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Acronyms and Abbreviations

AEROICESTUDY	Aerosol-Ice Formation Closure Pilot Study
AOS	aerosol observing system
APS	aerodynamic particle sizer
ARM	Atmospheric Radiation Measurement
CCN	cloud condensation nuclei
CFDC	continuous flow diffusion chamber
DRUM-CP	Davis rotating-drum unit for monitoring coupled with a cold plate
EC	elemental carbon
GIF	Guest Instrument Facility
IN	inorganic
INP	ice nucleating particles
IS	ice spectrometer
LAAPTOF	laser ablation aerosol particle time-of-flight mass spectrometer
MDFA	microfluidic droplet freezing array
MOUDI	multi-orifice uniform deposition impaction
NASA	National Aeronautics and Space Administration
OA	organic aerosol
OC	organic
OCIN	inorganic-organic particles
PINE	portable expansion chamber for ice nucleating particles measurement
PSD	particle size distribution
SGP	Southern Great Plains
SMPS	scanning mobility particle sizer
SP-AMS	soot-particle aerosol mass spectrometer
WIBS	wideband integrated bioaerosol sensor

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1.0 Summary

The aerosol-ice formation closure pilot study (AEROICESTUDY) was a field campaign conducted at the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility at the Southern Great Plains (SGP) observatory from October 7 to 28, 2019. The purpose of this campaign was to test a field observational approach for conducting an aerosol-ice formation closure study. In other words, the goal was to predict the number of ice nucleating particles (INPs) from the measurements of physical and chemical aerosol properties and compare those to measured INP number concentrations. This project was motivated by the fact that the aerosol community has widely conducted aerosol radiative closure, aerosol-cloud condensation nuclei (CCN), and CCN-droplet closure studies to test the physical models and parameterizations that cloud-resolving and climate models rely on to perform reliable simulations of the Earth system and energy budget. However, very few closure studies related to INPs have been conducted, and to our knowledge, none using robust, size-resolved, ambient aerosol composition measurements as model inputs.

The AEROICESTUDY project team reflects a collaborative approach to learn how to best achieve closure between predicted INP and measured INP number concentrations. Several institutions have been involved in this pilot study including Stony Brook University, Colorado State University, University of Illinois at Urbana-Champaign, West Texas A&M University, Purdue University, Carnegie Mellon University, the National Aeronautics and Space Administration (NASA) Goddard Institute of Space Sciences, and Texas A&M University, and the team included modelers and experimentalists.

During the AEROICESTUDY we measured, using online and offline techniques, ambient immersion freezing INPs and characterized the physicochemical properties of the aerosol population relevant to the cloud ice crystal formation rates by immersion freezing. The aerosol population data, in turn, serves as input for prediction of INP number concentrations using various state-of-the-art immersion freezing parameterizations and models, the agreement of which with the INP measurements can then be evaluated within quantified uncertainties.

This pilot study allowed us to evaluate procedures and measurement approaches on how to best achieve closure using ambient aerosol particles, i.e., identification of the necessary instrumentation, protocols, and analysis. This is crucial to address the overall scope of this campaign: to identify ice nucleation parameterizations that produce the most robust predictions of INP numbers and thus are best suited to be included in cloud and climate models. This objective can only be achieved by a suite of instrumentation that characterizes the physicochemical properties of the ambient aerosol population complemented with INP instrumentation (and select INP compositional analyses). A crucial prerequisite when attempting closure is that all instrumentation probe the same ambient air volume with accurate knowledge of the sampled ambient particle size distribution (PSD) by all instrumentation.

During the AEROICESTUDY we successfully installed two high-volume sampling stacks (with great help from ARM SGP site staff), powered by blowers in the bottom (Figure 1a), from which most employed instrumentation sampled the ambient aerosol population (Figure 1b). The stack sampled particles above the Guest Instrument Facility (GIF) roof line, at the height of the sampling platform (Figure 1a). During this campaign, the AEROICESTUDY team successfully operated the following equipment (located in SGP GIF, Figure 1c) for aerosol characterization and INP measurements allowing

for online and offline analyses: Scanning mobility particle sizer (SMPS), aerodynamic particle sizer (APS), laser ablation aerosol particle time-of-flight mass spectrometer (LAAPTOF), soot-particle aerosol mass spectrometer (SP-AMS), aerosol collection by multi-orifice uniform deposition impaction (MOUDI), Davis rotating-drum unit for monitoring coupled with a cold plate for size-resolved bulk immersion freezing (DRUM-CP), continuous flow diffusion chamber (CFDC) with alternating ambient concentrator followed by collection of ice residual, wideband integrated bioaerosol sensor (WIBS), ice spectrometer (IS) for bulk immersion freezing, microfluidic droplet freezing assay (MDFA), and portable expansion chamber for ice nucleating particles measurement (PINE).

Figure 1. Particle sampling and guest instrumentation employed during AEROICESTUDY. (a) High-volume sampling stack from which instrumentation sampled ambient aerosol particles via a port through GIF housing (b). (c) Guest instrumentation in GIF including PINE, CFDC, MOUDI, SMPS, APS, SP-AMS, and LAAPTOF.

These measurements were complemented by ARM SGP site aerosol measurements made with the aerosol observing system (AOS). Most of the guest instrumentation was 100% operational during the entire campaign period, though the first week of the campaign was used to refine measurement and particle collection protocols. Typically, online and offline measurements were conducted daily from about 8-9 am to 5-7 pm.

No unusual meteorological events were noticed, only typical incidences such as weather changes including precipitation. Sporadically, the influence of road dust suspended by cars and trucks driving in the sampling area and wind-blown soil emissions from fields was noticed.

2.0 Results

To achieve the goal of closure between predicted and measured INP concentrations, the main focus of the field deployment was to acquire sufficient and comprehensive data characterizing the ambient aerosol population complemented by INP measurements, all sampling from the same ambient air volumes. Application of the prototype high-volume sampling stack was proven to successfully sample

supermicron-sized particles. Except for collection of particles onto substrates for offline immersion freezing experiments (IS, MDFA[1, 2], DRUM[3-5]), all instrumentation was sampling continuously from the high-volume sampling stack. IS, MDFA, and DRUM were sampling very close to the intake of the sampling stack (Figure 1a) to minimize differences in collected particle population due to variation in air masses. Figure 2 shows example data from the online instrumentation spanning the entire duration of the campaign, demonstrating the reliability of employed experiments and measurement protocols. Figure 2A shows SMPS (14–750 nm particle diameter)- and APS (0.5–21 µm particle diameter)-derived particle number concentrations. During a cold frontal passage on 10/15, the morning showed larger amounts of supermicron-sized particles compared to the afternoon (green spike). This case will be discussed in our upcoming *Bulletin of the American Meteorological Society* campaign overview article. Figure 2B displays submicron aerosol composition determined by aerosol mass spectrometer. These data indicated enhanced organic aerosol (OA) concentration during the on 10/15, for which we detected some of the greatest organics concentrations coinciding with an increase of secondary inorganic particles (largest green spike). Figure 2C shows immersion freezing INP number concentrations measured by CFDC for the entire campaign duration and corresponding activation temperature (ranging from about -22 to -30 °C). Figure 2D displays INP concentrations determined by PINE for the campaign period with corresponding freezing temperatures ranging from about -12 to -30 °C).

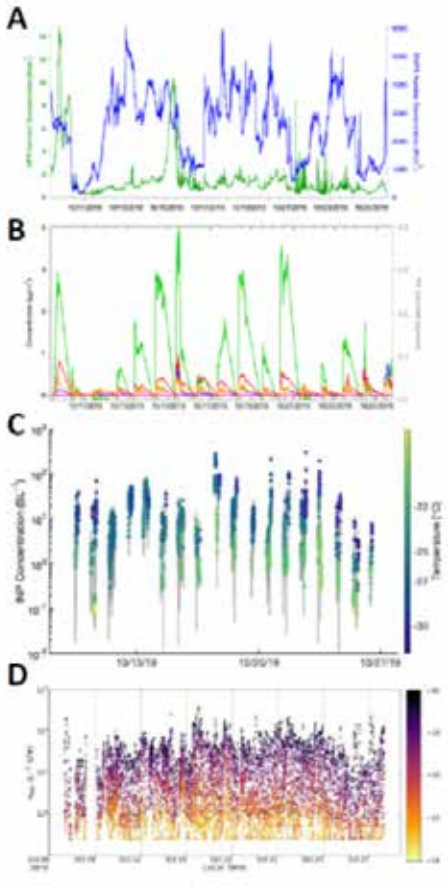

Figure 2. Online measurement data for entire field campaign. (A) SMPS- and APS-derived aerosol number concentrations; (B) AMS-derived aerosol composition; (C) and (D) immersion freezing INP number concentrations and corresponding activation temperatures determined by CFDC and PINE, respectively.

The online measurements were complemented by offline aerosol composition and INP number concentration analyses. Figure 3 shows example data sets including INP number concentrations derived by the IS and DRUM (A). Panel (A) also demonstrates the good agreement between online CFDC INP and offline IS and DRUM INP measurements. Offline single-particle micro-spectroscopic analyses of the ambient particle population are shown in panels (B) to (D)[6]. Those data complement online aerosol composition data by providing detailed mixing state analysis and extension of composition analysis to larger particle sizes. Panel (B) displays the ambient particle population composition applying a user-defined classification scheme derived from computer-controlled scanning electron microscopy with energy dispersive X-ray spectroscopy (CCSEM/EDX)[7]. These results indicate the dominance of organic particles accompanied by organonitrates, organosulfates, and mineral dust. Panels (C) and (D) show single-particle analysis performed by scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS)[8, 9]. Panel (C) displays the size-segregated organic volume fraction of the aerosol population on the afternoon of 10/15. STXM/NEXAFS indicates that a great number of particles were highly dominated by organic matter, present as pure OA or as a coating. Panel (D) shows the mixing state analysis of particles sampled during the afternoon on 10/15, expressed as false color particle maps. The particle population consists of a majority of inorganic-organic particles (OCIN, blue) and pure or highly OA-dominated particles (green).

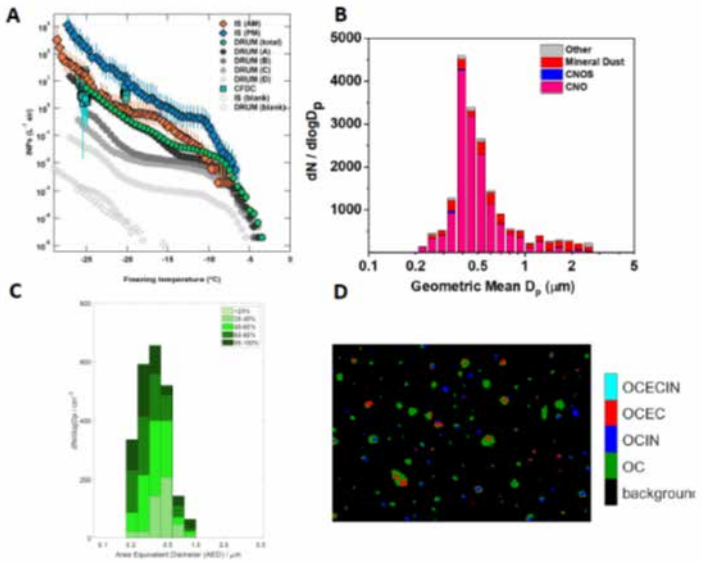

Figure 3. Overview of complementary offline measurements of INP number concentrations and particle composition for 10/15. Panel (A): IS- and DRUM-derived INP number concentrations and corresponding activation temperatures. IS shows morning and afternoon measurements whereas DRUM shows blank, size-resolved, and total INP number concentrations. Panel (A) also includes online CFDC data for comparison. Panels (B) displays particle population composition derived from CCSEM/EDX applying a user-defined classification scheme. Panels (C) shows STXM/NEXAFS-derived size-resolved organic volume fraction of the ambient particle composition. Panel (D) displays STXM/NEXAFS-derived mixing state analysis using false color particle maps. OC: organic, EC: elemental carbon, IN: inorganic.

The physicochemical aerosol properties serve as input into commonly applied immersion freezing parameterizations. Those include parameterizations by DeMott et al. (2010)[10], the soccer ball model[11], the

water-activity-based immersion freezing model[12], the ice nucleation active sites concept[13], and the heterogeneous ice nucleation mixing model.[14] In the first step of closure, the various data sets had to be merged, i.e., SMPS and APS data have to be converted to same-diameter type and acquisition time steps. The sampling time and PSD probed by each individual instrument have to be considered in addition to particle transmission losses between sampling stack and instrument inlet. Corresponding uncertainties have to be accounted for including measurement uncertainty, transmission losses, composition uncertainty, and uncertainties in immersion freezing parameterizations. After consideration of all these processes, our capability of closure can be assessed. The AEROICESTUDY team is currently addressing these points. A preliminary closure result is shown in Figure 4.

Figure 4 shows INP online measurements by PINE for four campaign dates and corresponding predictions of INP number concentrations applying the ice nucleation active sites immersion freezing parameterization assuming all particles resemble natural dust[15]. In this simplified closure exercise, i.e., incorporating only one type of particle composition, there is a clear difference between measured and predicted INP number concentrations, especially towards lower freezing temperatures. The AEROICESTUDY team is currently incorporating more realistic particle composition and evaluating all uncertainty contributions to finalize the closure analysis.

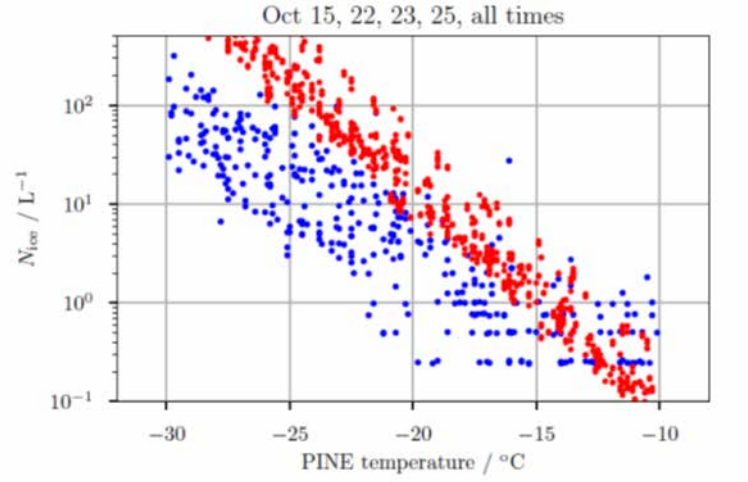

Figure 4. INP number concentrations measured by PINE (blue) and predicted by ice nucleation active sites parameterization of natural dust (red) for four campaign dates.

3.0 Publications and References

3.1 Publications

The AEROICESTUDY team is analyzing data and writing publications. Due to the COVID-19 pandemic, the theoretical and experimental data analyses have been delayed. We were invited to submit an overview article about this campaign to the *Bulletin of the American Meteorological Society* entitled "Aerosol–Ice Formation Closure: A Southern Great Plains Study". The AEROICESTUDY team is currently finalizing the results for this publication. Individual groups are working on more detailed manuscripts to be published in peer-reviewed community journals. The online measurement data have been completely analyzed and archived. Most offline measurement data have been analyzed and archived and the remainder will be continuously updated.

3.1.1 Publications Currently Under Review

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 Discussions*, https://doi.org/10.5194/amt-2020-307, in review.

3.1.2 Presentations

1. DeMott, PJ, CS McCluskey, SM Burrows, K Barry, E Järvinen, KA Moore, TCJ Hill, EJT Levin, JM Creamean, CH Twohy, DToohey, JL Stith, GM McFarquhar, J D'Alessandro, T Zaremba, W Wu, A Gettelman, J Fan, Y Lin, S Lasher-Trapp, X Zhao, X Liu, PL Lawson, AJ Heymsfield, and SM Kreidenweis. 2019. "Assessing the Roles of Primary and Secondary Ice Formation in Clouds Through Measurements and Modeling." Abstract A52G-01, American Geophysical Union Fall Meeting, December 9-13, San Francisco, California.

2. DeMott, PJ, CS McCluskey, GP Schill, TCJ Hill, Y Tobo, EJT Levin, JM Creamean, J Uetake, K Barry, KA Moore, E Järvinen, K Suski, JK Kodros, JR Pierce, G McMeeking, A Gettelman, S Burrows, and SM Kreidenweis. 2020. "How well do we understand and predict ice nucleating particle sources and concentrations around the world?" 100th AMS Annual Meeting, 12th Symposium on Aerosol Cloud Interactions, January 15, Boston, Massachusetts.

3. DeMott, PJ. 2020. "Identifying and predicting global ice nucleating particle sources." Texas A&M Atmospheric Sciences Seminar, April 15.

4. Hiranuma, N, HSK Vepuri, L Lacher, J Nadolny, and O Möhler. 2020. "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." EGU2020-12385, https://doi.org/10.5194/egusphere-egu2020-12385, EGU Sharing Geoscience Online, online, May 6.

5. 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, January 15.

6. Hiranuma, N. 2020. "Portable Ice Nucleation Experiment chamber (PINE): remote and semi-autonomous measurements of ice-nucleating particles at multiple atmospheric observatories." Texas A&M University ATMO seminar (invited), online, November 4.

7. 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, November 16–30.

3.2 References

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4.0 Lessons Learned

Our campaign highlights that for a successful attempt evaluating our ability to achieve closure between aerosol properties and INPs, greater efforts in particle sampling strategies have to be undertaken. Since ice formation by aerosol particles scales with the available number, surface area, and chemical composition of the ambient particles population, a large array of instrumentation is needed to cover this spectrum of information. This can include the size of macromolecules acting as INPs to supermicron-sized mineral, organic, and biological particles. The various instrumentation that can cover these different aspects of the aerosol population will need to operate at different conditions, such as sample intake (this can vary from 0.1 to 30 SLPM), size range of particles (submicron to 20 µm in diameter), power requirements, etc. In addition to this challenge, all involved instrumentation should sample the same air mass at the same time. Ideally, this calls for a sophisticated humidity- and temperature-controlled sampling stack with low losses for supermicron particles that can supply an isokinetic sampling flow to many instruments.

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