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Measurements of Aerosols, Radiation, and Clouds over the Southern Ocean (MARCUS) Ice Nucleating Particle Measurements Field Campaign Report

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Acronyms and Abbreviations

AAD	Australian Antarctic Division
ACAPEX	ARM Cloud Aerosol Precipitation Experiment
AMF	ARM mobile facility
AOS	aerosol observing system
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
BOM	Australian Bureau of Meteorology
CSU	Colorado State University
DOE	U.S. Department of Energy
GCSS	GEWEX Cloud Systems Study
GEWEX	Global Energy and Water Cycle Experiment
GPCI	GCSS Pacific Cross-section Intercomparison
INP	ice nucleating particle
MAGIC	Marine ARM GPCI Investigations of Clouds
MARCUS	Measurements of Aerosols, Radiation, and Clouds over the Southern Ocean
MICRE	Macquarie Island Cloud and Radiation Experiment
NSF	National Science Foundation
PI	principal investigator
SO	Southern Ocean
SOCRATES	Southern Ocean Clouds Radiation Aerosol Transport Experimental Study

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1.0 Summary

Evidence exists that sea-spray-sourced ice nucleating particles (INPs) are organic in origin, resulting from biological and biochemical processes in seawater and the sea surface microlayer, and represent a distinctly different and variable, but oftentimes much less effective, INP population in comparison to long-range transported desert dust (DeMott et al. 2016; McCluskey et al. 2018a). Over the Southern Ocean (SO), far from land sources, these INPs may dominate in the marine boundary layer (McCluskey et al. 2018b). Thereby, marine INPs may affect cloud phase and lifetime in this region where pervasive supercooled and mixed-phase boundary-layer clouds appear underrepresented in climate models. This motivated collection of ice nucleating particle concentration measurements during the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility Measurements of Aerosols, Radiation and Clouds over the Southern Ocean (MARCUS) deployment of the second ARM Mobile Facility (AMF2) on the *Aurora Australis* vessel from early November, 2017 (Austral spring) to late March, 2018 (Austral fall).

During this period, four voyages were made from Hobart, Tasmania to the Australian Antarctic Division bases at Casey, Davis, and Mawson, as well as a final resupply cruise to Macquarie Island, the sight of the related Macquarie Island Cloud and Radiation Experiment (MICRE) campaign. Aerosol filter collections of alternating 24- and 48-hour duration were made during all voyages. These filters were returned frozen for subsequent processing to determine variability in the temperature spectrum of the concentrations of INPs active via the immersion freezing mechanism and to conduct additional analyses to indicate the chemical and biological nature of INPs over the SO. The MARCUS data set provides the first spatial and seasonal picture of INP concentrations over the SO in more than four decades, complementing the full annual cycle of INP data we collected for a point location in MICRE.

Images of the filter sampler and its relation to the AOS trailer, other AMF measurements, and the ship stack are shown in Figure 1.

Figure 1. Instrumentation and filter positioning on the *Aurora Australis*. Left: the original mount of the filter unit (globe is a rain hat) along the front rail of the upper deck with other instruments. This position was problematic in snow situations. Middle: The mounting position after snow filled the rain hat during a storm in voyage 1 (V1). The ship stack is in the blue building structure to the aft. Right: View from the final filter position across the deck, with the AOS trailer (blue) just visible on the far-left side of the view.

The location of the sampler moved aft of the forward upper deck railing during the first voyage, following a blowing snowfall event that led to accumulation of snow in the updraft under the rain hat visible covering the filters. Both the filter samples and the instruments in the AOS trailer were located fore of the ship stack, but could be affected by ship exhaust under certain conditions. This is under evaluation presently. Pre-sterilized, open-faced polycarbonate filters were provided by our research team, along with training materials for the group of ARM technicians assisting AMF measurements on the *Aurora Australis*.

Filters were returned frozen to Colorado State University (CSU) at the end of the campaign following interim storage in an AAD freezer in Hobart. 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 67 sample filters were collected, including five blanks at selected intervals throughout the project. For archival and completion of tasks under this ARM proposal, 38 of the original filters were processed for basic temperature spectra, and 14 of these 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). All data have been added to the ARM Data Center.

Additional full processing of the remaining MARCUS filters will be accomplished under a proposal now 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) (distinguishes organic versus inorganic INPs)
- total organic carbon and nitrogen analyses
- ionic aerosol chemistry analyses, and
- genomic analyses of biological community diversity in aerosols.

This ASR study (Contract DE-SC0018929) also supports collaborative studies with other ARM, AAD, 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. The association of INP measurements with the AMF2 aerosol and remote-sensing suite in MARCUS, and the supplemental Wideband Integrated Bioaerosol Sensor measurements (Martin Schnaiter, PI), will provide the richest data set of all SO campaigns for investigating influences and impacts of INPs on SO clouds. Synergies will also be sought with the aircraft and ship measurements made during the shorter National Science Foundation (NSF)-funded Southern Ocean Clouds Radiation Transport Aerosol Transport Experimental Study (SOCRATES) in early 2018. The database developed will also provide comparison and contrast versus other marine INP samples collected during other DOE-funded (MAGIC and ACAPEX) studies from the Northern Hemisphere ocean regions. Measurements will

assist development of parameterizations of marine INP sources for use in regional- and global-scale modeling of the impacts of marine boundary-layer aerosols on clouds and climate.

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. Some first intriguing results are shown in Figures 2 and 3. In Figure 2, all INP number concentrations measured when out of port (some sampling was done at the Antarctic stations) are shown. The exponential spectra of INP concentrations mimic those seen in limited previous SO studies (McCluskey et al. 2018b), but show an especially large range of variation at single temperatures, and an indication of accentuated contributions of biological INPs at times (contrast between untreated versus heat-treated samples), overlaying less efficient non-heat-labile (but likely organic) INPs that are much less variable over time and space (red points).

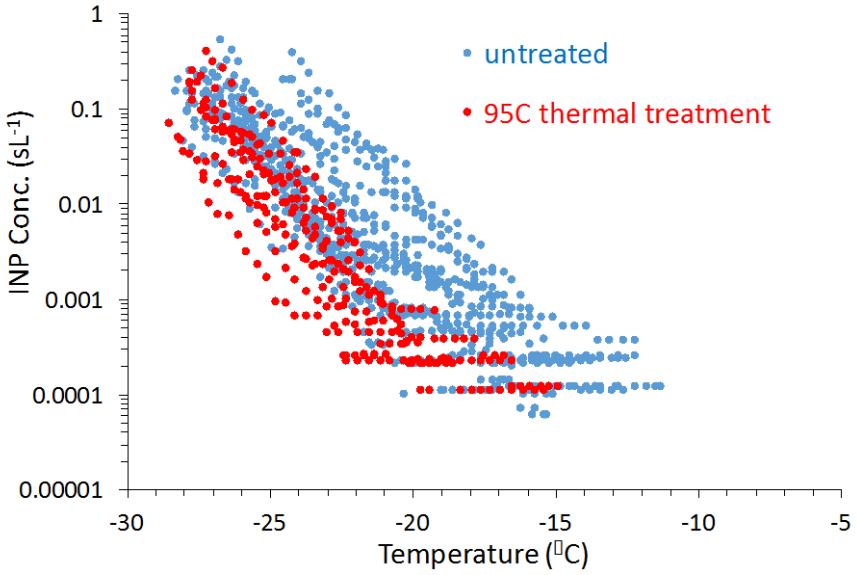

Figure 2. Ice nucleating particle number concentrations versus temperature for all currently processed MARCUS samples when away from port sites. Dates span the entire campaign, with latitudes from approximately -45 to -68°S and longitudes from approximately 60 to 160°. Sample processing is distinguished for no treatment of the aliquots (natural sample) and for heat treatment to test for the influence of microbial/proteinaceous types of INPs. A range of biological (microbial or proteinaceous) influences on INPs are clearly evident over open ocean during the course of MARCUS, never before so well demonstrated in shorter sampling efforts near or over oceans.

First spatial (latitudinal) analysis of MARCUS immersion freezing data is shown in Figure 3. In this representation of data, a few additional things stand out. First, the far Southern Ocean regions show a lower preponderance of heat-sensitive INPs. Then, north of 65°S there appears a general and average increase in INP concentrations at any temperature toward northern latitudes, but a rather constant population of non-heat-labile INPs. The consequent suggestion is that biological INPs more frequently

occur and provide a positive perturbation on INP concentrations in regions north of 60°S. Nevertheless, we must emphasize that these are preliminary analyses that will require evaluation after completion of processing of the full set of physical samples and consideration of insights given by other aerosol data sets.

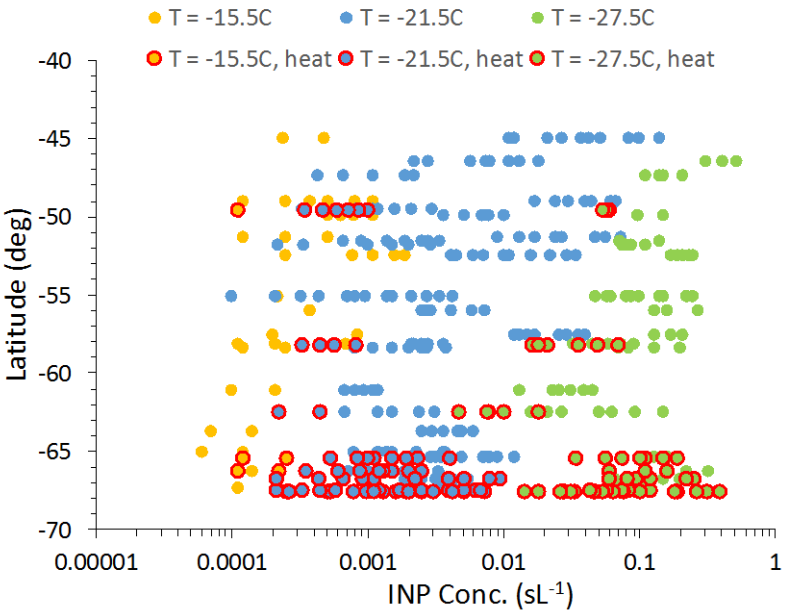

Figure 3. INP concentrations at three temperatures (midpoints of 3°C temperature ranges) and latitude for all MARCUS voyages at non-port locations. Red-circled points reflect thermal treatment of a sample at 95°C prior to immersion freezing measurements.

Ship stack influence on INP measurements has not been fully explored at this point, although preliminary analysis suggests no or limited influence on MARCUS INP measurements, consistent with McCluskey et al. (2018b). This is the case despite noted soot deposits on some of the filters (6 of the 38 so far). Nevertheless, coordination among investigators using aerosol measurements during MARCUS is occurring to evaluate the best means to flag stack-influenced data. After that point, we will revisit INP data to specifically analyze for stack particle impact on INPs, in order to isolate this from marine and terrestrial-sourced particles.

Further research opportunities have been mentioned in the Summary above. The MARCUS INP measurements constitute part of a rich data set toward SO aerosol-cloud-climate studies.

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 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, MARCUS, and MICRE Science Teams. 2019. "Ice nucleating particles and their impacts on clouds over the Southern Ocean." Invited presentation at American Meteorological Society Annual Meeting, Workshop, 11th Symposium on Aerosol–Cloud–Climate Interactions, Abstract 12.1, Phoenix, Arizona.

3.2 References

DeMott, PJ, TCJ Hill, CS McCluskey, KA Prather, DB Collins, RC Sullivan, MJ Ruppel, RH Mason, VE Irish, T Lee, CY Hwang, T. S. Rhee, JR Snider, GR McMeeking, S Dhaniyala, ER Lewis, JJB Wentzell, J Abbatt, C Lee, CM Sultana, AP Ault, JL Axson, M Diaz Martinez, I Venero, G Santos-Figueroa, MD Stokes, GB Deane, OL Mayol-Bracero, VH Grassian, TH Bertram, AK Bertram, BF Moffett, and GD Franc, 2016. "Sea spray aerosol as a unique source of ice nucleating particles." *Proceedings of the National Academy of Science* 113(21): 5797–5803, https://doi:10.1073/pnas.1514034112

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: 402–409, https://doi:10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2

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