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Examining the Ice-Nucleating Particles from the Southern Great Plains Field Campaign Report

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Acronyms and Abbreviations

APS	aerosol particle sizer
ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
CSU-SFDC	Colorado State University Continuous Flow Diffusion Chamber
DOE	U.S. Department of Energy
ENA	Eastern North Atlantic
ExINP-SGP	Examining the Ice-Nucleating Particles from Southern Great Plains
INP	ice-nucleating particles
INSEKT	Ice Nucleation Spectrometer of the Karlsruhe Institute of Technology
NSA	North Slope of Alaska
PINE	Portable Ice Nucleation Experiment
SGP	Southern Great Plains
SMPS	scanning mobility particle sizer

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1.0 Summary

The recent U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility field campaign, named Examining the Ice-Nucleating Particles from Southern Great Plains (ExINP-SGP), targeted experimentally characterizing abundance and other properties of ambient ice-nucleating particles (INPs) at the SGP site in Oklahoma (36° 36′ 18″ N, 97° 29′ 6″ W) during the period of October 1 to November 14, 2019. This campaign was funded through the DOE Office of Science Early Career Research Program (DE-SC0018979) as one of three major campaigns to be conducted at three ARM observatories, including the Eastern North Atlantic (ENA) station in 2020−2021 and North Slope of Alaska (NSA) site in 2021−2023, besides SGP. Different INP episodes were assessed to develop an SGP-relevant ice nucleation parameterization that would help understand convective and mixed-phase cloud systems typically observed in this region. The overall scopes of this campaign included:

- Elucidating sources, abundance, chemical composition, and ice nucleation processes of high-temperature (*T*) INPs above -15 °C at the SGP site,
- Examining if immersion and/or condensation freezing, requiring a water saturation condition or cloud condensation nuclei (CCN) activation prior to ice nucleation, is a more predominant ice nucleation mechanism at the SGP site, and
- Determining what ice nucleation pathway is the most sensitive to the chemical mixing state of ambient aerosol at the SGP site.

These research scopes are directly aligned with the research topic under the DOE Earth and Environmental System Sciences program about enhancing the unique capabilities of the ARM facility to advance the frontiers of earth and environmental science and addressing science gaps regarding Earth's radiative energy balance. This field campaign opportunity provided a foundation to advance the mission of ARM particularly related to aerosol-cloud ice interactions. Currently, ice formation processes are very poorly represented in the climate models (Ghan and Schwartz 2007). To complement this current deficit, we conducted 45 days of online INP measurements at the SGP site during the reported campaign. Such state-of-the-art representations of INPs from SGP and other ARM sites to the atmospheric modeling community might improve knowledge of ice nucleation processes and its formulation in the Earth's energy balance.

This report describes field results of ambient INP measurements by the commercialized version of the Portable Ice Nucleation Experiment chamber (PINE) from the ExINP-SGP campaign. Images of our experimental setup at the site and remote operation interface are shown in Figure 1. PINE allows simulating an adiabatic expansion process in a ≈10 L aluminum vessel, where artificial cloud is formed at *T*s relevant to atmospheric heterogeneous ice nucleation with ±1.5 °C *T* uncertainty. A time resolution of ≈8 minutes for each expansion experiment is suitable to continuously monitor ambient INPs and study atmospheric ice nucleation (Möhler et al. in review). With its optical particle detection capability of ≈0.3 INP L^{-1}, we conducted remote INP measurements for most of the campaign period. Overall, the PINE's ability to make remote INP measurements has been demonstrated during this campaign. As a result, a set of two INP parameterizations, such as cumulative number concentration of INPs per volume of air at a certain T ($n_{INP}(T)$, e.g., DeMott et al. 2017) and ice nucleation active surface site density as a function of T ($n_s(T)$ e.g., Connolly et al. 2009), have been developed. Furthermore, we analyzed the

relationship between aerosol particle size and INP abundance. At SGP, we found an increase in INPs with pulse episodes of super-micron particles, especially for diameters of > 2 µm, across a wide range of heterogeneous freezing T examined by PINE (-35 °C < T < -10 °C). To complement the online PINE measurements, atmospheric aerosol particles have been collected on filter and/or in an impinger. Collected aerosol particles were analyzed for immersion freezing with multiple offline freezing techniques. With some further analyses, the water activity-based freezing parameterization (e.g., Knopf and Alpert 2013) will be described in the future.

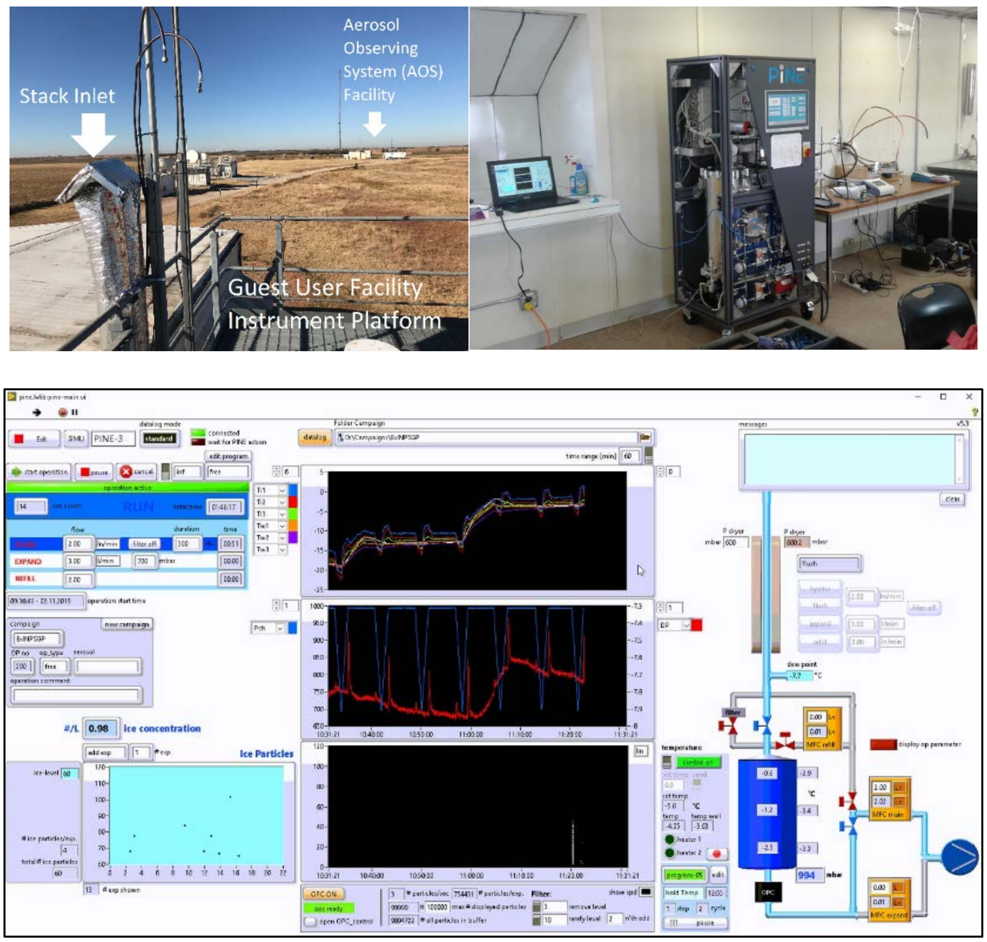

Figure 1. The ExINP-SGP campaign was carried out at the ARM SGP Central Facility using the Guest User Facility. A homemade quasi-laminar sampling stack was mounted to the instrument platform (a), allowing an intake of particle-laden air by PINE deployed inside the Guest User Facility (b). For most of the campaign period, the PINE chamber was remotely controlled using the LabView interface through the BeyondTrust remote-access console (c).

2.0 Results

The remote operation of PINE at SGP was successful throughout the campaign. Overall, the ExINP campaign produced more than 30,000 meaningful PINE-measured INP data points. Continuous measurements of INPs for >40 days, including more than two weeks of complete remote operations from West Texas, without any substantial down time, is quite remarkable in the cloud microphysics research community. All results are at a preliminary stage of evaluation. However, the overall success of the

ExINP-SGP campaign is represented in Figure 2. As seen in the figure, PINE is susceptive to the *T* below -10 °C for INP> 0.2 L^{-1}. The *T* distribution in the vessel was carefully assessed, and only reasonable INP data collected with the *T* deviation ≤ ±0.4 °C were used in Figure 2.

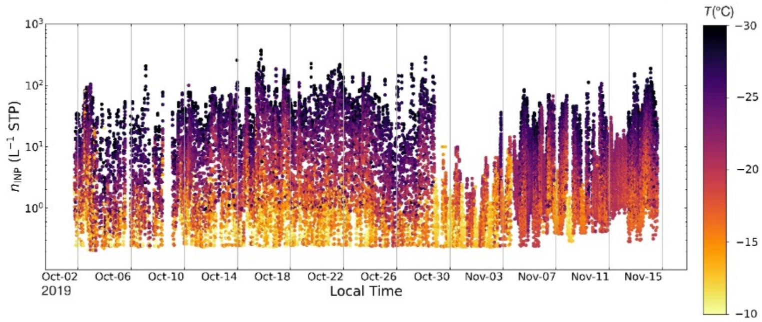

Figure 2. Time series of the measured chamber *T* and associated INP concentration with a *T* interval of 0.5 °C from October 1 to November 14, 2019. PINE is equipped with three vertical *T* sensors (i.e., top, middle, and bottom) to measure its air *T*s across its expansion vessel, and the color scale represents the chamber vessel middle air *T*.

A compilation of INP concentration spectra as a function of *T* in the range of -30 °C < *T* < -10 °C is presented in Figure 3. For simplicity in visual appearance of the figure, only INP concentrations at the end of individual expansions are shown for PINE. Note that the PINE data density can be substantially increased if all 0.5 °C *T*-resolved data during each expansion are included (Figure 2). Figure 3 also offers a qualitative comparison between the ExINP INP spectra to the Colorado State University Continuous Flow Diffusion Chamber (CSU-CFDC) data set of the Southern Great Plains Ice Nuclei Characterization Experiment campaign held in May to June, 2014. Though the measurement techniques and measured seasons were different in these campaigns, a reasonable consistency of INP concentrations, ranging from ≈0.1 to 100 at -15 °C and -30 °C, respectively, was found at overlapping *T*s. Again, this comparison is only qualitative and not conclusive. It is, however, noteworthy that there has been only one online INP measurement campaign carried out at SGP prior to the 2019 ExINP campaign.

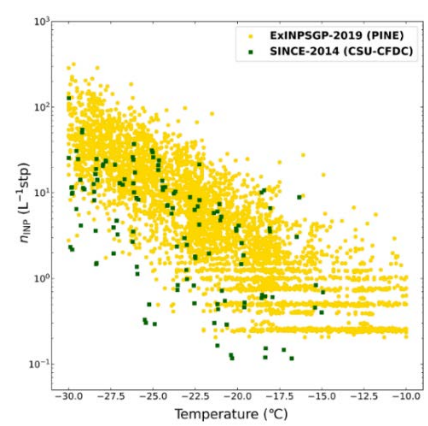

Figure 3. Comparison of the processed ExINP-SGP data to the previous SGP INP data from the Southern Great Plains Ice Nuclei Characterization Experiment held in 2014. The CSU-CFDC data are adapted from the DeMott et al. (2015).

Next, Figure 4 shows the preliminary result of $n_s(T)$ over the ExINP campaign period. For this ice nucleation parameterization, our PINE-measured INP concentrations (Figure 2) were scaled to the total surface area measured by a combination of ARM's scanning mobility particle sizer (SMPS) and aerosol particle sizer (APS) instruments. The data of these two size-distribution-measuring instruments were merged in a volume-equivalent-diameter metric by presuming a constant dynamic shape factor of 1.3 and aerosol particle density of 2.5 g cm^{-3}, which may represent the ExINP condition (personal communication with Dr. Konrad Kandler with TU Darmstadt). The total surface area values were computed based on the 5-minutes time-resolved ARM Discovery data and were interpolated to each $n_{INP}(T)$ measurement time of PINE's individual expansion experiments (i.e., median time while flushing/injecting aerosol particles into the chamber vessel – see the README file of our PINE data).

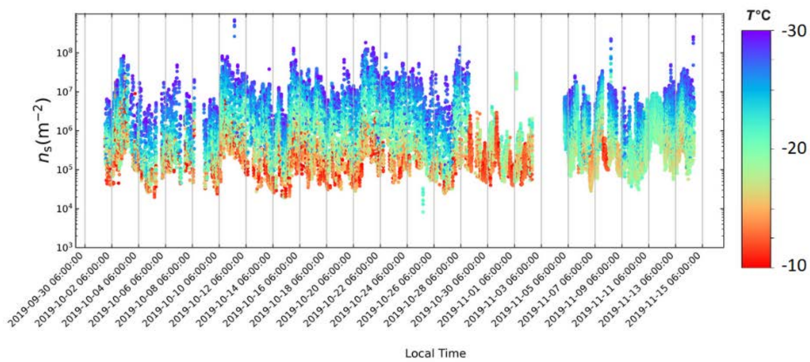

Figure 4. Time series of $n_s(T)$ with a T interval of 0.5 °C from October 1 to November 14, 2019. The color scale represents the chamber vessel middle air T.

As seen in Figure 4, a number of lower and upper outlier data points appearing in the $n_{INP}(T)$ plot reduces after normalizing to the integrated surface, suggesting that aerosol particle surface and associated large particles may play a crucial role in ice nucleation at SGP. Due to non-negligible particle loss of >5 µm in the PINE system, our preliminary n_s analysis was conducted by accounting the ARM-APS-measured aerosol particle size up to 5 µm in diameter. Note that the particle loss of aerosol particles to PINE, SMPS, and APS must be carefully assessed further, and the final n_s values may alter because of these loss adjustments. Some suspicious n_s spikes appear on our preliminary plot; e.g., 10/11, 10/25, 11/3, 11/8, 11/12, and 11/14. As our INP concentrations (Figure 2) do not capture these spikes, the ARM instruments might be responsible for these outliers. The Principal Investigator is currently scheduling a data discussion meeting with an ARM data translator to determine the cause of these spikes.

Due to PINE's continuous INP-measuring capability and its high time resolution, $n_s(T)$ spectra can be generated over several hours. For instance, Figure 5 displays the compilation of 12 hours $n_s(T)$ spectra (0:00-11:59 and 12:00-23:59) for two days; 10/21 and 10/25 in 2019. These two days were chosen as snapshot examples because these days had contrasting ambient particle abundances. Briefly, abundant super-micron particles persisted throughout the day on 10/21, and a relatively clean condition with less particles was observed on 10/25 after some precipitation. The observed average aerosol particle surface area concentrations were ≈10^4 µm^2 cm^{-3} and ≈10^3 µm^2 cm^{-3} on the 21st and 25th, respectively.

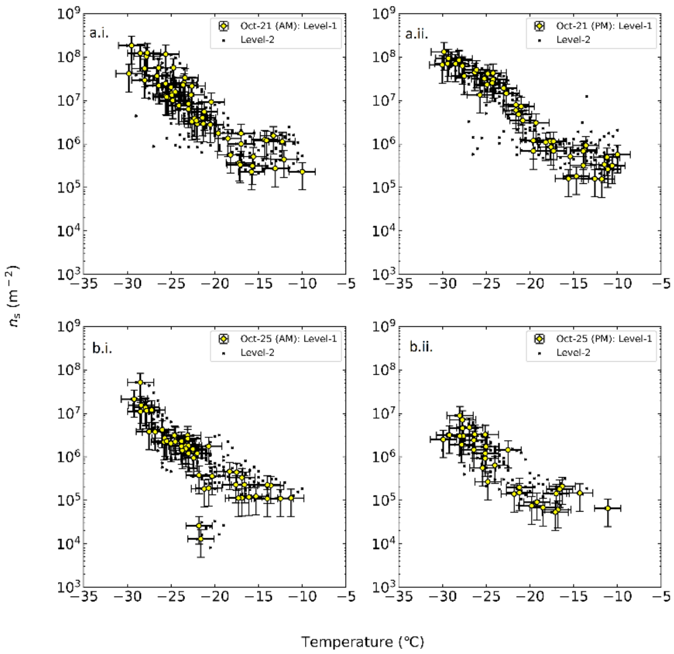

Figure 5. A summary of 12 hours $n_s(T)$ spectra for two days: 10/21 (a) and 10/25 (b) in 2019 (0:00-11:59, i, and 12:00-23:59, ii). The current PINE uncertainty with respect to T is ± 1.5-ºC (based on the laboratory tests with aerosol particles that have known compositions and ice nucleation properties). The systematic error for the PINE-estimated $n_s(T)$ is approximately 62%. The Poisson statistical INP mean concentrations and errors can be defined in terms of a 95% confidence interval (subject to be updated).

As seen in Figure 5, the $n_s(T)$ values from 10/25 are overall higher than 10/21 across the examined heterogeneous freezing Ts (i.e., -10 ºC to -30 ºC), suggesting that the surface abundance, or aerosol particle surface area, is not a sole factor determining ice nucleation efficiency for these days at SGP in 2019. Exploring other factors (e.g., aerosol particle composition, mixing state, etc.) and examining these for the entire campaign period or for SGP in general are ongoing efforts with further careful interdisciplinary analyses.

Offline-droplet freezing assays were conducted to assess immersion freezing abilities of aerosol particles collected on 47-mm polycarbonate filters and in impinger suspension samples. Our preliminary $n_{INP}(T)$ results (Figure 6) shows a reasonable agreement between PINE and offline freezing results, suggesting that immersion freezing was the dominant ice nucleation mechanism at the SGP site. To verify this hypothesis, we are currently investigating the relationship between the mole fraction of water in aerosol particles for the observed ambient conditions at SGP and the INP data derived from sub-saturated conditions (Chamber vessel T > Dew Point T). Until now, we compared our INP data to the ARM's cloud condensation nuclei (CCN) data, but we found no substantial statistical correlation between CCN at supersaturation of 0.2% and n_{INP}(-20 °C) during our ExINP-SGP period (r = 0.002), suggesting that CCN activation is not a significant prerequisite for ice nucleation at the SGP site. In addition, we analyzed the relationship between various aerosol particle size ranges and INP abundance. At SGP, we found an increase in the PINE-measured $n_{INP}(T)$ at -20 °C with the number concentration of super-micron particles, especially for diameters > 2 µm (r = 0.564). This reasonable correlation might indicate the predominant existence and importance of super-micron INPs at SGP in 2019.

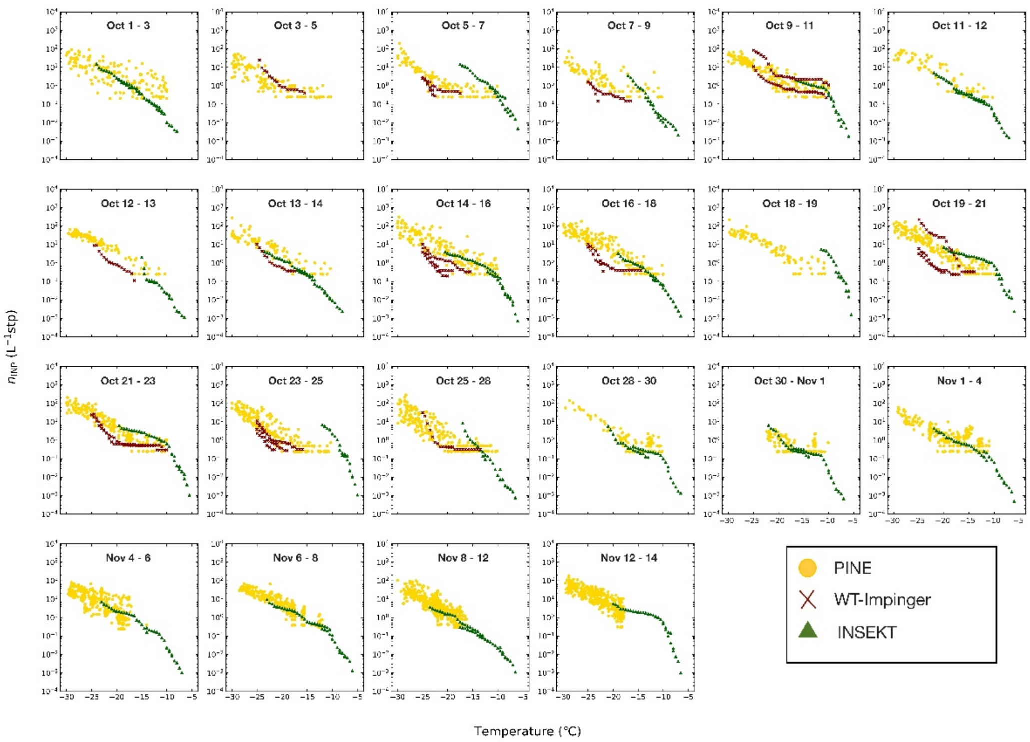

Figure 6. Combined $n_{INP}(T)$ spectra of online PINE-INP and offline INP data. Individual panels represent different measurement/sampling intervals, determined based on the Ice Nucleation Spectrometer of the Karlsruhe Institute of Technology (INSEKT)-filter sampling length (longest amongst three techniques). Both impinger and INSEKT data are shown for only a subset of panels for overlapping periods. The general specifications and data uncertainties of all measurement techniques are overviewed/described in the Data Abstract as well as the Attribute Accuracy Report, which are available in our campaign data archive.

Overall, our field campaign results well demonstrated the PINE's ability of making remote INP measurements, promising future long-term operations including at isolated locations, as well as the usefulness of other complementary measurements. We will estimate the particle loss through our sampling tubes and apply them on top of our system particle losses to calculate more representative total surface area values. Applying this correction will allow us to convert $n_{INP}(T)$ to $n_s(T)$ for all online and offline INP data for further comparison analyses. Lastly, the DOE Office of Biological and Environmental Research report on the vision of ARM over the next decade in 2014 (U.S. Department of Energy 2014), mentions the importance of ice nuclei measurements at other ARM observatories. Based on the feasibility-validation of the new INP measurement method at SGP, its first international remote application is currently ongoing at the ENA site (39° 5′ 29.76″ N, 28° 1′ 32.52″ W; September, 2020–current). Also, the long-term application of PINE at NSA (71° 19′ 22.8″ N, 156° 36′ 32.4″ W) is planned from 2021 to 2023.

3.0 Publications and References

3.1 Publications

One publication has been accepted by *Atmospheric Measurement Techniques* at the time of this report:

Möhler, O, M Adams, L Lacher, F Vogel, J Nadolny, R Ullrich, C Boffo, T Pfeuffer, A Hobl, M Weiß, HSK Vepuri, N Hiranuma, and BJ Murray. 2020. "The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles." *Atmospheric Measurement Techniques*, preprint, https://doi.org/10.5194/amt-2020-307

Science meeting/conference presentations have already featured some of the results. These include upcoming virtual presentations at the 3rd International Electronic Conference on Atmospheric Sciences and the Texas A&M ATMO seminar (invited):

Hiranuma, N, HSK Vepuri, L Lacher, J Nadolny, and O Möhler. "The Portable Ice Nucleation Experiment chamber (PINE): laboratory characterization and field test for its semi-automated ice-nucleating particle measurements in the Southern Great Plains." 2020. EGU2020-12385, https://doi.org/10.5194/egusphere-egu2020-12385, EGU Sharing Geoscience online.

Hiranuma, N, HSK Vepuri, L Lacher, J Nadolny, and O Möhler. 2020. "Characterization of a new Portable Ice Nucleation Experiment chamber (PINE) and first field deployment in the Southern Great Plains", Earth and Space Science Open Archive, https://doi.org/10.1002/essoar.10502526.1, 100th AMS Annual Meeting, 12th Symposium on Aerosol Cloud Interactions, Boston, Massachusetts.

Hiranuma, N. 2020. "Portable Ice Nucleation Experiment (PINE) chamber: remote measurements of ice-nucleating particles (INPs) at multiple atmospheric observatories," Texas A&M University ATMO seminar (invited), online.

Vepuri, HSK, L Lacher, J Nadolny, O Möhler, and N Hiranuma. 2020. "Online ice-nucleating particle measurements in the Southern Great Plains (SGP) using the Portable Ice Nucleation Experiment (PINE) chamber," 3rd International Electronic Conference on Atmospheric Sciences, online.

3.2 References

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DeMott, PJ, TCJ Hill, KJ Suski, and EJT Levin. 2015. Southern Great Plains Ice Nuclei Characterization Experiment Final Campaign Summary. U.S. Department of Energy. DOE/SC-ARM-15-012.

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Knopf, DA, and PA Alpert. 2013. "A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets." *Faraday Discussions* 165: 513–534, https://doi.org/10.1039/C3FD00035D

Möhler, O, M Adams, L Lacher, F Vogel, J Nadolny, R Ullrich, C Boffo, T Pfeuffer, A Hobl, M Weiß, HSK Vepuri, N Hiranuma, and BJ Murray. 2020. "The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles." *Atmospheric Measurement Techniques*, preprint, https://doi.org/10.5194/amt-2020-307

U.S. Department of Energy. 2014. Atmospheric Radiation Measurement Climate Research Facility Decadal Vision, DOE/SC-ARM-14-029.

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