary layer to determine INP relations to investigate cloud processing of INPs. Flights were based from Broomfield, Colorado. The time period covered is May 29 to June 25, 2021. A total of 10 research flights were done with filter collections at latitudes between 33.7 and 45.2 degrees and longitudes -105.2 to -94.8 degrees, and a maximum altitude of 13123 meters.

2.0 Instrument Description:

The Colorado State University Ice Spectrometer (IS) emanates from the developments of Hill et al. (2014; 2016) and is described in the approximate present form by Hiranuma et al. (2015) and DeMott et al. (2018). Immersion freezing temperature spectra are obtained in the IS following dispensing 32 or 48 aliquots of 50 μL suspensions of aerosols into sterile wells that are isolated in a cooled device that is purged by ultra-clean nitrogen gas. Temperature is lowered at $0.33^\circ\text{C min}^{-1}$ and frozen wells are counted at $0.2\text{-}1^\circ\text{C}$ degree intervals to a limit of approximately -29°C . Cumulative numbers of INPs mL^{-1} of suspension are estimated on the basis of Vali (1971) using,

$$c_{IN}(T) = -\frac{\ln(f_{unfrozen}(T))}{V_{drop}} \quad (1)$$

where $c_{IN}(T)$ is the concentration of INPs per unit volume of water (mL^{-1}), $f_{unfrozen}$ is the fraction of unfrozen drops at T , and V_{drop} is the population-median drop volume. Volumetric INP concentrations in air ($C_{INP}(T)$) are calculated via,

$$C_{INP}(T) = \frac{c_{IN} \cdot f \cdot V_{imp}}{V_a} \quad (2)$$

where V_{imp} is the total volume rinsed from a collection filter (mL), f accounts for any dilution of the suspension ($f = 1$ for undiluted), and V_a is the air volume collected into liquid (L). Seven filter blanks were collected and processed in a similar manner as aerosol samples to obtain a mean background INP spectrum.

Filter samples were obtained on the GV using 47-mm diameter in-line aluminum filter holders fitted with a $0.2 \mu\text{m}$ diameter pore Nuclepore polycarbonate membranes. These were located within the CFDC instrument rack. Sampling was from the HIMIL (same as used for the CFDC) at all times. For re-suspension of particles, filters are placed in sterile pre-rinsed 50 mL Falcon polypropylene tubes and 8 mL of suspension solution ($0.1 \mu\text{m}$ filtered deionized water) is added. Particles are re-suspended by tumbling end-over-end at $60 \text{ cycles min}^{-1}$ for 20 min. Ultra-filtered water used in experiments contained on average 1.2 INPs mL^{-1} at -25°C , and this sets typical lower detection limits. Correction is then applied for the number of INPs processed from rinsed filter blanks that were carried onto the GV during several flights.

A filter rinse produces sufficient volume for standard processing as described above, and alternate treatments prior to freezing tests to give inference to INP compositions, which will be provided in a later update. These treatments include thermal exposure of suspensions at 95°C for 20 minutes to destabilize biological ice nucleating proteins (typically destroying their activity as INPs while not affecting inorganic INPs) or digestion of all organic carbon through application of hydrogen peroxide (Suski et al., 2018). The latter test distinguishes total organic versus inorganic INPs.

3.0 Data Collection and Processing:

IS filter collections represent integrated volumes collected over extended times of sampling at standard flow rates that varied by altitude from several to 15 lpm. Oftentimes, collections included multiple passes at altitudes and locations considered representative of the environmental air seeding the developing cumulus clouds.

The primary data are INP concentrations as a function of temperature and the 95% negative and positive confidence widths. Binomial sampling confidence intervals (95%) are derived for all data using the formula (no. 2) recommended by Agresti and Coull (1998):

$$CI_{95\%} = \left(\hat{p} + \frac{1.96^2}{2n} \pm 1.96 \sqrt{\left[\hat{p}(1 - \hat{p}) + \frac{1.96^2}{4n} \right] / n} \right) / \left(1 + \frac{1.96^2}{n} \right) \quad (3)$$

where \hat{p} is the proportion of droplets frozen and n is the total number of droplets. Using this formula, a single well frozen out of 32 aliquots the $CI_{95\%}$ ranges from 18% to 540% of the estimated INP concentration, while for 16/32 wells frozen it is 68-132% of the INP concentration.

4.0 Data Format:

IS data are reported in non-standard ICARTT format to account for the fact that filters were often integrated over multiple passes at different locations to increase sample volumes. Bounding times and midpoint latitudes and longitudes for each pass are provided for each filter collection. The primary variables and units are given in the data file header but are repeated here. Processed data appear as arrays, led by a list of secondary overall filter start and stop times (not continuous sampling), location and altitude variables. Additionally, a flag is included for sample treatment on the liquid suspension. The list of variables and units are given in the data file header but are repeated here.

Time_Start_Leg1, seconds, Time_Start, seconds_past_midnight_UTC

Time_Stop_Leg1, seconds, Time_Stop, seconds_past_midnight_UTC

Time_Mid_Leg1, seconds, Time_Mid, seconds_past_midnight_UTC

LAT_Leg1, degree, Platform_Latitude_InSitu_None, Midpoint aircraft latitude during Leg1

LON_Leg1, degree, Platform_Longitude_InSitu_None, Midpoint aircraft longitude during Leg1

ALT_Leg1, meter, Platform_AltitudeMSL_InSitu_None, Midpoint aircraft altitude during Leg1

Time_Start_Leg2, seconds, Time_Start, seconds_past_midnight_UTC

Time_Stop_Leg2, seconds, Time_Stop, seconds_past_midnight_UTC

Time_Mid_Leg2, seconds, Time_Mid, seconds_past_midnight_UTC

LAT_Leg2, degree, Platform_Latitude_InSitu_None, Midpoint aircraft latitude during Leg2

LON_Leg2, degree, Platform_Longitude_InSitu_None, Midpoint aircraft longitude during Leg2

ALT_Leg2, meter, Platform_AltitudeMSL_InSitu_None, Midpoint aircraft altitude during Leg2

Time_Start_Leg3, seconds, Time_Start, seconds_past_midnight_UTC

Time_Stop_Leg3, seconds, Time_Stop, seconds_past_midnight_UTC

Time_Mid_Leg3, seconds, Time_Mid, seconds_past_midnight_UTC

LAT_Leg3, degree, Platform_Latitude_InSitu_None, Midpoint aircraft latitude during Leg3

LON_Leg3, degree, Platform_Longitude_InSitu_None, Midpoint aircraft longitude during Leg3

ALT_Leg3, meter, Platform_AltitudeMSL_InSitu_None, Midpoint aircraft altitude during Leg3

Time_Start_Leg4, seconds, Time_Start, seconds_past_midnight_UTC
Time_Stop_Leg4, seconds, Time_Stop, seconds_past_midnight_UTC
Time_Mid_Leg4, seconds, Time_Mid, seconds_past_midnight_UTC
LAT_Leg4, degree, Platform_Latitude_InSitu_None, Midpoint aircraft latitude during Leg4
LON_Leg4, degree, Platform_Longitude_InSitu_None, Midpoint aircraft longitude during Leg4
ALT_Leg4, meter, Platform_AltitudeMSL_InSitu_None, Midpoint aircraft altitude during Leg4
Time_Start_Leg5, seconds, Time_Start, seconds_past_midnight_UTC
Time_Stop_Leg5, seconds, Time_Stop, seconds_past_midnight_UTC
Time_Mid_Leg5, seconds, Time_Mid, seconds_past_midnight_UTC
LAT_Leg5, degree, Platform_Latitude_InSitu_None, Midpoint aircraft latitude during Leg5
LON_Leg5, degree, Platform_Longitude_InSitu_None, Midpoint aircraft longitude during Leg5
ALT_Leg5, meter, Platform_AltitudeMSL_InSitu_None, Midpoint aircraft altitude during Leg5
Num_Temps, unitless, none, Number of temperatures in data record
Total_Vol, liters, Total_Volume_Filtered, Total volume of air passed through filter (ambient temperature and pressure)
IS_Temp_C[], degrees Celsius, AerMP_INP_Insitu_Bulk_NumConcSTP, Measurement temperature of IS
IS_N_INP[], number per liter, AerMP_INP_Insitu_Bulk_NumConcSTP, Number of ice nucleating particles per liter of air (ambient temperature and pressure)
IS_Lower_CI[], number per liter, AerMP_INP_Insitu_Bulk_NumConcSTP, 95% lower confidence interval width array for number of ice nucleating particles per liter of air (ambient temperature and pressure)
IS_Upper_CI[], number per liter, AerMP_INP_Insitu_Bulk_NumConcSTP, 95% upper confidence interval width array for number of ice nucleating particles per liter of air (ambient temperature and pressure)
IS_Treatment_Flag[], unitless, Treatment_Flag, 0=Untreated, 1=Heat Treated (95 degrees C for 20 minutes), 2=Peroxide Treated (10% H2O2 at 95 degrees C for 20 minutes under UV-B, then H2O2 decomposed with catalase)

The file names archived as “preliminary” are:

SPICULE-IS_20210529_R0_RF01-F1.ict
SPICULE-IS_20210601_R0_RF02-F1.ict
SPICULE-IS_20210601_R0_RF02-F2.ict
SPICULE-IS_20210602_R0_RF03-F1.ict
SPICULE-IS_20210602_R0_RF03-F2.ict
SPICULE-IS_20210605_R0_RF04-F1.ict
SPICULE-IS_20210605_R0_RF04-F2.ict
SPICULE-IS_20210609_R0_RF05-F1.ict
SPICULE-IS_20210611_R0_RF06-F1.ict
SPICULE-IS_20210617_R0_RF07-F1.ict
SPICULE-IS_20210620_R0_RF08-F1.ict
SPICULE-IS_20210621_R0_RF08-F2.ict
SPICULE-IS_20210624_R0_RF09-F1.ict
SPICULE-IS_20210625_R0_RF10-F1.ict

SPICULE-IS_20210625_R0_RF10-F2.ict

Final file versions will have a different version number (Rx). Values that are below the detection limit of the IS are reported as -8888, and any missing values are reported as -9999.

5.0 Data Remarks

The complex and integrated nature of the IS samples has been discussed above. Due to the limited volumes of samples compared to fixed-site collections that typically last for 24 hours or more, background corrections are significant for this data set. Therefore, caution should be exercised when using this dataset, and it is encouraged to reach out to the PI before use.

6.0 References

Agresti, A., and Coull, B. A.: Approximate Is Better than "Exact" for Interval Estimation of Binomial Proportions, *The American Statistician*, 52, 119-126, 10.2307/2685469, 1998.

DeMott, P. J., et al.: The Fifth International Workshop on Ice Nucleation phase 2 (FIN-02): laboratory intercomparison of ice nucleation measurements, *Atmos. Meas. Tech.*, 11, 6231-6257, <https://doi.org/10.5194/amt-11-6231-2018>, 2018.

Hill, T. C. J., Moffett, B. F., DeMott, P. J., Georgakopoulos, D. G., Stump, W. L., and Franc, G. D.: Measurement of Ice Nucleation-Active Bacteria on Plants and in Precipitation by Quantitative PCR, *Applied and Environmental Microbiology*, 80, 1256-1267, 10.1128/aem.02967-13, 2014.

Hill, T. C. J., DeMott, P. J., Tobo, Y., Fröhlich-Nowoisky, J., Moffett, B. F., Franc, G. D., and Kreidenweis, S. M.: Sources of organic ice nucleating particles in soils, *Atmos. Chem. Phys. Discuss.*, 2016, 1-37, 10.5194/acp-2016-1, 2016.

Hiranuma, N., and coauthors: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, *Atmos. Chem. Phys.*, 15, 2489–2518, doi:10.5194/acp-15-2489-2015, 2015.

Suski, K. J., Hill, T. C. J., Levin, E. J. T., Miller, A., DeMott, P. J., and Kreidenweis, S. M.: Agricultural harvesting emissions of ice-nucleating particles, *Atmos. Chem. Phys.*, 18, 13755-13771, <https://doi.org/10.5194/acp-18-13755-2018>, 2018.

Vali, G.: Quantitative Evaluation of Experimental Results on the Heterogeneous Freezing Nucleation of Supercooled Liquids, *Journal of the Atmospheric Sciences*, 28, 402-409, 10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2, 1971.