Atmospheric System Research (ASR) Cloud-Aerosol-Precipitation Interactions Science Plan

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Introduction

The goal of the Atmospheric Systems Research (ASR) Program is to improve understanding and model representation of a variety of atmospheric processes and their interactions. As described in the ASR science plan, ASR research is grouped into three areas: aerosol life cycle, cloud life cycle, and cloud-aerosol-precipitation interactions. The strategy for improving understanding and representation of atmospheric processes in climate models is illustrated in Figure 1, taken from the ASR science plan. It involves:

- collecting data from field studies and laboratory experiments
- analyzing the data to improve understanding of atmospheric processes
- developing process models based on that understanding
- applying the process models to high-resolution atmosphere models
- evaluating the high-resolution models using data from the field studies
- using the high-resolution models to guide the development of parameterizations suitable for coarser resolution global climate models (GCMs).

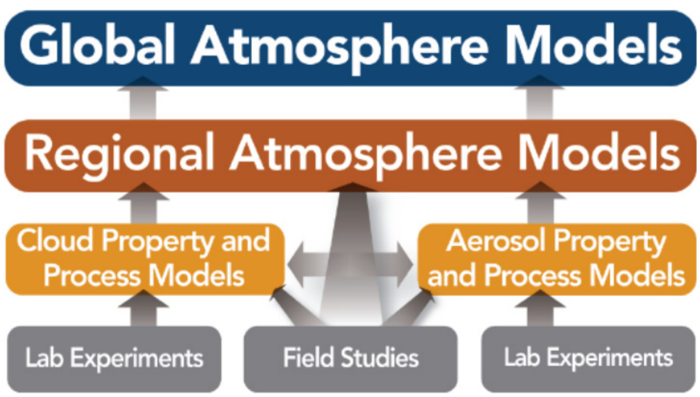

Figure 1. The ASR strategy for improving understanding and representation of atmospheric processes in climate models.

This science plan for the Cloud-Aerosol-Precipitation Interactions (CAPI) component of ASR describes the three primary science questions being addressed by the CAPI Working Group, and recommends field experiments, designed to collect the measurements needed to address them; analysis that uses measurements to improve understanding of CAPI processes associated with the questions; and the process modeling and parameterizations used to represent that understanding in climate models. The purpose of this plan is to provide a framework for focusing the efforts of the CAPI Working Group. It also represents the consensus of the CAPI Science Steering Committee following discussions with the CAPI Working Group. While the U.S. Department of Energy (DOE) ASR Program Managers have read the plan and consented to its publication, it does not necessarily represent their views on CAPI science. A CAPI Work Plan will subsequently be developed to identify and coordinate specific tasks and scientist assignments dedicated to achieving the objectives described in this plan.

Each primary science question is addressed through three objectives:

- Building measurement and modeling capacity.
- Improving understanding of microphysical processes.
- Improving understanding of the sensitivity of cloud systems to aerosol perturbations.

The CAPI Working Group approach to addressing each science question is described in terms of achieving each of these three objectives.

CAPI Primary Science Questions

1. What processes control diversity in the sensitivity of warm low clouds to aerosol perturbations, and why do GCMs seem to overestimate the sensitivity?

Microphysical, structural, and dynamical properties of low, liquid-phase clouds all show sensitivity to aerosol loading, but the responses are not uniform. In general, an increase in aerosol concentration increases cloud droplet concentration and reduces droplet size. These changes impact cloud albedo, precipitation, and cloud dynamics. However, the magnitude and even the sign of the response of various cloud-field characteristics (such as depth, liquid water path, cloud fraction) appear to depend upon cloud type and meteorological regime, cloud-field organization (itself a function of precipitation), and aerosol loading in the unperturbed clouds (Stevens and Feingold 2009; Ghan et al. 2013; Carslaw et al. 2013). Understanding which cloud regimes are more or less resilient to aerosol perturbations is fundamental to understanding the attendant radiative forcing.

A large fraction of global model estimates of the top-of-atmosphere radiative forcing from the aerosol indirect effect (AIE) is more negative than -1.5 W m^{-2}. Such values are difficult to reconcile with observed 20th century temperature records and with estimates of warming caused by the combination of increasing greenhouse gases and the direct effect of increasing aerosol (Kiehl 2007). The strongly negative AIE values tend to be associated with large predicted increases in cloud liquid water path (LWP) with increasing aerosol (Wang et al. 2012). However, cloud-resolving model simulations often find small or (under some common conditions) negative responses of LWP to increasing aerosol (Ackerman et al. 2004; Bretherton et al. 2007), and a recent analysis of satellite retrievals of aerosol and precipitation frequency (Wang et al. 2012) suggests that the sensitivity of precipitation occurrence and LWP to aerosols is overestimated in most global models.

Key challenges for the CAPI Working Group are to provide observational constraints on the AIE using Atmospheric Radiation Measurement (ARM) Climate Research Facility observations, understand why climate models produce a much stronger increase in LWP than do cloud models, and identify physically based ways to produce sensitivities to aerosols in global models that are more in line with high-resolution models and observations. By applying ARM observational facilities and ASR state-of-the-art numerical modeling expertise, the CAPI Working Group can address the challenge of understanding the diversity of warm low cloud responses to aerosol perturbations, and represent the sensitivity more realistically. The strategy will largely follow that illustrated in Figure 1, pursuing the three primary objectives.

Objective 1.A. Building Measurement and Modeling Capacity

The first challenge is measuring and characterizing the meteorological parameters that control cloud formation, a task in common with the Cloud Life Cycle Working Group. Second, researchers need to characterize cloud microphysics (droplet effective radius and number concentration), cloud macrophysics (LWP, cloud depth, cloud-base, precipitation), vertical velocity and its moments, aerosol parameters such as sub-cloud condensation nuclei, and radiation (spectral direct and diffuse solar radiation). Third, researchers need these measurements across a variety of aerosol and meteorological conditions to isolate the cloud responses to changes in aerosol within the context of the cloud controlling parameters.

A variety of ARM field studies have produced such data. The Variability of the American Monsoon System (VAMOS) Ocean-Cloud-Atmos-Land Study (VOCALS) study over the Southeast Pacific Ocean collected in situ data from aircraft flying above, below, and within stratocumulus clouds in a region with strong spatial gradients in aerosol concentration. The deployment of an ARM Mobile Facility at the Azores provided 21 months of remote sensing retrievals of stratocumulus clouds. Similarly, the Cumulus Humilis Aerosol Processing Study (CHAPS) study sampled cloud and aerosol within, below and above continental shallow cumulus clouds for one month, and Routine ARM Aerial Facility Clouds with Low Optical Water Depths (CLOWD) Optical Radiative Observations (RACORO) extended the CHAPS measurements for 6 months. The Indirect and Semi-Direct Aerosol Campaign (ISDAC) sampled aerosol and cloud properties in Arctic stratocumulus from aircraft, and the Two-Column Aerosol Project (TCAP) provided both aircraft and surface-based and aircraft remote sensing of cloud and aerosol off Cape Cod during summer and winter. Marine ARM GPCI[1] Investigation of Clouds (MAGIC) Pacific Cross-section Intercomparison) offered the first deployment of the ARM Mobile Facility on a cargo ship traveling repeatedly between Los Angeles and Hawaii.

Several additional field studies needed to collect useful data are being proposed, including an aircraft campaign over the Azores to validate microphysics, vertical velocity and entrainment retrievals for low warm clouds, and a campaign to characterize clouds, aerosols and their interactions in a pristine atmosphere, such as in the Southern Hemisphere storm track.

Data from past and future field studies and from the fixed ARM sites can be used to address this science question, but additional retrieval products are needed to characterize aerosol effects on low warm clouds. Additional effort is required to better retrieve cloud condensation nuclei (CCN) concentration at cloud-base, retrieve droplet number concentration and droplet effective radius from the surface and from satellite, retrieve cloud LWP for thin clouds and for drizzling clouds, retrieve light drizzle, estimate subadiabaticity in LWP, measure the entrainment rate above stratocumulus clouds, retrieve updraft velocity at cloud-base, estimate precipitation susceptibility for low clouds, calculate the sensitivity of the probability of precipitation to CCN concentration (S_{pop}), determine aerosol-cloud interaction (ACI) metrics in different low cloud regimes, and estimate the aerosol scavenging and precipitation efficiencies.

A variety of models are also needed to address this science question across multiple spatial scales. Detailed physically based process models and process parameterizations both represent key processes such as droplet formation and collision-coalescence in a simple dynamic framework. Large Eddy Simulation (LES) models explicitly resolve the dominant scales of processes involved with cloud

[1] GPCI: Global Energy and Water Cycle Experiment (GEWEX) Cloud System Studies (GCSS).

formation and aerosol-cloud-precipitation interactions using either bin or double-moment representations of both the aerosol and droplet size distributions. LES simulations can provide benchmark results to guide the development of cloud and aerosol modules for global models. Regional models provide more realism and a larger range of scale interactions, albeit at reduced resolution. Global models simulate the global life cycle of clouds, aerosols, and their interactions using either physically based parameterizations or embedded explicit reduced-dimension models that can be run in either single-column or global mode. Most global models employ a diagnostic rather than prognostic treatment of precipitation, and neglect covariance of cloud water and rain water as well as aerosol effects on shallow cumulus clouds.

The ARM Facility is investigating the operation of a high-resolution cloud-resolving or LES model dedicated to regular simulations of clouds over the Southern Great Plains (SGP) site. While the details of the model configuration are not yet determined, the model is expected to produce benchmark simulations of boundary layer clouds and interactions with aerosol that could be used to guide the development of parameterizations of clouds, aerosols, and their interactions in coarser resolution global models.

Objective 1.B. Improving Understanding of Microphysical Processes in Boundary Layer Clouds

The data from aircraft measurements and remote sensing should be used to improve understanding of key microphysical processes in boundary layer clouds such as the influence of turbulent mixing/entrainment on droplet formation and growth, cloud processing of the aerosol through activation, collision-coalescence, aqueous chemistry, and resuspension or removal, precipitation efficiency, and radiative closure.

Retrieved droplet number concentration should be compared with number estimated from retrieved cloud-based CCN spectrum and updraft velocity and models of droplet nucleation to test understanding of droplet formation. Turbulence and other measures of variability simulated by the models should be compared with retrievals and in situ measurements. Measured and modeled relationships between aerosol and cloud properties and dynamics (updraft speed, entrainment rates, rainfall frequency and rate) should be compared to ground-based, aircraft, and satellite retrievals to test understanding of the droplet and drizzle formation processes that drive the relationships. The fraction of precipitation from autoconversion simulated by cloud-resolving models (CRMs) and GCMs should be compared with averaged measurements and with LES simulations using bin microphysics to determine the influence of subgrid variations in droplet number and cloud liquid water and rain water on the relative importance of autoconversion and accretion, and the dependence on whether rain is treated diagnostically or prognostically. Spectral direct and diffuse solar radiation measurements should be used to evaluate the sensitivity of cloud and aerosol optical signatures to variations in cloud and aerosol microphysical properties.

Objective 1.C. Improving Understanding of the Sensitivity of Boundary Layer Cloud Systems to Aerosol Perturbations

Although validation of the representation of individual processes is important, it is the behavior of the fully coupled cloud-aerosol system that determines the sensitivity of low clouds to changes in aerosol sources. Data and modeling should be used to understand the resilience of different cloud regimes to aerosol perturbations and to provide observational constraints on the AIE with careful consideration of the meteorological drivers. Particular attention to the scale-dependence of relationships is needed. Differences in precipitation formation and impacts on the moisture and energy budgets, cloud dynamics and ultimately cloud optical depth, LWP, and cloud cover should be explored. Retrieved and simulated ACI metrics should be systematically examined to determine how they change with cloud dynamics and spatio-temporal scale. The retrieved susceptibility of precipitation to aerosol effects, and to droplet number concentration should be compared with simulations by CRMs and single-column models (SCMs). The value of the sensitivity of precipitation probability to aerosol (S_{pop}) simulated by LES and GCMs should be compared with retrieved values. Simulations by SCMs and the LES model driven by the same boundary conditions from a GCM should be compared with each other, with other observations, and then analyzed to determine the causes of the inferior simulation by most SCMs, and to provide a pathway toward improving aerosol effects on warm shallow clouds in GCMs.

2. **What aerosol-related processes control deep convective cloud properties relevant to climate (precipitation, cloud radiative forcing, latent heating profiles)?**

Deep convection has a powerful impact on the Earth's energy balance and water cycle. Recent work (Li et al. 2011; Tao et al. 2012) suggests that under some conditions atmospheric aerosol loadings can modify the convective characteristics, precipitation intensity and radiative impacts of these clouds. Cloud-resolving (or cloud system-resolving) models have been used to identify several potential pathways by which aerosols could affect deep convective clouds, involving complex interactions between microphysics and dynamics. Changes in aerosol loading may impact cloud droplet number concentrations and thereby modify condensation rates, collision-coalescence, evaporation/sublimation, condensate loading, updraft glaciation, and ice-growth pathways. The net result of a CCN perturbation on convective strength, precipitation, and radiative forcing likely depends on cloud-base temperature, environmental wind shear and relative humidity, and many other factors. A perturbation in ice nucleus (IN) concentration could also potentially affect anvil size and lifetime and hence cloud radiative forcing. Several studies (e.g., Koren et al. 2005; Rosenfeld et al. 2008) have identified an aerosol "invigoration effect" whereby aerosol loading leads to a reduction of droplet collision-coalescence and reduced rainout, lofting of additional liquid water above the freezing level, and enhanced latent heating and updraft strength, especially under conditions of weak environmental wind shear. However, other studies (e.g., van den Heever and Cotton 2007; Morrison 2012) have shown reduced convective strength under other conditions. Moreover, the multiscale dynamical and thermodynamical response of the cloud system might be quite different from the response of individual deep convective clouds because of feedbacks between clouds and their environment (Grabowski 2006). The radiative impacts due to changes in anvil characteristics from aerosol loading can also be distinct from changes in convective characteristics. For example, modeling studies (Morrison and Grabowski 2011; Fan et al. 2013) have shown an increase in anvil extent and optical thickness even with a decrease in convective intensity because of a decrease in mean ice particle size with aerosol loading.

The major challenges for the CAPI Working Group within this topic are to provide observational constraints on aerosol effects on deep convective clouds using ARM observations, to understand specific causes for the large spread of model simulations of these effects, and to develop physically based parameterizations of aerosol impacts for GCMs consistent with observations and high-resolution models. ASR provides a unique opportunity to address these issues through ARM infrastructure and current ASR-funded modeling efforts. Specific issues related to the three broad objectives outlined above are described in detail below.

Objective 2.A. Building Measurement and Modeling Capacity

Key observations include quantities related to aerosol, cloud microphysics and radiative properties: CCN and IN profiles, droplet and ice crystal spectra, ice particle properties (e.g., density and fallspeed), longwave and shortwave radiative flux profiles, and integrated microphysical quantities such as LWP and ice water path (IWP). Additional observations are needed that specifically address the dynamics of deep convective systems including: three-dimensional (3D) vertical velocity fields; cold pool characteristics (measured through the depth of the cold pool); convective draft morphology, size, and number per area; profiles of latent heating and condensate loading in convective drafts; and spatial heterogeneity of surface latent and sensible heat fluxes. Satellite retrievals of cloud microphysics and morphology are also important given the multiscale aspects of the problem and the need for large data sets for statistical robustness.

The goal is to develop comprehensive data sets of aerosol, cloud, thermodynamic, and dynamic properties from these measurements and value-added products (VAPs) from recent and future field experiments focused on deep convection as well as long-term monitoring sites. Such data sets are needed to characterize relationships between aerosols, clouds, and convection observationally, and also to evaluate model simulations of deep convection for case studies as well as longer-duration simulations. To address this objective, the CAPI Working Group recommends that additional effort be made to design future experiments and perform long-term monitoring. Previous ARM field campaigns were not specifically designed to study aerosol influences on deep convection. Although long-term ARM data have been useful for studies of aerosol effects on deep convection, it is missing critical elements needed to address this science question.

In terms of cloud-resolving modeling, the magnitude and even sign of changes in updraft strength and surface precipitation with aerosol loading often vary depending on the particular model and microphysics parameterization used, even for the same initial and forcing conditions. In particular, recent studies (Lebo and Seinfeld 2011; Tao et al. 2012; Fan et al. 2013) have shown large differences in simulation of aerosol effects on deep convection using bulk versus bin microphysics parameterizations. While comparatively less studied, considerable influence of aerosols on anvil characteristics and radiative impacts with different representations of microphysics is also likely. This finding reflects the general sensitivity of CRM deep convection simulations to microphysics, with different microphysics schemes (or parameter settings within a single scheme) often producing large differences in storm structure, dynamics, precipitation, and anvil characteristics. Other studies have documented considerable sensitivity of deep convection to CRM grid spacing, even for horizontal grid spacings less than 1 kilometer. These microphysical and dynamical sensitivities have made it difficult to define “benchmark” simulations.

It is reasonable to believe that improved understanding and ability to simulate aerosol effects on deep convection will require reduced uncertainty in simulating deep convective systems more generally. This issue could be addressed by investigating the sensitivity of CRM deep convection simulations to parameterization of microphysics, model grid resolution, subgrid-scale turbulence/mixing schemes, along with other model components and detailed comparisons with observations. Continued parameterization development efforts focused on improvements in aerosol and cloud microphysics schemes and subgrid dynamics/turbulence schemes should be a key part of this work.

Owing to the spread of model simulations of deep convection and biases relative to observations documented in recent studies, a focused model-observation intercomparison may be needed to understand the specific causes of inter-model differences and model biases. A simplified modeling framework may be required to achieve this goal given the complexity of microphysics-dynamics interactions in deep convection. This effort could include testing different microphysics parameterizations in a constrained dynamical framework with a specified flow field mimicking observed convective drafts, isolating specific microphysical processes, and neglecting complications from feedbacks between the microphysics and dynamics. Once understanding is gained using a simplified framework, complexity can be added up to the point of comparing fully coupled 3D dynamical-microphysical models driven by realistic large-scale boundary conditions.

Only a few global models currently represent aerosol effects on deep convective clouds. These models can be broadly categorized into two types: 1) high-resolution models that explicitly simulate deep convection without use of a convection parameterization (e.g., Wang et al. 2011) (global CRMs and multiscale-modeling framework (MMF) with CRMs embedded in a larger-scale model); and 2) traditional scale GCMs that include aerosol effects on microphysics in deep convection parameterizations (e.g., Song and Zhang 2011). While able to "resolve" deep convective motion, global CRMs and MMF are computationally expensive and hence limited to short integrations. On the other hand, microphysics-dynamics interactions for deep convection are entirely parameterized in traditional GCMs. Nearly all traditional GCM estimates of aerosol effects on convective clouds report a negative aerosol indirect forcing. However, a conceptual model (Rosenfeld et al. 2008), cloud model simulations (Morrison and Grabowski 2011; Fan et al. 2013), and observational analysis (Li et al. 2011) suggest that increasing aerosol loading could lead to a positive top-of-atmosphere radiative forcing by decreasing ice particle size, raising cloud tops, or expanding anvil area. Reducing uncertainty in global estimates of aerosol effects using GCMs will require an improved representation of cloud-aerosol interactions in deep convection parameterizations, including improved treatments of aerosol physics (scavenging and transport) as well as cloud microphysics. For example, comparisons of simulations using bulk and bin cloud microphysics indicate biases in the cloud response to aerosol in bulk schemes that need to be addressed. Observations and CRM simulations will be critical in developing these improved parameterizations. Testing could be done for specific case studies in an SCM framework, allowing for a direct comparison with CRM simulations, as well as longer-term SCM and global simulations.

Objective 2.B. Improving Understanding of Microphysical and Dynamical Processes in Deep Convective Clouds

Understanding and quantifying interactions between microphysics and dynamics for deep convective cloud systems is especially challenging because of the myriad processes, feedbacks, and close coupling of microphysics and dynamics through buoyancy and cold pools. These effects are also strongly linked across multiple spatial and temporal scales. This work has strong links with several foci of the Cloud Life Cycle Working Group pertaining to deep convective clouds. Key specific objectives include understanding the:

- effects of ice microphysics on latent heating and buoyancy, effects of sublimation, melting, and evaporation by altering cold pool dynamics and convective downdrafts
- interactions between microphysics, turbulence, and entrainment, including entrainment of IN and CCN from the environment

- links between convective updraft characteristics, detrainment, and anvil microphysical, macrophysical, and radiative characteristics
- impacts of microphysics on upscale growth and mesoscale organization
- effects of microphysics on precipitation efficiency and partitioning of stratiform/convective precipitation
- processing of aerosols in deep convective clouds and subsequent impacts on cloud microphysics and dynamics.

Progress on these specific questions will require close coordination of observations and modeling. This work should leverage the observational data sets and modeling capacity developed as part of objective 2A.

Objective 2.C. Improving Understanding of the Sensitivity of Deep Convective Clouds to Aerosol Perturbations

Similar to other regimes, a key challenge in quantifying aerosol effects on deep convective clouds from observations is the difficulty of separating correlation from causality. Analysis of long-term ARM measurements has revealed correlations between aerosol and convective cloud properties (Li et al. 2011), but this signal is difficult to untangle from co-variability with meteorology. A key concept is to identify specific regimes where aerosols might have the greatest influence on convective clouds, which recent studies (Khain et al. 2008; Khain 2009; Fan et al. 2009) have suggested is strongly dependent upon meteorological conditions (environmental shear and relative humidity in particular). Understanding aerosol effects on convection will also require understanding the key meteorological controls on convective characteristics, which is needed to separate the influence of meteorology from aerosols. It is also important to understand the role of multiscale feedbacks between deep convective clouds and their environment, and how aerosol effects vary across spatial and temporal scales. Recent studies (Morrison and Grabowski 2011) have suggested that such feedbacks can exert a dominant control over large spatio-temporal scales for some deep convective regimes. This finding also concerns the impacts of meso- and larger-scale convective organization on aerosol effects (Lee 2012) and those of surface characteristics and surface feedbacks.

Models play a critical role in understanding and quantifying aerosol effects on deep convective clouds, which should involve an integration of studies of aerosol effects on deep convection using the high-resolution model dedicated to the SGP site, other LESs, CRMs, SCMs, global CRMs and MMF, and traditional GCMs with representation of cloud-aerosol interactions within parameterized deep convection. These modeling tools should be used to investigate aerosol effects over a range of spatial and temporal scales, from individual convective clouds to planetary-scale phenomena. Detailed testing is required to determine if models show fidelity in simulating observed case studies. These same models, however, also need to demonstrate an ability to reproduce long-term ARM observations and data sets from satellite analyses. The high-resolution simulations at SGP will be especially useful for this task. If models reproduce observed long-term correlations between aerosols and thermodynamic, cloud, and convective characteristics, they can then be used to address causation versus correlation. For such studies, it is critical to develop robust statistics from detailed, long-term data sets. This may be especially true for deep convective regimes given the relatively low level of inherent predictability at deep convective scales of motion, relative to larger scales (i.e., large data sets are needed to distinguish robust signals from noise).

3. **What processes control ice nucleation and its impact on ice-containing clouds (e.g., Arctic stratus, altostratus, cirrus, convective clouds)?**

Ice nucleation processes involving aerosols are key to the formation and microphysical and optical properties of ice and mixed-phase clouds. Ice nucleation plays a strong role in determining the ice crystal number concentration and size distribution in ice-containing clouds, the liquid/ice partitioning of mixed-phase clouds, and cloud glaciation, which can significantly impact cloud optical depth, cloud fraction, and precipitation. There are two main aerosol and cloud drop freezing pathways: homogeneous and heterogeneous ice nucleation (Pruppacher and Klett 1997). Homogeneous nucleation occurs efficiently only at temperatures below approximately -38 ºC, where hydrated aerosol or cloud droplets are sufficiently supercooled to freeze spontaneously. This process likely plays a dominant role in cirrus clouds, on a global scale, and is fairly well understood (e.g., Heymsfield and Miloshevich 1995; Koop et al. 2000). Conversely, heterogeneous ice nucleation involves a variety of poorly understood ice nucleation pathways, and much remains unknown about the concentrations and properties of ice nuclei, their dominant modes of action, and competition between them, in part owing to a lack of suitable instrumentation to provide the necessary field measurements. The importance of heterogeneous nucleation in cirrus clouds, globally and in the relatively polluted Northern Hemisphere, is still unclear. Regardless of freezing mechanism, the impact of ice nucleation on ice-containing clouds is known to be strongly modulated by ice crystal properties such as habit and fall speed that are not well constrained by field measurements.

Mineral dust has long been known to be an efficient IN, but there is still a great deal of uncertainty regarding the ice nucleation efficiency of black carbon (BC, an important anthropogenic aerosol) (Kärcher et al. 2007) and organic materials (DeMott et al. 2003; Murray et al. 2010). It is unclear how ice nucleation efficiency changes when aerosols are internally mixed (e.g., mineral dust coated by sulfate and organics). Finally, the stochastic or deterministic nature of heterogeneous nucleation processes remains a subject of debate (Vali 2008). ASR laboratory experiments, ARM field measurements, and ASR modeling are needed to develop robust physically based parameterizations of ice nucleation in terms of physical and chemical properties of the aerosol (DeMott et al. 2010), to provide constraints on ice crystal properties, and to develop validated ways to represent subgrid variability in supersaturation with respect to ice, ice nucleation, and crystal growth in conditions saturated and unsaturated with respect to liquid water.

Objective 3.A. Building Measurement and Modeling Capacity

Data from several past ARM field studies have proven useful for addressing this science question. Mixed-Phase Cloud Radiative Properties (M-PACE) collected ice nuclei concentration measurements by a continuous flow diffusion chamber (CFDC) on an aircraft flying near and within mixed-phase Arctic clouds. ISDAC also had a single-particle mass spectrometer (SPMS), as well as a CFDC sampling from an aerosol inlet outside Arctic mixed-phase clouds and from a counterflow virtual impactor (CVI) operating inside clouds. Small Particles in Cirrus (SPARTICUS) extensively used a new ice probe that is relatively free of the ice-shattering artifact that has biased in situ measurements of ice crystal number concentration in previous field studies.

The first order of importance for ice nucleation studies is to quantify uncertainties in IN measurements by instruments with different designs and with different methods (e.g., CFDC versus filter sample). Participating in the international workshops on the IN instrument intercomparison in laboratories (e.g., Aerosol Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber in Germany) and in the field would be useful to determine the limitations/biases of different IN instruments, and thus, is highly recommended. New instruments are needed to more clearly distinguish modes of ice nucleation (deposition, immersion, contact), and a CVI that distinguishes between liquid droplets and ice crystals would be invaluable in studies of mixed-phase clouds.

Understanding of ice nucleation could be tested with IN closure experiments in laboratories with controlled aerosol and in the field with ambient aerosol, using a CFDC to nucleate ice crystals at specified temperature and supersaturation, followed by a CVI to isolate and an SPMS to characterize the nuclei. IN closure studies could be tested in the field (e.g., at the Barrow site) to understand mixed-phase cloud formation. Deployment of CFDC, CVI, and SPMS instruments on the aircraft to measure aerosol and IN properties above-cloud, inside- and below-cloud would provide necessary information for understanding IN sources and sinks. The dependence of ice nucleation on updraft velocity as well as temperature and ice nuclei concentration should be explored in crystal number concentration closure experiments using in situ data from past and future field experiments. Measurements of the abundance and properties of ice nucleation particles (e.g., dust) in the free troposphere are also highly valuable.

It is highly recommended that future in situ ice cloud experiments with the new ice probe design should include aircraft CVI and SPMS to determine the composition of ice crystal residues. Remote sensing and in situ measurement of ice crystal properties (number, shape and size distribution) as well as measurements of IN, aerosols, and environmental conditions (e.g., updraft velocity and temperature) are needed to characterize relationships between aerosols, ice nucleation and cloud microphysics and dynamics.

While sampling in the environment provides data on properties and processes in the real atmosphere, controlled laboratory experiments can be used effectively to: test new instruments; explore the time-, temperature- and/or relative humidity-dependence of ice nucleation on mineral dust, BC and biological aerosols; improve understanding of the effects of soluble coatings on activation of various types of mineral dust; and clarify the efficiency of contact ice nucleation.

The CAPI Working Group strongly recommends that dedicated IN instruments (e.g., CFDC, filter) be deployed at DOE ARM ground sites to provide long-term IN measurements. Through the analysis of relationships among IN, CCN, aerosol physical and chemical properties, and environmental conditions, the seasonal effect of different types of aerosols on ice nucleation can be identified and the robust relationships among these variables established.

Given the prominent role of dust as ice nuclei, modeling studies with parcel models, CRMs and GCMs are needed to understand ice nucleation effects on cold clouds and determine the importance of speciating dust for ice nucleation. Data from ISDAC, which provide aerosol composition information, could be used for such a study. Remote sensing retrievals and in situ observations of the relationship between aerosol and cloud properties (e.g., ice crystal number, ice water content, liquid/ice partitioning in mixed-phase clouds) could be useful for validation of modeled aerosol effects on cold clouds (e.g., mixed-phase and cirrus clouds).

Modeling studies should also be used to quantify the influence of dust coatings and of BC aerosol on ice nucleation, using parameterizations developed using controlled laboratory experiments. Intercomparisons of model results from parcel models, high-resolution models and GCMs are needed to quantify the source of diversity among the models for the simulated ice nucleation and aerosol effects on cold clouds.

Objective 3.B. Improving Understanding of Ice Nucleation Mechanisms and Their Relationship to Overall Aerosol Properties and Environmental Conditions

Although only a small fraction (often less than 1%) of all aerosol particles, atmospheric ice nuclei strongly affect cloud radiative properties and precipitation formation (DeMott et al. 2010). To determine this activation fraction, a mechanistic understanding of ice nucleation on BC, biological aerosols, and dust mineralogy is needed under a variety of environmental conditions. Although laboratory experiments with controlled conditions and aerosol compositions are essential for developing empirical parameterizations and constraining nucleation theory (e.g., classical nucleation theory), field measurements are also necessary to test them in natural conditions. Because different aerosol species are often internally mixed in the atmosphere, the effect of coating of soluble materials and associated chemical reactions on the aerosol surface needs to be explored. The modes of heterogeneous ice nucleation (e.g., immersion/condensation, deposition, and contact) and their relative importance in different environmental conditions need to be identified and quantified. The stochastic or deterministic nature of heterogeneous nucleation processes and the role of time dependence needs to be understood so that theoretical formulations for describing the ice nucleation in models can be established.

For sufficiently strong updrafts, the number of ice crystals formed from homogeneous ice nucleation depends on the number concentration of hygroscopic aerosol. This dependence provides a mechanism for anthropogenic aerosol effects on homogeneous ice nucleation (Liu et al. 2009). Although the dependence is understood well, the parameterizations depend strongly on updraft velocity, which is poorly resolved by global models. Although subgrid variations in updraft velocity can be related to parameterized turbulence under some conditions, other processes that drive subgrid vertical velocity are neglected in global models and need to be understood and represented. In cirrus clouds, heterogeneous nucleation can compete with the homogeneous nucleation to form ice crystals. The relative importance of the two mechanisms depends on the updraft velocity as well as the abundance of heterogeneous IN (Liu et al. 2012).

In addition to ice nucleation, ice multiplication takes place in cold clouds to form ice crystals. Its role at temperatures above -10°C, where IN number is usually low, and how this relates to the cloud droplet size and initial ice crystals needs to be explored.

Objective 3.C. Improving Understanding of the Sensitivity of Mixed and Cirrus Clouds to Aerosol Perturbations

The Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5) included an assessment of aerosol effects on cold clouds with significant uncertainties. Some of the estimated radiative forcing due to aerosol perturbations to cold clouds can be as large as those for warm clouds. Surface lidar and satellite observation (e.g., CALIPSO) indicated significant geographical differences in the phase partitioning of liquid versus ice water in mixed-phase clouds, influenced by different aerosol types (Choi et al. 2010). Hemispheric differences in cirrus cloud properties and threshold relative humidity for cirrus formation were identified from in situ aircraft observations in two hemispheres under similar dynamic conditions (Haag et al. 2003). Long-term data are needed to establish statistics of co-variability of cold cloud properties with aerosols and environmental conditions.

For many years, homogeneous nucleation was thought to be the main mechanism for ice formation in cirrus clouds owing to the scarcity of heterogeneous IN in the upper troposphere (Heymsfield and Miloshevich 1995). However, a recent analysis of ice residues collected in convective anvil (Cziczo et al. 2013) suggests the dominant role of heterogeneous nucleation. Because homogeneous and heterogeneous nucleation are impacted by different types of aerosols, the relative importance of homogeneous versus heterogeneous nucleation in cirrus clouds needs to be clarified. Long-term observations (such as SPARTICUS) of ice crystals, ice residues, and environmental conditions, for the relationships between ice number, ice nucleating aerosols, temperature, and relative humidity can provide additional understanding.

In complicated cloud systems with multiple dynamical and microphysical processes, the cloud perturbation by aerosols through ice nucleation may be dampened or amplified by other processes. For example, increases in heterogeneous IN may reduce the overall ice number concentration in cirrus clouds when homogeneous nucleation initially dominates the ice formation due to the competition of water vapor, thus causing a negative Twomey effect (Kärcher et al. 2006). Although increases in IN can likewise increase the initial ice number, reduce ice effective size and thus initially produce a stronger shortwave cloud forcing in mixed-phase clouds, more ice crystals will lead to the faster conversion of liquid water to ice water through the Bergeron-Findeisen process. Because ice crystals are larger and sediment faster than cloud droplets, this glaciation process will enhance precipitation efficiency; together with a reduced liquid amount, glaciation will reduce cloud albedo and cloud longevity (Lohmann 2002). For clouds in the upper troposphere, the impact of ice nucleation on longwave cloud forcing typically compensates the impact on shortwave cloud forcing, so the impact on net cloud forcing is smaller than the impacts on shortwave or longwave cloud forcing. The role of ice crystal properties such as habit and fall speed as well as the influence of preexisting ice crystals on ice nucleation through water vapor competition also needs to be quantified.

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