Ice Nucleating Particles, Aerosols and Clouds over the Higher Latitude Southern Ocean

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Final Technical Report

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Executive Summary

Improved understanding of aerosol-cloud-precipitation interactions is a key goal of the DOE-ASR program. Ice Nucleating Particles (INPs) are rare aerosol particles that trigger ice formation in clouds. By doing so they may initiate precipitation in clouds, and this impacts cloud lifetime. Nowhere may the role of INPs be manifested so dramatically as in the Southern Ocean (SO). This vast region was the focus of the DOE-ARM MARCUS (Measurement of Aerosols, Radiation and CloUds over the Southern Oceans) and MICRE (Macquarie Island Cloud and Radiation Experiment) campaigns. In the SO, a deficit of INPs has been a leading hypothesis for why liquid clouds persist, which underlays the bias of global climate models in predicting excess shortwave radiation reaching the ocean surface between 55°S and Antarctica.

In this study, we used the 6-month acquisition of INP data on four MARCUS cruises from Hobart, Tasmania, to Antarctica, and the single-location full annual INP cycle observed in MICRE, to gain a holistic, quantitative picture of the INP number concentrations as a function of temperature and their variability over SO latitudes from 43°S to Antarctica. Augmenting initial processing of filter collections of aerosol particles during MARCUS and MICRE, additional freezing studies following thermal treatment (to removes INPs associated with microbes) and peroxide treatment (to remove all organics), as well as ionic chemistry and total organic carbon analysis, were applied to samples to improve time and space resolution, and differentiate marine from terrestrial contributions. We also performed Next Generation Sequencing to determine that bacterial composition can tag periods of enhanced and degraded marine organic INPs and the relative lack of occurrence of land-sourced bio-particles. Through these and other analyses, INP data were categorized into representative source types (e.g., marine versus terrestrially-influenced). The products of our analyses are being used to parameterize ice formation for use in numerical modeling studies to determine if, and when, ocean-derived INPs control the microphysical composition and radiative balance of SO clouds in a manner not presently captured by global models.

The specific objectives and approaches proposed were largely accomplished, including:

1) Additional and value-added chemical and biological analyses of archived samples of aerosols were completed to categorize and quantify INP types and concentrations over the SO.
2) Analyses were completed to place INP data in meteorological and aerosol context.
3) Sea spray aerosol INPs were demonstrated to dominate the SO region in all but episodic events.
4) New parameterizations for INP sources relevant to the SO were constructed.
5) Collaborative modeling studies demonstrating the crucial role of INPs in determining cloud radiative and precipitation properties were conducted with MARCUS and MICRE partners. Collaborative publication submissions in this regard ensued within the two year study.

This work will advance the science of interactions of aerosols, clouds and precipitation, to improve their representation in regional and global climate models. Results have informed representation of primary ice crystal nucleation, give inference to the role of secondary processes, and improve investigations of aerosol influences on clouds via ice nucleation over SO high latitudes and similar vast ocean regions. Application will improve representation of such interactions for clouds in both regional and global climate models. The data base and analyses methods applied will continue to serve research studies in the future.

1.0 Introduction

The project was focused heavily around analyses to evaluate three central and related hypothesis to be addressed using data collected in 2017-2018 field campaigns. Specifically,

Hypothesis 1: Sea spray emissions represent the primary source of ice nucleating particles (INPs) over the Southern Oceans
Corollary 1: INP concentration variations follow factors controlling sea spray emission rates.
Corollary 2: Enhancements of marine biogenic INPs follow/lag Chl *a* and other microbial signatures of productivity.

*Hypothesis 2: Seasonal cycles and variability of INP concentrations are present in the SO, controlled by SSA emission rates and ocean biological activity, and not by long range transport of smoke and du**st***
Hypothesis 3: Primary ice nucleation dominates ice formation in post-frontal scenarios over the high latitude Southern Ocean
An extended set of objectives shape the approach to addressing these related hypotheses as the major goal of the study. These include (distilled from proposal):

1) Integrating existing DOE MARCUS (Measurement of Aerosols, Radiation and CloUds over the Southern Oceans) ship (R/V Aurora Australis) and MICRE (Macquarie Island Cloud and Radiation Experiment), and NSF-funded SOCRATES (Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study) ship and aircraft campaign INP archive measurements, and new measurements on previously unprocessed filters (archived frozen) across all studies, to develop descriptions of the seasonal, annual and spatial variability of INPs.
2) Utilize thermal and chemical pre-treatment of samples to infer the preponderance of proteinaceous/microbial and organic molecular contributions to the INP population measured over the Southern Ocean.
3) Support conclusions on local ocean versus long range transport sources of INPs to MBL-capping clouds through air mass source region back-trajectory analyses, relating these trajectories to satellite ocean color-derived Chl *a* to infer biological influences, integrating real-time bioaerosol measurements into this analysis for MARCUS and SOCRATES, and performing first-ever large scale DNA sequencing of aerosol samples over the Southern Ocean region.
4) Assist parameterization of INP emissions through unifying INP data sets on the basis of aerosol surface area using a variety of methods for specific projects. Additionally compare concentrations and compositions of INPs to other aerosol property measurements. Relatedly, as value-added products, analyze portions of aerosol samples for chemical composition (major inorganic ions, total organic carbon and nitrogen).
5) Perform meteorological categorization with respect to general features such as high/low pressure, frontal location, wind speed, temperature and atmospheric stability, and precipitation occurrence.
6) Coordinate with collaborators to explore INP relationships to cloud phase and precipitation formation processes, using remote sensing and in situ cloud property data for specific case studies.
7) Coordinate with numerical modeling collaborators to integrate new ice nucleation parameterizations in simulations over a range of scales.

2.0 Results

Most objectives of the proposal were met, although emphases were placed mostly in the area of fully characterizing INPs in relation to other aerosols, and providing input to some initial numerical modeling studies. Advanced analyses, expanded parametrization studies, additional numerical modeling and additional publication efforts will continue under new and related DOE-ASR funding.

Hypothesis 1 was evaluated positively based on analyses of the INP compoositions in both MICRE and MARCUS, and additional aerosol chemistry and gas phase data collected at the sites. I depth analyses provided a much more complex picture of relationships between INPs and environmental factors than imagined, and mining of the data set and completion of publications is continuing. A strong signature of INPs at Maquarie Island in concert with vastly greater microbial diversity there in comparison to other areas of the Southern Ocean hints at a biogenic origin for INPs at most times for all of the region south and west of Australia, whereas modest influences of transports of highly active continental dust aerosols are extremely episodic.

Hypothesis 2 is confirmed in line with Hypothesis 1, especially by the only episodic and modest influence of long range transported INPs over the region, and the clear influence of biological activity on INPs active at the warmest supercooled temperatures in the MICRE data set. These results are highlighted in the rest of this section.

For MARCUS, forty-five filter samples were ultimately processed, some for multiple analyses, for MARCUS. Use of treatments for segregating INP types is demonstrated in Fig.1. Striking is that although there is a clear impact of heat at times, implying presence of proteinaceous ice nucleation elements, the impact is relatively restricted, with the removal of INP concentrations only in the > -20C regime, and there are many cases where the heated sample results are indistinguishable from the unamended samples. A factor at play could be atmospheric oxidation impacts on these most labile of emitted marine INPs, a topic that has yet to be explored in either laboratory or atmospheric studies. In contrast to the clear but limited impact of thermal treatments, peroxide digestions prior to freezing indicated the strong contributions of organic INPs nearly throughout the campaign, albeit influence being strongest at the higher temperatures. That result could be a realization of the expected stronger impact of any mineral aerosols present on INPs only at the lower temperatuers. Examples of two limiting cases are shown in Fig. 2, demonstrating, in Fig. 2, a larger presence of heat labile, more proteinaceous INPs and limited organic INPs, versus in Fig. 2b a limited or negligible role of proteinaceious versus predominantly organic INPs. Understanding the sources of these different profiles is a subject of continuing research. Nevertheless, note that in no case was an inorganic source found to dominate INPs in the Southern Ocean region, as might be expected for strong mineral dust transport effect. This may relate to scavenging effects during transport events to the regions of MARCUS and MICRE measurements.

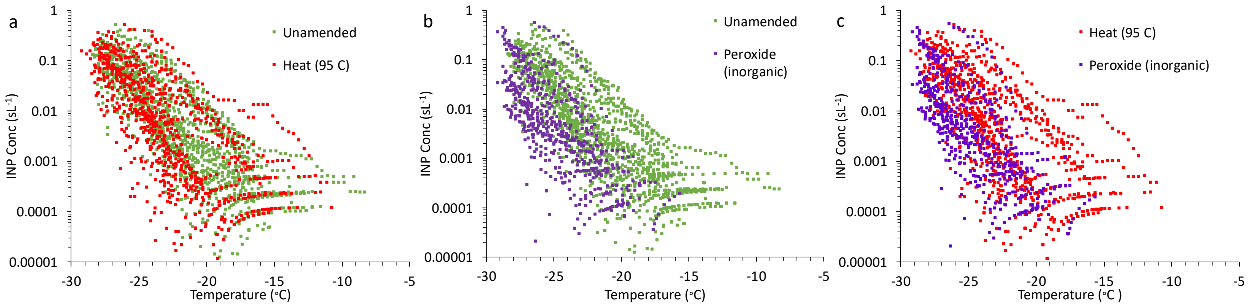

Figure 1. MARCUS filter data shown without regard to date or latitude, to compare and contrast the impact of treatments across the full data set. Results show the preponderance of organic INPs as indicated by strong removal of INPs by peroxide digestion (b), but more selective involvement of purely biological INPs as indicated by removal due to 95°C heating alone (a).

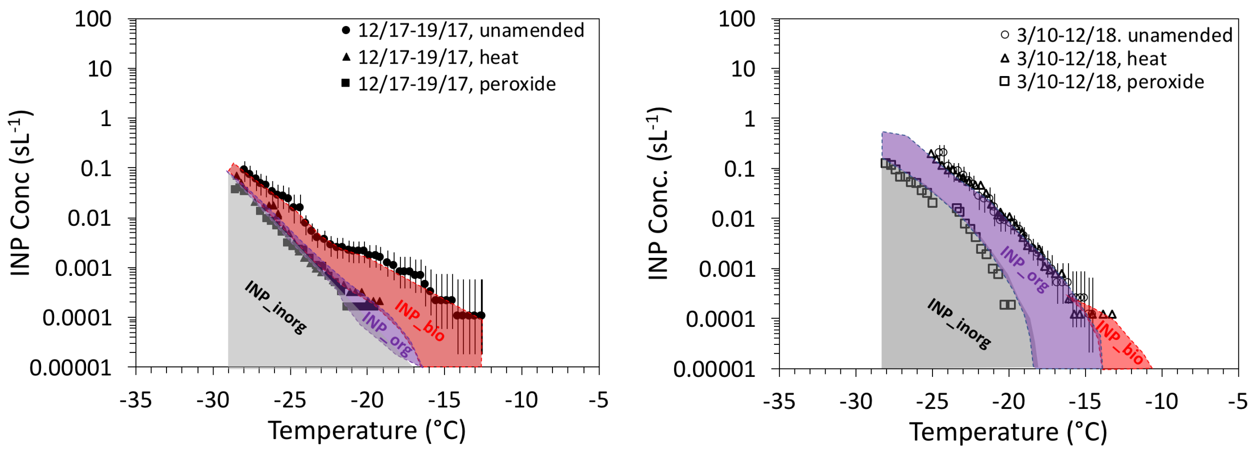

Figure 2: Cases with both treatments that emphasize cases indicating similar signatures of INP types found in the latitude-limited earlier studies of McCluskey et al. (2018b). In a), a period demonstrating the presence and dominance of biological INPs, as indicated by the depth of influence of thermal treatment (red shading) is shown. In b) little or no presence of heat labile biological INPs is noted, but instead organic INPs dominate (purple shading). Case b dominated in the MARCUS data set overall.

An inference of the roles of pure marine versus transported sources of INPs was also be gleaned from analysis of surface active site denstity derivations using the MARCUS and MICRE data, and comparison to similar analyses published for similar and other marine regions. In Figure 3, we have utilized surface areas derived from AOS nephelometer data (DeMott et al., 2016) to normalize INP concentraitons during MARCUS. For select MICRE days with rare clear sky, AERONET inversion data were used to derive average surface area for altitudes from the surface to 1000 m. These data are compared to similar calculations (albeit with aerosol size disgribution data as the source for surface area calculations) for the NSF SOCRATES/CSIRO CAPRICORN-2 campaign that overlapped with MARCUS in the general region south of Australia in January and February of 2018, calculations of McCluskey et al. (2018b) for the CAPRICORN-1 (nephelometer surface area derivation) study of 2016, and the parameterization for marine organic INPs based on Northern Hemisphere study of McCluskey et al. (2018a). These comparison results will be published by Moore et al. (2020, in preparation). An additional publication (Moore et al., 2021, in preparation) will detail the use of different instrumental data (nephelometer, lidar-derived aerosols, and single particle aerosol sizing data) to derive the surface area data needed to calculate the n_s parameter. All results in Fig. 3 excepting MICRE data show both remarkable similarity, and more than order of magnitude variability (approximate 3°C equivalent) variability in n_s over a broad temperature range. MICRE data show a significant positive bias versus the Northern Hemisphere "clean marine" parameterization. All are vastly distinct n_s populations as compared to continental dust INPs.

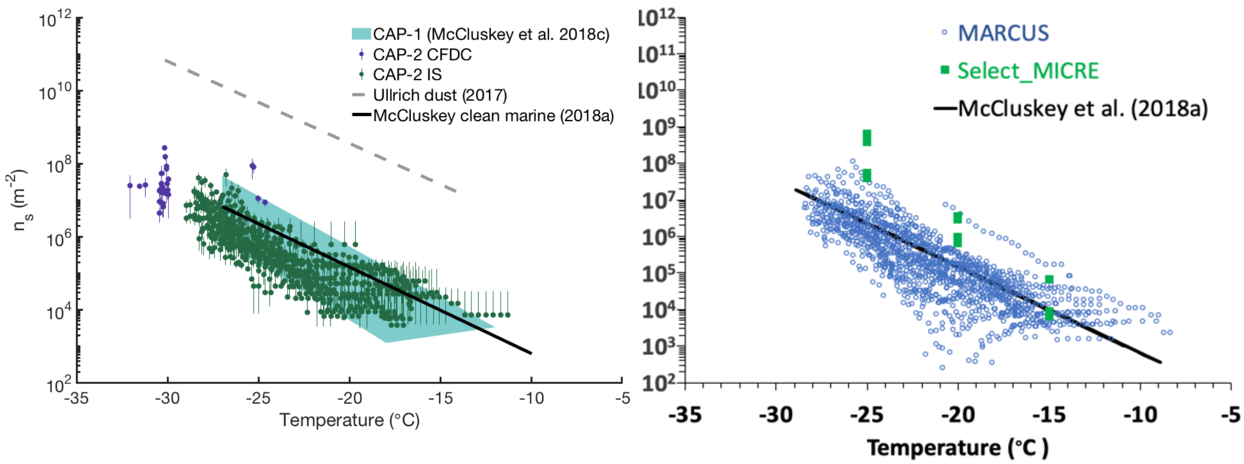

Figure 3. Active site density (n_s) and comparison for Southern Ocean studies in 2016-2018, including CSIRO/NSF funded CAPRICORN-1(2016) and CAPRICORN-2 (2018) studies on left (Moore, 2020), and MARCUS and select MICRE (see text) data on right. Parameterizations of n_s are shown based on Northern Hemisphere marine organic INPs from McCluskey et al. (2018a) and mineral dust INPs (Ullrich et al., 2017).

Finally, new representation of latitudinal disctributions of INPs over Southern Ocean regions were greatly augmented through final MARCUS analyses. Figure 4, from the accepted article of McFarquhar et al. (2020), shows the modest differences with latitude, and equal temporal and spatial variability at any particular latitude in recent investigations. All recent study values differ strongly from the results of Bigg (1973) in ship studies over this wider region. Potential reasons for these discrepancies remain to be determined, but possibilities are discussed in McCluskey et al. (2018a) and Welti et al. (2020), the latter of which also included data from the present study. Investigations of influences of wind, wave height and ocean biology (as inferred from satellite data) are underway but not completed at the time of this writing. These activities are related to new DOE funding that continues the analyses of Southern Ocean project data as part of development of unified understanding and parameterization of INPs using DOE ARM databases.

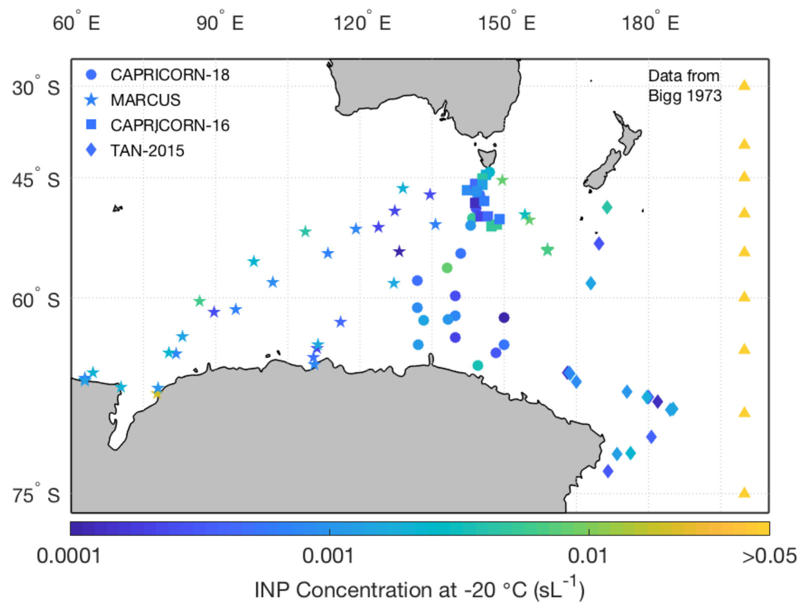

Figure 4. Compilation (to be published in McFarquhar et al., 2020) of INP concentrations (at -20°C) measured in the marine boundary layer from the large array of recent studies, including MARCUS, over Southern Ocean regions that have gone into advanced analysis in this work. The CAPRICORN and CAPRICORN II studies were supported by CSIRO and NSF in 2016 and 2018, respectively. The TAN1502 studies were conducted on the NIWA R/V Tangaroa. Each data point represents the mid-point position of a single filter collection. Historical data from Bigg (1973) are shown at right for context (each is the mean of numerous measures at that latitude); all are the same color since all were >0.1 sL^{-1}.

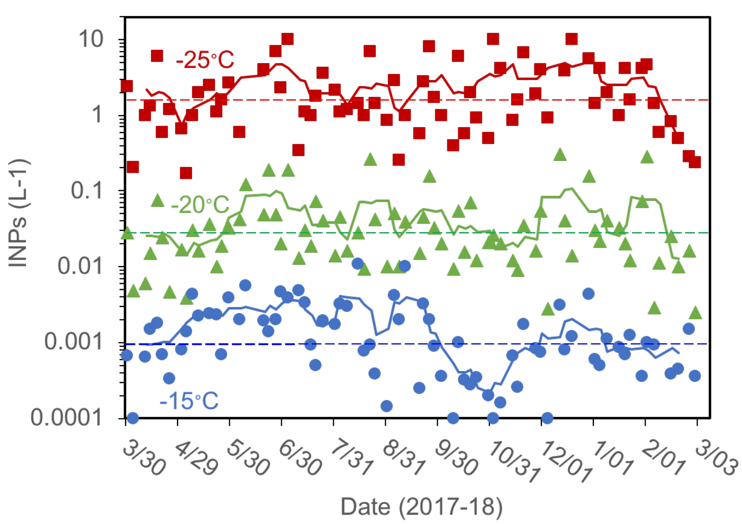

Figure 5. MICRE timeline of INP concentrations at three temperatures, highlighting that short term variability spans values that exceed any apparent annual cycle, except perhaps for INPs active at -15°C, where the weak cycle is suggestive of a potential solar influence. Such an influence could come via oxidative processing of particles in the longer days outside of winter, although a link to wind/wave action is also evident.

Seventy six filter samples from an annual cycle were ultimately processed for MICRE, including nearly 200 total processes that included treatments to indicate INP types, ionic chemical analyses, and DNA analyses. MICRE analyses allowed for clearer evaluation of seasonal INP variations, the focus of Hypothesis 2. Data in Fig. 5 do not support the existence of a strong seasonal cycle in general, excepting a clear wintertime maximum in the most active INP populations (data at -15°C). The lack of a general INP seasonal cycle may reflect the fact that winds in the latitude range of Macquarie Island have only a modest seasonal cycle, with highest values in the Fall/Winter transition.. Figure 6 demonstrates the dominance of organic INPs compared to inorganic ones in the annual cycle on the basis of peroxide elimination of organic INPs. Inorganic INPs, perhaps indicative of long range transports, are noted to dominate only episodically in the warm season, and the are never associated with the highest INPs observed. Hence, either transports of continental aerosols include highly organic INPs or, more likely, enhanced episodes are associated with special marine organic INP emissions at Macquarie Island. The modest influence of continental transports in this region at 55°S is also evident in the absence of a relation of INPs to radon gas data, a tracer of air of continental origin, as shown in Figure 7. Figure 7 also presents investigations of a variety of single correlations of INP data to various meteorological and other factors. No key single relation was evident to explain INP variability in any temperature range.

Two factors do stand out in the MICRE observations that deserve additional mention. INP concentrations and surface active site densities were elevated in MICRE compared to other ocean regions (even nearby MARCUS measurements), which may be a feature of either the ehanced wave breaking region at the island edge or the enhanced biological production of INPs in the ocean regions around the island. Figure 3 makes clear that normalization by surface area alone (n_s) does not explain all of the difference in MICRE. Therefore, biological factors are likely at play. The distinct aerobiology of the MICRE region is evindent in Figure 8, where the vastly greater diversity of bacterial aerosols in comparison to open ocean regions in SOCRATES is indicated by differences in the number of amplicon sequence variants (ASVs) (see Uetake et al., 2020) phylum level taxonomy is indicated in the aerosol DNA sequences. Hence, a strong link between INPs and ocean/atmosphere biology is indicated in MICRE.

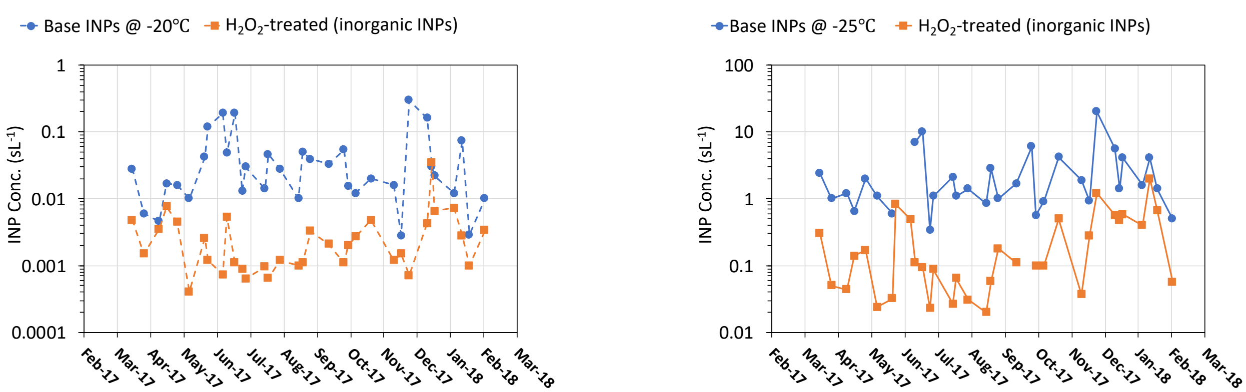

Figure 6. INP concentrations as a function of time at -20 and -25°C on days with sample treatments to isolate INP types, showing a stronger influence of inorganic INPs only in the Austral spring through fall, and implying a preponderance therefore of marine organic INPs at nearly all times, and most notably in periods that represent the highest INP concentrations at each respective temperature.

These results allow for a possible explanation of the winter maximum and summer minimum apparent in the -15°C data. This might be hypothesized to reflect the absence of photochemical impacts on SSA INPs, especially biogenic ones over the winter season, a possibility supported by separate laboratory studies in preparation for submission (DeMott et al., 2021). However, such impacts should spread over the entire INP temperature spectral range, suggesting there are more complex combinations of effects determining the temperature dependence of seasonal INP cycles. Nevertheless, advanced analysis underway indicate a strong role of the solar cycle on the most effective INPs active at modest supercooling, where microbial INPs are known to play a major role. Finally, Fig. 9 shows also that biology was at play in leading to aerosol differences (in ocean and air) that affected INP degradations in the November period at the onset of greater solar insolation. These aerosol samples in November stand out for their unique biology. All factors influencing INPs will be described in a future publication.

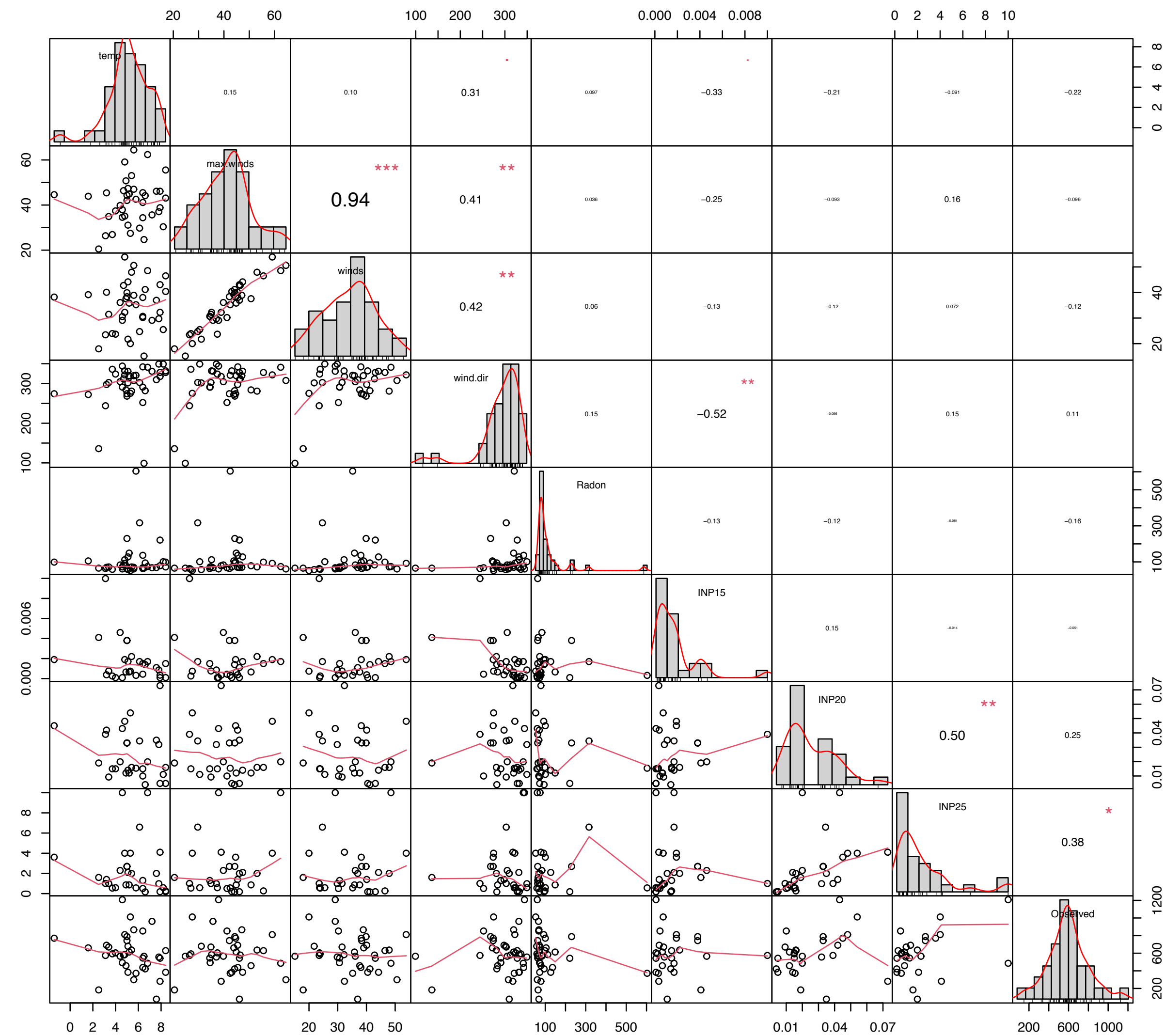

Figure 7. Correlation matrix of INPs at select temperatures in the MICRE data set with mean and maximum winds, radon as a tracer of continental orgins, and operational taxonomic units (OTUs) in air based on genomic analyses (last column). In this figure, the diagonal column represents (from left to right and downward) the temperature (°C), average wind speeds (km hr^{-1}), the maximum wind speeds (km hr^{-1}), wind direction (degrees), radon concentrations (mBq m^{-3}), INPs at -15, -20 and -25(°C (sL^{-1}), and OTUs.

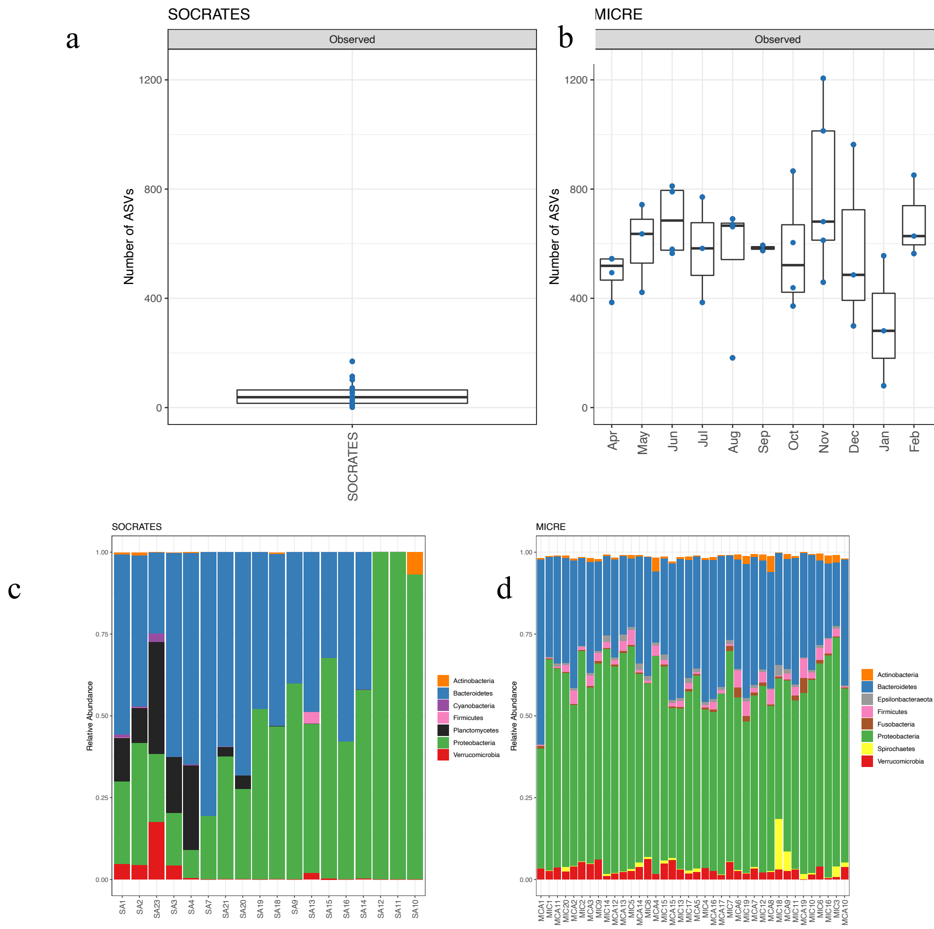

Figure 8. Comparison of amplicon sequence variants (ASVs) in SOCRATES/CAPRICORN-2 ship samples (Uetake et al., 2020) versus MICRE in a) and b), and relative abundance of phylum level taxonomy in c) and d). SA7 and SA 19 in c) are the closest in latitude to the MICRE .

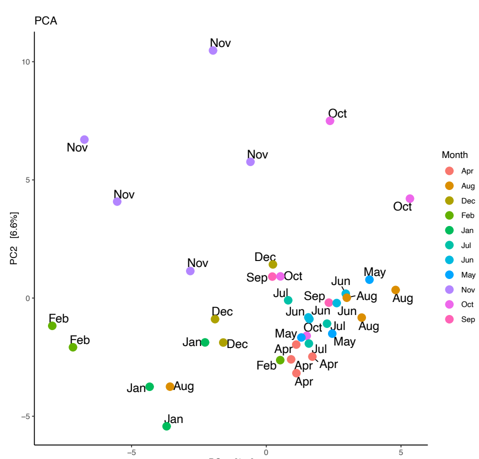

Figure 9. Principle component analysis (PCA) analysis with biologicals: These appear to show high grouping with different periods of INP concentrations, especially demarking biological particle differences demarking the low INP abundance period for warm temperature INPs in the November period in Figure 3.

Hypothesis 3 was partially evaluated, but not conclusively at this time. While primary ice nucleation was observed to be a major controlling factor in proper simulations of Southern Ocean clouds, and secondary ice formation was not critical for producing water distributions in one case, more numerical modeling studies are needed to determine the critical roles of primary versus secondary ice formation. Overall, good progress was made on numerical studies to begin to evaluate the implications of INPs for cloud and precipitation properties over the campaign regions. Studies also advanced analytical tools for relating INP to large aerosol data sets for evaluation and improvement of parameterizations. This was accomplished while simultaneously assisting promotion of professional development of young scientists at multiple institutions.

Collaboration is ongoing with Dr. Susannah Burrows of Pacific Northwest National Laboratory for numerical modeling studies of aerosol impacts on clouds and precipitation, and publication of these efforts. This effort has initially included development of specific parameterization of marine versus other INPs, where our data were used to support and evaluate parameterization of marine organic INPs as emanating from marine gels, or transparent exopolymer particles. This work was conducted by PNNL postdoctoral scientist, Isabelle Steinke.

For NCAR's Andrew Gettleman, Christina McCluskey and Laura Riihimaki (DE-SC0020098; "Freezing Processes in Southern Ocean Mixed Phased Clouds") we have collaboratively provided and advised on use of MARCUS and MICRE INP measurements to assess simulated INPs and aerosol quantities in the Community Atmosphere model version 6 (CAM6). The MARCUS data is critical for a focus on higher latitudes that were not accessed in previous field campaigns, providing insight into a different regime of the Southern Ocean that is closer to the ice edge with relatively high biological activity. The parsing out of purely primary biological and marine organic INPs present over the Southern Ocean region is allowing for evaluation of the utility of previously developed parameterizations that do not account for all marine INP types and further develop/improve these parameterizations. Finally, NCAR collaborators are exploring the utility of the refractory INPs (those remaining after H_2O_2 treatment) as an indicator and validation parameter for mineral dust INPs over the Southern Ocean, which further aids in fforts to improve simulated INPs and mineral dust aerosol predictions in CAM6. Ultimately, these efforts will extend to online simulations to explore varied INP source effects on Southern Ocean cloud microphysics, for which other data sets exist for validation to examine if understanding is "closed".

]An unanticipated collaboration has resulted in studies evaluating of the role of primary versus presently known secondary ice formation processes in Polar WRF simulations. These studies were led by Etienne Vignon (French National Center for Atmospheric Research) and included MARCUS colleagues Simon Alexander (Australian Antarctic Division) and Roj Marchand (U. Washington), as well as colleagues Athanasios Nenes and Alexis Berne (École Polytechnique Fédérale de Lausanne). MARCUS INP data collected around a polar frontal system close to Antarctica, was used in comparison to standard nucleation routines in WRF simulations of mixed-phase clouds over the high-latitude Southern Ocean, and simulations wereevaluated with remotely-sensed data from the MARCUS. Account for the low observed concentration of ice nucleating particles was found to be critical to simulate observed aspects of the cloud system, in particular the important supercooled liquid water layers at cloud top. This great improvement in simulations allowed revelation of other needed improvements for WRF in order to represent such cloud systems, in particular parameterization developments targeting the convection at cloud top in order to reproduce critical turbulence-microphysics interplay.

New analyses as originally proposed will continue, especially of wind/wave relations and relation to satellite-derived ocean biology, under funding of DE-SC0021116 ("Understanding the natural sources of aerosols and their impacts on cloud formation and climate across hemispheres"), which expands this successful project to multiple regions of study utilizing DOE data. A template for equivalent re-analyses of all studies including MARCUS and MICRE was arranged under this proposal through mentorship (April 2020 to August 2020) of exchange student, Baptiste Testa during completion of his M.S. studies at the University of Lyon. Because of the ease of use of data from the CACTI campaign, this was the focus of the INP partitioning analyses and coding development, which will now be applied to MARCUS and MICRE data under new funding.

DOE-ARM data archives for MARCUS and MICRE have been updated to reflect all new analyses. Only chemical analyses are being finalized and prepared for archival at the time of this report.

Major publications were delayed so that Kathryn Moore (Ph.D. candidate under her NSF graduate fellowship) can fold the analyses in this project into a more overarching set of publications on all Southern Ocean studies. This will be more impactful when completed by early 2021.

3.0 Publications and References

Refereed publications

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