Final Technical Report for
***"Ice nuclei relation to aerosol properties: Data analysis and model parameterization for IN in mixed-phase clouds"* (SC00002354)**

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Abstract. Clouds play an important role in weather and climate. In addition to their key role in the hydrologic cycle, clouds scatter incoming solar radiation and trap infrared radiation from the surface and lower atmosphere. Despite their importance, feedbacks involving clouds remain as one of the largest sources of uncertainty in climate models. To better simulate cloud processes requires better characterization of cloud microphysical processes, which can affect the spatial extent, optical depth and lifetime of clouds. To this end, we developed a new parameterization to be used in numerical models that describes the variation of ice nuclei (IN) number concentrations active to form ice crystals in mixed-phase (water droplets and ice crystals co-existing) cloud conditions as these depend on existing aerosol properties and temperature. The parameterization is based on data collected using the Colorado State University continuous flow diffusion chamber in aircraft and ground-based campaigns over a 14-year period, including data from the DOE-supported Mixed-Phase Arctic Cloud Experiment. The resulting relationship is shown to more accurately represent the variability of ice nuclei distributions in the atmosphere compared to currently used parameterizations based on temperature alone. When implemented in one global climate model, the new parameterization predicted more realistic annually averaged cloud water and ice distributions, and cloud radiative properties, especially for sensitive higher latitude mixed-phase cloud regions. As a test of the new global IN scheme, it was compared to independent data collected during the 2008 DOE-sponsored Indirect and Semi-Direct Aerosol Campaign (ISDAC). Good agreement with this new data set suggests the broad applicability of the new scheme for describing general (non-chemically specific) aerosol influences on IN number concentrations feeding mixed-phase Arctic stratus clouds. Finally, the parameterization was implemented into a regional cloud-resolving model to compare predictions of ice crystal concentrations and other cloud properties to those observed in two intensive case studies of Arctic stratus during ISDAC. Our implementation included development of a prognostic scheme of ice activation using the IN parameterization so that the most realistic treatment of ice nuclei, including their budget (gains and losses), was achieved. Many cloud microphysical properties and cloud persistence were faithfully reproduced, despite a tendency to under-predict (by a few to several times) ice crystal number concentrations and cloud ice mass, in agreement with some other studies. This work serves generally as the basis for improving predictive schemes for cloud ice crystal activation in cloud and climate models, and more specifically as the basis for such a scheme to be used in a Multi-scale Modeling Format (MMF) that utilizes a connected system of cloud-resolving models on a global grid in an effort to better resolve cloud processes and their influence on climate.

Project background and overview

The formation of ice in clouds is of vital importance, as ice formation is one of the key processes initiating precipitation. At temperatures warmer than about -38˚C, the freezing of cloud droplets is initiated by heterogeneous ice nucleation, triggered by ice nuclei (IN) that possess surface properties favorable to lowering the energy barrier to crystallization. Only a tiny fraction of all particles serve as IN in the atmosphere, typically on the order of 1 in 10^5, and IN number concentrations can vary dramatically both spatially and temporally. The key aerosol particle types contributing to atmospheric IN include populations such as mineral dusts and biogenic nuclei, whose source strength may be highly variable and significantly impacted by weather and climate feedbacks as well as by anthropogenic disturbances or land-use change. Ice nuclei exert an influence on cold cloud microphysical processes that is disproportionate to their low number concentrations. For example, the concentrations of IN needed to explain observed precipitation rates range from as small as 10^{-3} per standard liter at -10˚C (*1*) to more typical estimates of a few IN per standard liter at -20˚C (*2*). Studies have shown that variations in heterogeneous IN can have significant effects on both dynamical and microphysical processes in clouds and can impact precipitation (e.g. (*3, 4*)), and results from a general circulation model suggest that predicted aerosol indirect effects are sensitive to parameterizations of IN (*5*).

In this project, we have derived a simple, observationally-based parameterization of IN number concentrations. We combine, for the first time, reanalyzed data from 9 field studies occurring in a variety of locations over 14 years. We show, using simultaneous measurements of total ambient aerosol size distributions and IN that a correlation exists between IN concentrations active in supercooled water clouds and the number concentrations of particles larger than 0.5 μm diameter. This size dependence is expected from theoretical considerations (e.g.(*6*)), but also may be due to the fact that particles which are efficient at nucleating ice (e.g. dust and bacteria) tend to fall into this size range. Considering temperature and particle size reduces the spread of potential errors in predicting IN concentrations from a factor of ~1000 to ~10. This improvement leads to significantly more realistic and well-constrained descriptions of aerosol-ice formation relationships.

We additionally explore the validity of the new IN parameterization using a new IN data set collected specifically in the vicinity of Arctic stratus clouds. Finding consistency of the parameterization with this overall IN data set from the 2008 Indirect and Semi-Direct Aerosol Campaign (ISDAC) *(7),* we explore the application of the parameterization in a the cloud-resolving model for predicting cloud properties in ISDAC special observational cases, as well as a prior Mixed-Phase Arctic Cloud Experiment (M-PACE) case that has been the subject of a number of model inter-comparison studies. Within these modeling efforts that constitute the topic of a student thesis, we also explore the general sensitivity of Arctic stratus to changes in IN populations, and the fundamental importance of modeling such clouds using prognostic versus diagnostic ice nucleation schemes. Our results confirm the strong potential sensitivity of Arctic cloud lifetimes and properties to IN abundance, and demonstrate good capability for prediction of ice activation in Arctic stratus using the simple parameterized relation to aerosol concentrations, although challenges remain to explain a general tendency to underestimate ice activation on the basis of known IN properties. This latter fact may reflect IN sampling issues, poor understanding or simulation of mixing processes, or failure to assess all primary and secondary ice formation processes in mixed-phase clouds.

The remainder of this final report consolidates multiple progress reports, and so is intended to be read as a progress report.

Year 1 Objectives and Accomplishments

The major product of first year efforts was a manuscript submitted for publication to *Proceedings of the National Academy of Sciences*. Specific objectives outlined in the proposal are listed here, as well as a more detailed description of progress during this stage of research.

1) Merge aerosol and IN data sets from 10 field programs.

Full methods are outlined in the final publication *(8)*. IN and aerosol data from 11 separate field campaigns were analyzed, and ultimately data from 9 of those studies were utilized. IN number concentration data are all from the Colorado State University Continuous Flow Diffusion Chamber (CFDC). These data came from studies in locations as distinct as the Arctic and the Amazon Basin. The remaining two studies (CRYSTAL-FACE and FIRE-ACE/SHEBA) were not used because the data did not meet the criteria described below. We focused on measurements applicable to mixed-phase clouds. We thus isolated IN concentrations measured at relative humidity with respect to water exceeding 100%, where contributions from all known heterogeneous ice nucleation mechanisms are possible, and at temperatures between -9 and -35˚C. Although we did not specifically address deposition nucleation, studies have shown that deposition nucleation primarily contributes to ice nucleation at temperatures below about -30˚C (*9*), just a few degrees warmer than the lower temperature limit of the data used here.

Counting statistics determined that IN number concentrations lower than 0.3 per liter in standard inlet sampling and 0.01 per liter using a counterflow virtual impactor (CVI) inlet were below levels of quantification and were removed. Lower-valued IN number concentration data were included from the WISP-94 study, for which 200 L grab samples were collected from the aircraft in large conductive bags, and then processed in our laboratory. Additional criteria applied to the screening of IN data were that the measurements during a selected sample period occurred within a 500 m altitude layer and that the CFDC processing temperature and RH over the period did not vary more than 5˚C or 3%, respectively. Cloudy periods were defined on the basis of other microphysical measurements and were removed from consideration, except when sampling was specifically made using a CVI. Finally, IN data were only included if ambient aerosol size distributions were available over the same periods. Both aerosol and IN concentrations were corrected to standard temperature and pressure conditions (STP; 273.15K, 1013.5 mb).

2) Develop an aerosol-dependent IN parameterization as a function of temperature and supersaturation.

Variability in IN concentration at any temperature T ($n_{IN,T}$, std L^{-1}) must relate to spatio-temporal changes in IN particle types, and these changes may mirror changes in the overall atmospheric aerosol population. As such, we explored a parameterization of IN that considered temperature alone versus one that also considered aerosol concentrations. We found significant improvement in predictive ability of a parameterization of $n_{IN,T}$ that links IN number concentrations to both temperature and total aerosol number concentrations exceeding 0.5 μm ($n_{aer,0.5}$, scm^{-3}) diameter. To specify the new relationship, we aggregated our multi-study data set

into 3˚C temperature intervals for fitting the relation between $n_{IN,T}$ and $n_{aer,0.5}$. Correlation coefficients for power law fits were >0.6 (to 0.8). Correlations for individual projects over similar temperature ranges were sometimes much higher. The power law coefficients were then used to determine the overall size and temperature dependencies of IN active under mixed-phase cloud conditions, that is, for activation by condensation/immersion freezing nucleation in the water-supersaturated conditions needed to form supercooled water clouds:

$$n_{IN,T_k} = a(273.16 - T_k)^b (n_{aer,0.5})^{(c(273.16 - T_k)+d)} \quad . \qquad (1)$$

Here a = 0.0000594, b = 3.33, c = 0.0264, d = 0.0033, and T_k is cloud temperature in Kelvin. The new parameterization predicts 62% of the measured IN within a factor of 2 and aligns the overall data set along a 1:1 predictive line (8). In contrast, a previous parameterization (*10*) which is based on humidity alone predicts only 16% of the observed points within a factor of 2 and is biased above the 1:1 line. Based on our prior studies quantifying the compositions of the detected IN (*11, 12*), we expect that the remaining variability can be attributed to variations in chemical composition that are not captured by this parameterization. Nevertheless, a greatly improved degree of constraint on IN number concentrations is achieved, which is needed to model the initiation of ice formation and to thereby represent the main microphysical features of liquid and ice phase distributions in low- and mid-level supercooled clouds in global climate models.

3) Incorporate this new parameterization into models.

Although this objective was expected to begin in Year 2, collaboration with Dr. X. Liu began in Year 1. We incorporated the new ice nucleation parameterization into the CAM3 version including the two-moment microphysics scheme of Liu et al. (*13*) to treat liquid and ice microphysics, thereby predicting both mass and number mixing ratio of cloud particles and explicitly treating liquid and ice mass partitioning in mixed-phase clouds. An aerosol module implemented in CAM3 was used to predict size-resolved aerosol mass and number concentrations. For the aerosol number calculation used in the IN parameterization, sea salt contributions to aerosol concentrations were excluded since these particles are not believed to be an important source for IN. The CAM3 global simulations were run for a 5 year period using climatological sea surface temperatures in addition to the present-day (Year 2000) aerosol emissions. The simulation using the new IN parameterization predicted large increases (10-30 g m^{-2}) in annually-averaged liquid water path, decreases in ice water path up to 10 g m^{-2}, and stronger (5-10 W m^{-2} more negative) shortwave cloud forcing at storm track latitudes and at higher latitudes that is not balanced by positive changes in longwave cloud forcing, compared to simulations with a previous parameterization (*10*). These changes are a consequence of reduced IN number concentrations in regions with low predicted number concentrations of particles larger than 0.5 μm diameter, which in turn inhibits liquid conversion to ice in mixed-phase clouds in these regions. Consequently, cloud cover increases by 5-10% at high latitudes, and downwelling shortwave radiation reaching the surface is reduced in the annual zonal mean *(8)*.

The implied climate sensitivity from these studies is a ~1 W m^{-2} per decade increase of IN number concentration. The strong sensitivity of climate to IN suggests that long-range import of IN from dust storms, boreal biomass burning and anthropogenic pollution could lead to feedbacks on mixed-phase clouds, impacting precipitation and radiation *(8)*.

The major conclusions of the parameterization development and global model simulations are summarized in the appended research highlight submitted to the DOE-ARM website (http://www.arm.gov/science/highlights). Additional simulations using the single-column version of the global model (SCAM-3) were also performed for a well documented case from the Mixed-Phase Arctic Cloud Experiment (M-PACE). The long lived liquid-dominated cloud structure observed for the October 10, 2004 case was well-reproduced in simulations. These simulation results were presented at DOE program meetings.

Year 2 Objectives and Accomplishments

Year 2 of the project focused on analysis needed for independent testing of the new IN parameterization, and steps toward implementing it into further models. Specific objectives listed in the proposal, and the associated activities and accomplishments included:

1) *Test the parameterization against data collected during ISDAC.*

For the parameterization evaluation, ISDAC IN data were provided by Mr. Andrew Glen and Dr. Sarah Brooks of Texas A&M University, whose CFDC flew on the Canadian Convair aircraft and sampled via a forward facing and approximately isokinetic air inlet. Aerosol data from a wing-mounted Passive Cavity Aerosol Spectrometer Probe (PCASP) were provided by Dr. Peter Liu and Mr. Michael Earle of Environment Canada. Analyses followed procedures used in the parameterization development (8). Cloudy periods were first omitted from consideration using a cloud mask and cloud phase analysis provided by Dr. Greg McFarquhar and Mr. Robert Jackson of the University of Illinois as part of a value added ARM PI product (*14*). IN data during cloud-free periods were then combined for all flights and further segregated for times when the CFDC was processing aerosol above water saturation. Passive Cavity Aerosol Spectrometer Probe (PCASP) aerosol concentrations above 0.48 μm (closest size lower bin limit to the parameterization size condition) were determined for the same time periods that satisfied the cloud-free and CFDC water supersaturated conditions required for inter-comparison.

Figure 1 shows a time series of data from one flight segment from ISDAC Flight 17. Predicted IN are shown in comparison to observed IN data prior to application of the water supersaturation and cloud-free masks. Only periods of aircraft passage through precipitating ice crystals are included and the CFDC was operating in supersaturated conditions through the entire period shown. Good agreement is seen between observed and predicted IN values, and the presence of precipitating ice crystals in the sample appears to have only a modest influence on the intercomparison. Similar results are shown for a period on April 26, 2008 in Figure 2. In this case, CFDC water saturation values deviated substantially from supersaturated conditions at times, although at most times was within 5% of water saturated conditions (100% *RH*). Agreement in predicted and observed IN is again reasonably good, easily within the uncertainty involved in the parameterization. This bears well for use of the parameterization to describe IN in these specific case studies, and for generalized use to overcome the limitations of case study IN data that are seldom comprehensive as regards relevance to the thermodynamic and cloud conditions that occurred in each case. An IN instrument can only measure at one set of thermodynamic conditions at one time and an aircraft can only be in one place at one time. This is the advantage to validating an IN parameterization for general use in simulating cloud case studies.

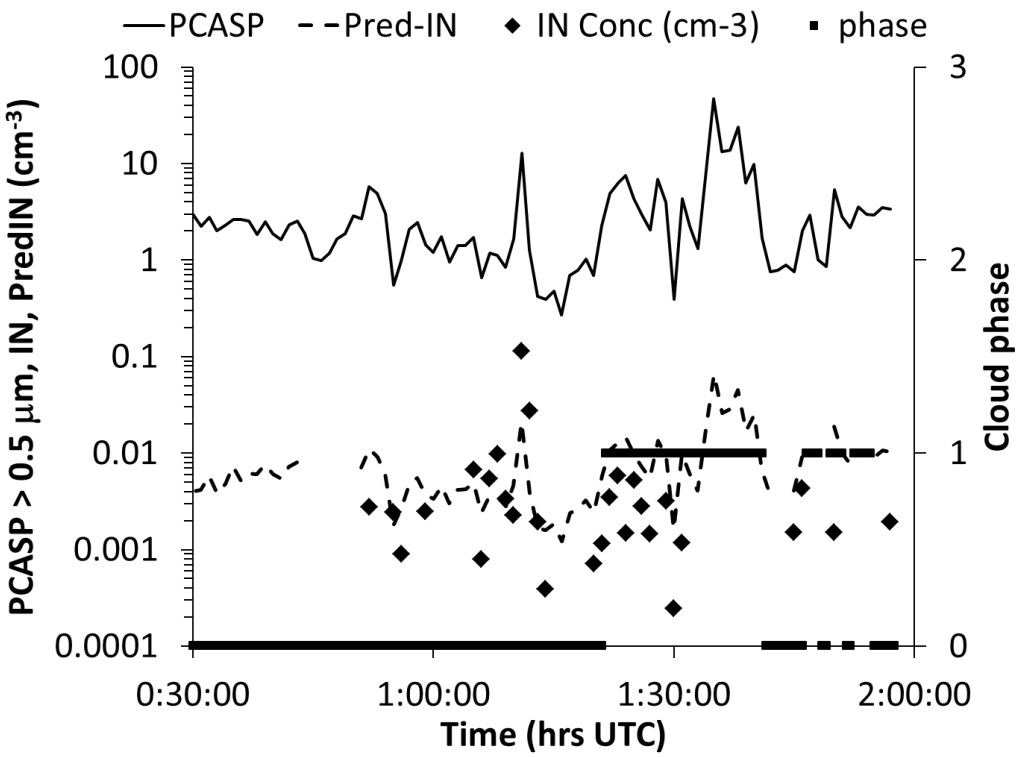

Figure 1. Comparison of observed IN versus IN predicted on the basis of Eq. (1) (dashed curve) for ISDAC Flight 17 on April 8, 2008 (local time). UTC time is for the beginning of April 9, 2008. IN data are for one minute integral periods. IN processing temperatures ranged from -27 to -30˚C during the period shown. PCASP aerosol concentrations at sizes above 0.5 μm and cloud phase (0 = no cloud; 1 = subcloud ice present) are also shown. An ambient pressure range from the surface to 450 mb is represented.

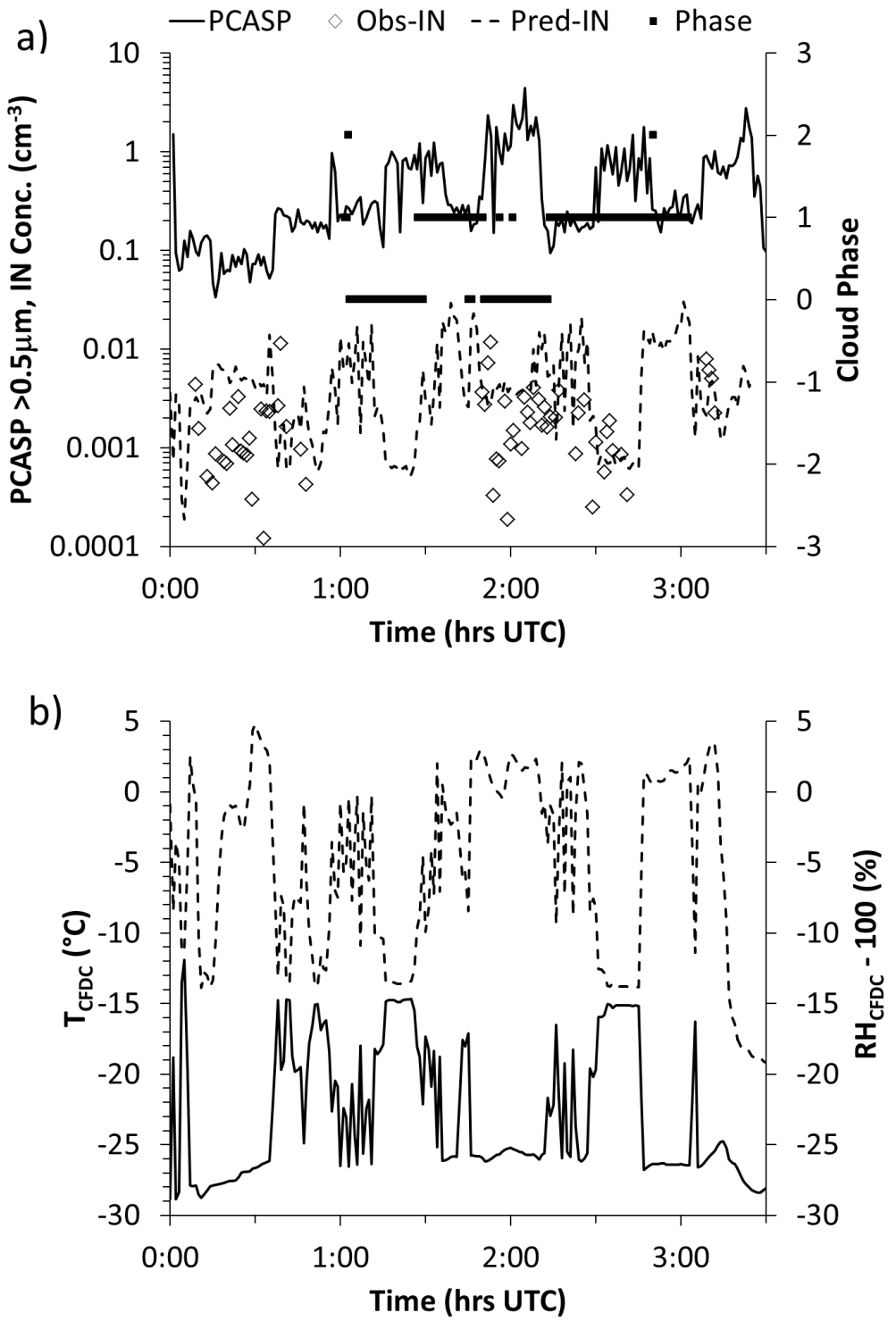

Figure 2. Comparison of observed IN versus predicted IN in a) for a portion of ISDAC Flight 31 on April 26, 2008. Gaps in IN data are periods of sampling cloud particles from a counterflow virtual impactor inlet. PCASP aerosol concentrations at sizes larger than 0.5 μm and cloud phase are also shown. CFDC IN processing temperatures and RH are shown in b). A mix of above (~910 mb, highest T_{CFDC}) and below (~970 mb, lowest T_{CFDC}) cloud sampling is represented.

This research was consolidated with Year 1 results in an acknowledged highlight of the DOE-ASR Cloud, Aerosol, and Precipitation working group for 2011 (see http://asr.science.energy.gov/science/researchhighlights/accomplishments/CAPI_Accomplishments_2011.pdf).

2) Incorporate the new parameterization into models.

The focus during Year 2 turned to implementation within the System for Atmospheric Modeling (SAM), the cloud-resolving model that serves as the core model within the Multiscale Modeling Framework (MMF) that is the basis for the global modeling efforts of the Center for Multiscale Modeling of Atmospheric Processes (CMMAP), and NSF Science and Technology Center at Colorado State University. SAM is a cloud-resolving model with the dynamical framework of a large-eddy simulation model (*15*). This model is widely used and for the CMMAP concept the SAM is used to represent cloud processes separately within each grid cell of a global model called the SP-CAM (Super-Parameterized Community Atmospheric Model). This revolutionary concept is a means of overcoming one of the weakest components of global climate models, the parameterization of cloud processes (*16*). The use of the SAM will permit more explicit treatment of aerosol indirect effects on clouds and climate, through incorporating aerosol-microphysical links such as the simplified IN parameterization developed in the present work.

In a collaborative effort, a CMMAP student (Mr. James Carpenter) was assigned to our project to take on simulations for meeting his M.S. thesis requirements. Mr. Carpenter worked to implement the IN parameterization in the SAM model, to test it for a well-documented previous case from M-PACE, and begin setting up simulations of ISDAC "golden days" (key cases that appear to meet ISDAC objectives to describe single-layer Arctic clouds) (7). Initial simulations were set up for diagnostic prediction of ice nuclei activation, meaning that although initial aerosol concentrations are specified, the concentrations are not affected by cloud activation. Thus, ice activation was allowed in all cases to achieve a limit determined by Eq. (1) as a function of temperature. Sensitivity of Arctic stratus simulations to IN number concentrations in the diagnostic implementation were performed for the Flight 16 (April 8) case of ISDAC to show that the lifetime of Arctic clouds can potentially be sensitively controlled by IN, as proposed previously (*17*). If IN were increased by an order of magnitude, a dominant ice process took place that greatly increased precipitation, but reduced liquid cloud lifetime, which an order of magnitude IN number decrease shut off precipitation and led to gross underestimates of cloud ice number and mass. Preliminary reports of these results were made at DOE Science Team meetings, as listed in the presentation list later in this report.

Year 3 (No-cost Extension) Objectives and Accomplishments

No-cost extension of this grant was merited on the basis of the continuation of numerical modeling studies as part of Mr. Carpenter's thesis, and the intent to include the comprehensive ISDAC IN analyses with his work in a final publication (submission planned for December 2012).

1) Test the parameterization against data collected during ISDAC

A project summary comparison of ISDAC IN data with the new IN parameterization for conditions meeting all criteria was completed, shown in Figure 3. In keeping with the single case examples shown in Figures 1 and 2, Figure 3 indicates that the ISDAC project data set as a whole is consistent with the compendium of data used for the parameterization development (*8*). Only a very few data points fall outside the bounds of a plus or minus factor of 5 range about the predicted values. Data points shown in blue in Figure 3 are one-minute data from above cloud top at around 2200 to 2230 UTC on April 8, 2008 when the Texas A&M CFDC was processing at below 90% RH. The highest IN concentrations during this period were used as constraint on maximum IN number concentrations in recently reported cloud-resolving model simulations of the April 8 case (*18*). Based on our analyses, these data represent extreme under-prediction outliers for the aerosol concentrations present and CFDC processing temperatures (-21 to -23˚C) used at the time, and exceed IN concentrations measured above cloud at similar aerosol concentrations, but at much lower processing temperatures, just a few hours later (Figure 1). Further implications are discussed by Carpenter (M.S. thesis in preparation, Fall 2012).

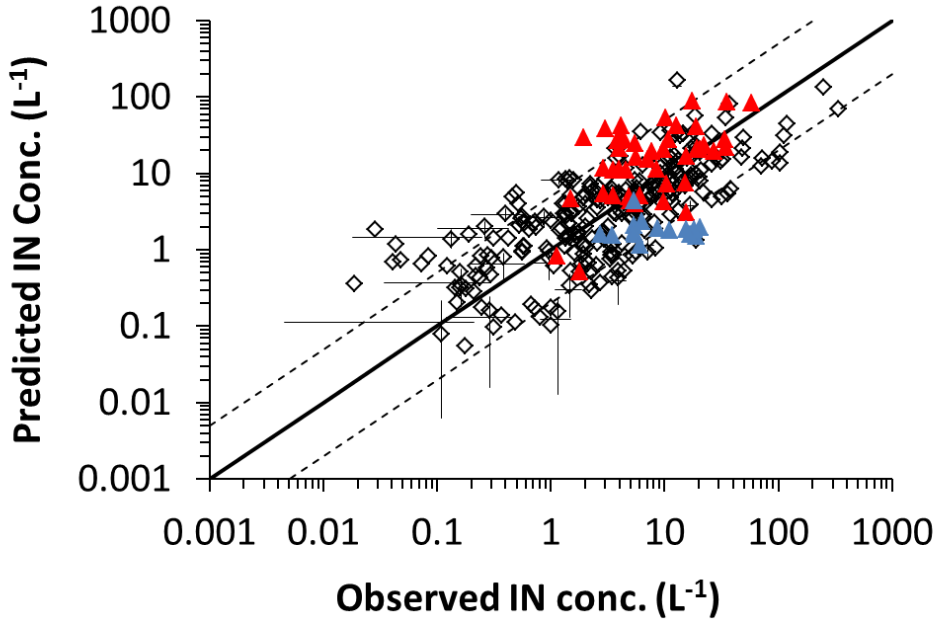

Figure 3. Summary of ISDAC IN number concentration (red points) relation to IN concentrations predicted using ISDAC aerosol data in Eq. (1) overlaying original parameterization development data (*8*). All data are for standard temperature and pressure conditions for 3 to 20 minute time periods. The dashed lines parallel to the solid line (1:1) bracket a factor of 5 times the standard relation from Eq. (1). Points highlighted in blue are observed 1 minute data from the time period on April 8 (Flight 16) utilized by Avramov et al. (2011), who took values exceeding 10 per liter to constrain IN number concentrations in numerical simulations of the April 8 case from ISDAC. These blue data periods do not satisfy the conditions for processing in the water supersaturated regime (RH was below 90%).

In coordination with laboratory and field research funded by the National Science Foundation, we also began to explore the potential development of composition-specific IN

parameterizations in the style of the one developed in this research (*8*). It was hypothesized that some of the remaining unexplained variance shown in Figure 3 is due to variations in dominant IN composition for species that are more or less active than the global average (*8*). In Figure 4 are shown data from both laboratory and field measurements on mineral dust aerosols that show both a remarkable uniformity for different dust types, and a consistently higher activity than predicted by the global IN parameterization in (*8*). Excellent description of these results (dashed lines in Fig. 4) is possible using a slightly modified parameterization of the form,

$$n_{IN,T_k} = (n_{aer,0.5})^a 10^{(b(27316-T_k)+c)} \quad (2)$$

We have additionally examined such relations for other dominant species such as smoke, sea spray, and biological aerosols in laboratory and field situations. These results will be summarized in a paper in preparation (DeMott et al. 2013, unpublished). This research holds great promise for providing parameterizations for the condensation/immersion freezing activity of IN of specific compositions for use in cloud and climate models that carry different aerosol types.

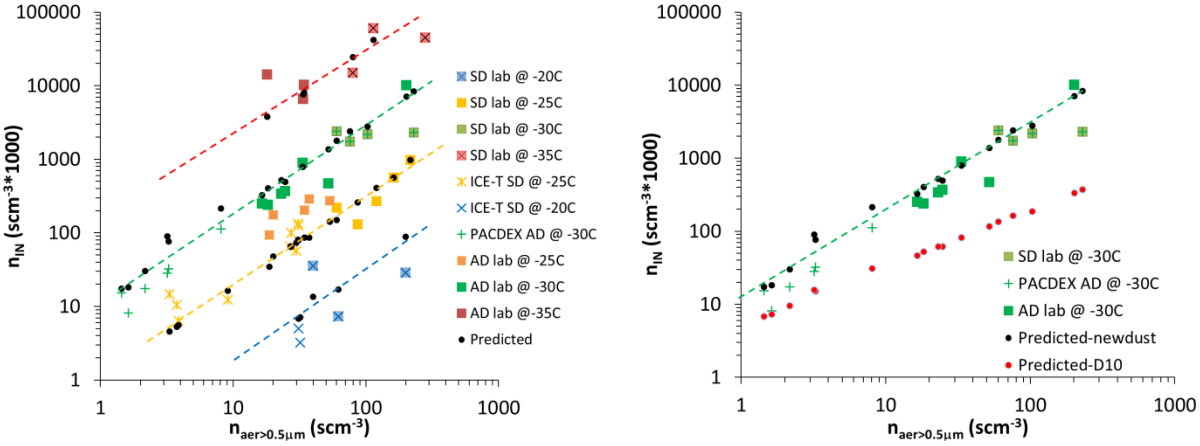

Figure 4. Relations between ice nuclei number concentrations active in the condensation/immersion freezing mode (above water saturation) and total aerosol concentrations larger than 0.5 μm for sampling of specific mineral dust types in the laboratory or within atmospheric plumes. Dust types are classified generally as Saharan (SD) or Asian (AD). Best fit predictions of coefficient fits to Eq. (2) are shown by black data points. Specific temperatures for processing are highlighted by different colors of data and parameterization fits. The left panel summarizes data for a 15˚C range of temperatures, while the right panel shows the under-prediction of IN by the global parameterization from (8), termed D10 here, compared to the mineral dust-specific parameterization ("newdust") valid when mineral dusts are dominant as IN.

2) Incorporate this new parameterization into models.

Two types of simulations have now been performed for ISDAC "golden day" cases. First, the SAM model was run in diagnostic mode as described in Year 2 accomplishments, and then model revisions were made to perform simulations with prognostic prediction of ice formation, including sinks and sources of aerosols that have an explicit connection to IN via Eq. (1). For this purpose, tracer fields were implemented in SAM. Rather than monitoring the total aerosol

field to trigger ice activation, we have begun with a primitive prognostic treatment, in which the available ice nuclei field is the tracer, and is limited to a value representative of the expected IN concentration at the coldest cloud temperatures. As ice nuclei activation occurs, the number activated at each time step is subtracted from this initial pool, and these may be returned due to ice crystal sublimation. We expect to advance this prognostic scheme to multiple bins and dynamic aerosol fields in the future. Details will be provided in a future publication.

Simulations performed for the ISDAC flights on 8 April 2008 indicated an excellent ability to reproduce cloud properties in diagnostic simulations using the new IN parameterization (*8*). Figure 5 shows a long-lived cloud dominated by liquid in the upper layers, as observed (7), and cloud ice number concentrations approaching a sustained value of about 0.00035 cm^{-3}, in agreement with observations ice crystal number concentrations that ranged from 0 to 0.0008 cm^{-3} (*14*). Surface precipitation values approaching 0.2 mm hr^{-1} are only modestly less than average rates of 0.34 mm hr^{-1} measured at the DOE-ACRF-NSA site near Barrow, Alaska.

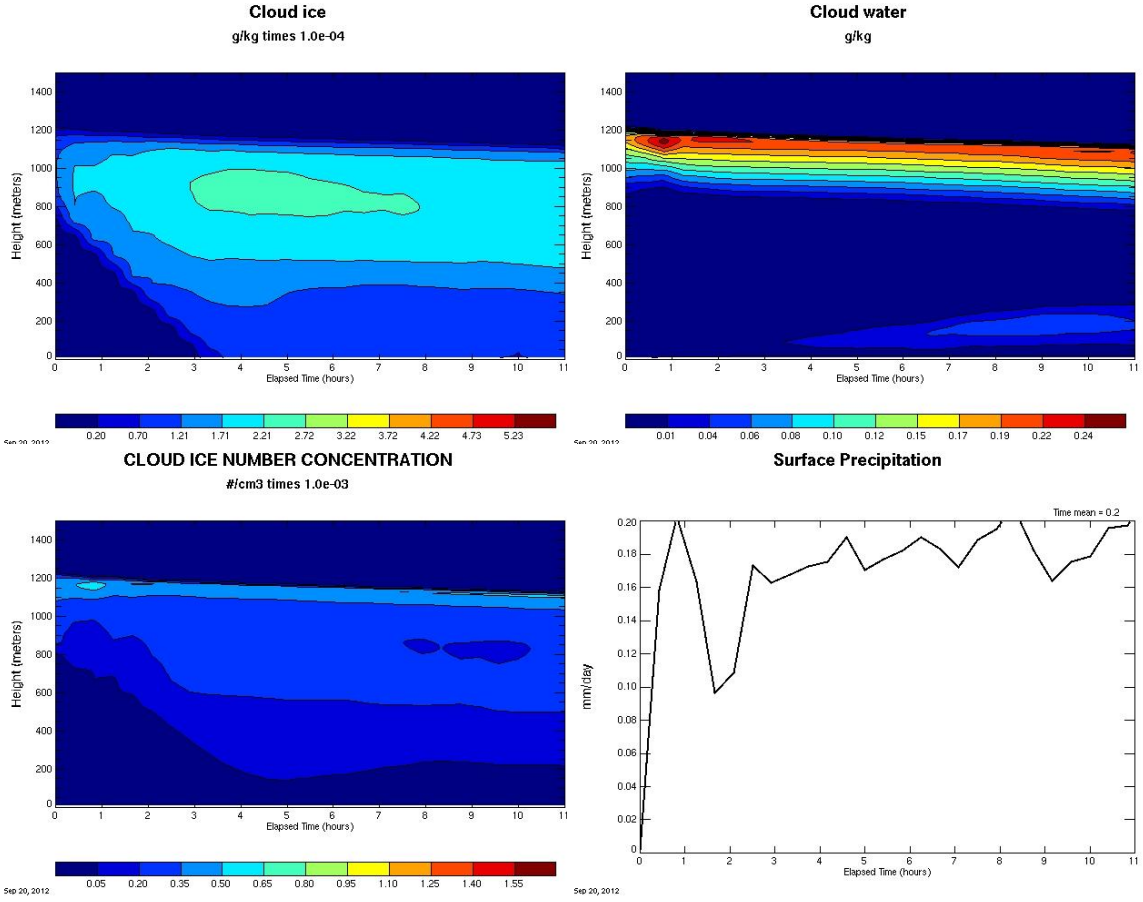

Figure 5. Selected SAM output for the April 8, 2008 ISDAC cloud case from a diagnostic implementation of Eq. (1). The horizontal axis is time, and the vertical axis is height. From the upper left, clockwise: cloud ice mass in g/kg, cloud water mass in g/kg, surface precipitation rate in mm/day, and cloud ice number concentration in #/cm^3. Adapted from Carpenter (2012).

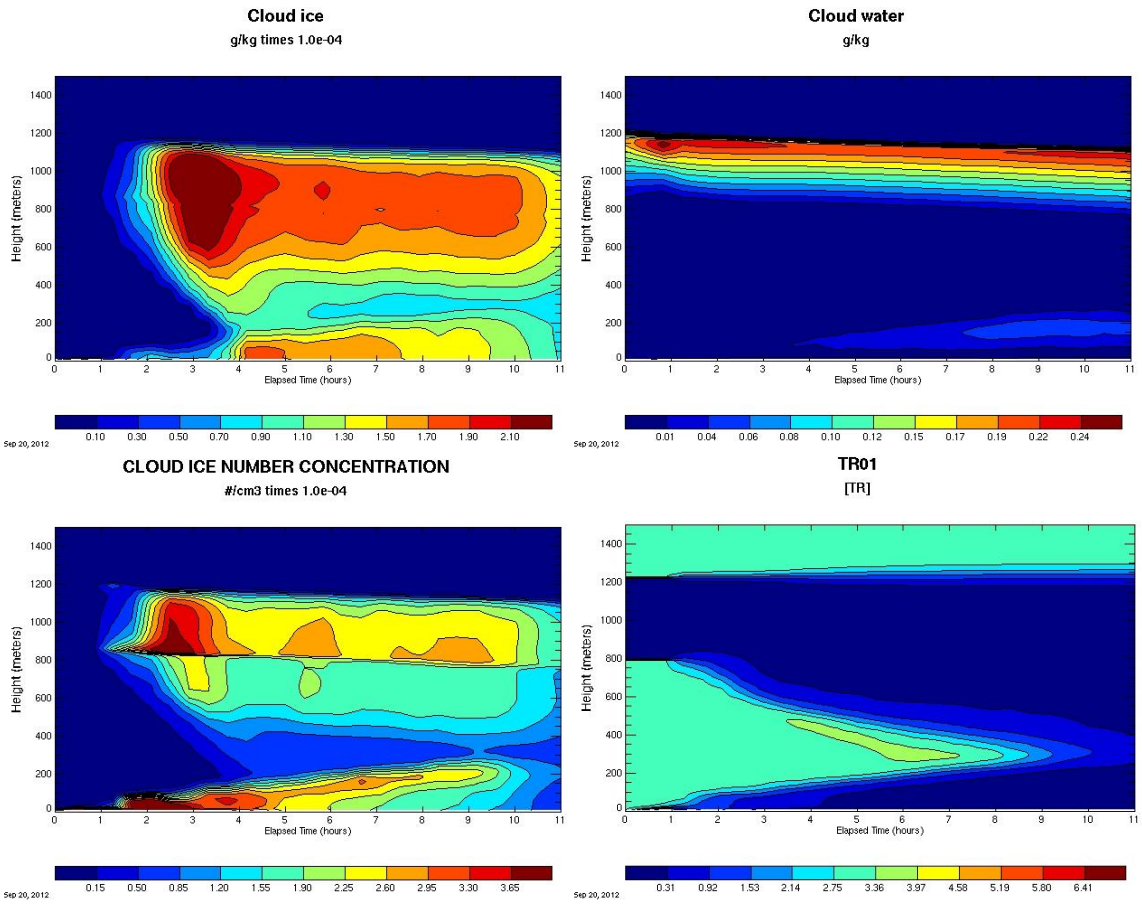

Figure 6. Selected SAM output from a prognostic IN simulation with IN specified at 5x the global IN parameterization (*8*), and including IN recycling due to all sublimating ice species. The horizontal axis is time, and the vertical axis is height. From the upper left, clockwise: cloud ice mass in g/kg, cloud water mass in g kg^{-1}, potential IN concentration in L^{-1}, and cloud ice number concentration in cm^{-3}. Adapted from Carpenter (2012)

Prognostic treatments of IN were tested in two basic forms. The first simulations were set up to run with IN present in the layer in which the cloud forms, similar to the diagnostic form, with the exception that activated nuclei were depleted from the initial pool. The results of these simulations show that activation occurs very efficiently, rapidly depleting the pool of IN, leading to a short-lived cloud that rapidly dissipates. This is fully consistent with other recent cloud-resolving model studies of Arctic stratus ice nucleation (*19*).While it takes time for the activated crystals to grow to sufficient size to settle out of cloud, no additional activation occurs after the initial time steps, resulting in no other possible outcome than cloud dissipation. In order to simulate ice activation as would be expected to occur deep within a cloud system where IN enter the cloud within the limitations of entrainment and mixing from below, additional simulations were run where IN initially populated only the layers above and below cloud top. In simulations using this case, activation prognostically becomes much less efficient, such that predicted ice crystal concentrations with the standard scheme (Eq. 1) remain several times less than observed, consistent with expectations from other studies (19), but inconsistent with conclusions that IN

"closure" with ice concentrations was readily achieved in ISDAC (18). At face value, these results appear to require a missing primary or secondary ice activation source, or the presence of an IN population that is drastically different than that represented in the new IN parameterization. We therefore have explored sensitivity studies of altering IN within reasonably expected bounds. In Figure 6 are shown prognostic simulation results of the April 8 case for which the IN number concentrations in Eq. (1) were increased a factor of 5 times to account for the maximum uncertainty in the global IN parameterization (see Figure 3), a situation that may also be explained by the possible presence of mineral dust aerosols as the predominant IN type at times in the Arctic Spring. In this case, a cloud is simulated in much better correspondence with microphysical observation, at least in terms of ice crystal concentrations. Studies are continuing at this time to explore other case simulations, and reasons for typical underestimates of ice water contents compared to observations. Results will appear in Mr. Carpenter's thesis and will be reported in a publication acknowledging this grant, likely in early 2013.

Publications and Outreach

Publications acknowledging this grant

Carpenter, J. M., 2012: Simulations of Arctic mixed-phase clouds using a new aerosol-linked ice nuclei parameterization in a prognostic ice prediction scheme, M.S. Thesis, Colorado State University (in preparation).

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DeMott, P. J., A. J. Prenni, X. Liu, T. Eidhammer, C. H. Twohy, J. L. Stith, D. C. Rogers, M. D. Petters, S. M. Kreidenweis, M. S. Richardson, R. Subramanian, and A. J. Heymsfield, Multi-project analyses of ice nuclei relation to other aerosols and ice in clouds, *MOCA-09 Joint Assembly: Our Warming Planet*, Montreal, Canada, 2009.

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Related studies partially motivated by this work

Xie, S., X. Liu, C. Zhao, and Y. Zhang, Sensitivity of CAM5 Simulated Arctic Clouds and Radiation to Ice Nucleation Parameterization, *Journal of Climate*, in revision, 2012.

DOE highlights involving this grant

A highlight of Year 1 studies is published on the DOE-ARM website (http://www.arm.gov/science/highlights).

A highlight of Year 2 studies is published on the DOE-ASR website: (http://asr.science.energy.gov/science/researchhighlights/accomplishments/CAPI_Accomplishments_2011.pdf).

These documents are attached to this report as Appendix A.

Other

Dr. DeMott and Dr. Prenni actively participated in DOE science and working group meetings, and Dr. DeMott helped form a thematic focus group on ice nucleation within the CAPI group, in collaboration with Dr. Liu and Dr. Wang. Research highlights were also provided, as noted.

Collaborators on this research

Dr. Xiaohong Liu, Pacific Northwest National Laboratory
Mr. Mark Branson, Colorado State University
Dr. Sarah Brooks and Mr. Andrew Glen, Texas A&M University
Dr. Peter Liu and Mr. Michael Earle, Atmospheric Environment Service of Canada
Dr. Greg McFarquhar and Mr. Robert Jackson, University of Illinois Urbana-Champaign
Zhien Wang, University of Wyoming

Education

Mr. James C. Carpenter has participated in this research at no cost to the grant, via collaboration with the NSF-funded CMMAP program at Colorado State University. The research conducted on this grant was used toward partial fulfillment of Mr. Carpenter's M.S. degree requirements, with completion scheduled for December 2012.

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Appendix A: DOE Highlights

Ice Nucleation Link to Aerosols for Global Models

Research Highlight

The formation of ice in clouds is of vital importance to life on Earth, as ice formation is one of the key processes for precipitation initiation. Since ice nucleation is tied to the action of specific aerosol particles (ice nuclei: IN) representing only 1 in 10^5 or less ambient particles, natural and human impacts on ice nucleation can alter the energy and hydrological cycles. This study has addressed the urgent need for descriptions of complex ice formation processes that encapsulate the environmental dependencies of ice formation, but also include a link to aerosol properties. This paper describes an observationally based, yet simple, parameterization of IN number concentrations active for mixed-phase cloud conditions as a function of temperature and aerosol number concentrations (DeMott et al. 2010). Strong differences compared to a commonly applied ice nucleation parameterization without links to aerosol properties are demonstrated in simulations with the NCAR CAM3 (Community Atmospheric Model-3) global climate model with two-moment microphysics.

Ice nuclei number concentration data from the Colorado State University continuous flow diffusion chamber for activation conditions in mixed-phase clouds were assembled from nine field studies occurring at locations from the Arctic through the mid-latitudes to the Amazon. As much as three orders of magnitude variation of IN number concentrations were observed at any temperature, consistent with previous single-parameter IN parameterizations (Figure 1a). Using simultaneous measurements of total ambient aerosol size distributions, it was shown that a correlation exists between observed IN concentrations and the number concentrations of (non-sea salt) particles larger than 0.5 micron diameter (DeMott et al. 2010) that reduces the spread of potential errors in predicting IN concentrations as a function of temperature to less than a factor of 10, and predicts about two-thirds of values within a factor of two (Fig. 1b). This relation likely reflects a predominant role of larger particles such as mineral dusts as IN.

The developed parameterization was compared to the previous parameterization of Meyers et al. (1992) to examine the implications of including sensitivity to both temperature and aerosol concentrations determined from the expanded IN database in five-year global climate simulations using prescribed present-day aerosol fields. The new IN parameterization predicts large increases (10-30 g m^-2) in annually averaged liquid water path (Figure 2a) and stronger (5-10 W m^-2 more negative) shortwave cloud forcing at storm track and higher latitudes (Figure 2b) that is not completely balanced out by positive changes in longwave cloud forcing, such that a net, globally averaged cloud forcing change of 1.3 W m^-2 occurs. These changes are a consequence of reduced IN number concentrations in regions with low predicted number concentrations of non-sea salt particles larger than 0.5 micron diameter; the lower IN concentrations inhibit liquid conversion to ice in mixed-phase clouds in these regions, increasing high latitude cloud cover and reducing annual zonal mean downwelling shortwave radiation at the surface.

The developed parameterization significantly improves constraint and representation of aerosol impacts on cold clouds for immediate use in global climate simulations. Based on presented modeling studies, it appears just as important, for the accurate representation of cloud forcing, to properly simulate the lack of available IN as it is to simulate the presence of IN. Further improvements in parameterizations should follow from future improvements to IN measurement methods and the more challenging collection of enough data to account for variations in IN chemical composition that are not captured by the new parameterization.

Additional submitter: Anthony J Prenni

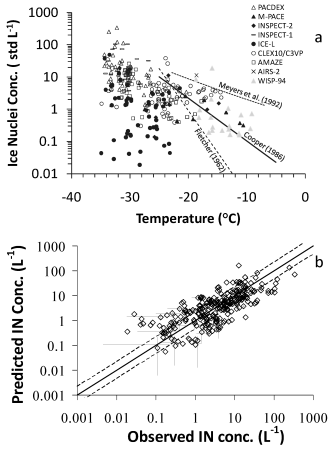

Figure 1. (a) IN number concentration for mixed-phase cloud conditions versus temperature from nine studies. Data points are 5–30 minute averaged data. Single variable (T, RH) parameterizations (see DeMott et al. 2010). are labeled and plotted over the data. b) Relation between observed IN (from panel (a) to IN predicted by the new parameterization linking temperature and aerosol concentrations larger than 0.5 microns. Dashed lines indicate observations (62%) within a factor 2 of 1:1 line.

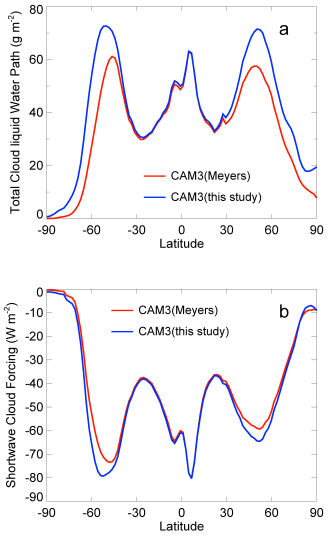

Figure 2. CAM3 global model simulation results using the IN parameterization of Meyers et al. (1992), referred to as CAM3 (Meyers), and the new aerosol-linked parameterization (this study). Shown are annual zonal averages of (a) tropospheric integrated liquid water path, and (b) shortwave cloud forcing.

Ice Nucleation Link to Aerosols for Global Models

Reference(s)
DeMott PJ, AJ Prenni, X Liu, SM Kreidenweis, MD Petters, CH Twohy, MS Richardson, T Eidhammer, and DC Rogers. 2010. "Predicting global atmospheric ice nuclei distributions and their impacts on climate." Proceedings of the National Academy of Sciences, 107(25), doi:10.1073/pnas.0910818107.

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Working Group(s)
Aerosol Life Cycle, Cloud-Aerosol-Precipitation Interactions

Ice Nucleation Link to Aerosols: Improving Modeling of Aerosol Impacts on Clouds and Precipitation

Paul J. DeMott, Anthony J. Prenni /Colorado State University
Xiaohong Liu / Pacific Northwest National Laboratory

ASR Atmospheric System Research

Science Question

Do simplified relations exist between aerosol physical and chemical properties and the number concentrations of **ice nuclei (IN)** for improving global modeling of mixed phase clouds and precipitation?

Approach

- Use large data base of IN measurements with co-sampled aerosol data to parameterize IN number concentration as a power law function of aerosol concentration at sizes >0.5 μm and temperature
- Use DOE ISDAC (Arctic) study data to test robustness of parameterization
- Implement in global model simulations

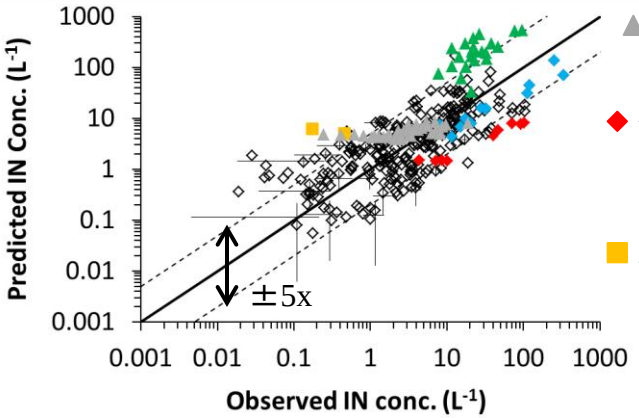
Results

- Parameterization agrees with ISDAC IN data. Need to incorporate chemical speciation of IN.
- Inclusion of aerosol size and T sensitivities strongly impacts mixed phase cloud properties and forcing in global simulations.

Publication

DeMott, P.J., A. J. Prenni, X. Liu, et al.,, 2010: Predicting global atmospheric ice nuclei distributions and their impacts on climate, Proc. Natnl. Acad. Sci., 107 (25), 11217-11222.

New understanding of IN variability in the atmosphere

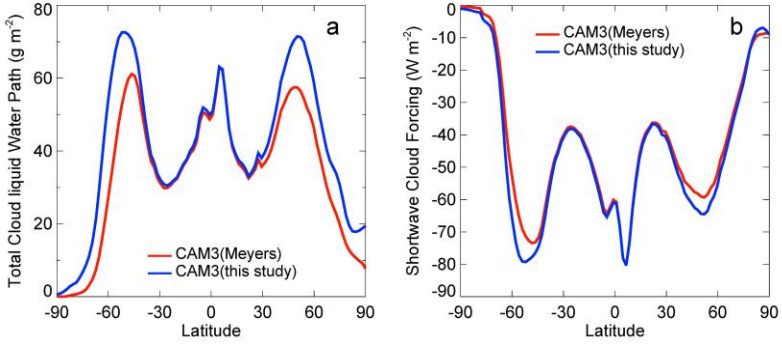


ISDAC IN conc. consistent with global parameterization

IN conc. under-predicted in Saharan (red) and Asian (blue) dust plumes.

IN conc. over-predicted in pollution (yellow) and smoke (green) plumes.

Strong sensitivity to IN in global model simulations



Net globally-averaged cloud forcing decreases compared to a previous aerosol-independent IN scheme. Reduced [IN] in regions with low concentrations of large particles inhibits cloud liquid conversion to ice, increasing high latitude cloud cover and reducing annual zonal mean downwelling shortwave radiation at the surface.