Final Technical Report

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Final Report: DOE DE-SC0020006 – "Small field campaign: aerosol – ice formation closure pilot study"

Overview

The goal of this project is conducting a field-based pilot study at the Atmospheric Radiation Measurement (ARM) user facility at Southern Great Plains (SGP) to evaluate our capability to predict the number concentration of aerosol particles that serve as ice-nucleating particles (INPs) in the immersion freezing mode. Immersion freezing, where INPs initiate freezing in supercooled water droplets, is recognized as one of the atmospherically dominant ice formation pathways in mixed-phase and cirrus clouds. Successful prediction of INP number concentrations is also termed "closure". This field-observational approach represents a first-of-its kind attempt of an aerosol–ice formation closure study (AEROICESTUDY). Very few closure studies related to INPs have been conducted, and to our knowledge, none using robust size-resolved ambient aerosol composition measurements as a starting point. Achievement of aerosol–ice formation closure relies on our ability to characterize the ambient aerosol population with respect to particles size and composition and to determine INP number concentrations for specified freezing temperatures. This requires numerous online and offline instrumentation resulting in this pilot field campaign being a multi-institutional and community-collaborative effort. We chose the ARM SGP megasite for this first aerosol-ice formation closure pilot study due to its significant measurement capabilities available to obtain detailed physical characterization of the local aerosol population including size distribution, mass loading, and chemical composition of non-refractory aerosol particles.

The overall objective of this project is 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 allows us to answer several important questions regarding our predictive capability of INP number concentrations in the atmosphere:

i) What are the crucial aerosol physicochemical property measurements needed to accurately guide ice nucleation representations in models and long-term INP measurements?

In response to this question, our initial closure analysis published in an article in the *Bulletin of the American Meteorological Society* (Knopf et al., 2021) concludes that the advancement of our predictive capability for atmospheric immersion freezing is greatly assisted by size-resolved aerosol composition analysis, including the coarse mode and refractory particles, and accompanying online and offline INP measurements. This includes improved speciation of the organic species (e.g., secondary organic species, soil-derived organic and biological matter) and mineral dust types and efficiency in the analysis of larger sized particles. This approach will elucidate sources of bias in immersion freezing parameterizations.

ii) What level of parameter details needs to be known to achieve aerosol–ice formation closure?

Our closure exercise concludes that surface area particle size distribution (PSD) and the size-resolved major particle types are crucial information to achieve closure within measurement and parameterization uncertainties. However, this requires that the immersion freezing parameterizations for identified INP types are available.

iii) What are the leading causes for climate model bias in INP predictions?

From the initial aerosol–ice formation closure exercise, we conclude that for any meaningful INP number concentration predictions by cloud and climate models, the aerosol fields (PSD and particle composition) have to be sufficiently accurate to apply immersion freezing parameterizations. For example, mass-based information of airborne mineral dust particles may not allow sufficiently accurate derivation of the surface area of the PSD. This in turn results in large uncertainties of predicted INP number concentrations. Missing and misrepresented INP types, e.g., the multitude of organic aerosol particles species, also leads to discrepancies between predicted and observed INP number concentrations.

Overall, as further outlined below, we found that the advances in our understanding of immersion freezing garnered over the last 20 years allowed us to yield partial and full closures of atmospheric immersion freezing from ambient aerosol particles (Knopf et al., 2021). When the aerosol population is physicochemically complex and parameterizations for representative INP types are not yet available, we still struggle to accurately predict INP number concentrations. This project clearly demonstrates that with more laboratory and field measurements that are accompanied by particle composition analysis, the necessary datasets to achieve aerosol–ice formation closure for various locations will emerge, thus providing a robust foundation for guiding the representation of INPs in cloud and climate models.

Significance

Prediction of atmospheric ice formation from aerosol particles by heterogeneous nucleation represents one of the grand challenges in atmospheric science. Our insufficient predictive understanding of primary ice formation is the reason that climate models typically do not include heterogeneous ice nucleation with subsequent effects on climate uncertainty. Mixed-phase clouds, where supercooled water droplets and ice crystals coexist play globally an important role regulating climate. This is especially the case for the Arctic region that experiences the greatest warming due to climate change compared to other regions in the world (Hahn et al., 2021;Morice et al., 2021). Immersion freezing initiated by INPs is recognized as the dominant primary ice formation pathway in mixed-phase cloud regimes (Ansmann et al., 2009;de Boer et al., 2011;Westbrook and Illingworth, 2013). For this reason, it is crucial to evaluate our capability to predict immersion freezing for a given ambient aerosol population. Modern climate models like NASA GISS Model E (Schmidt et al., 2014;Schmidt et al., 2006) or CAM6 (Danabasoglu et al., 2020) include modules such as MATRIX (Bauer et al., 2008) and MAM4 (Liu et al., 2016) that describe different aerosol particle types and respective particle size distributions (PSDs). This information allows for a bottom-up prediction of INP number concentrations via respective ice nucleation parameterizations.

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 (Rosenfeld et al., 2014b;Rosenfeld et al., 2014a;Broekhuizen et al., 2006;Medina et al., 2007;Wang et al., 2008;Lance et al., 2009;Cubison et al., 2008;Chang et al., 2007). 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 a starting point to provide a bottom-up prediction of INP number concentrations.

The objectives of the aerosol–ice formation closure pilot study are directly relevant to the missions of U.S. Department of Energy (DOE), Office of Science, Office of Biological and Environmental Research (OBER), Climate and Environmental Sciences Division (CESD) and the ARM User Facility by improving our predictive understanding of complex environmental systems for energy and infrastructure security, independence, and prosperity. This project delivered fundamental science on the physics and chemistry of aerosol particles that govern clouds and precipitation interactions thereby enabling major scientific developments in earth system-relevant atmospheric process and modeling research. Prediction of atmospheric ice formation represents one of the grand challenges in atmospheric sciences. Ice formation by INPs is not physically represented in most current climate models due to our insufficient understanding but is increasingly recognized as an important factor for realistic maintenance of radiatively important cloud liquid water. The observational data acquired through this aerosol–ice formation closure study is available to the climate research community that is dedicated to advancing understanding of the interaction between aerosol and ice crystals, designed to improve representation of clouds and aerosols in climate and earth system models. The advanced knowledge from this project is meaningful to allow policymakers to plan sustainable energy production, resources, and mitigation strategies.

The choice of the SGP ARM megasite for the proposed endeavor is in line with the decadal vision of ARM to use this site for development of a unique integration of high-density measurements with routine high-resolution models. This aerosol–ice formation closure study addresses several science themes (microphysics and radiative properties of mixed-phase and ice clouds) and manifests an observation-model testbed methodology. Our study’s objectives generated data products and analysis tools that strengthen the evaluation of models using ARM data. The garnered experience and results of this pilot study will inform future aerosol-focused field campaigns conducted at SGP and other ARM sites regarding the design, construction, and testing of required infrastructure such as aerosol inlets and sampling lines that reliably deliver the same aerosol sample to a suite of online and offline instruments.

Project Outcomes

We successfully conducted a three-week field campaign at the US DOE ARM SGP site (10/6 to 10/27/19). The closure study concept is straightforward to test any physical model: measure all model inputs as well as predicted outputs, and then evaluate whether the model can predict the measured outputs when

Table 1. Atmospheric Radiation Measurement (ARM) site and guest instrumentation, online and offline, for physicochemical characterization of aerosol population and measurement of ice-nucleating particles. PSD refers to particle size distribution. From (Knopf et al., 2021).

Investigator	Instruments/methods	Measurement	Particle size range	Sampling rate	Measurement frequency
Online					
ARM Site	Scanning mobility particle sizer (SMPS)	PSD	~0.01–0.8-μm diameter	0.1–0.3 LPM (liters per minute)	5 min
ARM Site	Aerodynamic particle sizer (APS)	PSD	~0.5–20-μm diameter	5 LPM	1 s
Colorado State University (CSU)	Continuous Flow Diffusion Chamber (CFDC) with alternating ambient concentrator	Immersion-mode INP concentration at –15° and –30°C	Up to ~2.5 μm, 50% cut point	1.5 LPM	Typically integrated 3–5 min
CSU	Wideband Integrated Bioaerosol Sensor (WIBS model 4A)	Fluorescence and PSD of biological particles	~0.5–20 μm	0.3 LPM	Continuous
Carnegie Mellon University (CMU)	SMPS	PSD	~0.01–0.8-μm diameter	0.3 LPM	4 min
CMU	APS	PSD	~0.5–20-μm diameter	5 LPM	1 s
CMU	Laser Ablation Aerosol Particle Time-of-Flight Mass Spectrometer (LAAPTOF)	Size-distributed single-particle aerosol composition/type	0.2–3 μm	0.1 LPM	30 min
CMU	Soot-Particle Aerosol Mass Spectrometer (SP-AMS)	Size-distributed single-particle aerosol composition/type	0.05–0.8 μm	0.1 LPM	4 min
West Texas A&M University (WTAMU)	Portable Ice Nucleation Experiment chamber (PINE-c)	Immersion-mode INP concentration at –15° and –30°C	0.35–5 μm	2–5 LPM	5 min
Offline					
Stony Brook University/Purdue University (SBU/PU)	Aerosol collection by multi orifice uniform deposition impaction (MOUDI)	Size distributed aerosol composition/type of aerosol	0.15–16 μm	30 LPM	1–4 h
SBU	Multi Orifice Uniform Deposition Impaction Droplet Freezing Technique (MOUDI-DFT)	INP concentration, frozen fraction	0.15–16 μm	30 LPM	1–4 h
CSU	Davis Rotating-drum Unit for Monitoring coupled with a Cold Plate (DRUM-CP) for size-resolved bulk immersion freezing	INP concentration, frozen fraction	0.13–12 μm	26–30 LPM	24 h
CMU	Microfluidic Ice Nucleation Technique (MINT)	INP concentration, frozen fraction	All into filter	16–18 LPM	4+ h
CSU	Ice Spectrometer (IS) for bulk immersion freezing with heat labile and organic INP analyses	INP concentration, frozen fraction	All into filter	16–18 LPM	1–4 h

accounting for input and output measurement uncertainties. Here, we determine INP number concentrations by two online INP instrumentation. We apply measured particle sizes and composition to predict INP number concentrations. We account for measurements uncertainties including transmission losses in sampling lines, instrument operation conditions, e.g., range of particle sizes sampled, and uncertainties in immersion freezing parameterizations. When predicted INP number concentrations inclusive derived uncertainties matches online-derived INP number concentrations, we achieve closure. Table 1 lists the numerous, successfully operating online and offline instrumentation and groups involved. In the first

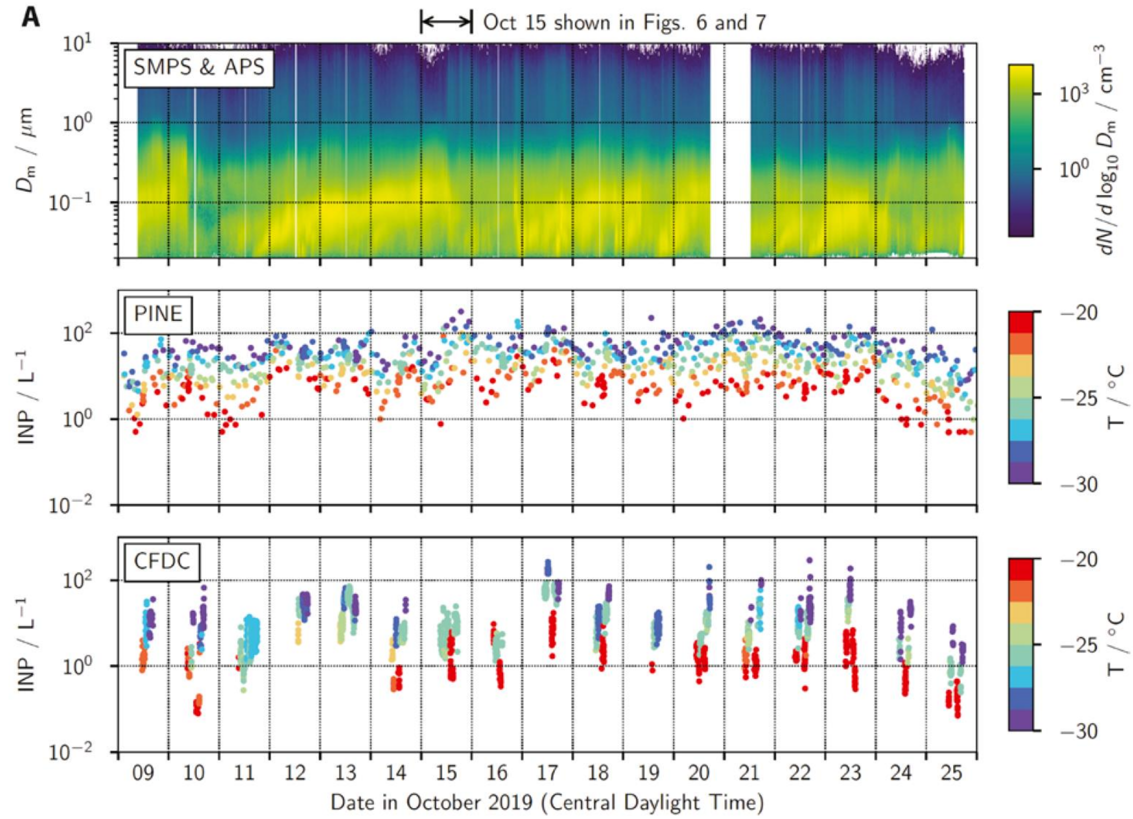

Figure 1. Overview of online measurements for entire campaign period. Upper panel shows particle size distributions from combined measurements by scanning mobility particle sizer (SMPS) spectrometer and aerodynamic particle sizer (APS) spectrometer. INP number concentrations with associated freezing temperatures measured by PINE and CFDC are displayed in middle and lower panels, respectively. INP measurements were done for specific daily time periods and defined temperatures for closure exercises. From (Knopf et al., 2021).

closure calculation attempt, we used measured aerosol PSD, online aerosol composition measurements, offline single particle micro-spectroscopic analytical techniques to determine particle types in the ambient particle population, and two online INP instrumentation, complemented by offline INP measurements to assist in interpretation. Figure 1 shows the continuous measurements of ambient aerosol PSD by SMPS and APS (Table 1) and the two online INP instrumentation, PINE and CFDC, detecting about 1 to 100 INP/L for freezing temperatures between -20 to -30 °C. The measured aerosol PSD shows the presence of supermicron-sized particles. We published our first aerosol–ice formation closure calculations for the case of October 15 where a cold front passed through the site resulting in different aerosol PSDs between the morning and afternoon hours.

For the October 15 case study, aerosol PSDs indicate that the morning was dominated by submicrometer-sized particles and the afternoon, with stronger winds present, was dominated by supermicrometer-sized particles which also showed more fluorescence indicative of the presence of organic and biological particles. Figure 2 displays an overview of the aerosol composition derived by online and offline instrumentation. LAAPTOF, analyzing particles up to 3 μm in aerodynamic diameter, indicated that mixed, aged inorganic–organic carbon particles dominated the ambient particle population in the morning with decreasing numbers toward afternoon while mineral-organic particle numbers displayed an increasing trend. The SP-AMS measurements indicate that, during the morning, the submicron aerosol population was dominated by aged/oxidized organic particles with decreasing concentrations in the afternoon. Both online aerosol composition measurements suggest the presence of aerosol particles that were highly aged, secondary in nature, and mixed. CCSEM/EDX analysis shows the dominance of carbonaceous organic (CO), inorganic-organic (CNO, COS, CNOS), and soot, elemental carbon (EC), particle types during the

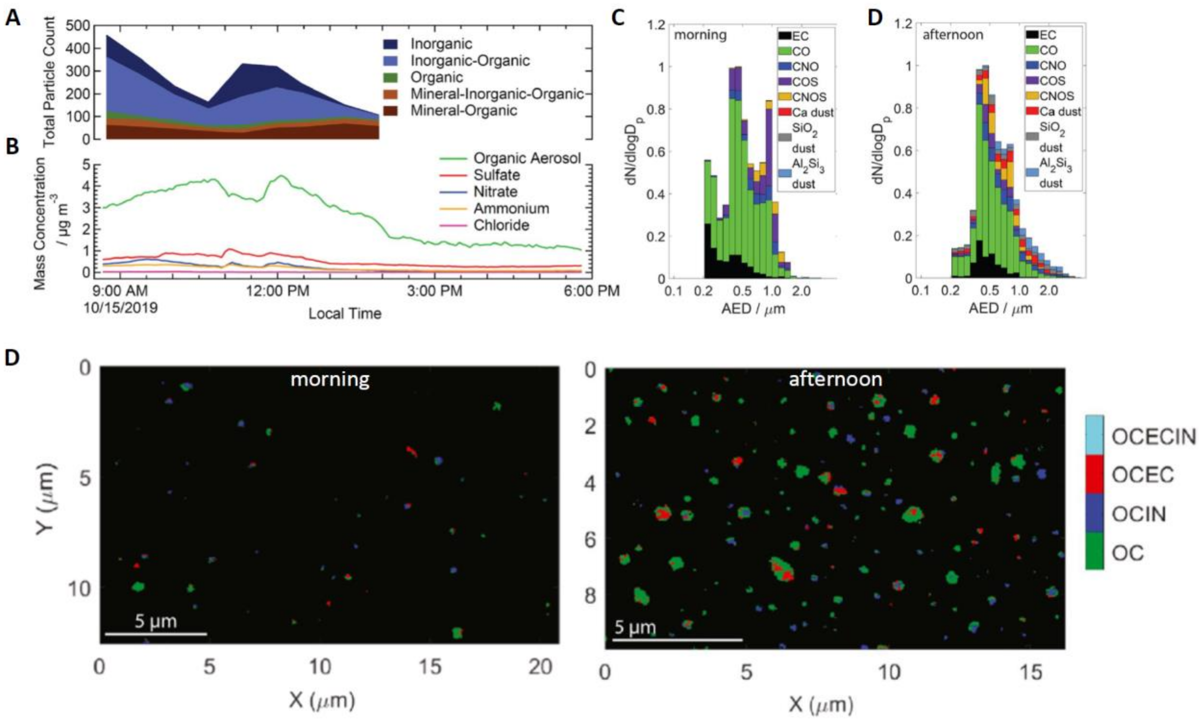

Figure 2. Ambient particle composition for frontal passage closure case study on 15 October determined by online and offline instrumentation. A, B: Time evolution of particle mixing state and composition analysis by Laser Ablation Aerosol Particle Time-of-Flight Mass Spectrometer (LAAPTOF) and of nonrefractory submicrometer aerosol composition derived by aerosol mass spectrometer (SP-AMS), respectively. C, D: Size-resolved, in area equivalent diameter (AED), single-particle micro-spectroscopic analyses by computer-controlled scanning electron microscopy with energy dispersive X-ray analysis (CCSEM/EDX) where elemental particle composition are: EC: elemental carbon; CO: carbon, oxygen; CNO: carbon, nitrogen, oxygen; COS: carbon, oxygen, sulfate; CNOS: carbon, nitrogen, oxygen, sulfate. D: False-color chemical imaging of ambient particles by scanning transmission X-ray microscopy with near-edge X-ray absorption fine structure spectroscopy (STXM/NEXAFS) where IN: inorganic; EC: elemental carbon; OC: organic carbon.

morning (Fig. 2C). Those particles are likely secondary in nature. Also, almost no traces of mineral dust particles are present in the morning. In contrast, in the afternoon (Fig. 2D), larger particles were present and the fraction of mineral particle types (e.g., Ca, SiO_2, and Al_2Si_3 dust) was greater. We performed STXM/NEXAFS to infer the size-resolved particle mixing state and organic volume fraction of the aerosol population (not shown). Figure 2E provides false-color STXM derived images of the morning and afternoon particle samples. A clear difference between particles collected during morning and afternoon hours is visible, were in the afternoon greater winds resuspended soil-dust from the agricultural fields. STXM demonstrates that no pure mineral dust was present but that it was associated with organic matter. These analyses demonstrate the significant differences in the morning and afternoon samples, thus allowing for testing our predictive capability of immersion freezing INPs.

Aerosol PSD (Fig. 1) and composition (Fig. 2) allow for an aerosol population representation from which INP number concentrations can be predicted. In this first attempt, we distinguish mineral dust, organic, and soot particles. We established a closure calculation that uses as input aerosol PSD and composition and accounts for measurement uncertainties, transmission losses, and online INP instrument operation conditions. Three immersion freezing parameterizations typically employed in the community were integrated in this calculation. Those include: i) particle number based parameterization by (DeMott et al., 2010) and (DeMott et al., 2015) to be applied to atmospheric particles in general (DM2010) and mineral dust (DM2015) specifically. ii) ice-nucleation active sites (INAS) based on number and surface area of aerosol for mineral dust (Niemand et al., 2012), organic particles (China et al., 2017) and soot (Schill et al., 2020). iii) classical nucleation theory derived water activity based immersion freezing model (ABIFM) for mineral dust (Alpert and Knopf, 2016), organic particles (Knopf and Alpert, 2013), and soot (Knopf et al., 2021). To more accurately represent soil-dust INPs, especially during afternoon periods, we apply the mineral dust parameterization to the organic particle fraction. It has been shown that Wyoming soil dust possesses similar immersion freezing efficacies as described by the mineral dust parameterization (Tobo et al., 2014). Offline INP measurements (not shown here) clearly demonstrate significant contribution of

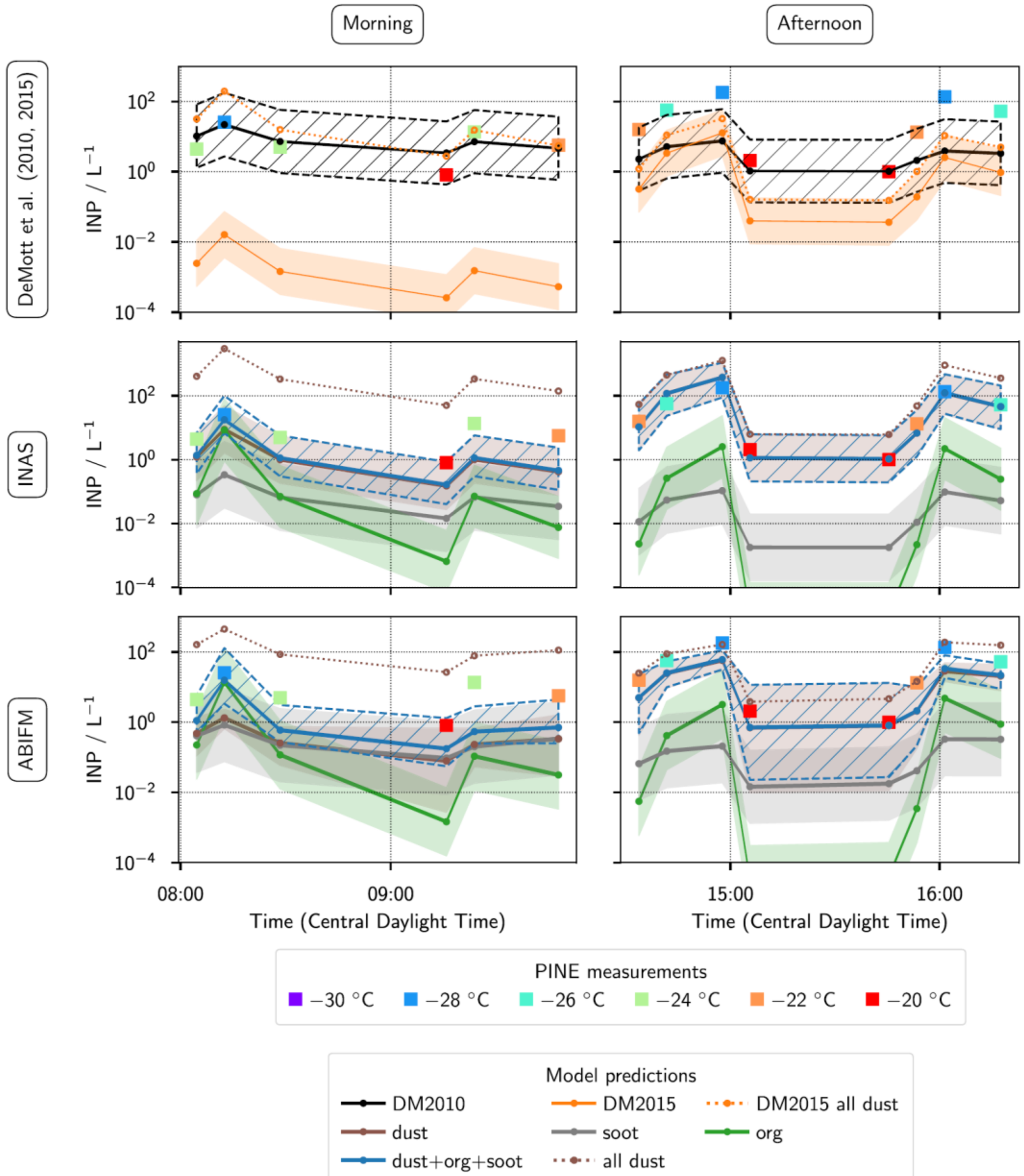

Figure 3. INP number concentrations measured by PINE at different freezing temperatures for closure case study on 15 October for morning and afternoon periods (large colored squares). Uncertainties in measured INP number concentrations are about ±20%. Solid lines, small circle symbols, and corresponding shading represent predicted INP number concentrations. (top) INP number concentrations predicted by DeMott et al. (2010, 2015) parameterizations as black and orange lines, respectively. The dotted orange line represents the prediction by the DeMott et al. (2015) parameterization assuming all particles larger than 0.5 μm are acting as mineral dust INPs. (middle) INP number concentration predictions by the ice nucleation active sites model (INAS) applying parameterizations for organic (green), soot (gray), and mineral dust (brown) INPs. Blue line represents total INP number concentrations from all individual INP types. Dotted brown line displays INP number concentrations when all particles are assumed to be mineral dust particles. (bottom) INP number concentration predictions by the water activity–based immersion freezing model (ABIFM) where lines are the same as for the INAS case in middle panels. From (Knopf et al., 2021).

organic and biological INPs to the overall INP pool. Those INPs stem most likely from the suspended agricultural soil dust.

Figure 3 shows the closure calculation for October 15 for the case of PINE INP measurements. Three immersion freezing parameterizations are evaluated (lines) for their ability to represent measured INP number concentrations (squares). In the morning, within uncertainties DM2010 captures PINE INP measurements very well. In the afternoon, both DM2010 and DM2015 underestimate observed INP number concentrations, though DM2010 fairs better. In the morning, INAS mineral dust dominates total predicted INP numbers, although very few mineral dust particles are present in the morning. Overall INAS underestimates INPs. In the afternoon, INAS mineral dust dominates and, within uncertainties, captures the INP observations. When accounting for missing INP numbers by soil-derived organic and biological INPs (using the mineral dust INP parameterization to describe organic particles), predicted INP numbers are overpredicted. In the morning ABIFM underestimates INP numbers similar to INAS, though mineral dust INPs are less dominating the total INP numbers. In the afternoon, ABIFM underestimated total INP numbers. However, when accounting for missing soil-dust INPs (brown dotted line), for the most part the INP predictions agree with observations.

In the morning, the particle population is dominated by aged inorganic–organic carbon particles, likely secondary in nature. For those specific particle types, we do not have the correct immersion freezing parameterizations. In this case, the field derived DM2010 performs best. In the afternoon, organic and mineral dust particles dominate the PSD. Assigning those organic particles the freezing efficacies of humid acid INPs, which likely underestimates INP prediction, INAS predictions agree with observations while ABIFM predictions underestimate observations. Accounting for offline immersion freezing experiments and assigning the organic particles the freezing efficacies of soil-dust INPs, INAS overestimates INP numbers while ABIFM mostly predicts INP numbers correctly. When having an incomplete description of the freezing capability of the organic particles, one would expect the closure calculation to yield an underestimation of INPs, as in the case of ABIFM. We conclude that each parameterization achieved closure but for different reasons, though more evaluations have to be conducted to assess the role of missing soil-dust particles acting as INPs. It is interesting to note that all mineral dust freezing parameterizations are based on the same laboratory experiment and explain those laboratory results equally well. However, when applied to the ambient particle population, they predict distinctively different INP number concentrations.

The results of this pilot closure study strongly suggest that if the ambient aerosol population is well characterized in terms of size distribution and particle types, INP number concentrations can be predicted from aerosol particle properties when immersion freezing parameterizations are available. However, when the aerosol population is physicochemically complex and parameterizations for representative INP types are not yet available, accurate prediction of INP number concentrations is still challenging. Below we summarize our lessons learned.

The first aerosol–ice formation closure calculations described here have been published (Knopf et al., 2021) and all campaign data has been uploaded to the U.S. DOE ARM Data Archive (AEROICESTUDY) to allow further closure analyses by the community.

Lessons learned

This field-based aerosol–ice formation closure pilot study allowed the community to acquire novel experience and insights how to conduct a bottom-up evaluation of our predictive capability of immersion freezing. In addition to general conclusions outlined in the overview section above, we learned:

- Since only few aerosol particles serve as INPs and larger particles are likely to exert greater ice nucleation efficiencies, the entire aerosol PSD including the coarse mode has to be sampled without significant particle transmission losses. In addition, the particle size range and corresponding sampling efficiency of each analytical instrument has to be accurately known.
- Aerosol composition and mixing state analyses should accompany aerosol PSD measurements and should include the coarse mode particle sizes.

- To improve immersion freezing parameterizations and identification of INP types, long term INP measurements would benefit to be accompanied by concurrent measurements of aerosol PSD and composition.
- This closure pilot study identified a couple of INP types for which immersion freezing parameterizations are not known or are uncertain. Laboratory studies should focus on characterizing secondary inorganic and organic aerosol and organic and biological matter from soil-dust serving as INPs. Further development of methods is needed to differentiate these influences in field measurements.
- The closure calculation demonstrates how immersion freezing parameterizations based on the same laboratory study diverge in their prediction of INPs when applied to ambient particles. Laboratory studies typically do not reflect every atmospheric condition exactly. Hence scaling analysis and different parameterization approaches should be applied to evaluate the range in INP predictions.
- The aerosol fields provided by climate models need to be sufficiently accurate in terms of aerosol particle numbers and sizes to predict INP number concentrations.

Involvement in ASR Program Activities

The PI and co-PIs have always been actively involved at DOE ARM/ASR User and PI Meetings. In each meeting during the award's time period (2019-2022), several poster and platform presentations have been given. Furthermore, the PI co-organized the Ice Nucleation Breakout Session (2020) and High latitude Marine Clouds (2021). He is also participating in the ACE-ENA and Polar Mixed-Phase Clouds working groups. The PI and co-PIs have communicated on the progress and findings of this aerosol - ice formation closure pilot field study by means of ARM and ASR newsletters and highlights. All campaign data has been submitted to the ARM Data Center in a timely manner.

Publications from this grant

Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A. L., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Riemer, N., Laskin, A., DeMott, P. J., Liu, X., Aerosol–Ice Formation Closure: A Southern Great Plains Field Campaign, *B. Am. Meteorol. Soc.*, 102 (10), E1952–E1971, 2021.

Swanson, B. E., DeMott, P. J., Hill, T. C. J., Barry, K. R., Levin, E. J. T., Suski, K. J., Testa, B., Creamean, J. M., Freney, E., Sellegri, K., Knopf, D. A., Riemer, N., Kreidenweis, S. M., Commonalities and differences in key ice nucleating particle types and their relation to aerosol physical properties at three continental locations in North America, South America and Europe, *in preparation* for submission to *J. Geophys. Res.*, 2022.

Partly supported:

Möhler, O., Adams, M., Lacher, L., Vogel, F., Nadolny, J., Ullrich, R., Boffo, C., Pfeuffer, T., Hobl, A., Weiß, M., Vepuri, H. S. K., Hiranuma, N., and Murray, B. J.: The Portable Ice Nucleation Experiment (PINE): a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles, *Atmos. Meas. Tech.*, 14, 1143–1166, https://doi.org/10.5194/amt-14-1143-2021, 2021.

Patade, S., P., Phillips, V. T. J., Amato, P., Bingemer, H. G., Burrows, S. M., DeMott, P. J., Goncalves, F. I. T., Knopf, D. A., Morris, C. E., Alwmark, C., Artaxo, P., Pöhlker, C., Schrod, J., Weber, B., Empirical

formulation for multiple groups of primary biological ice nucleating particles from field observations over Amazonia, *J. Atmos. Sci.*, 78, 7, 2195–2220, 2021.

Silber, I., Fridlind, A. M., Verlinde, J., Ackerman, A. S., Cesana, G. V., Knopf, D. A., The prevalence of precipitation from polar supercooled clouds, *Atmos. Chem. Phys.*, 21, 3949–3971, 2021.

Student theses resulting from this grant

Yijie Lu, Micro-Spectroscopic Analysis of Ambient Particles and Their Impact on Atmospheric Ice Formation, Stony Brook University, Department of Chemistry (2021).

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Conference and Seminar Presentations

DeMott, P. J., “Investigating atmospheric organic and biological ice nucleating particles". Presented at Molecular Understanding of Organic Atmospheric Aerosols, Lake Arrowhead, CA, May 15-20, 2022. (invited talk)

Knopf, D. A., “On the interpretation of immersion freezing relevant for cold clouds: Its impact on laboratory, field, and cloud modeling studies”. Presented at Brookhaven National Laboratory Environmental and Climate Sciences Department Seminar, April 7, 2022. (invited seminar)

Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A. L., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Riemer, N., Laskin, A., DeMott, P. J., Liu, X., “A Southern Great Plains Pilot Field Campaign to Evaluate a Field-Observational Approach to Aerosol–Ice Formation Closure”. Presented at the annual meeting of the American Meteorological Society, January 27, 2022. (talk)

Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A. L., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Riemer, N., Laskin, A., DeMott, P. J., Liu, X., “Field-observational approach to conduct an aerosol-ice formation closure study using physicochemical particle characteristics”. Presented at the International Chemical Congress of Pacific Basin Societies (PACIFICHEM), Honolulu, December 20, 2021. (invited talk)

Wilbourn, E. K., Hiranuma, N., Vepuri, H. S. K., Lacher, L., Nadolny, J., and Möhler, O.: Comparing online and offline measurements of ice nucleating particles from two autumn field campaigns, American Association of Aerosol Research (AAAR) Annual Conference, Online, Oct. 21, 2021. (talk)

Hiranuma, N., Vepuri, H. S. K., and Wilbourn, E. K.: An abundance of ice-nucleating particles in the Atlantic sector of the Arctic and the mid-latitude sites, TAMU-ATMO seminar, College Station, TX, USA, Sept. 8, 2021. (invited seminar)

Wilbourn, E. K., Hiranuma, N., Vepuri, H. S. K., Lacher, L., Nadolny, J., and Möhler, O.: A comparison of aerosol particle sources and ice-nucleating particle properties from the Eastern North Atlantic and U.S. Southern Great Plains, European Aerosol Conference 2021, Online, Aug. 31, 2021. (talk)

Lacher, L., Vogel, F., Nadolny, J., Adams, M. P., King, L., Boffo, C., Pfeuffer, T., Hobl, A., Hiranuma, N., Vepuri, H. S. K., Murray, B. J. and Möhler, O.: Characterization and application of the Portable Ice Nucleation Experiment PINE: A novel instrument to monitor INP concentrations, International Conference on Clouds and Precipitation 2021, Online, Aug. 4, 2021. (poster)

DeMott, P. J., C. S. McCluskey, G. P. Schill, T.C.J. Hill, K. R. Barry, K. A. Moore, R. J. Perkins, Y. Tobo, E. J. T. Levin, J. M. Creamean, C. H. Twohy, J. K. Kodros, J. R. Pierce, S. Burrows, E. Freney, L. Lacher, O. Möhler, D. Knopf, X. Liu, N. Reimer, and N. Hiranuma, "Combining observations and modeling to understand global ice nucleating particle sources and impacts". Presented at the International Conference on Cloud Physics, Pune, India (virtual), August 4, 2021. (talk)

Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A. L., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Riemer, N., Laskin, A., DeMott, P. J., Liu, X., "AEROICESTUDY: An ARM Southern Great Plains Pilot Study to Assess a Field-Observational Approach to Conduct Aerosol-Ice Formation Closure". Presented at the DOE ARM/ASR PI meeting, June 24, 2021. (poster)

Lacher, L., Vogel, F., Nadolny, J., Ullrich, R., Büttner, N., Adams, M., Boffo, C., Pfeuffer, T., Hobl, A., Weiß, M., Vepuri, H. S. K., Wilbourn, E. K., Hiranuma, N., Murray, B. J., and Möhler, O.: Characterization and first applications of the Portable Ice Nucleation Experiment (PINE), 10th virtual INP Colloquium, Online, Feb. 12, 2021. (invited seminar)

Knopf, D. A., Barry, K., Brubaker, T., Jahl, L., Li, J., Lu, Y., Monroe, L., Moore, K., Rivera-Adorno, F., Sauceda, K., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Levin, E., Creamean, J., Hill, T., China, S., Moffet, R. C., Hiranuma, N., Sullivan, R., Fridlind, A., West, M., Riemer, N., Laskin, A., DeMott, P., Liu, X., "A Field-Observational Approach to Aerosol–Ice Formation Closure". Presented at the annual meeting of the American Meteorological Society, January 14, 2021. (talk)

DeMott, P. J., Testa, B., Hill, T. C. J., Creamean, J. M., Barry, K. R., Moore, K. A., Levin, E. J. T., Uetake, J., Hare, H., Hume, C., Knopf, D. A., Freney, E., Kreidenweis, S. M., "Ice-Nucleating Particles in Midlatitude Continental Regions of North America, South America, and Europe". Presented at the annual meeting of the American Meteorological Society, January 14, 2021. (talk)

Fridlind, A., Silber, I., Verlinde, J., Cesana, G., Knopf, D. A., "On Precipitation from Polar Mixed-Phase Clouds". Presented at the annual meeting of the American Meteorological Society, January 13, 2021. (invited talk)

Möhler, O., Adams, M., Lacher, L., Vogel, F., Nadolny, J., Ullrich, R., Boffo, C., Pfeuffer, T., Hobl, A., Weiß, M., Vepuri, H. S. K., Hiranuma, N., and Murray, B. J.: The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles, 101th American Meteorological Society Annual Meeting, 13th Symposium on Aerosol–Cloud–Climate Interactions, Online, Jan. 13, 2021. (poster)

Hill, T., Barry, K. R., Testa, B., Creamean, J., Uetake, J., Levine, E. J. T., Suski, K., Rauker, A. M., Miller, A., Hare, H., Hume, C., Kreidenweis, S. M., DeMott, P. J., "Ice nucleating particles above agricultural landscapes: linking sources with environment and land management". Presented at the American Geophysical Union Meeting, December 10, 2020. (poster)

Patade, S. G., Phillips, V. T., Amato, P., Bingemer, H., Burrows, S. M., DeMott, P. J., Teixeira Gonçalves, F. T., Knopf, D. A., Morris, C. E., Alwmark, C., Artaxo, P., Pöhlker, C., Schrod, J., Weber, B., "Empirical formulation for multiple groups of primary biological ice nucleating particles from field observations over Amazonia". Presented at the American Geophysical Union Meeting, online, December 9, 2020. (talk)

Silber, I., Fridlind, A. M., Verlinde, H., Ackerman, A. S., Cesana, G., Knopf, D. A., Jackson, R. C., Collis, S. M., "The Prevalence of Precipitation from Polar Supercooled Clouds in Observations and Models". Presented at the American Geophysical Union Meeting, online, December 7, 2020. (talk)

Vepuri, H. S. K., Lacher, L., Nadolny, J., Möhler, O., and Hiranuma, N.: Online ice-nucleating particle measurements in the Southern Great Plains (SGP) using the Portable Ice Nucleation Experiment (PINE) chamber, The 3rd International Electronic Conference on Atmospheric Sciences, Online, Nov. 16, 2020. (poster)

Hiranuma, N. and PINE-c Team: Portable Ice Nucleation Experiment (PINE) chamber: remote measurements of ice-nucleating particles (INPs) at multiple atmospheric observatories, TAMU-ATMO webinar, College Station, TX, USA, Nov. 4, 2020. (invited seminar)

Naruki Hiranuma, Hemanth S. K. Vepuri, Larissa Lacher, Jens Nadolny, and Ottmar 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, EGU2020-12385, DOI: https://doi.org/10.5194/egusphere-egu2020-12385, EGU Sharing Geoscience Online, May 6, 2020. (invited talk)

DeMott, P.J., Identifying and predicting global ice nucleating particle sources, Texas A&M Atmospheric Sciences Seminar, April 15, 2020. (invited seminar)

Naruki Hiranuma and Hemanth S. K. Vepuri: Atmospheric ice-nucleating particles (INPs), ACS West Texas Chapter Seminar, March 3, 2020. (invited seminar)

Naruki Hiranuma, Hemanth S. K. Vepuri, Larissa Lacher, Jens Nadolny, and Ottmar Möhler: 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, DOI: https://doi.org/10.1002/essoar.10502526.1, 100th AMS Annual Meeting, 12th Symposium on Aerosol Cloud Interactions, January 15, 2020, Boston, MA. (poster)

DeMott, Paul J., Christina S. McCluskey, Gregory P. Schill, Thomas C. J. Hill, Yutaka Tobo, Ezra J. T. Levin, Jessie M. Creamean, Jun Uetake, Kevin Barry, Kathryn A. Moore, Emma Järvinen, Kaitlyn Suski, John K. Kodros, Jeffrey R, Pierce, Gavin McMeeking, Andrew Gettelman, Susannah Burrows and Sonia M. 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, 2020, Boston, MA. (talk)

DeMott, Paul J. Christina S. McCluskey, Susannah M. Burrows, Kevin Barry, Emma Järvinen, Kathryn A. Moore, Thomas C. J. Hill, Ezra J. T. Levin, Jessie M. Creamean, Cynthia H. Twohy, Darin Toohey, Jeffery L. Stith, Greg M. McFarquhar, John D'Alessandro, Troy Zaremba, Wei Wu, Andrew Gettelman, Jiwen

Fan, Yun Lin, Sonia Lasher-Trapp, Xi Zhao, Xiaohong Liu, Paul L. Lawson, Andrew J. Heymsfield, and Sonia M. 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, 2019, San Francisco, CA. (talk)

References

Alpert, P. A., and Knopf, D. A.: Analysis of isothermal and cooling-rate-dependent immersion freezing by a unifying stochastic ice nucleation model, *Atmos. Chem. Phys.*, 16, 2083-2107, 10.5194/acp-16-2083-2016, 2016.

Ansmann, A., Baars, H., Tesche, M., Mueller, D., Althausen, D., Engelmann, R., Pauliquevis, T., and Artaxo, P.: Dust and smoke transport from Africa to South America: Lidar profiling over Cape Verde and the Amazon rainforest, *Geophys. Res. Lett.*, 36, L11802, 10.1029/2009gl037923, 2009.

Bauer, S. E., Wright, D. L., Koch, D., Lewis, E. R., McGraw, R., Chang, L. S., Schwartz, S. E., and Ruedy, R.: MATRIX (Multiconfiguration Aerosol TRacker of mIXing state): an aerosol microphysical module for global atmospheric models, *Atmos. Chem. Phys.*, 8, 6003-6035, 10.5194/acp-8-6003-2008, 2008.

Broekhuizen, K., Chang, R. Y. W., Leaitch, W. R., Li, S. M., and Abbatt, J. P. D.: Closure between measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol compositions in downtown Toronto, *Atmos. Chem. Phys.*, 6, 2513-2524, 10.5194/acp-6-2513-2006, 2006.

Chang, R. Y. W., Liu, P. S. K., Leaitch, W. R., and Abbatt, J. P. D.: Comparison between measured and predicted CCN concentrations at Egbert, Ontario: Focus on the organic aerosol fraction at a semi-rural site, *Atmos. Environ.*, 41, 8172-8182, 10.1016/j.atmosenv.2007.06.039, 2007.

China, S., Alpert, P. A., Zhang, B., Schum, S., Dzepina, K., Wright, K., Owen, R. C., Fialho, P., Mazzoleni, L. R., Mazzoleni, C., and Knopf, D. A.: Ice cloud formation potential by free tropospheric particles from long-range transport over the Northern Atlantic Ocean, *J. Geophys. Res.*, 122, 3065-3079, 10.1002/2016jd025817, 2017.

Cubison, M. J., Ervens, B., Feingold, G., Docherty, K. S., Ulbrich, I. M., Shields, L., Prather, K., Hering, S., and Jimenez, J. L.: The influence of chemical composition and mixing state of Los Angeles urban aerosol on CCN number and cloud properties, *Atmos. Chem. Phys.*, 8, 5649-5667, 10.5194/acp-8-5649-2008, 2008.

Danabasoglu, G., Lamarque, J. F., Bacmeister, J., Bailey, D. A., DuVivier, A. K., Edwards, J., Emmons, L. K., Fasullo, J., Garcia, R., Gettelman, A., Hannay, C., Holland, M. M., Large, W. G., Lauritzen, P. H., Lawrence, D. M., Lenaerts, J. T. M., Lindsay, K., Lipscomb, W. H., Mills, M. J., Neale, R., Oleson, K. W., Otto-Bliesner, B., Phillips, A. S., Sacks, W., Tilmes, S., van Kampenhout, L., Vertenstein, M., Bertini, A., Dennis, J., Deser, C., Fischer, C., Fox-Kemper, B., Kay, J. E., Kinnison, D., Kushner, P. J., Larson, V. E., Long, M. C., Mickelson, S., Moore, J. K., Nienhouse, E., Polvani, L., Rasch, P. J., and Strand, W. G.: The Community Earth System Model Version 2 (CESM2), *J. Adv. Model. Earth Syst.*, 12, 35, 10.1029/2019ms001916, 2020.

de Boer, G., Morrison, H., Shupe, M. D., and Hildner, R.: Evidence of liquid dependent ice nucleation in high-latitude stratiform clouds from surface remote sensors, *Geophys. Res. Lett.*, 38, L01803, 10.1029/2010gl046016, 2011.

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, *Proc. Natl. Acad. Sci. U. S. A.*, 107, 11217-11222, 10.1073/pnas.0910818107, 2010.

DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Mohler, O., Snider, J. R., Wang, Z., and Kreidenweis, S. M.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, *Atmos. Chem. Phys.*, 15, 393-409, 10.5194/acp-15-393-2015, 2015.

Hahn, L. C., Armour, K. C., Zelinka, M. D., Bitz, C. M., and Donohoe, A.: Contributions to Polar Amplification in CMIP5 and CMIP6 Models, *Front. Earth Sci.*, 9, 17, 10.3389/feart.2021.710036, 2021.

Knopf, D. A., and Alpert, P. A.: A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets, *Faraday Discuss.*, 165, 513-534, 10.1039/C3FD00035D, 2013.

Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Laskin, A., DeMott, P. J., and Liu, X.: Aerosol–Ice Formation Closure: A Southern Great Plains Field Campaign, *B. Am. Meteorol. Soc.*, 102, E1952–E1971 10.1175/BAMS-D-20-0151.1, 2021.

Lance, S., Nenes, A., Mazzoleni, C., Dubey, M. K., Gates, H., Varutbangkul, V., Rissman, T. A., Murphy, S. M., Sorooshian, A., Flagan, R. C., Seinfeld, J. H., Feingold, G., and Jonsson, H. H.: Cloud condensation nuclei activity, closure, and droplet growth kinetics of Houston aerosol during the Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS), *J. Geophys. Res.*, 114, D00f15, 10.1029/2008jd011699, 2009.

Liu, X., Ma, P. L., Wang, H., Tilmes, S., Singh, B., Easter, R. C., Ghan, S. J., and Rasch, P. J.: Description and evaluation of a new four-mode version of the Modal Aerosol Module (MAM4) within version 5.3 of the Community Atmosphere Model, *Geosci. Model Dev.*, 9, 505-522, 10.5194/gmd-9-505-2016, 2016.

Medina, J., Nenes, A., Sotiropoulou, R. E. P., Cottrell, L. D., Ziemba, L. D., Beckman, P. J., and Griffin, R. J.: Cloud condensation nuclei closure during the International Consortium for Atmospheric Research on Transport and Transformation 2004 campaign: Effects of size-resolved composition, *J. Geophys. Res.*, 112, D10s31, 10.1029/2006jd007588, 2007.

Morice, C. P., Kennedy, J. J., Rayner, N. A., Winn, J. P., Hogan, E., Killick, R. E., Dunn, R. J. H., Osborn, T. J., Jones, P. D., and Simpson, I. R.: An Updated Assessment of Near-Surface Temperature Change From 1850: The HadCRUT5 Data Set, *J. Geophys. Res.*, 126, 28, 10.1029/2019jd032361, 2021.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles, *J. Atmos. Sci.*, 69, 3077-3092, 10.1175/jas-d-11-0249.1, 2012.

Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., de Leeuw, G., Donovan, D. P., Kahn, R., Kinne, S., Kivekas, N., Kulmala, M., Lau, W., Schmidt, K. S., Suni, T., Wagner, T., Wild, M., and Quaas, J.: Global observations of aerosol-cloud-precipitation-climate interactions, *Rev. Geophys.*, 52, 750-808, 10.1002/2013rg000441, 2014a.

Rosenfeld, D., Sherwood, S., Wood, R., and Donner, L.: Climate effects of aerosol-cloud interactions, *Science*, 343, 379-380, 10.1126/science.1247490, 2014b.

Schill, G. P., DeMott, P. J., Emerson, E. W., Rauker, A. M. C., Kodros, J. K., Suski, K. J., Hill, T. C. J., Levin, E. J. T., Pierce, J. R., Farmer, D. K., and Kreidenweis, S. M.: The contribution of black carbon to global ice nucleating particle concentrations relevant to mixed-phase clouds, *Proc. Natl. Acad. Sci. U. S. A.*, 117, 22705-22711, 10.1073/pnas.2001674117, 2020.

Schmidt, G. A., Ruedy, R., Hansen, J. E., Aleinov, I., Bell, N., Bauer, M., Bauer, S., Cairns, B., Canuto, V., Cheng, Y., Del Genio, A., Faluvegi, G., Friend, A. D., Hall, T. M., Hu, Y. Y., Kelley, M., Kiang, N. Y., Koch, D., Lacis, A. A., Lerner, J., Lo, K. K., Miller, R. L., Nazarenko, L., Oinas, V., Perlwitz, J., Perlwitz, J., Rind, D., Romanou, A., Russell, G. L., Sato, M., Shindell, D. T., Stone, P. H., Sun, S., Tausnev, N., Thresher, D., and Yao, M. S.: Present-day atmospheric simulations using GISS ModelE: Comparison to in situ, satellite, and reanalysis data, *J. Clim.*, 19, 153-192, 10.1175/jcli3612.1, 2006.

Schmidt, G. A., Kelley, M., Nazarenko, L., Ruedy, R., Russell, G. L., Aleinov, I., Bauer, M., Bauer, S. E., Bhat, M. K., Bleck, R., Canuto, V., Chen, Y. H., Cheng, Y., Clune, T. L., Del Genio, A., de Fainchtein, R., Faluvegi, G., Hansen, J. E., Healy, R. J., Kiang, N. Y., Koch, D., Lacis, A. A., LeGrande, A. N., Lerner, J., Lo, K. K., Matthews, E. E., Menon, S., Miller, R. L., Oinas, V., Oloso, A. O., Perlwitz, J. P., Puma, M. J., Putman, W. M., Rind, D., Romanou, A., Sato, M., Shindell, D. T., Sun, S., Syed, R. A., Tausnev, N., Tsigaridis, K., Unger, N., Voulgarakis, A., Yao, M. S., and Zhang, J. L.: Configuration and assessment of the GISS ModelE2 contributions to the CMIP5 archive, *J. Adv. Model. Earth Syst.*, 6, 141-184, 10.1002/2013ms000265, 2014.

Tobo, Y., DeMott, P. J., Hill, T. C. J., Prenni, A. J., Swoboda-Colberg, N. G., Franc, G. D., and Kreidenweis, S. M.: Organic matter matters for ice nuclei of agricultural soil origin, *Atmos. Chem. Phys.*, 14, 8521–8531, 10.5194/acp-14-8521-2014, 2014.
Wang, J., Lee, Y. N., Daum, P. H., Jayne, J., and Alexander, M. L.: Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect, *Atmos. Chem. Phys.*, 8, 6325-6339, 2008.
Westbrook, C. D., and Illingworth, A. J.: The formation of ice in a long-lived supercooled layer cloud, *Q. J. Roy. Meteor. Soc.*, 139, 2209-2221, 10.1002/qj.2096, 2013.