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Marine Ice Nuclei Collections – MAGIC (MAGIC-IN) Final Campaign Summary

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ARM
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Executive Summary

This campaign augmented measurements obtained via deployment of the Atmospheric Radiation Measurement (ARM) Climate Research Facility's ARM Mobile Facility (AMF) in the Marine ARM GPCI[1] Investigation of Clouds (MAGIC) field campaign. The measurements, comprised of shipboard aerosol collections obtained during the five legs of the summer 2013 cruises, were sent for offline processing to measure ice nucleating particle (INP) number concentrations. The forty-three sample periods each represented, nominally, 24-hour segments during outbound and inbound transits of the Horizon Spirit. The samples were collected at locations between Los Angeles and Hawaii. Eight samples have been analyzed for immersion freezing temperature spectra thus far, using funding from other grants. Remaining samples are being frozen until support for further processing is obtained. Future analyses will investigate the inorganic/organic proportions of ice nuclei, in addition to determining the genetic composition of the overall biological community associated with INPs. Resulting correlations will be compared with other archived aerosol quantities, meteorological and ocean data (e.g., temperature, wind speed, sea surface temperature, etc…) and satellite ocean color products. These findings will ultimately aid in parameterizing oceanic (e.g., sea spray) INP emissions in regional and global scale models, when illustrating aerosol connections to cloud phases and properties. Independent future analyses of frozen filter samples, as proposed by collaborating investigators at the time of this report, will include single particle analyses of marine boundary layer aerosol compositions and morphology. The MAGIC-IN data are considered representative of the oligotrophic, low Chlorophyll-a (with the exception of near-shore) ocean regions, which exist along the MAGIC transect. Current analyses suggest that INP numbers in the marine boundary layer over this region are typically low, compared to existing measurements over marine areas and those collected in the laboratory as the result of realistic sea spray particle generation. These findings, along with separate studies, confirm the existence of highly variable emission sources for INP from oceans, (though weaker than land-based emissions at modestly cooled temperatures).

[1] GPCI = GCSS Pacific Cross-section Intercomparison, a working group of GCSS
GCSS = GEWEX Cloud Systems Study
GEWEX = Global Energy and Water Cycle Experiment, a core project of the World Climate Research Programme.

Acronyms and Abbreviations

DOE	U.S. Department of Energy
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric Systems Research
MAGIC	Marine ARM GPCI Investigation of Clouds
GPCI	Global Energy and Water Cycle Experiment Cloud Systems Study Pacific Cross-section Intercomparison
NSF	National Science Foundation
NASA	National Aeronautics and Space Administration
CSU	Colorado State University
CFDC	continuous flow diffusion chamber
INPs	ice nucleating particles
IS	ice spectrometer
ACAPEX	ARM Cloud Aerosol Precipitation Experiment
CalWater-2	Moniker for multiagency campaign studies of California precipitation

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1.0 Background

Ice nucleating particles (INPs) trigger the initial formation of ice crystals in the mixed phase (liquid and ice) regions of clouds, thereby impacting solar and thermal energy transfer and precipitation formation processes, in many clouds. Advances in quantification methods which measure INP concentrations in different weather and aerosol scenarios, contribute to improved mixed phase cloud representations in regional and global climate models, via more accurate parameterization development.

Varied natural and human-caused sources of INPs remain poorly quantified. Thus, their impact on clouds and climate remains poorly understood. In this work, samples were collected and processed to more accurately understand the contribution of sea spray aerosols over oceans as INPs, and identify circumstances in which they may predominate over long-range-transported INPs (i.e. those from desert soil dusts). A working hypothesis, supported by inferences from data collected over oceans more than forty years ago (e.g., Bigg 1973), suggests sea spray aerosols represent a modest emission source of INP to the atmosphere. However, even this modest contribution could nevertheless control INP number concentration levels reaching mixed-phase clouds over expansive ocean areas, in some circumstances, either directly or after transport and convective lofting overhead (e.g., Burrows et al. 2013). To evaluate this hypothesis, INP data collections should be analyzed from varied ocean regions, including regions of more or less active marine biological activity, as inferred from Chlorophyll-a (Chl-a) concentrations observed from space.

Adding INP measurements expanded the suite of measurements made during the Marine ARM (Atmospheric Radiation Measurement) GPCI[1] Investigation of Clouds (MAGIC) field campaign; a study with a focus on warm cloud properties. Samples were collected during the summer of 2013, surrounding and overlapping with a MAGIC intensive operations period. Using filter samples was especially suitable, as these are now archived frozen (to limit changes in biological aerosols), until support is available for offline processing. Analysis entails rinsing collected particles in liquid to measure the temperature spectrum of immersion freezing INP (those that freeze within liquid cloud droplets) concentrations, per volume of air sampled (Hill et al. 2014). Uniquely, the MAGIC INP collections represent critical in-situ data for ocean regions with typically low concentrations of Chlorophyll-a.

Once all samples have been processed, the coexistence of the AMF2 data set, obtained during MAGIC, will ultimately permit comparisons with other aerosol parameters, meteorological data (e.g., wind speed, T) and satellite ocean color products. Furthermore, genomic analyses of extracted DNA from aerosols, (performed under NSF funding), will permit important INP markers and air mass characteristics attributable to each sample to be identified either directly, or indirectly, via association with particular microbial consortia. Finally, the data will be useful in developing sea spray-produced INP parameterization measures. This parameterization is pivotal for illustrating marine aerosol impacts on climate and radiation, via aerosol-indirect effects on mixed phase clouds, in regional and global models.

Study contributors included Dr. Paul DeMott (Senior Research Scientist, PI) and Dr. Thomas Hill (Research Scientist), in close coordination with MAGIC PI, Dr. Ernie Lewis. DOE-ARM funding

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supported shipping and sampling materials costs, including financing a small pump necessary for drawing samples. All samples were collected by AMF personnel, with Michael Ritsche as the designated point of contact. A select number of samples were analyzed through pre-existing studies, conducted under National Science Foundation grants. No other support was available from federal funding.

2.0 Notable Events or Highlights

During sampling periods, total particle numbers in the air experienced typical decay toward more normative marine boundary layer concentrations (measured by the AMF-2 condensation particle counter) of 150-300 particles cm-3. Forty three distinct sample periods characterized INP number concentrations, in relation to aerosol properties measured by the AMF-2 aerosol suite, over a three-month period (as illustrated in Table 1. Significant wind speed variability was experienced during these sampling periods.

Table 1. MAGIC-IN sample log and status of analysis (processed in green)

Leg	Samp	Start Time (UTC)	End Time (UTC)	Start Location	End Location	Total Min	Status
13	P01	2013-06-22, 17:14	2013-06-23, 16:54	(32.91, -119.67)	(31.04, -128.00)	1420	
13	P02	2013-06-23, 17:14	2013-06-24, 17:26	(31.04, -128.01)	28.89, -137.03)	1452	
13	P03	2013-06-24, 18:37	2013-06-25, 18:35	(28.78, -137.42)	(26.07, -145.88)	1438	
13	P04	2013-06-25, 18:39	2013-06-26, 18:10	(26.07, -145.88)	(22.96, -153.75)	1411	
13	P05	2013-06-26, 18:20	2013-06-27, 05:01	(22.96, -153.75)	(21.36, -157.33)	641	
13	P06	2013-06-28, 20:14	2013-06-29, 21:34	(21.38, -157.31)	(24.85, -149.60)	1520	
13	P07	2013-06-29, 21:36	2013-06-30, 21:35	(24.85, -149.60)	(27.65, -142.21)	1439	
13	P08	2013-06-30, 21:39	2013-07-01, 21:22	(27.65, -142.19)	(30.05, -134.39)	1423	
13	P09	2013-07-01, 21:33	2013-07-02, 21:33	(30.05, -134.39)	(31.97, -126.25)	1440	
13	P10	2013-07-02, 21:45	2013-07-03, 21:21	(31.97, -126.25)	(33.58, -118.47)	1416	
14	P11	2013-07-08, 20:51	2013-07-09, 20:53	(30.58, -127.97)	(27.78, -137.86)	1442	
14	P12	2013-07-09, 21:19	2013-07-10, 21:45	(27.73, -137.25)	(24.88, -146.31)	1466	
14	P13	2013-07-10, 22:12	2013-07-11, 22:29	(24.83, -146.47)	(21.99, -155.26)	1457	
14	P14	2013-07-13, 21:30	2013-07-14, 21:33	(22.38, -155.19)	(25.33, -148.39)	1443	
14	P15	2013-07-14, 21:54	2013-07-15, 21:28	(25.38, -148.28)	(27.96, -141.29)	1414	
14	P16	2013-07-15, 21:48	2013-07-16, 21:29	(27.99, -141.17)	(30.19, -133.87)	1421	
14	P17	2013-07-16, 21:49	2013-07-17, 20:25	(30.22,-133.76)	(31.91, -126.50)	1356	
14	P18	2013-07-17, 21:02	2013-07-18, 20:33	(31.95, -126.30)	(33.47, -118.67)	1411	
15	P19	2013-07-20, 21:37	2013-07-21, 21:12	(32.58, -121.25)	(30.02, -129.79)	1415	
15	P20	2013-07-21, 21:44	2013-07-22, 21:16	(29.96, -129.99)	(27.33, -138.53)	1412	
15	P21	2013-07-22, 21:41	2013-07-23, 22:00	(27.28, -138.68)	(24.54, -147.36)	1459	
15	P22	2013-07-23, 22:28	2013-07-24, 22:34	(24.49, -147.52)	(21.79, -155.87)	1446	
15	P23	2013-07-26, 22:37	2013-07-27, 22:31	(22.05, -155.89)	(24.60, -150.18)	1434	
15	P24	2013-07-27, 22:53	2013-07-28, 22:27	(24.64, -150.09)	(26.95, -144.18)	1414	
15	P25	2013-07-28, 22:45	2013-07-29, 22:20	(26.98, -144.08)	(29.31, -137.01)	1415	
15	P26	2013-07-29, 22:35	2013-07-30, 22:18	(29.33, -136.93)	(31.21, -129.74)	1423	
15	P27a	2013-07-30, 22:39	2013-07-31, 22:15	(31.24, -129.64)	(31.69, -122.27)	1416	
16	P27b	2013-08-03, 18:00	2013-08-04:1800	(33.10, -119.49)	(30.40, -128.40)	1440	
16	P28	2013-08-04, 18:00	2013-08-05, 18:00	(30.30, -128.50)	(27.72, -137.29)	1440	
16	P29	2013-08-05, 18:07	2013-08-06, 18:01	(27.69, -137.36)	(24.99, -145.95)	1434	
16	P30	2013-08-06, 18:25	2013-08-07, 18:07	(24.94, -146.09)	(22.30, -154.32)	1422	
16	P31	2013-08-10, 18:15	2013-08-11, 19:05	(24.97, -149.28)	(27.38, -142.97)	1490	
16	P32	2013-08-11, 19:30	2013-08-12, 20:45	(27.38, -142.97)	(29.58, -136.06)	1515	
16	P33	2013-08-12, 21:07	2013-08-13, 21:46	(29.58, -136.06)	(31.29, -129.39)	1479	

16	P34	2013-08-13, 22:00	2013-08-14, 23:10	(31.29, -129.39)	(32.66, -122.41)	1510	
18	P35	2013-08-31, 22:10	2013-09-01, 22:40	(32.47, -121.65)	(29.79, -130.54)	1470	
18	P36	2013-09-01, 23:01	2013-09-02, 22:42	(29.75, -130.68)	(27.16, -139.06)	1421	
18	P37	2013-09-02, 23:00	2013-09-03, 22:40	(27.13, -139.17)	(24.54, -147.35)	1420	
18	P38	2013-09-03, 23:00	2013-09-04, 22:45	(24.50, -147.47)	(21.89, -155.57)	1425	
18	P39	2013-09-06, 22:58	2013-09-07, 22:46	(22.37, -155.28)	(25.02, -149.17)	1428	
18	P40	2013-09-07, 23:08	2013-09-08, 22:45	(25.06, -149.07)	(27.48, -142.66)	1417	
18	P41	2013-09-08, 23:08	2013-09-09, 22:45	(27.52, -142.56)	(29.58, -136.05)	1417	
18	P42	2013-09-09, 23:05	2013-09-10, 22:45	(29.61, -135.95)	(31.35,-129.16)	1420	
18	cntrl	2013-09-10, 23:11	2013-09-11, 22:45	(31.37, -129.04)	(32.72, -122.06)	1414	

3.0 Lessons Learned

An extremely positive lesson learned was the capability of AMF-2 personnel, following training, in conducting ship-based filter sampling collections while simultaneously exercising excellent hygiene/cleanliness protocol when handling and storing the filters (frozen). The sampling arrangement is shown in Figure 1. This sampling procedure has been reproduced on three subsequent ship cruises led by other international partners, and served as the basis for specifying sampling on the NOAA RV Ron Brown ship during the DOE-ARM Cloud, Aerosol, and Precipitation Experiment (ACAPEX) conducted January-February 2015.

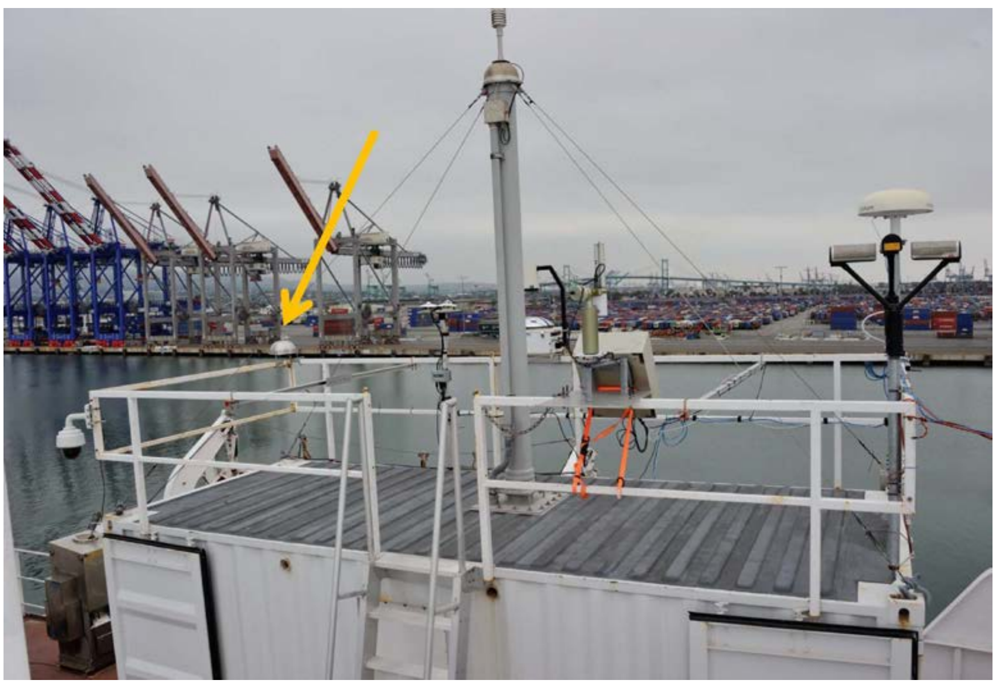

Figure 1. Siting of filter sampler (gold arrow pointing to the rain “hat” over the filter sampler) on the Horizon Spirit, atop one of the AMF-2 sea containers.

4.0 Results

INP number concentrations from MAGIC-IN showed significant variability, though typically lower than the range of values measured over oceans, and from sea spray aerosols generated in the laboratory.

Figure 2 shows INP number concentrations (per liter) measured during two periods of cruise Leg 15 of the MAGIC study. One sampling period occurred close to the U.S. mainland on transit toward Hawaii, while the second sampling period was closer to Hawaii. Data from these sampling periods are compared to laboratory measurements of isolated sea spray particles generated by a laboratory wave flume (Prather et al. 2013) using similar immersion freezing methods and using the real-time CFDC instrument (DeMott et al. 2010) on three different days. MAGIC-IN Leg 15 cruise data are also compared with coastal and aircraft collections in the marine boundary layer around Puerto Rico and St. Croix, USVI, in addition to data from a ship-based sample collected in the Bering Sea. All comparisons are illustrated in Figure 2. Uncertainties representing 95% confidence limits for the immersion freezing data, and twice the Poisson sampling error for CFDC data, are shown as vertical bars in the figure. Finally, the gold arrow shows the range of INP concentrations over remote oceans from Bigg (1973). Total aerosol concentrations in all cases were between 100 and 300 cm^{-3}.

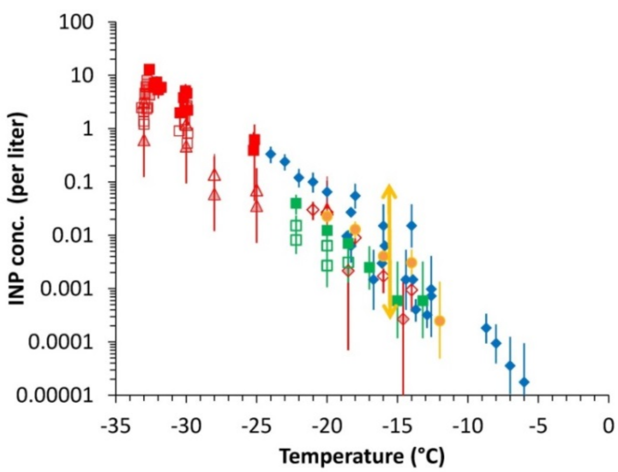

Figure 2. INP concentrations in the marine boundary layer for MAGIC cruise periods P19 (filled green symbols) and P22 (open green symbols), compared to laboratory measurements (red triangles and diamonds), coastal and aircraft collections in the marine boundary layer around Puerto Rico and St. Croix, USVI (blue data points), and a ship-based sample collected in the Bering Sea (gold data points).

Figure 3 shows a map of the general sampling locations for the data shown in Figure 1. Analyses and visualizations used in Figure 3 were produced with the Giovanni online data system, developed and maintained by the NASA GES DISC.

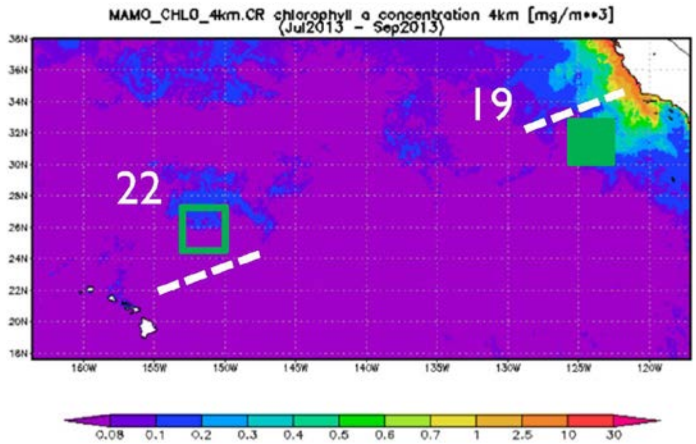

Figure 3. Plot of Chl-a concentrations (mg m^{-3}) integrated over all periods of the MAGIC-IN sampling study.

Comparison to sea spray aerosol samples in the laboratory and other marine boundary layer INP measurements indicate consistency with the overall range of INP number concentrations first measured over forty years ago. Furthermore, MAGIC-IN data processed thus far are relatively consistent with laboratory data collected from aerosols generated using water sampled from the Scripps Institution of Oceanography pier. However, the data fall within the lower range of INP data collected in the air over regions with higher phytoplankton activity. It remains to be determined if this behavior is consistent throughout the MAGIC-IN sample period, thereby indicating a possible connection between phytoplankton blooms and INP releases with sea spray. As evident in Figure 4, the MAGIC-IN samples processed thus far exhibit reasonable agreement with open Pacific ocean measurements in the SHIPPO project, while filters collected closer to land masses may simply reflect continental transport of INP.

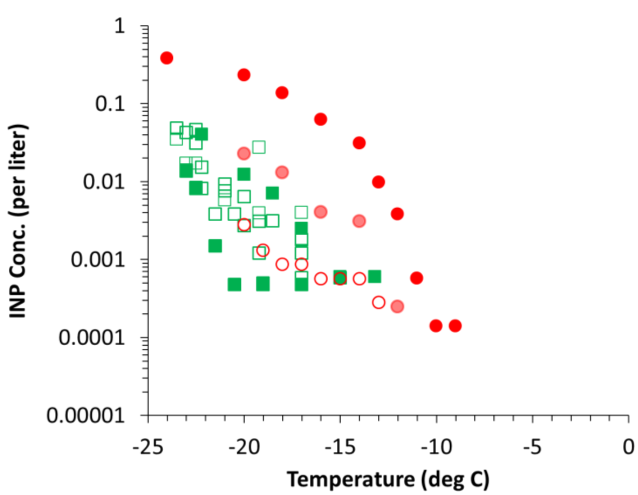

Figure 4. Plot of INP number concentrations versus temperature for MAGIC-IN samples closer to the U.S. mainland (filled green square symbols) and closer to Hawaii (open green symbols) in comparison to samples collected during the SHIPPO (SHIp-borne Pole to Pole Observations) project in July 2012, from the Sea of Japan (red circles), an open ocean location south of the Aleutian Islands chain (open red circles), and the central Bering Sea (filled pink circles).

Data from filters analyzed thus far will be archived in the DOE-ARM archive before April 30, 2015. Further analyses are planned, pending other proposal support. Although an initial proposal to DOE-ASR including these analyses was not funded in 2014, a new proposal is pending under the 2014 DOE-ASR proposal call. If funded, sample analyses will be submitted to the ARM archive within a six month period of funding approval. These data sets will assist in defining climatological INP number concentrations over the north east Pacific, as well as enhancing understanding of INP sources in marine air which contribute to storms supplying the majority of annual precipitation to the Western U.S. coastal regions. Thus, MAGIC-IN data will supplement INP data collected over similar regions during the recent wintertime ACAPEX and CalWater-2 campaigns. Additional use of segments of the MAGIC-IN filters for single particle analyses has also been proposed by Dr. Andrew Ault of the University of Michigan, and MAGIC PI Ernie Lewis, to gain information on the chemical composition of marine aerosols, relevant to cloud activation properties of marine boundary layer clouds that were the emphasis of the MAGIC science plan.

5.0 MAGIC-IN Publications

5.1 Journal Articles/Manuscripts

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

5.2 Meeting Abstracts/Presentations/Posters

DeMott, PJ, KA Prather, TC Hill, T Lee, CY Hwang, Y Tobo, DB Collins, MJ Ruppel, J Axson, C Lee, C Sultana and B Moffett. 2013. "Studies on the relation of ice nuclei from sea spray to ocean biological cycles." Presented at the *American Association for Aerosol Research Annual Meeting*. Portland, Oregon.

DeMott, PJ, TC Hill, MJ Ruppel, KA Prather, C Hwang, et al. 2013. "Investigations of ice nucleating particles from sea spray." Presented at the *DOE-ASR CAPI Fall Working Group Meetings*. Rockville, Maryland.

DeMott, TC Hill, MJ Ruppel, KA Prather, DB Collins, JL Axson, T Lee, CY Hwang; RC Sullivan, GR McMeeking, R Mason, AK Bertram, OL Mayol-Bracero, and ER Lewis. 2013. "Measurements to fill knowledge gaps on ice nucleating particle sources over oceans." 2013. Presented at the *AGU Fall Meeting*. San Francisco, California.

DeMott, PJ, TC Hill, MJ Ruppel, KA Prather, DB Collins, JI Axson, T Lee, CY Hwang, RC Sullivan, GR McMeeking, R Mason, AK Bertram, OL Mayol-Bracero, and E Lewis. 2014. "Investigations of marine ice nucleating particles." In *Sixth Symposium on Aerosol-Cloud-Climate Interactions, Ninety-Fourth Annual Meeting of the American Meteorological Society.* Atlanta, Georgia.

DeMott, PT Hill, K Prather, C McCluskey, R Sullivan, E Lewis, and S Kreidenweis. 2014. “Field and laboratory explorations of marine ice nuclei.” Presented at the *DOE Atmospheric Systems Research Science Team Meeting*. Potomac, Maryland.

DeMott, PJ, TC Hill, CS McCluskey, EJ Levin, KA Prather, DB Collins, G Cornwell, RC Sullivan, MJ Ruppel, R Mason, C Sultana, C Lee, T Lee, CY Hwang, JI Axson, AP Ault, MD Martinez, OL Mayol-Bracero, A Bertram, O Laskina, VH Grassian, and ER Lewis. 2014. “Evaluating ocean sources of ice nucleating particles. Presented at the *Fourteenth American Meteorological Society Conference on Cloud Physics*. Boston, Massachusetts.

DeMott, P. J., T. C. J. Hill, Y. Tobo, C. S. McCluskey, E. J. T. Levin, K. Suski, D. B. Collins, G. Cornwell, C. Lee, C. Sultana, J. Axson, F. Malfatti, K. A. Prather S. M. Kreidenweis, O. Laskina, J. Trueblood, V. H. Grassian, A. Bertram, and R. Mason. 2014. “Hunting Sources of Biogenic Ice Nucleating Particles in Soils, Sea Spray and Air,” *American Association for Aerosol Research Annual Meeting*, Oct. 22, 2014.

DeMott, PJ, KA Prather, TC Hill, CS McCluskey, EJT Levin, KJ Suski., J Creamean, DB Collins, A Martin, G Cornwell, H Al-Mashat, D Rosenfeld, LR Leung, JM Comstock, JM Tomlinson, SM Kreidenweis and MD Petters. 2014. “Ice nucleating particles and their role in California winter clouds.” Presented in the *American Geophysical Union Fall Meeting*.

6.0 References

Bigg, EK. 1973. “Ice nucleus concentrations in remote areas.” *Journal of Atmospheric Science* 30: 1153-1157, doi:http://dx.doi.org/10.1175/1520-0469(1973)030%3C1153:INCIRA%3E2.0.CO;2.

Burrows, SM, C Hoose, U Pöschl, and MG Lawrence. 2013. “Ice nuclei in marine air: Biogenic particles or dust?” *Atmospheric Chemistry Physics Journal* 13: 245–267, doi:10.5194/acp-13-245-2013**.**

DeMott, PJ, AJ Prenni, X Liu, MD Petters, CH Twohy, MS Richardson, T Eidhammer, SM Kreidenweis, and DC Rogers. 2010. “Predicting global atmospheric ice nuclei distributions and their impacts on climate.” *Proc. National Academy of Science Journal* 107(25): 11217-11222, doi:10.1073/pnas.0910818107.

Hill, TCJ, BF Moffett, PJ DeMott, DG Georgakopoulos, WL Stump, and GD Franc. 2014. “Measurement of ice nucleation-active bacteria on plants and in precipitation by quantitative PCR.” *Applications of Environmental Microbiology Journal* 80(4):1256-1267, doi:10.1128/AEM.02967-13.

Prather, KA, TH Bertram, VH Grassian, GB Deane, MD Stokes, PJ DeMott, LI Aluwihare, B Palenik, F Azam, JH Seinfeld, RC Moffet, MJ Molina, CD Cappa, FM Geiger, GC Roberts, LM Russell, AP Ault, J Baltrusaitis, DB Collins, CE Corrigan, LA Cuadra-Rodriguez, CJ Ebben, SD Forestieri, TL Guasco, SP Hersey, MJ Kim, W Lambert, RL Modini, W Mui, BE Pedler, MJ Ruppel, OS Ryder, N Schoepp, RC Sullivan, and D Zhao. 2013. “Bringing the ocean into the laboratory to probe the chemical complexity of sea spray aerosol.” *Proc. National Academy of Science Journal* 110(19): 7550-7555, doi:10.1073/PNAS.1300262110.

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