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Measurements and analysis of ice nuclei relevant to west coast U.S. precipitation

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Final Technical Report

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Executive Summary

Improved understanding of the interactions of natural and human produced aerosols with clouds, and ultimately their influences on precipitation and climate is a key science need, and a goal of the DOE Atmospheric Systems Research program. This study focused on characterizing the numbers and types of ice nucleating particles (INPs), the rare particles in air that possess properties to trigger the formation of ice crystals in cold clouds, which are the initial seeds for wintertime snowfall, and for rainfall in many parts of the Earth. In this study, data were analyzed from field studies of INP numbers, types and spatial distributions feeding winter clouds on the West Coast of the U.S., so that regional scale and climate model simulations performed by other DOE-funded scientists can be improved in the future.

Data collected in DOE-funded and other aircraft and ground-based studies in California and the upstream Pacific Ocean regions between 2011 and 2015 were used to test the hypothesis that INPs from desert mineral dust/biological aerosol layers transported intercontinental distances at higher altitudes provide the primary control over precipitation efficiency in winter cloud systems over the Sierra Nevada Mountains. This hypothesis was rejected on the basis of finding that, while marine aerosols generated over upstream ocean regions that are lifted by winds over the Sierras often support deep and highly supercooled cloud layers due to a relative deficiency of INPs., INPs produced by ocean sea spray do provide sufficient seeds for ice crystal and subsequent precipitation formation. Significantly, pollution aerosols were not found to be sources of INPs for winter storm systems. Additionally, variability of INP number concentrations collected from marine air over broad regions feeding the West Coast of the U.S. was explored. While no seasonal and inter-annual variability of INP number concentrations in marine air was found, these concentrations can vary over three or more orders of magnitude at any given temperature below 0°C. Such variability reflects varied emissions from ocean sea spray, possibly in response to ocean biological processes, as well as influences of transports of particles by winds from terrestrial regions that enhance INP concentrations.

Data on the makeup of cloud particles (droplets and ice crystals) were also analyzed from aircraft research flights to find that nucleation from INPs is additionally supplemented by so-called secondary ice formation processes, which describes the additional formation of new ice crystals due to interactions of ice and liquid particles subsequent to nucleation by INPs. An example is the formation of ice splinters when supercooled cloud droplets collide and freeze onto ice crystals. The results from the present study are being implemented into numerical models of clouds and precipitation in order to better elucidate the ultimate role of these different processing in precipitation formation.

The proposed work advances the science of the interactions of aerosols, clouds and precipitation, with direct application to improve representation of such interactions for clouds in regional and global climate models. The data base will continue to serve research studies in the future.

1.0 Introduction

This project supported analyses of data collected primarily during DOE-funded atmospheric campaigns, particularly the ARM Cloud Aerosol Precipitation Experiment (ACAPEX) and associated NOAA and NSF campaigns termer "CalWater", toward understanding the sources and concentrations of ice nucleating particles (INPs), the relatively rare particles in air that possess properties to trigger the formation of ice crystals in cold clouds, the initial seeds for wintertime snowfall and rainfall in many parts of the Earth. These campaigns focused on the characterizing aerosols and impacts on precipitation in winter cloud systems on the west coast of the U.S. due to the overarching importance of winter precipitation on water resources in this region of the world. The ultimate goal of studies was to distill information so that related regional scale and climate model simulations performed by other DOE-funded scientists can be improved. This final report builds on annual reports previously submitted.

Study objectives were to:

1. Analyze and categorize ice nucleating particle (INP) number concentration and chemical composition data collected in CalWater-1 (2011) and ACAPEX (2015) flight campaigns, in combination with surface-based (NSF-supported coastal site at Bodega Bay Marine Laboratory) INP data from the Winters of 2014 and 2015, and ship-based (NOAA RV Ron Brown with AMF-2) INP and aerosol data from ACAPEX in 2015, to quantify INP distributions around California winter clouds and their relation to other aerosol properties.

2. Perform case study and conceptual winter storm analyses of INP-cloud interactions utilizing the INP, aerosol, and cloud particle data sets from all sites/platforms, and develop aerosol-specific (e.g., marine versus mineral dust versus other, such as pollution) INP parameterizations for use in collaborator-led numerical modeling.

3. As a secondary objective of the overall effort, the latitudinal, seasonal, and inter-annual variability of marine boundary layer INPs affecting the U.S. West Coast will be investigated through analyses of other archived ship-based filter samples from over subtropical source regions from which wintertime Atmospheric Rivers emanate.

Specific hypotheses evaluated were:

H1: Long range transported dust is the key particle type controlling ice concentrations and precipitation in winter orographic storms over the Sierra Nevada Mountains.

H2: An oceanic source of INP determines the concentrations of INP in the marine boundary layer in the absence of continental aerosol influences, and number concentrations are consistently low enough for deep cloud supercooling to persist when clean marine layers are lofted.

H3: Pollution particles and other boundary layer aerosols are associated with enhanced INP number concentrations relative to INP number concentrations in other air masses

2.0 Results

Nearly all objectives of the proposal were met, although emphases were placed mostly in the area of characterizing INP influences occurring during the ACAPEX and CalWater-2 (2015) experimental periods at all sites, as well as in completing processing of relevant INP data collected over different ocean regions. The key results of this study thus far have been submitted for publication by Levin et al. (2019). A particularly important conclusion was the rejection of hypothesis H1. In contrast to CalWater-1, when the role of long-range transported dust entering cloud systems near cloud top was emphasized (Creamean et al., 2013), marine aerosol was the dominant aerosol type and marine INPs were dominant at cloud activation temperatures (sampled in air aloft and from cloud particle residuals via the G-1 counterflow virtual impactor inlet) during the major atmospheric river (AR) event during ACAPEX. It was also the apparent case that marine INPs were a common feature in instances of air being lofted from offshore over the Sierra Mountains. While marine INPs possess low ice active site density (n_s, m^{-2}) in comparison to more highly efficient ice nucleating particles such as certain mineral dusts (**Fig. 1**), it can be the case that mineral dust number loadings are often extremely small in air entering orographic cloud systems. Marine INPs can still provide sufficient ice nucleation to stimulate precipitation formation. Mineral dust transports still can and do have a strong influence on INP and subsequent primary ice concentrations, when they occur, and this competition is likely evidenced in **Fig. 1** by the large n_s variations exceeding those explainable by marine aerosols alone. Both aerosol types can therefore be important for wintertime orographic precipitation, and modeling studies combined with observational data from ACAPEX and future studies should continue to shed light on the implications for cloud microphysics and precipitation. As a corralary, hypothesis H2 was accepted. Figure 1 also demonstrates that although the winter season of CalWater-1 was observed previously to be more episodic in terms of the influence of mineral dust transports, it overall reflected quite similar ranges of INP concentrations (n_{INP}) and n_s values.

Hypothesis 3, regarding the potential direct role of anthropogenic pollution particles as important INPs for winter orographic clouds in California was also rejected. This was based on the fact that, although there was considerable scatter in the vertical profiles of n_{INP}, there was overall no strong relation with altitude. Also, when n_{INP} was normalized by either total aerosol concentration or aerosol surface area (to obtain n_s), both active fraction and n_s values were found to increase slightly with altitude, likely indicating that higher concentrations of anthropogenic aerosols near the surface (most often in the Central Valley region or over the San Francisco and Sacramento metro areas) were poor sources of INPs.

While n_{INPs} in the ACAPEX region were generally at their lowest during the major AR event, strong increases in n_{INPs} active at temperatures higher than -20 °C were documented in the boundary layer over the region after storm passage (**Fig. 2**). As discussed in Levin et al. (2019), these increases mimicked those expected for biological INPs enhanced following rain events in other locales. Association with biological particles types previously directly associated with INP emissions was specifically demonstrated for the Bodega Bay site at this time by Cornwell (2019). Enhancements in INPs were seen throughout the boundary layer for several days following this AR during ACAPEX suggesting that persistence could be of influence during periods of higher frequency storm events than occurred in 2015.

Capabilities for general direct studies of biological aerosols via genomic analyses was also advanced during this study, a first for our group that paid benefits for sampling and analysis during DOE Southern Ocean campaigns in 2018 (DeMott et al., 2018a,b). DNA from aerosol samples was initially shared and analyzed through a special grant from the DOE Joint Genomics Institute. Additional samples that jointly

served INP measurements with the IS instrument were targeted toward the objective of utilizing DNA to categorize air mass influences, and to dig down into specific sequences for ice nucleating biological organisms. This analysis could not be completed due to insufficient DNA extraction from marine aerosols in limited 24-hour samples. This finding led to the altered sampling protocol that was successful for Southern Ocean studies during 2018.

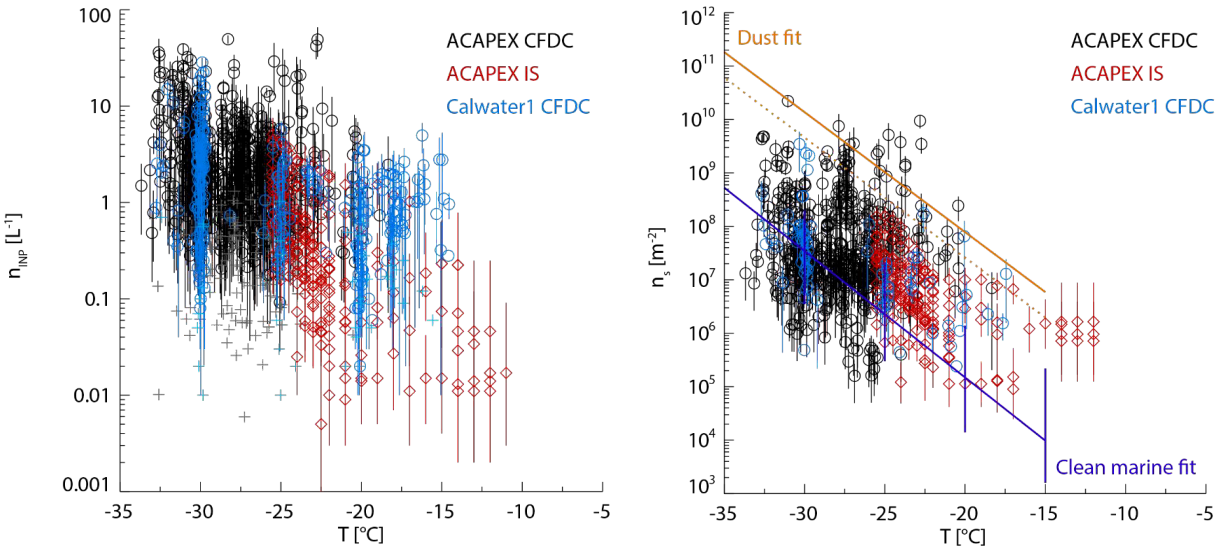

Figure 1: From Levin et al. (2019). **Left panel:** All airborne n_{INP} measurements (per standard liter) from the CFDC (continuous flow diffusion chamber; black) and IS (ice spectrometer immersion freezing device; red) during ACAPEX and CFDC measurements from CalWater-1 (blue) as a function of temperature. For the CFDC data, circles represent points above the 95% confidence limit and lighter colored crosses show measured points below this threshold. Uncertainty bars show the 95% confidence intervals. **Right panel:** Ice nuclei surface site density (n_s) as a function of temperature measured in clear air from the G-1 during ACAPEX from both the CFDC (black circles) and IS (red diamonds) and from the CFDC during CalWater-1 (blue). To add context the solid orange line shows a fit to laboratory data for dust INP [*Niemand et al.*, 2012] with the dashed orange line showing the same fit divided by a factor of 3, following DeMott *et. al.* [2015], to indicate expected CFDC measurements of dust n_s values. The purple line shows a fit to clean marine data [*McCluskey et al.*, 2018a] with the vertical purple lines showing the range of values found by DeMott *et. al.* [2016] for marine INPs from both field and laboratory experiments. These reference values would be valid if all particles dominating surface area and the INP population were of a single compositional type and behaved as INPs as in these previous studies. These values characterized the major proportion of ACAPEX and CalWater-1 data. Marine aerosols characterized about 1/3 of particles sampled during all flights, about 80% of aerosols sampled during the major AR event, and also 80% of all cloud particle residuals during the ACAPEX campaign.

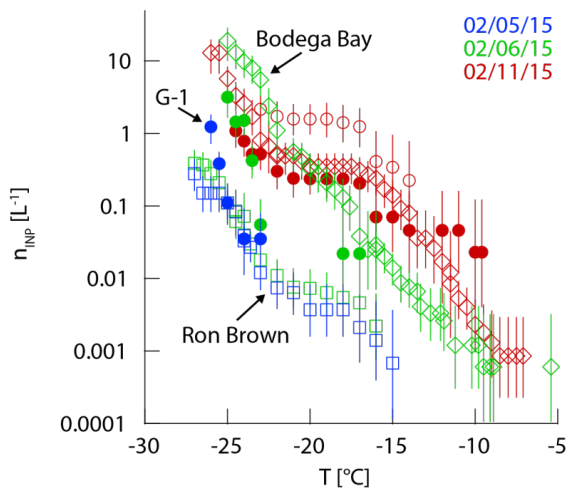

Figure 2: n_{INP} measured by the IS for aerosols rinsed from filters collected as the AR approached land (blue), made landfall (green) and after the event (red). Processed samples are distinguished for those collected aboard the Ronald H. Brown (squares), at the Bodega Bay surface site (diamonds), and from the DOE G-1 aircraft (circles). Building in of warmer temperature and likely biological INPs following the AR rains was an unexpected finding from these studies.

Only moderate progress was made on integrating INP data with cloud microphysical data in order to describe ice formation processes in winter orographic clouds in California. An M.S. student, Kevin Barry, began his studies with this topic in Fall 2017, but changed his thesis topic before Summer 2018. Nevertheless, he put in place an analysis scheme for use with G-1 cloud probe data that follows from the work of G. McFarquhar and colleagues, as documented in the University of Illinois Optical Array Probe (OAP) Processing Software scheme (https://github.com/weiwu5/UIOPS), with adjustments specific to ACAPEX clouds. Dr. DeMott plans to work with Mr. Barry to complete this task moving forward. Initial results from the AR cloud flight periods during ACAPEX were presented at the AMS Cloud Physics Conference in 2018, and some of these results are shown in **Fig. 3**. Secondary ice formation was found to be present in cases focused around the largest atmospheric river event. Analyses continue to characterize microphysical processes over the entire ACAPEX and CalWater-1 data sets. If secondary ice formation processes are ubiquitous in such winter storms, these processes could modulate aerosol impacts.

Collaboration is ongoing with Dr. Jiwen Fan on numerical modeling studies of aerosol impacts on clouds and precipitation, and publication of these efforts. This effort will include specific parameterization of marine versus mineral dust INPs, and evaluation of the role of known secondary ice formation processes in their spectral bin microphysics treatments. These efforts will extend on the idealized simulations of Fan et al. (2017) to treat realistic aerosol scenarios defined in this project.

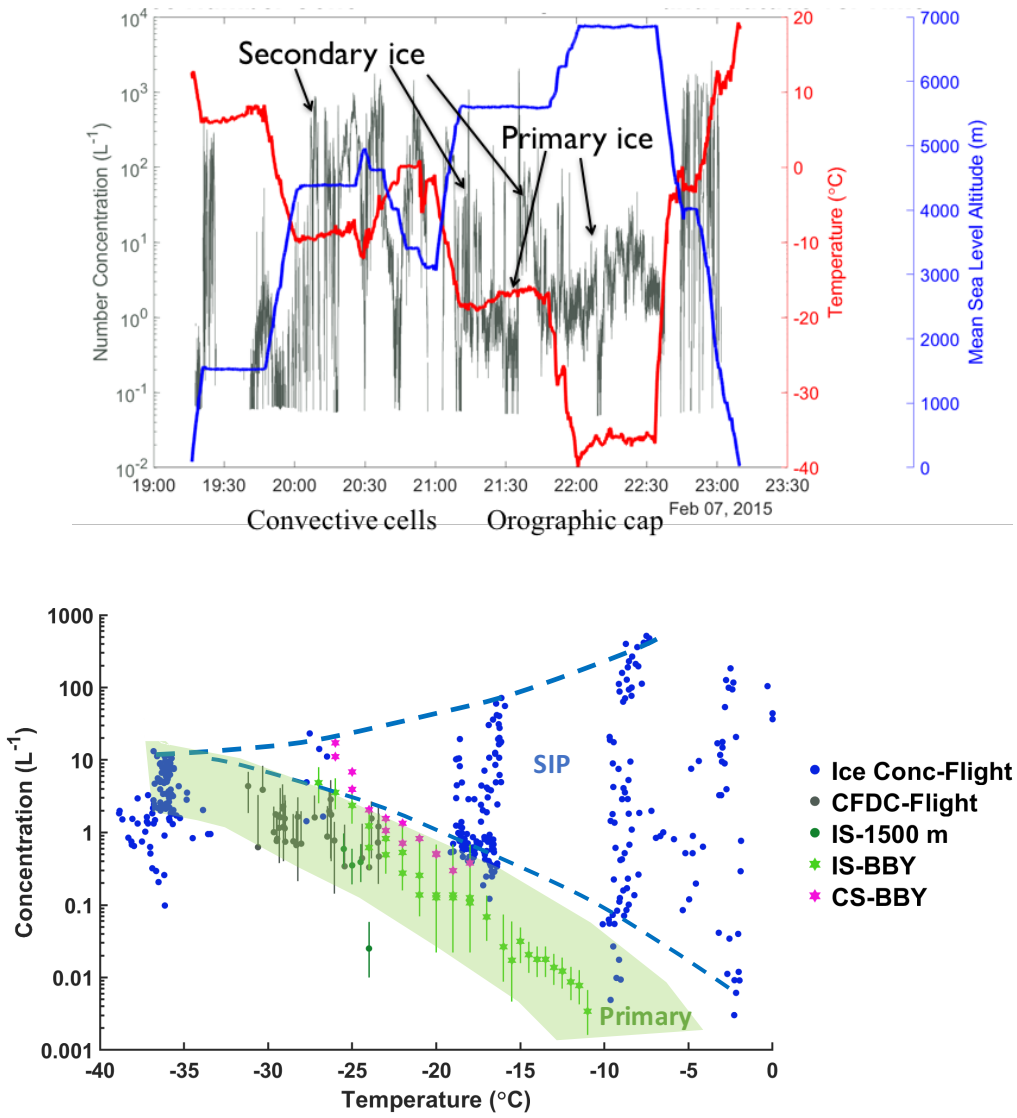

Figure 3. **Top panel:** Timeline of ice crystal number concentrations derived from 2DS cloud particle probe and related data for the flight on February 7, 2015 in the midst of the major ACAPEX AR event. Ambient temperature and altitude are also shown, and regions are identified where orographically-triggered convective clouds were penetrated (sometimes near their tops) upstream of the Sierra barrier crest versus regions over the barrier in the orographic cap cloud. Areas reflecting primary and secondary ice formation are suggested largely on the basis of knowledge of cloud top location in each region and information gleaned from the lower panel regarding measured primary INP concentrations. **Bottom panel:** A consolidation of INP concentration measurements and derived ice crystal concentrations for this case. INP measurements are from the CFDC and IS measurements on suspensions of aerosols collected during flight, and from the IS and NCSU cold stage (CS) immersion freezing measurements of aerosols collected at different tims on this day at Bodege Bay (BBY). The green shaded region is intended to identify the expected "Primary" ice concentrations on the basis of INP measurements. Ice concentrations (blue) are from the top panel. The occurrence of secondary ice production (SIP) can be inferred by the existence of ice concentrations in some cases exceeding INP concentrations by orders of magnitude.

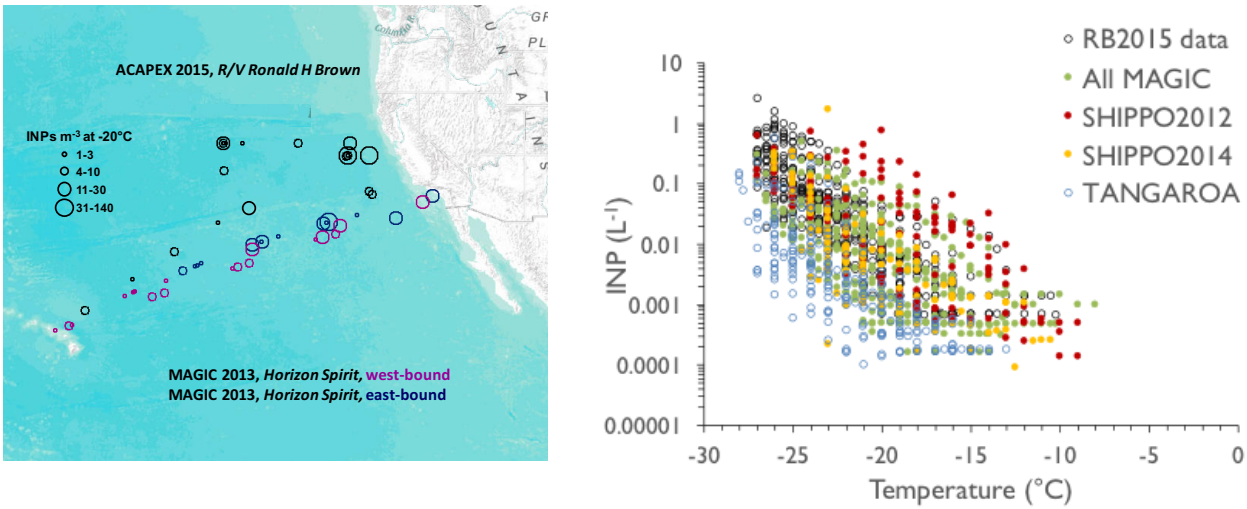

Figure 4. Left panel: INP concentration measurements processed at -20°C, from west of the California coast in two campaigns (ACAPEX and MAGIC), demonstrating little seasonal or interannual variability of INPs for over the marine boundary layer upstream. It is suggested that higher n_{INP} is consistently found close to the coast, which may result from a combination of terrestrial influences and ocean productivity influences on INPs closer to shore. **Right panel:** All INP temperature spectra are compared for the two studies. In addition, data from SHIPPO2012 (Korea to Nome, AK), SHIPPO2014 (Korea to Christchurch, New Zealand) and TANGAROA (Aukland, New Zealand to 72S latitude in 2015) are shown. Terrestrial influences are also inferred over the Western Pacific (SHIPPO2012) and enhancements of Northern versus Southern Hemisphere INPs are apparent in comparing to the SHIPPO2014 and TANGAROA data.

Finally, the potential variability of boundary layer INPs over upstream oceans was investigated via further processing of samples collected during the Marine ARM GCPI Investigation of Clouds (MAGIC): Marine Ice Nucleating Particle Collections (DeMott et al., 2014) samples to those from ACAPEX reveal strong consistency of the Ron Brown ACAPEX atmospheric river INP temperature spectra (DeMott et al., 2016) with comparable pristine MAGIC spectra. Thus, seasonal differences are not readily apparent over open ocean regions to the west of California (**Fig. 4**) that are not explicable via variations in marine INP emissions as described by the clean marine n_s data (c.f., **Fig. 1**). Elevated INP concentrations were associated with either terrestrial influences or enhanced ocean productivity near land regions. Additionally, substantial correspondence between these Northern Hemisphere data from the Eastern Pacific was found in comparison to summer season INP spectra obtained in the Western Pacific during the Korea Polar Research Institute SHIPPO2012 (SHIpborne Pole-to-Pole Observations 2012) campaign's latitudinal transect from Incheon, Korea to Nome, AK (**Fig. 4**). However, distinct reductions in INP concentrrations over oceans were noted in the cross-hemispheric SHIPPO2014 cruise (Korea to Christchurch, New Zealand) and the deep Southern Ocean TANGAROA (NIWA's 2015 Austral summer) cruise. These latter results are consistent with inter-hemispheric differences noted by McCluskey et al. (2018b), and imply a remarkable 4 order of magnitude difference in INP concentrations active at -20°C over global ocean regions.

Data archives from the MAGIC, CALWATER-1 and ACAPEX studies are available in the DOE-ARM archive. Data from the Bodega Bay Marine Laboratory in CalWater-2 may be found in the Colorado State University digital repository (https://mountainscholar.org/handle/) under the archive name "*Calwater2 ice nucleating particle measurements at Bodega Bay Marine Laboratory*". Further processed INP samples over oceans during this study, including SHIPPO2012, SHIPPO2014, and TANGAROA are available in

the Colorado State University digital repository under the archive title "*Additional shipborne ice nucleating particles data processed under contract DE-SC0012354.*" DOIs for these digital archives were not available at the time of this report.

Two additional publications are anticipated from this research, focused around the results shown in Figures 3 and 4, on topics of the distribution of, 1) INPs seasonally and spatially over ocean regions of influence to orographic precipitation regions, and 2) ice formation processes and relations to ice nucleating particles in winter precipitation events over California's Sierra Mountains.

Numerous opportunities were provided for professional development during the course of this study.

1) Dr. Kaitlyn Suski worked as a flight scientist during ACAPEX in 2015. She contributed analyses and assisted data archival efforts of the overall CalWater efforts during Year 1 of this grant. Dr. Suski was subsequently hired to a position within EMSL at the DOE Pacific Northwest National Laboratory in July 2016.

2) Dr. Ezra Levin was mentored as a postdoctoral scientist by Dr. DeMott in his major analyses and archival of the G-1 aircraft data sets, and was supported to attend the American Geophysical Union conference in 2015. Dr. Levin was hired as a research scientist during the time frame of this grant and continued efforts until the term of this grant.

3) Dr. Jun Uetake, a specialist in microbiology and sequencing analyses of environmental samples, was hired as a postdoctoral scientist during the proposal period, and contributed toward aerosol DNA analyses duing the final year of study.

4) Dr. Christina McCluskey joined the project for the last stages of her PhD research. She participated in data collection during 2015 studies, collaborated on publications with UCSD and NCSU colleagues, successfully applied for an SCGSR award to extend her research in modeling studies under the mentorship of Dr. Susannah Burrows at PNNL during 2017, and is presently a postdoctoral scientist at NCAR.

5) Mr. Kevin Barry contributed toward cloud microphysical analyses during the initial stages of his M.S. studies, and will ultimately contribute to the publication that ensues from this effort.

6) Ms. Katie Rocci worked in our laboratory as an undergraduate summer intern of the NSF Center for Multiscale Modeling of Atmospheric Processes, and was mentored by Dr. Hill for conducting INP measurements that included some samples from the Bodega Bay and Ron Brown data bases. She presented hr results at the AGU Fall Meeting in 2015. She is attending Colorado State University toward a Ph.D. in Environmental Ecology.

7) Ms. Anne Marie Rauker worked as an undergraduate student hourly employee from 2016 to 2018, conducting analyses on all ice spectrometer data sets. She graduated to attend Indiana University for combined studies in Atmospheric Sciences and Environmental Policy.

3.0 Publications and References

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