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Cloud, Aerosol, and Complex Terrain Interactions (CACTI) ARM Mobile Facility (AMF) Measurements of Ice Nucleating Particles Field Campaign Report

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ARM

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Acronyms and Abbreviations

AAF	ARM Aerial Facility
AMF	ARM Mobile Facility
AOS	aerosol observing system
ARM	Atmospheric Radiation Measurement
CACTI	Cloud, Aerosol, and Complex Terrain Interactions
CSU	Colorado State University
DI	deionized
G-1	Gulfstream-159 aircraft
INP	ice nucleating particle
IS	ice spectrometer
PCR	polymerase chain reaction
SL	standard liters

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1.0 Summary

During the Cloud, Aerosol, and Complex Terrain Interactions (CACTI) Experiment, a project with the overarching goal to improve understanding of cloud life cycle and organization in relation to environmental conditions so that cumulus, microphysics, and aerosol parameterizations in multi-scale models can be improved, our group was tasked with providing and assisting the collection of aerosol filter samples for measuring ice nucleating particle (INP) concentrations. This included ground-based and aircraft measurements in north central Argentina. This report details the efforts and results from the Atmospheric Radiation Measurement (ARM) first Mobile Facility (AMF1).

The INP measurements were especially focused around research needs for addressing the major CACTI science questions regarding the role of aerosols as one factor affecting the properties and life cycles of orographically generated cumuli, and the initiation of deep convection and mesoscale organization. As the primary means for first initiation of the ice phase in clouds, absent remnant ice particles from prior convection or overseeding from higher clouds (cirrus) where homogeneous freezing can occur, the abundance of INPs can play a vital role in the formation of precipitation. The INP data collected should ultimately find use in future investigations, especially in relational analyses to other Gulfstream-159 (G-1) aircraft measurements of aerosol properties and location with respect to storm systems. This data can serve as the basis for developing and improving numerical model parameterizations of ice nucleation.

Within efforts to measure INPs in CACTI, the AMF1 INP measurements provided the unique ability to capture an extended seasonal cycle of INPs over austral spring to fall. The AMF1 site was located near Villa Jacanto, Argentina, (32.12°S, 64.75°W), approximately 20 km east of the highest ridge top in the Sierras de Córdoba range at an elevation of approximately 1150 m. Within this region, a vast array of aerosol influences was expected to be encountered at various times/seasons, from local soil and plant emissions, long-range-transported desert dusts, regional pollution, and biomoass burning.

This report describes the installation, collections, processing, and archiving of data from this effort. Images of the filter sampler and its mounting on the aerosol observing system (AOS) trailer are shown in Figure 1. Pre-cleaned and pre-sterilized 47-mm Nuclepore polycarbonate filters (0.2 μm pore size, backed by clean 0.45 μm pore size filters) were mounted in plastic holders that were open to the atmosphere. Single-use filter units were provided in sealed plastic by our research team (Figure 2), along with training materials for the group of ARM technicians assisting AMF1 measurements. Filters were typically drawn for an 8-hour period, totaling 6000 liters sampled on average, measured with a mass flow meter. A total of 83 sample filters were collected over seven months, including six blanks (installed with no flow) at intervals throughout the project. Filters were stored temporarily in sealed petri dishes in a -20°C freezer prior to return to Colorado State University (CSU) at the end of the campaign with a dry nitrogen shipper (Cryoport.com).

Figure 1. AMF1 site, looking northwest toward the Sierras de Córdoba range behind the trailers (left), with the filter unit in place atop the AOS trailer, underneath the half-spherical rain hat. In this position, it sat only 2 m below the AOS inlet.

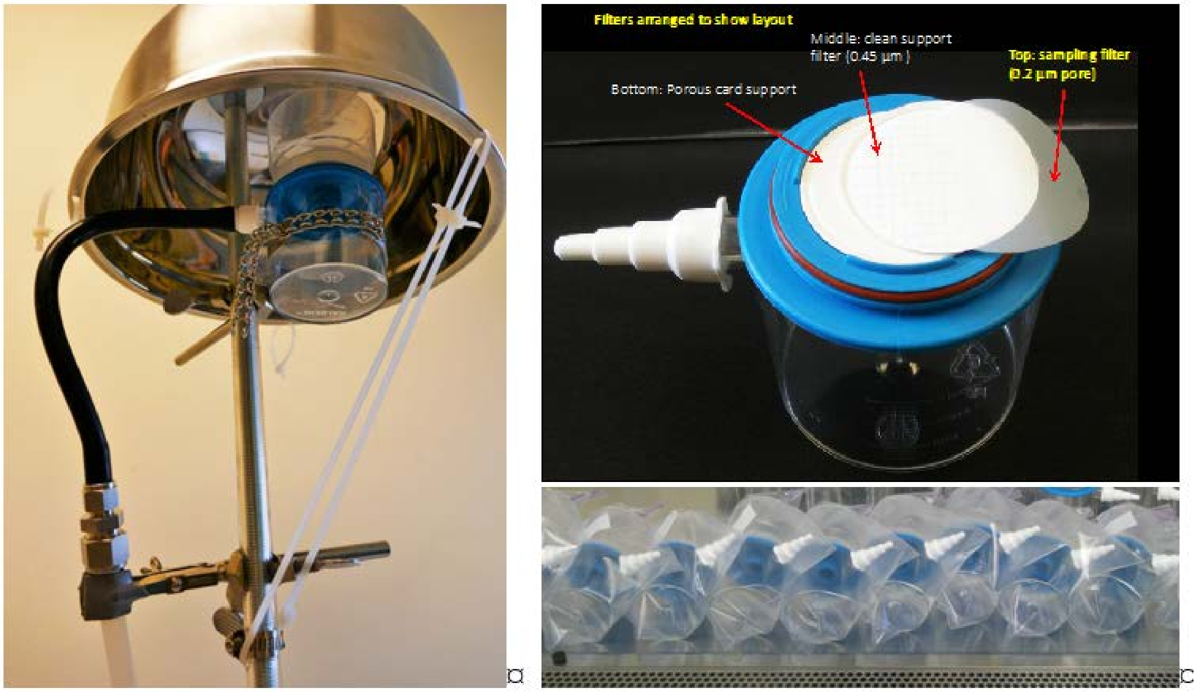

Figure 2. Filter sampler unit close up within rain hat (left), pre-cleaned and pre-sterilized filter configuration as arranged before shipping (upper right) and cleaned and bagged units as shipped to Argentina prior to opening for sampling each day (bottom).

Initial processing to obtain spectra of INP number concentration active via the immersion freezing mechanism versus temperature was conducted using the CSU ice spectrometer (IS) instrument system (McCluskey et al. 2018). For processing, each filter was placed into a 50 mL Falcon polypropylene centrifuge tube with 7-8 mL of 0.1 µm-filtered deionized (DI) water and shaken in a Roto-Torque rotator for 20 min to create a suspension. Thirty-two aliquots of 50 µL (i.e., 1.6 mL) of each sample, plus a series of dilutions, were then dispensed into polymerase chain reaction (PCR) trays that were then fitted into aluminum blocks in the IS. Samples were cooled at a rate of approximately 0.33°C min^{-1}. Freezing temperatures of wells were recorded using a camera and software system on each of three IS instrument systems. The lowest freezing temperature archived for each sample was defined by the temperature for which the number of sample wells frozen significantly exceeded those frozen in a 32-well, 0.1 µm-filtered DI water blank tested simultaneously in the same tray. This final temperature was generally between -26 and -29°C for the CACTI AMF1 sample set. Cumulative INP concentrations were determined by first calculating the INPs per mL of suspension based on Vali (1971) and then converting to concentration per standard liter of air using the proportion of the total liquid sample dispensed and the air sample volumes. The number of INPs on the average of all blank filters that had been handled and processed identically, with exception of air flow, were subtracted from the calculated number of INPs on each sample filter (Figure 3) before the conversion to number concentration per standard liter. The large sample volumes of filters led to numbers collected on filters that exceeded blank filter background numbers by a minimum of two orders of magnitude. Confidence intervals (95%) for binomial sampling were calculated based on Agresti and Coull (1998).

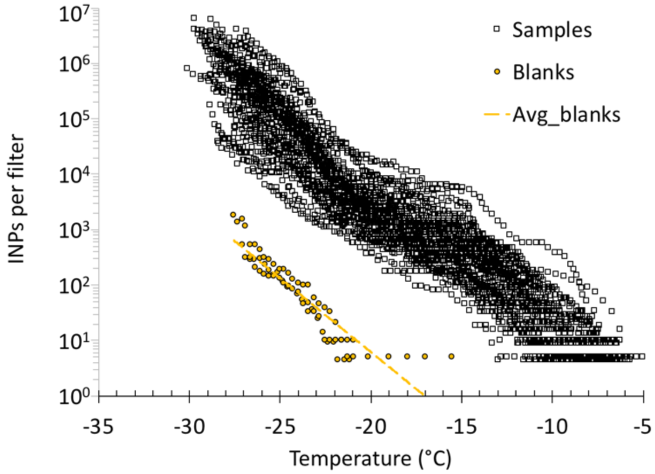

Figure 3. INP number per filter for all unamended samples and blanks, and the average fit to correct data over the study period.

To gain insights in the biological proportion of INPs, a portion of a selected number of original suspensions was heated to 95°C for 20 min, prior to determining the immersion freezing temperature spectra. This thermal treatment should denature most heat-labile organics, such as proteins. Hydrogen peroxide (H_2O_2) digestions were also performed on a selected proportion of suspensions to remove all organic carbon INPs following methods detailed in McCluskey et al. (2018) and Suski et al. (2018). This was typically done for the same filter samples for which thermal treatments were done. The peroxide treatment is also done at 95°C, incrementally removing all remaining organics. The difference in the INP concentrations at a particular temperature after heat or H_2O_2 treatment determines the contributions of biological and organic INP types, respectively, for each sample period.

For archival and completion of tasks under this ARM proposal, 60 of the 83 original filter particle collections were processed for basic temperature spectra, with 29 of these 60 also tested for thermally removing microbial/proteinaceous contributions toward INPs and 28 of those 29 also tested for removal of all organic carbon (Table 1). Metadata for processed filters is shown in Table 2. All data have been added to the ARM Data Center.

Table 1. Comparison of the planned collections to those obtained and processed.

Campaign		Base	Blanks	95 C	H_2O_2	Processes
CACTI AMF	Anticipated	90				
	Promised	60		29*	28*	
	Obtained	83	6			
	Processed	60	4	29	28	121

* = promised processing of 1/3 of total collected

Table 2. Metadata for processed CACTI AMF1 filters, where SL is standard liters collected.

Latitude: -32.126306

Longitude: -64.728514

Altitude (m): 1147 (at height of rain hat)

Date (US)	Filter no.	Start day	Start mo.	Start year	Start time (UTC)	Stop time (UTC)	Total SL (1013 hPa, 273 K)	Weather notes at start of sampling period
10/5/18	1	5	10	2018	11:30:00	20:30:00	7162	Cloudy; temp = 11C; Wind =3.8 m/s-23degree; RH =50%
10/8/18	2	8	10	2018	11:30:00	20:30:00	6741	Sunny; temp =26C; Wind =1.3m/s-300degree; RH =21%
10/11/18	3	11	10	2018	11:30:00	20:30:00	7212	Cloudy/small rain; temp =6.5C; Wind =5m/s-30degree; RH =60%
10/14/18	4	14	10	2018	11:00:00	20:00:00	7179	Cloudy; temp =14.9C; Wind =3.2m/s-228.9degree; RH =61.4%
10/17/18	5	17	10	2018	11:30:00	20:30:00	6944	Partly cloudy/drizzling; temp =20C; Wind =4m/s-270degree; RH =50%
10/20/18	6	20	10	2018	11:00:00	20:00:00	7218	Fog; temp =13C; Wind =5m/s-30degree; RH =91%
10/23/18	7	23	10	2018	11:00:00	20:00:00	7556	Fog; temp =10.6C; Wind =1.9m/s-291.2degree; RH =97.6%
10/26/18	8	26	10	2018	11:30:00	20:30:00	6416	Cloudy; temp =13.8C; Wind =2.4m/s-300.3degree; RH =84.5%
10/30/18	9	30	10	2018	11:30:00	20:30:00	6416	Fog; temp =14C; Wind =2.8m/s-190degree; RH =90%
11/2/18	11	2	11	2018	11:30:00	20:30:00	6784	Clear Sky; temp =16.3C; Wind =3.8m/s-350.4degree; RH =73.1%
11/2/18	12	2	11	2018	20:30:00	8:30:00	9511	Clear Sky; temp =22C; Wind =3.9m/s-110degree; RH =62%
11/6/18	15	6	11	2018	11:55:00	16:15:00	6556	Fog; temp =16.3C; Wind =2.1m/s-172.4degree; RH =99.5%
11/10/18	16	10	11	2018	12:00:00	16:00:00	6186	Clear Sky; temp =24.4C; Wind =2.4m/s-78.7 degrees; RH =64.6%
11/12/18	18	12	11	2018	14:00:00	20:00:00	4661	Raining; temp =16.4C; Wind =3.8m/s-220.5 degrees; RH =100.1%
11/14/18	19	14	11	2018	14:00:00	18:00:00	3356	Sunny; temp =19.1C; Wind =8m/s-230 degrees; RH =22%
11/15/18	20	15	11	2018	13:00:00	17:00:00	3208	Clear Sky; temp =24.0C; Wind =1.9m/s-131.9 degrees; RH =23.4%
11/16/18	21	16	11	2018	14:00:00	18:00:00	3184	Mostly Cloudy; temp =16.4C; Wind =2.7m/s-64.9 degrees; RH =88.9%
11/17/18	22	17	11	2018	12:00:00	16:15:00	3409	Partly Cloudy; temp =26.3C; Wind =7.2m/s-269.3 degrees; RH =39.8%
11/20/18	23	20	11	2018	16:00:00	20:00:00	3249	Clear Sky; temp =25C; Wind =2m/s-150 degrees; RH =40%
11/21/18	24	21	11	2018	15:00:00	19:00:00	3208	Clear Sky; temp =26.6C; Wind =3.2m/s-44.5 degrees; RH =43%
11/22/18	25	22	11	2018	14:00:00	18:00:00	3257	Raining; temp =14C; Wind =7.2m/s-181 degrees; RH =75.6%
11/24/18	26	24	11	2018	16:00:00	20:00:00	3208	Partially Cloudy With Sun ; temp =18.1C; Wind =2.8m/s-120 degrees; RH =55.6%
11/25/18	27	25	11	2018	16:00:00	20:00:00	3251	Partially Cloudy with Sun ; temp =21C; Wind =7.7m/s-57 degrees; RH =29.1%
11/26/18	28	26	11	2018	13:00:00	17:00:00	3263	Raining ; temp =10C; Wind =3.1m/s-5 degrees; RH =94%
11/28/18	30	28	11	2018	14:00:00	19:00:00	3965	Partially cloudy ; temp =17C; Wind =3.5m/s-73 degrees; RH =70%.
11/29/18	31	29	11	2018	12:00:00	17:00:00	4010	Partially cloudy ; temp =16C; Wind =4m/s-350 degrees; RH =77%
12/1/18	33	1	12	2018	14:00:00	18:00:00	3247	Cloudy ; temp =12C; Wind =3.6m/s-120 degrees; RH =50%
12/2/18	34	2	12	2018	12:00:00	17:00:00	4315	Cloudy ; temp =9C; Wind =3m/s-25 degrees; RH =72.3%
12/3/18	35	3	12	2018	16:00:00	20:00:00	3208	Clear Sky ; temp =19C; Wind =1.6m/s-155 degrees; RH =53.4%
12/4/18	36	4	12	2018	16:00:00	20:00:00	3208	Partially cloudy with sun ; temp =24.3C; Wind =6.4m/s-148 degrees; RH =31.6%
12/5/18	37	5	12	2018	12:00:00	16:23:00	3425	Mostly cloudy, no sun ; temp =16.5C; Wind =1.6m/s-262 degrees; RH =61.5%
12/7/18	38	7	12	2018	15:00:00	19:00:00	3131	Partially cloudy with sun ; temp =19C; Wind =2.3m/s-19 degrees; RH =50.3%
12/8/18	39	8	12	2018	16:00:00	20:00:00	3057	Sunny ; temp =21.5; Wind =9m/s-45 degrees; RH =45.6%
12/13/18	40	13	12	2018	12:00:00	20:00:00	6231	Mostly cloudy, no sun; temp =18.2C; Wind =5.3m/s-15 degrees; RH =93.8%
12/17/18	42	17	12	2018	12:00:00	20:00:00	6368	Partially cloudy; temp =20.3C; Wind =4m/s-170 degrees; RH =65%.
12/20/18	43	20	12	2018	12:00:00	20:50:00	6964	Partially cloudy; temp =19C; Wind =3.5m/s-100 degrees; RH =69%
12/24/18	44	24	12	2018	13:00:00	21:00:00	6174	Sunny; temp =23.5C; Wind =5.1m/s-56 degrees; RH =37.4%
12/31/18	46	31	12	2018	12:30:00	20:30:00	6157	Partially cloudy; temp =18.2C; Wind =6.2m/s-22 degrees; RH =34.8%
1/3/19	47	3	1	2019	14:00:00	20:00:00	4622	Mostly cloudy; temp =18.3C; Wind =2m/s-70 degrees; RH =29.9%
1/7/19	49	7	1	2019	12:00:00	20:00:00	6416	Clear sky; temp =20C; Wind =1m/s-173 degrees; RH =50%
1/14/19	51	14	1	2019	12:00:00	20:00:00	6426	Mostly Cloudy, No Sun; temp =17C; Wind =0m/s-99 degrees; RH =80%
1/17/19	52	17	1	2019	12:00:00	20:00:00	6622	Raining; temp =10C; Wind =7m/s-171 degrees; RH =92%
1/21/19	53	21	1	2019	12:00:00	20:00:00	6416	Clear sky; temp =22.5C; Wind =4.4m/s-154 degrees; RH =50.4%
1/24/19	54	24	1	2019	12:00:00	21:00:00	6484	Fog; temp =21.6C; Wind =3.6m/s-344 degrees; RH =99.5%
1/29/19	55	29	1	2019	12:00:00	20:30:00	6817	Cloudy =19C; Wind =4.5m/s-120 degrees; RH =85%
2/1/19	56	1	2	2019	12:00:00	20:00:00	6042	Clear sky; temp =24.2C; Wind =1.5m/s-226 degrees; RH =69.9%
2/7/19	59	7	2	2019	13:00:00	20:00:00	5503	Clear sky; temp =21.4C; Wind =0.7m/s- 11degrees; RH =50%
2/14/19	61	14	2	2019	12:00:00	20:00:00	6158	Clear sky; temp =19C; Wind =1.2m/s- 185degrees; RH =51%
2/21/19	63	21	2	2019	11:30:00	19:30:00	6084	Clear sky; temp =22.8C; Wind =3.2m/s- 41degrees; RH =75.6%
3/1/19	65	1	3	2019	12:00:00	20:00:00	6053	Clear sky; temp =24.9 C; Wind =2m/s-55degrees; RH =26%
3/7/19	68	7	3	2019	12:00:00	20:00:00	6416	Clear sky; temp =23 C; Wind =6.4m/s-160degrees; RH =60%
3/16/19	70	16	3	2019	12:00:00	20:00:00	6960	Partly cloudy, sun; temp =16.8 C; Wind =3.5m/s-41.degrees; RH =75.1%
3/19/19	71	19	3	2019	12:00:00	20:00:00	6621	Mostly cloudy, no sun; temp =12.7 C; Wind =3.3m/s-131.1degrees; RH =91.6%
3/26/19	73	26	3	2019	12:00:00	20:00:00	6641	Cloudy, no sun; temp =8.5 C; Wind =2.4m/s-73degrees; RH =93.2%
3/31/19	75	31	3	2019	12:00:00	21:00:00	7218	Fog; temp =17.5 C; Wind =8m/s-19degrees; RH =94.3%
4/2/19	77	2	4	2019	12:00:00	20:00:00	6167	Sunny; temp =24 C; Wind =3.2m/s-19degrees; RH =46%
4/8/19	79	8	4	2019	13:00:00	22:00:00	7477	Sunny; temp =19.2 C; Wind =2.6m/s-45degrees; RH =30.1%
4/12/19	81	12	4	2019	12:00:00	22:30:00	7071	Sunny; temp =17.2 C; Wind =0.2m/s-165degrees; RH =54.7%
4/24/19	85	24	4	2019	12:00:00	20:00:00	6416	Drizzling; temp =9.1 C; Wind =4.3m/s-341degrees; RH =93.3%
4/28/19	89	28	4	2019	12:00:00	20:00:00	8096	Sunny; temp =19 C; Wind =2.1m/s-210degrees; RH =20%

2.0 Results

Results are at an early stage of evaluation, as final processing was completed just prior to the drafting of this report. In Figure 4, the overall campaign INP temperature spectral results are represented for samples that were unamended or treated to indicate INP compositions. This represents one of the most comprehensive such databases accumulated for a continental site anywhere. The results reveal a preponderance of apparent biological INPs present at the surface in this region of Argentina, with a special role by them in accounting for immersion freezing INPs active in the temperature regime higher than -20°C. These biological INPs are largely responsible for the “hump” in INP activity that leads the INP spectra to diverge positively from exponential at these higher temperatures, consistent with other reports attributing such impact to this category of INPs (Hill et al. 2018, O’Sullivan et al. 2018). Other organic INPs contribute to a lesser degree, accounting for the rest of the >-20°C hump and (surprisingly) dominating over inorganic INPs (presumed as those left after peroxide treatments of suspensions) in the temperature range somewhat lower than -20°C (on average to -22°C). The INP spectra of the inorganic populations of INPs, indicated by the INPs remaining after H_2O_2 treatments in Figure 4, are highly exponential versus temperature, with an approximate one-order-of-magnitude increase in atmospheric concentrations for each 4°C of cooling. The temperature spectra, represented by the Δ[INP]/dT following treatments, are remarkably consistent with laboratory measurements made on Argentinian soil dust from La Pampa province, the province just south of Córdoba province, reported by DeMott et al. (2018).

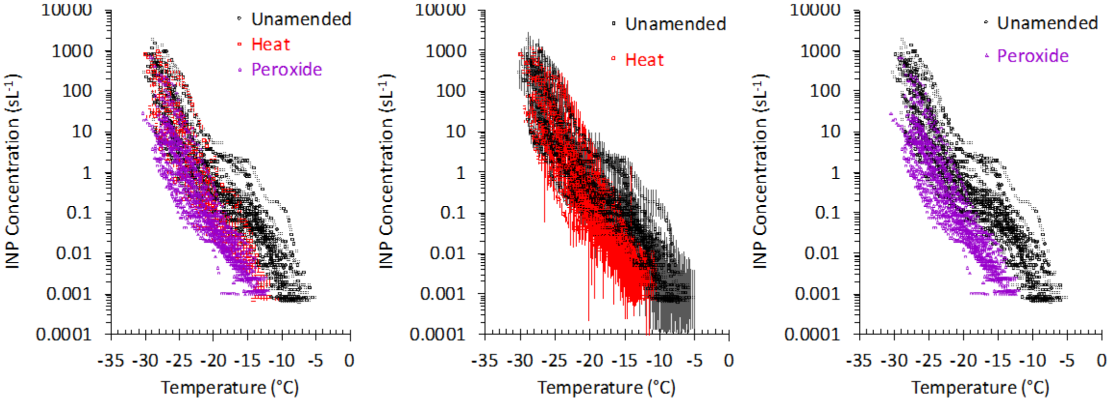

Figure 4. Ice nucleating particle number concentrations versus temperature for all multiple-processed samples from the AMF1 (November 2018 to March2019). From left to right are all processes, unamended and thermally treated (95°C) only, and unamended and H_2O_2 treated only processes. Uncertainties are shown only for the middle panel. Results show the ubiquitous presence of biological and organic INPs in different temperature regimes, as discussed in the text.

Figure 5 shows all results obtained by month. The most intensive sampling and processing occurred for the months of the AAF aircraft campaign in November and December of 2018. These results demonstrate that the overall campaign results are reflected in every month. Plotting a timeline of INP concentrations at 5°C intervals, as in Figure 6, indicates that although some of the highest INP concentrations were achieved during the AAF intensive period in late spring, there was no apparent seasonal cycle at the AMF1 site at any processing temperature. This may in part reflect the moderate climate of the region, and the roles of agriculture (predominance of pasture), evergreen native shrubland, and wind action on INP populations.

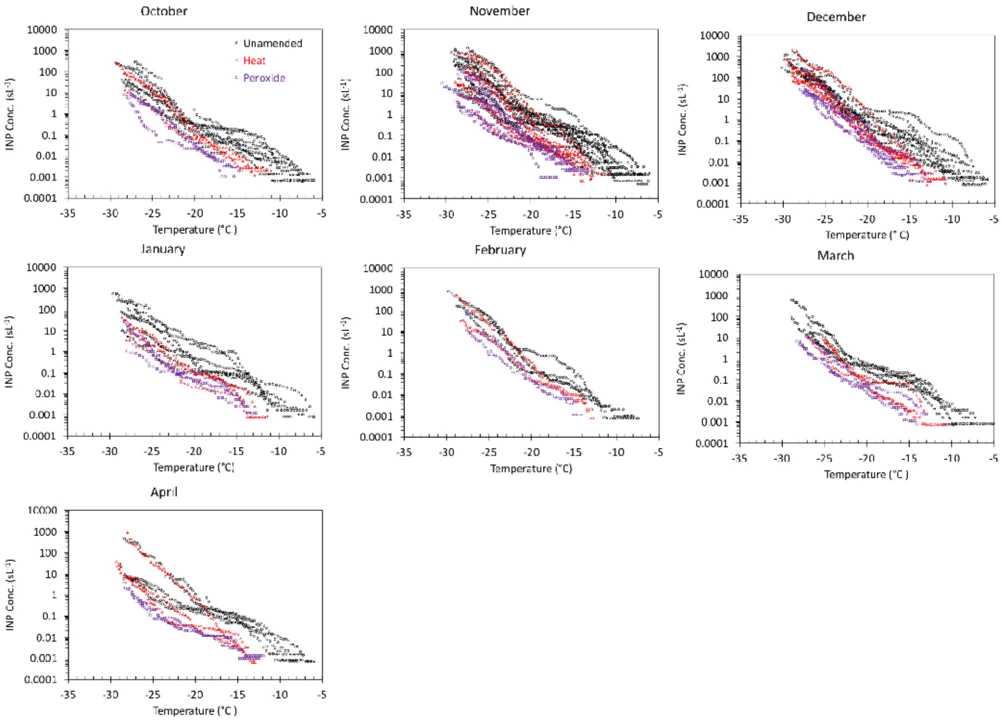

Figure 5. INP spectra by month, including all treatments. More observations were made during the AAF intensive operational period in November through December.

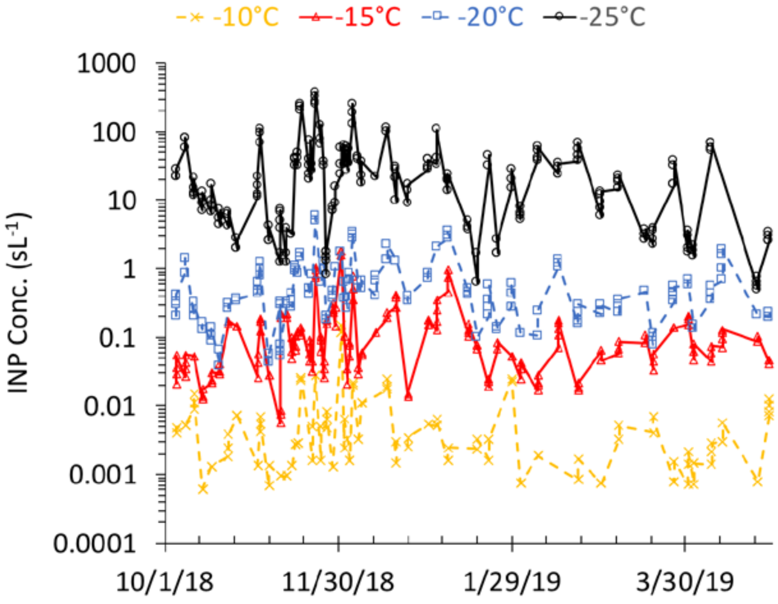

Figure 6. Timeline of INP concentrations at selected temperatures, indicating high variability at any temperature over time, but no clear seasonal cycle. The highest INP concentrations of the project occurred during the late spring period, in the midst of the intensive AAF campaign and more frequent sampling at the AMF1.

Finally, in Figure 7 we provide a first, deeper look at how the data set could serve for research into the makeup and variability of the INP population, and how this might ultimately inform advanced observational and numerical modeling studies using the CACTI AMF1 data set. Here we have defined the different INP types and their regimes of contribution on the basis of treatments. The INP_bio, INP_org, and INP_inorg categories are noted in Figure 7 as those reduced by heat, peroxide, and remaining after the final peroxide treatment, respectively. Future work could dissect these contributions and analyze for the factors that influence each. Notable is the presence of INP_bio in both the austral spring and fall.

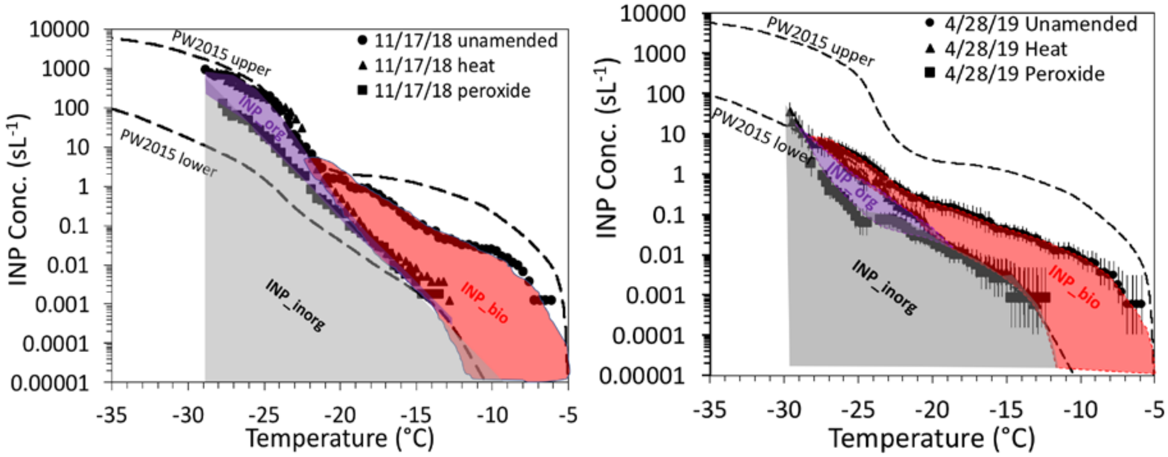

Figure 7. Single-day INP spectra from spring (left) and fall (right) filter samples, with interpreted segments of the populations that are biological (INP_bio), other organic (INP_org), and inorganic (INP_org) INPs. Overlain are the immersion freezing bounding curves for precipitation and cloud water samples compiled by Petters and Wright (2015), and assuming cloud water content of 0.4 g m^{-3}.

The smaller contribution of INP_org, primarily at lower temperatures, is also seen uniformly by season in the overall data set. Baseline inorganics, presumably from regional airborne soil dust, are also nearly constant. For reference, the bounding values of immersion freezing INPs made on the basis of measurements made on precipitation and cloud water by Petters and Wright (2015) are also shown. The CACTI data tended to fill the bounding curves. These are the first such comprehensive data from southern South America for comparison to global compilations.

3.0 Publications and References

No publications have been prepared at the time of this report. First presentation of results is planned for the Department of Energy ARM/Atmospheric System Research Principal Investigators meeting in June 2020. Publications are in preparation, and advanced analyses have been proposed at the time of this report. References follow.

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