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Macquarie Island Cloud and Radiation Experiment (MICRE) Ice Nucleating Particle Measurements Field Campaign Report

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Acronyms and Abbreviations

AAD	Australian Antarctic Division
ACRE	Antarctic Clouds and Radiation Experiment
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
BOM	Australian Bureau of Meteorology
CSIRO	Commonwealth Scientific and Industrial Research Organization
CSU	Colorado State University
DOE	U.S. Department of Energy
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
MSL	mean sea level
NSF	National Science Foundation
SO	Southern Ocean
SSA	sea spray aerosol

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1.0 Summary

The U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility Macquarie Island Cloud and Radiation Experiment (MICRE) Ice Nucleating Particle Measurements campaign was aligned as a supplemental activity with the second year of the overall MICRE campaign. The purpose of the supplemental campaign was to provide a first-ever annual cycle of ice nucleating particle (INP) measurements at a Southern Ocean (SO) site, toward the investigation/characterization of sources of INPs in the marine boundary layer as being from the production or primary sea spray aerosol (SSA) emissions versus other long-range transport influences. Furthermore, measuring the variability in INPs seasonally in this region, as this may relate to meteorology and ocean biological processes, is expected to help constrain (via parameterizations) numerical model simulations of cloud phase and regional climate. In concert with other SO studies during the time frame of the campaign (March 2017 to March 2018), MICRE sought to evaluate the hypotheses that SSA INPs represent the major INP source in the SO and, due to their generally less efficient INP activation properties compared to terrestrial sources, that they may directly influence the persistence of supercooled clouds in a unique manner for this climatic region.

These measurements supplemented the original deployment of primarily radiation measurements provided in the ARM MICRE request and the associated Australian Antarctic Division (AAD) Antarctic Clouds and Radiation Experiment (ACRE). Assistance and collaboration was obtained from the AAD, the Commonwealth Scientific and Industrial Research Organization (CSIRO) and the Australian Bureau of Meteorology (BOM). Follow-up plans will include investigations with their additional aerosol, cloud, and radiative transfer measurements. AAD also saw to the transport to Australia and storage of filters at the end of the campaign. The added effort involved an average of twice-weekly collections (a 48-hour and a 72-hour collection) of pre-sterilized, open-faced, polycarbonate filter samples for post-processing of INP number concentrations and tests to analyze the nature of INP sources. Collections were made by AAD technician Emry Thaggard Crocker after online training from our team. Images of the filter sampler and site are shown in Figure 1.

Sampling occurred from an approximate elevation of 4m MSL. Filters were returned frozen to Colorado State University (CSU) at the end of the campaign. Initial processing to obtain INP temperature spectra (0 to -28°C) of the concentrations of INPs active via the immersion freezing mechanism were conducted using the CSU ice spectrometer (IS) instrument systems (Hiranuma et al. 2015; McCluskey et al. 2018). In this method, particles are first removed from filters using ultrapure water while under mechanical agitation. Arrays of the particle suspensions from a portion of filter rinses are then cooled to obtain immersion freezing data per volume of water, and finally are related to sampling volumes to obtain numbers per volume of air following the analysis methods of Vali (1971). A total of 103 filters were collected, including blanks at selected intervals throughout the project. For archival and completion of tasks under this ARM proposal, nearly half of the filter collections (46) were processed for basic temperature spectra, and half of these (23) were also tested for microbial/proteinaceous contributions toward INPs via 95°C pre-treatment of portions of the original suspensions.

In this type of analysis, we inspect for removal of INP activity by the thermal treatment as an indication of the presence of such biological INPs (McCluskey et al. 2018a, b). We note that eight of the heated samples suffered a contamination issue at the point of processing that was related to improper laboratory

cleaning protocol by summer interns working on another study. These lost sample analyses will be supplemented through processing additional samples under ASR funding, as described below. All data have been added to the ARM Data Center.

The MICRE study provides the annual context to other ARM support for our participation in the Measurements of Aerosols, Radiation and Clouds over the Southern Ocean (MARCUS) project. The latter project provides a more seasonal and spatial investigation of INP influences using the same methods as used for MICRE. Additional full processing of the remaining MICRE filters (and MARCUS filters) is now occurring under a proposal funded by the DOE Atmospheric Systems Research (ASR) program. This will include:

- additional use of frozen particle suspensions for thermal processing of more samples
- similar chemical treatments using hydrogen peroxide to remove organic components prior to freezing analyses (McCluskey et al. 2018; Suski et al. 2018), thereby revealing organic versus inorganic INPs
- total organic carbon and nitrogen analyses
- ionic aerosol chemistry analyses, and
- genetic analyses of biological community diversity in aerosols.

This ASR study (Contract DE-SC0018929) also supports collaborative studies with other ARM (Marchand), AAD (Alexander), and BOM collaborators toward remote-sensing analyses of aerosol and cloud properties. These further analyses will permit normalization of INP data versus aerosol surface area, a critical step toward development of model parameterizations, and exploring INP relations to supercooled cloud properties over the site.

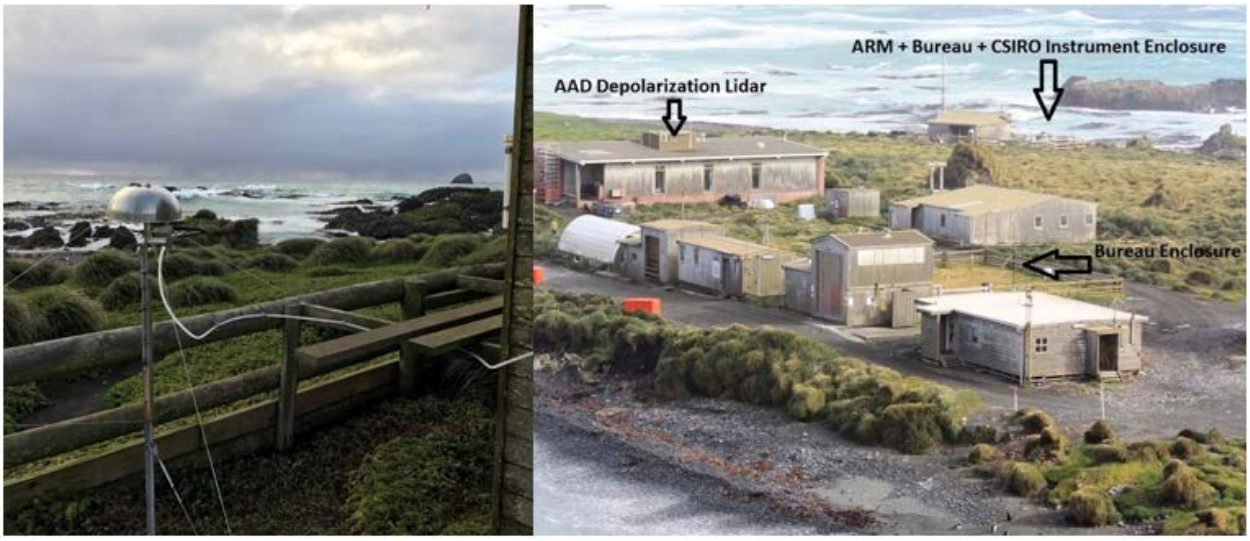

Figure 1. View of filter sampler site at the Macquarie Island station (-54.499699 lat, 158.934406 long). The sampler (left) was located inside the "ARM+Bureau+CSIRO" instrument enclosure (right), and within a short distance of the shoreline under prevailing winds.

2.0 Results

Results are at an early stage of evaluation, as final shipment of samples did not occur until well into May 2018 and processing of the first level of samples was not completed until October 2018. Nevertheless, the uniqueness and strong potential of the new INP data set are demonstrated already in Figure 2. Variation of INP concentrations at any temperature range over an order of magnitude or more. Distinct maxima in INP concentrations occurs at some temperatures, especially in the -15 to -20°C range, during the winter season, with minima in the spring and summer months. This is the first time such a cycle has been observed.

Since this report is being prepared at the same time as the MARCUS INP project report, it is also instructive to check for consistency between the MARCUS and MICRE data sets. This is important due to possible concerns about the representativeness of the island site for the surrounding oceans. A first inspection of this issue in shown in Figure 3. There, MARCUS data from a few-degree latitude range around Macquarie Island is plotted along with MICRE data collected in the time frame of MARCUS voyages. A general correspondence of data from the two studies is seen, especially at more modestly supercooled temperatures, although higher INP concentrations are evident at the lowest temperatures in the MICRE data set. This will be the subject of further investigation, as the specific influence only at lower temperatures could reflect either enhanced production of aerosol surface and lower temperature INPs due to wave breaking in the island surf zone, or it could potentially reflect the influence of higher biological productivity in the near-shore regions. DNA analyses in comparison to open oceans will aid this study.

We have also investigated (not shown) the influence of thermal treatments performed thus far on the MICRE samples. In contrast to MARCUS, a non-heat-labile INP population appears to dominate at Macquarie Island over most times, at least below -20°C.

The MICRE INP data set, in concert with MARCUS data, represent an unprecedented and rich database for investigating the life cycle of INPs in the remote SO region. Aerosol properties will be more challenging to characterize in MICRE, since a large aerosol suite was not located on the island. Through collaborations with Australian group colleagues, we will have total particle number concentrations, and we will use the sun photometer and BOM lidar to explore retrieval of aerosol concentration and surface areas during occasional clear-sky periods. Chemical and biological analyses of aerosols funded under the DOE ASR contract could provide additional clues to aerosol property differences at times.

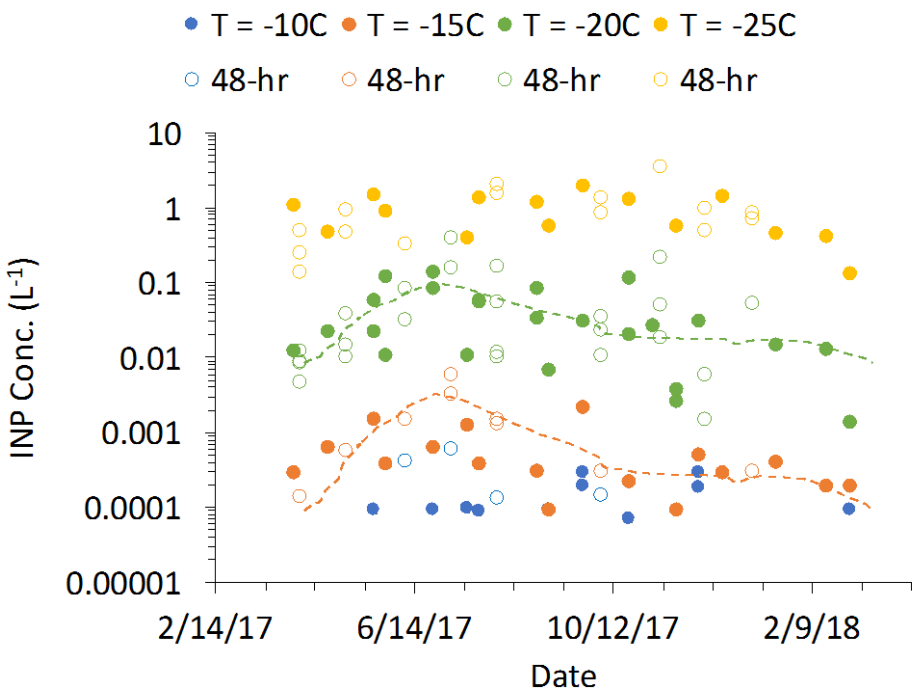

Figure 2. Initially processed archive of INP concentration versus date to capture the annual cycle of INPs at Macquarie Island. Filled data points are 72-hour collections while open points are 48-hour samples. While not consistent across temperatures, there appears a winter maximum at moderate supercooling and a span of average INP concentrations at these temperatures of an order of magnitude change during the annual cycle. The dashed lines plotted for -15 and -20°C data are simply to guide the eye. A completed annual cycle of data will be produced under ASR funding, as discussed in the text.

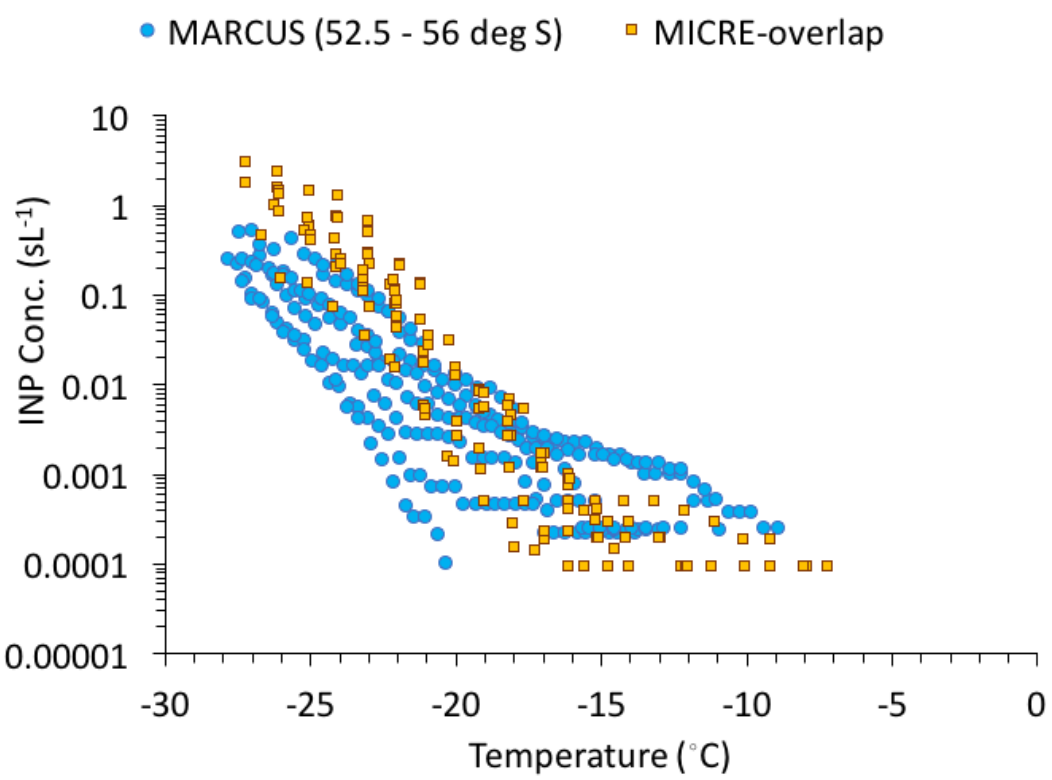

Figure 3. Comparison of MICRE data that has undergone standard processing during the equivalent time period of the MARCUS campaign, and for all MARCUS data within the latitude band of 52.5 to 56°S latitude. This includes data from the fourth MARCUS voyage that sat near-shore at Macquarie Island.

Further research opportunities have already been mentioned at the end of Section 1. Relation of INP data to cloud phase, precipitation and radiative properties will occur with collaborating partners on this proposal, and numerical modeling studies using the information gained on INP populations will be pursued with other collaborators on separate DOE and NSF proposals.

3.0 Publications and References

No publications have been prepared at the time of this report. Science meeting/conference presentations have already featured some of the results. These include upcoming talks at the American Geophysical Union Fall Meeting and the American Meteorological Society Annual Meeting.

3.1 Presentations

DeMott, PJ, TCJ Hill, KJ Moore, CS McCluskey, EJT Levin, KR Barry, MD Petters, GC Cornwell, JL Stith, GM McFarquhar, SP Alexander and SM Kreidenweis. 2018. “Ice nucleating particles over marine regions (and their potential impact on clouds).” Invited presentation at the Telluride Science Research Center’s workshop on Aerosols and Clouds: Connections from the Laboratory to the Field to the Globe, Telluride, Colorado.

DeMott, PJ, C McCluskey, K Moore, T Hill, E Levin, C Twohy, L Russell, D Toohey, B Rainwater, G McFarquhar, A Protat, R, Humphries, G Mace, M Keywood, R Marchand, C Wolff, J Stith, and S Kreidenweis, 2018. "The concentrations, spatial distribution, and compositions of ice nucleating Particles in and around stratiform clouds over the Southern Ocean", Presented at the 10th International Aerosol Conference, St. Louis, Missouri.

DeMott, PJ. 2018. "Measuring the atmospheric diversity and distribution of ice nucleating particles to predict their impacts on clouds and climate." Invited presentation at LaMP Seminar, Clermont-Ferrand, France.

DeMott, PJ, KA Moore, CS McCluskey, AM Rauker, TCJ Hill, EJT Levin, CH Twohy, D Toohey, B Rainwater, JL Stith, GM McFarquhar, R Marchand, C Bretherton, R Wood, SP Alexander, A Gettleman, Y Huang, S Siems, A Protat, RH Humphries, J Ward, MD Keywood, SM Kreidenweis, and the SOCRATES and MICRE Science Teams. 2018. "Spatial and temporal distributions of ice nucleating particles over the Southern Ocean." Presented at the American Geophysical Union Fall Meeting, Abstract A11B-06, Washington, D.C.

DeMott, PJ, KA Moore, CS McCluskey, AM Rauker, TCJ Hill, EJT Levin, CH Twohy, D Toohey, B. Rainwater, JL Stith, GM McFarquhar, R Marchand, C Bretherton, R Wood, SP Alexander, A. Gettleman, Y Huang, S Siems, A Protat, RH Humphries, J Ward, MD Keywood, SM Kreidenweis, and the SOCRATES, MARCUS, and MICRE Science Teams. 2019. "Ice nucleating particles and their impacts on clouds over the Southern Ocean," Invited presentation at the American Meteorological Society Annual Meeting, Workshop, 11th Symposium on Aerosol–Cloud–Climate Interactions, Abstract 12.1, Phoenix, Arizona.

3.2 References

Hiranuma, N, S Augustin-Bauditz, H Bingemer, C Budke, J Curtius, A Danielczok, K Diehl, K Dreischmeier, M Ebert, F Frank, N Hoffmann, K Kandler, A Kiselev, T Koop, T Leisner, O Mohler, B Nillius, A Peckhaus, D Rose, S Weinbruch, H Wex, Y Boose, PJ DeMott, JD Hader, TCJ Hill, ZA Kanji, G Kulkarni, EJT Levin, CS McCluskey, M Murakami, BJ Murray, D Niedermeier, MD Petters, D O'Sullivan, A Saito, GP Schill, T Tajiri, MA Tolbert, A Welti, TF Whale, TP Wright, and K Yamashita. 2015. "A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: A comparison of 17 ice nucleation measurement techniques." *Atmospheric Chemistry and Physics* 15(5): 2489–2518, https://doi.org/10.5194/acp-15-2489-2015

McCluskey, CS, J Ovadnevaite, M Rinaldi, J Atkinson, F Belosi, D Ceburnis, S Marullo, TCJ Hill, U Lohmann, ZA Kanji, C O'Dowd, SM Kreidenweis, and PJ DeMott. 2018a. "Marine and Terrestrial Organic Ice Nucleating Particles in Pristine Marine to Continentally Influenced Northeast Atlantic Air Masses." J*ournal of Geophysical Research: Atmospheres* 123(11): 6196–6212, https://doi.org/10.1029/2017JD028033

McCluskey, CS, TCJ Hill, RS Humphries, AM Rauker, S. Moreau, PG Strutton, SD Chambers, AG Williams , I McRobert , J Ward, MD Keywood, J Harnwell, W Ponsonby, ZM Loh, PB Krummel, A Protat , SM Kreidenweis, and PJ DeMott. 2018b. "Observations of ice nucleating particles over Southern Ocean waters." *Geophysical Research Letters* 45(21), https://doi.org/10.1029/2018GL079981

Suski, KJ, TCJ Hill, EJT Levin, A Miller, PJ DeMott, and SM Kreidenweis. 2018. "Agricultural harvesting emissions of ice-nucleating particles." *Atmospheric Chemistry and Physics* 18(18): 13755-13771, https://doi.org/10.5194/acp-18-13755-2018

Vali, G. 1971. "Quantitative evaluation of experimental results on the heterogeneous freezing nucleation of supercooled liquids." *Journal of the Atmospheric Sci*ences 28(3): 402–409, https://doi:10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2, 1971

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