The Ice in Clouds Experiment
- Tropical Field Campaign- (ICE-T)

-Research Plan-

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Contributing Authors:
Andy Heymsfield[1], Paul DeMott[2], Paul Field[3], Dave Rogers[1], Zhien Wang[4], Sonia Lasher-Trapp[5]

Affiliations: [1]NCAR, [2]CSU, [3]UK Met Office, [4]U. Wyoming, [5]Purdue U.

Table of Contents

1. Executive Summary

More than 50% of the earth's precipitation originates in the ice phase. Ice nucleation, therefore, is one of the most basic processes that lead to precipitation. The poorly understood processes of ice initiation and secondary ice multiplication in clouds result in large uncertainties in the ability to model precipitation production and to predict climate changes. Therefore, progress in modeling precipitation accurately requires a better understanding of ice formation processes.

Drawing upon the results of the successful Ice in Clouds-Layer (ICE-L) field campaign conducted in November-December 2007, this document plans the next phase of a study of ice processes acting in clouds. As with ICE-L, it includes field observations, laboratory experiments, and numerical modeling. Continued advances in observational tools, laboratory cloud simulation chambers, numerical models, and computer hardware are providing better capabilities to understand and model ice initiation processes. The objective of the Ice in Clouds Experiment (ICE) is to focus on the following long term scientific goal:

> To show that under given conditions, direct ice nucleation measurement(s), or other specific measurable characteristics of the aerosol, can be used to predict the number of ice particles forming by nucleation mechanisms and secondary processes in selected clouds. Improved quantitative understanding of the roles of thermodynamic pathway, location within the cloud, and temporal dependency are also sought.

This goal statement implies that ice nucleation is definable as the process responsible for at least the initial ice concentration in the selected clouds, that the specific ice nucleation path is identified, and that the parameters most important to governing the process are understood. In ICE-L, we focused on heterogeneous nucleation in clouds where secondary processes are not thought to occur. For ICE-T, we turn our attention to tropical convective clouds, where both primary and secondary ice formation processes may play significant roles.

During ICE-L, we sought to sample clouds with a strong aerosol-ice nucleation signal. Focus was on observational studies with high likelihood of showing a strong connection of aerosols to an effect on ice formation. The targets were layer clouds: lenticular wave clouds, extensive upslope stratiform mixed-phase decks. The thermodynamic and kinematic environments of lenticular wave clouds are relatively steady with lifetimes often longer than an hour, making these clouds an attractive target for study. Wave clouds provided a range of temperature, humidity, and vertical wind conditions in which first ice may form in a laboratory-like setting. ICE-L used airborne measurements of clouds, concentrating on the role of heterogeneous nucleation, along with coordinated ground-based radar measurements in mountainous locations that included the Front Range of Colorado and Wyoming. Table 1 lists the publications that have resulted from ICE-L.

Tropical maritime cumulus clouds are an important part of the global water cycle. ICE-T (tropical) will aim to understand the role of primary and secondary ice production in developing towering cumulus clouds. These clouds provide a relatively simple convective framework for studying the production of ice. During ICE-L, the influences of dust, pollution, and biomass burning aerosols on primary ice formation processes were examined. We expect ICE-T clouds to be subject to influences of clean maritime conditions and episodic mineral dust transport events, both including possible biological particle influences of oceanic or terrestrial origin, and possible influence of long range transports biomass burning particles.

In order to make progress towards the ICE scientific goal stated above ICE-T will:

1. Attempt to observe the conditions leading to glaciation of maritime cumulus with top temperatures warmer than -10°C.

2. Characterize the aerosol as CCN and IN and investigate the dependence on temperature, size and aging (special interest in dust and biological material).

3. Characterize the link between warm rain and primary and secondary ice processes as a function of time and environmental conditions. As part of this characterization, estimate the fraction of vapor flowing into cloud base (the cloud base mixing ratio) that arrives at the 0°C, -5°C and -10°C temperature levels in the form of vapor, supercooled liquid water and ice. How does dust affect these fractions? How does this depend on the cloud lifecycle?

4. Determine if primary ice nucleation can explain the onset and glaciation of maritime cumuli.

5. Determine whether secondary ice formation processes are critical to the glaciation of cumuli. If so, what concentration of primary IN are sufficient to trigger them and how does the process work?

6. Determine whether mid-level entrainment plays a role in feeding CCN and IN into maritime convective clouds.

7. Test primary and secondary ice nucleation schemes in models and evaluate them against observations.

Support for several facilities will be requested or included as a part of the basic ICE research package for ICE-T.

Components include:

- High-capacity cloud and aerosol physics aircraft, specifically the NCAR/NSF C-130. The C-130 offers a large payload capability and ten canisters for PMS-type probes. The C-130 can reach temperatures low enough to sample Towering Cu (see climatology). It can transport a multitude of instruments and investigators.
- The Wyoming Cloud Radar, looking both upwards and downwards.
- The Wyoming Cloud Lidar, looking both upwards and downwards.
- *In situ* instruments for high resolution measurement of small ice and other hydrometeors (SID-2) and for high resolution imagery of particles in the 10 to several hundred micron range (2D-S and CPI).
- A comprehensive set of *in situ* aerosol instrumentation designed to measure the chemical, physical, and cloud active properties of aerosol particles. The optimal payload includes a Continuous-Flow Diffusion chamber (CFDC) for IN measurements, a counter-flow virtual impactor (CVI) inlet, an aerosol mass spectrometer for size resolved composition measurements, electron microscope grid and filter sampling, CCN spectrometers, and aerosol size measurements that cover a

wide size range (~10 nm to >10 µm, electrical mobility and optical particle sizing instruments, CN).

2. Introduction and Scientific Overview

As ice develops in clouds, it influences all major cloud characteristics of interest: precipitation formation (Tao and Simpson 1993, Tao 2003), interactions with radiation (Ackerman et al., 1988, Ackerman, 1988, Martin et al. 2001, Liu et al. 2003, Toon et al. 1989), latent heat release and cloud dynamics (Willoughby et al. 1985, Simpson et al. 1967, Lord and Lord, 1988), chemical processes (Crutzen et al. 1999), charge separation (Sun et al. 2002, Tinsley et al. 2001, Tinsley and Heelis, 1993), water vapor content (Schiller et al. 1999, Gierens et al. 1999, Heymsfield et al. 1998), icing potential (Rasmussen, et al. 2001, Thompson, et al. 2002), particle scavenging (Heusel-Waltrop et al. 2003), precipitation efficiency and others. Many mid-latitude clouds have extensive supercooled (liquid existing at temperatures < 0 °C) regions, lending themselves to copious ice production that may eventually precipitate as frozen or liquid hydrometeors. Even deep tropical clouds, which have a strong warm rain process, produce a major fraction of their rainfall through the ice phase, as suggested by the strong correlation between rainfall rate and ice water path as retrieved from satellite-borne radiometers (e.g., Liu and Curry, 1999).

Ice formation has been known to be important since the early work of Bergeron (1935) and Findeisen (1938), yet scientific knowledge is lacking on important aspects of the problem (Cooper, 1991; Beard, 1992; Rasmussen, 1995; Khain, et al., 2000; Arakawa, 2004; Cantrell and Heymsfield, 2005). It is known that ice may nucleate as a result of drop freezing from an immersed ice nucleus, or external contact with an ice nucleus (Rasmussen et al. 1992, Stoelinga et al. 2003). Additional ice crystals can be produced through *secondary* processes like ice-splinter production during riming (Hallett and Mossop, 1974, Harris-Hobbs et al. 1987, Griggs and Choularton, 1983, Mason, 1998, Phillips et al., 2003) or fragmentation resulting from ice crystal collisions (Vardiman, 1978). There may be other significant secondary ice production mechanisms, but they have not yet been defined and characterized. Identifying where the first ice originates that can assist in determining the major nucleation process(es) has been difficult observationally, given the initially low ice crystal number concentrations, and the small sizes and nearly spherical initial shapes of ice crystals that makes them difficult to distinguish from water droplets with past instrumentation. Determining if the ice is forming by primary or secondary processes has also been a challenge in this regard. Laboratory studies, while insightful, are unable to completely simulate the composite impacts of aqueous chemistry, evaporative cooling, water vapor competition, and potential secondary ice production processes. These difficulties in field and laboratory studies have limited progress on the representation of ice in numerical models, and thus the ability to model precipitation production (Tao et al. 2003, McCumber et al. 1991) and to predict climate change (Fowler and Randall 2002, Zurovac-Jevtic and Zhang, 2003).

A particularly difficult, long-standing problem has been to explain observations in extra-tropical and tropical maritime clouds, acquired as early as the mid-1960’s (Fig. 2.1) but also in later field studies (Fig. 2.2), that exhibit far greater ice concentrations than expected, especially when cloud top temperatures were warmer than -10 °C (Mossop 1968; Mossop et al, 1970; Sax et al., 1979, Hobbs and Rangno 1985). Such observations appear to contradict even the most recent measurements of ice nuclei concentrations (Fig. 2.3). Intrusions of mineral dust, a well known terrestrial source of ice nuclei, into the clouds of such regions may partially explain these observations, but laboratory studies indicate that the dust only becomes active at temperatures less than -15 °C (see later discussion), a fact also supported in some lidar measurement studies of stratiform clouds (Ansmann et al. 2008). Biological material introduced into the clouds of these regions may be a potential candidate for ice nuclei active at -10°C, but the numbers of such particles in air is not well known, and may be less than 0.1 per liter (Bowers et al. 2009), significantly below the hundreds per liter of ice crystals sometimes found in the maritime clouds. If primary ice nucleation cannot explain the observed high ice concentrations in these clouds, then secondary ice production mechanisms must be considered, such as rime splintering (Mossop, 1985). An alternative mechanism to explain the observations of high ice concentrations is that the research aircraft making the in-situ measurements were responsible for the generation of large concentrations of ice (Rangno and Hobbs 1984).

This document describes fundamental observational, laboratory, and modeling studies that have addressed how ice may form in clouds, and it proposes a research plan to obtain observations that will fill gaps in current knowledge of ice nucleation. Such knowledge can be used to improve the current representations of ice formation in numerical models, yielding better estimates of precipitation. Heterogeneous ice nucleation initiates the ice phase in most clouds at temperatures from near 0°C to as low as -35°C, but the conditions under which ice particles first form, and upon which aerosol heterogeneous ice nucleation is favored, are not well known. Past measurements in clouds have lacked the temporal resolution and instrumentation to sample early ice in clouds adequately, or the nuclei that influence its formation. Entrainment of ice nuclei and ice crystals into cloud updrafts further complicates attempts to identify the ice initiation processes and their evolution, as does the possibility that observations may be affected by the sampling of the clouds by an aircraft itself. Fortunately, new observational tools and simulation techniques are now available to study ice formation mechanisms. After reviewing the major issues related to ice formation in clouds, new opportunities for addressing these difficulties are presented.

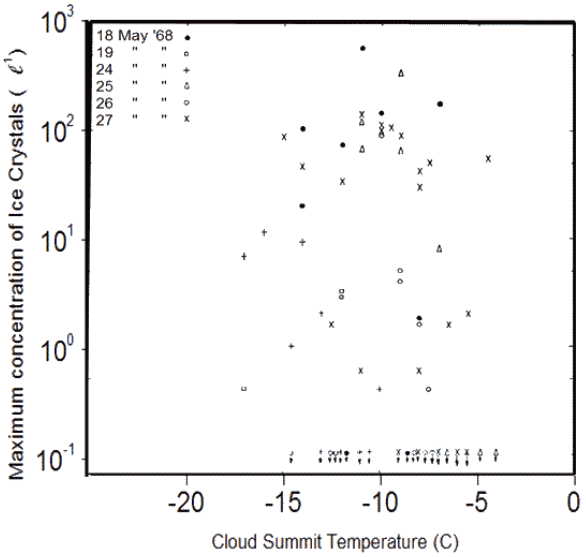


Figure 2.1 Cloud width plotted against cloud summit temperature. Clouds consisting entirely of water drops are indicated by solid circles, those containing ice particles by crosses. From Mossop et al., 1970.

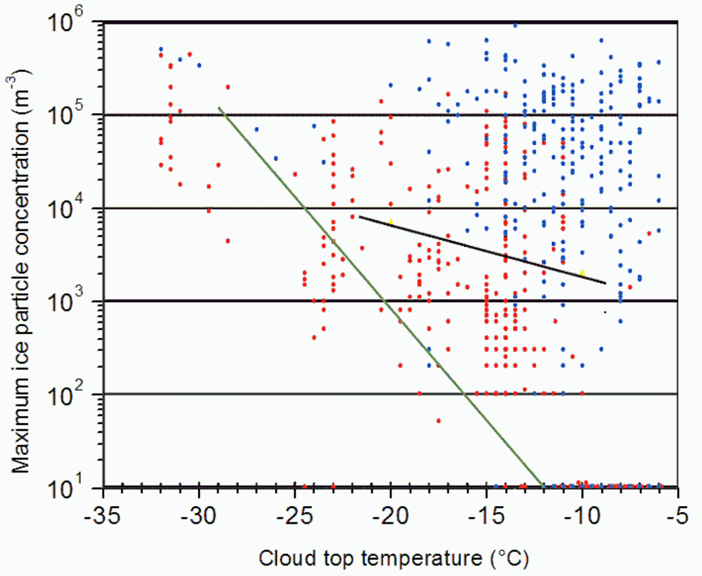


Figure 2.2 Ice particle number concentration versus cloud top temperature, compiled from data from several field programs by Wallace and Hobbs (2006). Ice crystal number concentrations measured in maritime clouds in blue, and in continental clouds in red. Expectations of ice crystal number concentrations from Fletcher curve (1962) and Meyers et al. (1992) parameterization (black) greatly underestimate the ice crystal number concentration in most of the clouds, but especially at higher temperatures and in the maritime clouds.

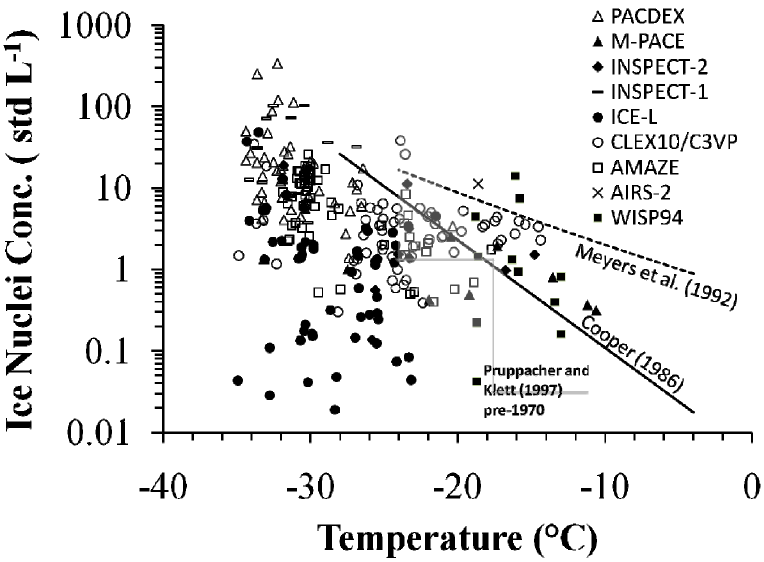


Figure 2.3 Summary of ice nuclei number concentration data measured by the continuous flow diffusion chamber method, adapted from DeMott et al. (2009) and Möhler et al. (2007). Data points represent 5 to 30 minute average samples at near constant temperature in the RH regime from 100 to 104% from several aircraft and ground-based studies (acronyms listed and referenced in DeMott et al.). Points falling below a level of 0.3 per liter (limit of quantification from aircraft ambient inlet samples) are from reprocessing cloud particle residuals from the CVI inlet during ICE-L and large volume bag samples collected during the WISP94 project. The shaded region represents the range of average ice nuclei concentrations measured by various methods from around the world before 1970. The solid curve represents an ice nucleus temperature spectrum based on Cooper's measurements of ice crystal concentration in clouds absent secondary ice formation processes. The dashed line follows the parameterization of Meyers et al. (1992) based on the first ever ground-based CFDC data and calculated at water saturation.

2.1 Primary (Heterogeneous) Ice Formation

The recent Ice in Clouds Experiment- Layer (ICE-L) field study was based upon the premise that advancing the understanding and predictability of ice crystal concentrations formed from primary nucleation processes required an improved assessment of ice nucleation dependence on temperature and supersaturation alone (e.g., Fletcher, 1962; Meyers et al. 1992) and additional linkage to aerosol properties and ice formation mechanisms. ICE-L provided significant steps in this direction.

In the right situation, clouds provide the ultimate measure of ice-forming activity. Cooper (1986) summarized aircraft observations of ice crystal concentrations in clouds where ice formation was attributed to primary nucleation. Although concentrations varied by up to a factor 10 at the same temperature, a clear trend of increasing concentration with decreasing temperature was found, and the results showed remarkable consistency from locale to locale. It seems likely that this variability could reflect the minimum or typical spatial and/or temporal variability of ice nuclei. Cooper noted that typical pre-1986 ice nuclei (IN) temperature spectra also showed a variability of about a factor 10, but IN concentrations were ~10 times less than ice crystals, even in cases where ground-based IN measurements were available at the same location. He also noted that most of the earlier IN measurements neither allowed separation of different nucleation modes nor reproduced through modeling any realistic cloud parcel conditions other than temperature. This situation seems partly ameliorated in comparing Cooper's relation to more recent ice nuclei data in Fig. 2.3, but there is a clear need for modern IN measurement methods to validate IN concentrations and their relation to ice in clouds at modest supercoolings warmer than -15°C. Furthermore, DeMott et al. (2009) identify variability of larger than one order in ice nuclei number concentrations at similar temperatures, attributing these variations to specific aerosol concentration differences in time and space.

Cloud observations confirm the role of heterogeneous ice nucleation in ice phase initiation. Eidhammer et al. (2009) found for one ICE-L case investigated, that IN number concentrations measured for air entering the clouds and from the residuals of cloud particles both compared well with measured ice crystal concentrations, evidence that the ice was formed from primary heterogeneous nucleation (most likely condensation or immersion freezing). They also showed that ice crystal concentrations were predicted well by two new ice nucleation parameterizations based upon measured aerosol properties (size distribution and chemical properties) as input parameters.

Ice concentrations in orographic wave clouds at temperatures of -24°C>T>-29°C were shown to be related to aerosol characteristics in nearby clear air during five research flights over the Rocky Mountains (Twohy et al., 2009). When clouds with influence from colder temperatures were excluded from the data set, mean ice nuclei and cloud ice number concentrations were very low, on order of 1 to 5 L^{-1}. In this environment, ice number concentrations were found to be significantly correlated with the number concentration of larger particles, those larger than both 0.1 μm and 0.5 μm diameter. A variety of complementary techniques were used to measure aerosol size distributions and chemical composition. For these five flights, correlations were observed between ice concentrations and the number concentrations of black carbon and biomass burning aerosols. Ice nuclei concentrations directly measured in biomass burning plumes were the highest detected during the project, indicating a potential role for biomass burning aerosol particles in ice formation.

Pratt et al (2009a) found that mineral dust and biological particles comprised the majority of cloud ice crystal residues within a cold wave cloud influenced by long-range transport of desert dust during ICE-L. Concurrent elevated ice nuclei concentrations suggested that biological and dust particles initiated ice formation in the sampled clouds, suggesting that biological particles can enhance the impact of desert dust storms on the formation of cloud ice. Pratt et al (2009b) identified aged biomass burning particles within two orographic wave cloud regions sampled over Wyoming during ICE-L. Enrichment of biomass burning particles internally mixed with oxalic acid in both homogeneously-frozen ice and cloud droplets suggests either preferential activation as CCN or aqueous phase cloud processing.

Field et al. (manuscript in preparation) used a 1-D kinematic framework to infer ice nuclei concentrations from the ICE-L wave cloud observations. Two isolated wave clouds were sampled at ~-30°C, one producing ice concentrations for particles larger than 100μm of up to 50 L^{-1} the other, less than 1 L^{-1}. The wave cloud producing more ice appears to extend to cold enough temperatures for homogeneous freezing to affect the observations in the downwind ice tail of the cloud, but heterogeneous ice nuclei concentrations of ~6 L^{-1} (ranging from 2 to

18 L^{-1}) are inferred from the modeling to match observations of ice crystal concentration within the clouds. The wave cloud with less ice is not affected by homogeneous freezing and ice nuclei concentrations of ~0.5 L^{-1} (0.2 to 1.5 L^{-1}) are inferred from the model. These concentrations are broadly in agreement with those obtained from an airborne ice nuclei counter, but lower than those predicted by commonly used primary heterogeneous nucleation parameterizations.

Despite the successes of the ICE-L field campaign other questions regarding primary ice nucleation processes in clouds remain. For glaciated maritime, continental and Arctic Canadian clouds, Gultepe et al. (2001) showed typical concentrations of ice particles were near 1-10 L^{-1} (independent of temperature), for particles greater than 125 μm, as measured with a PMS 2D-C, and 1-10 cm^{-3} as measured with a PMS FSSP. The FSSP measurements clearly need to be revisited because this probe was not designed to count small ice particles. Although there are many uncertainties, the above measurements suggest that "average" ice particle concentrations in stratiform clouds are independent of temperature and geographic location. However, it is difficult to explain such observations using the currently accepted physical processes of primary and secondary nucleation. Instrumentation issues may be a major part of these puzzling observations. For instance, recently Korolev (pers. Comm., 2009) has shown that many small ice particles as observed with PMS 2D-C probes have shattered ice particles, thus likely leading to too high concentration measurements of smaller ice particles in many prior studies. It will be critical for ICE-T to deploy 2D-probes with new probe tips designed to minimize such ice particle shattering; the changes are relatively easy to accomplish but have not yet been implemented.

It is unlikely that all primary and secondary ice forming processes have been identified. The source of very high ice concentrations in some small precipitating cumulus clouds remains a mystery (Hobbs and Rangno, 1985; Rangno and Hobbs, 1994). Hobbs and Rangno (1990) summarized 10 years of field observations that show rapid formation of high ice concentrations in slightly supercooled cumulus clouds (~ -10°C). They hypothesized that a succession of processes, including coalescence growth of droplets, contact-freezing nucleation of drizzle drops, and in small regions, the activation of large numbers of IN at high water supersaturations (~15%) was responsible for the such observations of high ice concentrations. The latter two hypotheses processes were found quantitatively inadequate on subsequent analysis (Baker 1991a; Baker 1991b). Rogers et al. (1994) explored this idea on the basis of extrapolating IN measurements, but could not explain ice crystal concentrations exceeding a few tens per liter. Nevertheless, Hobbs and Rangno (1990) suggested that "*more information is needed on the supersaturation dependence of atmospheric ice nuclei (extending up to water supersaturation on the order of 10%).*", and such data would assist in understanding primary nucleation processes in cumuliform clouds, as well as other cloud types.

In more recent airborne studies of Arctic clouds, Lawson et al. (2001) and Rangno and Hobbs (2001) concluded that ice concentrations were generally much higher in Arctic stratus clouds than predictions of simple equations based on temperature (Fletcher 1962; Meyers et al., 1992). Lawson et al. (2001) examined two cloud regions with high ice concentrations, one that met the Hallett-Mossop secondary ice production criteria, and another at –12°C that could not be explained by the Hallett-Mossop process. Rangno and Hobbs (2001) found that higher concentrations of ice crystals were associated with conditions when cloud droplets

Table 1

ICE-L Investigations

Baker, B., 2010a: Comparisons between ICE-L wave cloud data and earlier wave cloud data sets. Part 1: Kelvin-Helmholtz waves within gravity waves. Resubmission planned*, J. Atmos. Sci.*

Baker, B., 2010b: Comparisons between ICE-L wave cloud data and earlier wave cloud datasets. Part 2: Sudden high ice concentrations with evaporation Resubmission planned*, J. Atmos. Sci.*

DeMott, P.J., A. J. Prenni, X. Liu, M. D. Petters, C H. Twohy, M. S. Richardson, T. Eidhammer, S. M. Kreidenweis, and D. C. Rogers, 2009: Predicting global atmospheric ice nuclei distributions and their impacts on climate, Submitted to *Proc. Natnl. Acad. Sci*.

Eidhammer et al. 2009: Ice initiation by aerosol particles: Measured and predicted ice nuclei concentrations versus measured ice crystal concentrations in an orographic wave cloud. *J. Atmos. Sci., ICE*-L Special Issue, in press.

Field et al. 2010: Contrasting the ice nucleation in two lee wave clouds observed during the ICE-L campaign. Article in preparation.

Heymsfield, A. J., P. C. Kennedy, S. Massie, C. Schmitt, Z. Wang, S. Haimov and A. Rangno, 2010: Aircraft-Induced Hole Punch and Canal Clouds: Inadvertent Cloud Seeding. *Bull. Amer. Meteor. Soc.*, in press.

Hudson, J., 2009: Comparisons of CCN with Supercooled Clouds. Resubmitted, *J. Atmos. Sci.*, ICE-L Special Issue.

Pratt, K.A., P.J. DeMott, J.R. French, Z. Wang, D.L. Westphal, A.J. Heymsfield, C.H. Twohy, A.J. Prenni and K.A. Prather. 2009. In-situ detection of biological particles in cloud ice crystals. *Nature Geoscience*. **2**: 398-401.

Pratt, K.A. and K.A. Prather, 2010. Aircraft measurements of vertical profiles of aerosol mixing states. *J. Geophys. Res.,* in press.

Pratt, K.A., A.J. Heymsfield, C.H. Twohy, S.M. Murphy, P.J. DeMott, J.G. Hudson, R. Subramanian, Z. Wang, J.H. Seinfeld and K.A. Prather. In-situ chemical characterization of aged biomass burning aerosols impacting cold wave clouds. .*J. Atmos. Sci.* special issue*,* in press.

Pratt, K.A., C.H. Twohy, S.M. Murphy, R.C. Moffet, A.J. Heymsfield, C.J. Gaston, P.J. DeMott, P.R. Field, D.C. Rogers, T.R. Henn, M.K. Gilles, J.H. Seinfeld and K.A. Prather, 2010. Observation of playa salts as nuclei of orographic wave clouds. *J. Geophys. Res.,*in press.

Pratt, K.A., S.M. Murphy, R. Subramanian, P.J. DeMott, G.L. Kok, D.C. Rogers, A.J. Heymsfield, J.H. Seinfeld and K.A. Prather. Flight-based chemical characterization of biomass burning aerosols within two prescribed burn smoke plumes. Atmos. Chem. & Phys.. In preparation.

Twohy, C. H., P. J. DeMott, K. A. Pratt, R. Subramanian, G. L. Kok, S. M. Murphy, T. Lersch, K. A. Prather, J. H. Seinfeld, A. J. Heymsfield and Z. Wang, Relationships of biomass burning aerosols to ice in orographic wave clouds. *J. Atmos. Sci.* special issue, in press.

exceeded a threshold size and number concentration. They hypothesized several ice multiplication mechanisms, corresponding to different temperature regimes. These mechanisms involved rime splintering and shattering when large drops froze. Fridlind et al. (2007) and similar recent modeling and remote sensing studies (e.g., Fan et al. 2009; van Diedenhoven et al. 2009) have supported that hypothesized enhanced droplet freezing or ice nuclei formation during evaporation could explain sustained ice concentrations exceeding 1 per liter at -10 to -12°C in an Arctic stratus case, but enhance ice formation in evaporation regions in ICE-L clouds were only found associated with homogeneous freezing processes (Pratt et al. 2009b). Thus, this issue remains enigmatic. These observations need to be revisited with recently developed particle probes that are less susceptible to measurement errors.

2.2 Secondary Ice Formation Processes

After primary ice has formed in a cloud, the concentration of ice crystals can be increased through secondary production mechanisms. Such mechanisms include rime-splintering (Hallett and Mossop, 1974), fragmentation during crystal-crystal collisions (e.g., Vardiman, 1978), and fragmentation during evaporation (Oraltay and Hallett, 1989). Field observations within cumulus clouds have shown consistency with laboratory measurements of the Hallett-Mossop mechanism (e.g. Harris-Hobbs and Cooper, 1987). With optimal conditions, the rime-splintering process can rapidly generate high concentrations of ice crystals in supercooled water clouds, but the onset depends on a number of specific pre-existing conditions, including graupel in the presence of supercooled droplets with certain sizes and concentrations, and within a narrow temperature range (-3 to -8 °C). Secondary processes have been found to be quite important in some types of cumulus clouds (e.g., Blyth and Latham, 1993; Rangno and Hobbs, 1991) and in winter California orographic storms (Gordon and Marwitz, 1986, Marwitz, 1987), but less important in orographic storms with colder cloud base temperatures (e.g., Cooper and Saunders 1980; Rauber and Grant, 1987). Secondary processes are thought to be especially important in tropical clouds (Hallett et al., 1978), although the effects are modulated by the strength of the updraft (e.g. Lopez et al., 1985).

The enhancement of ice concentrations through secondary processes can promote the ice production process. Secondary processes have been identified only for narrow temperature regimes. Evidence exists, however, that other secondary mechanisms operate outside of these limits. For example, it is known that ice multiplication can be very important for generating ice in maritime cumulus in certain restricted temperature ranges when large ice particles are present, but it is not known how significant the process or other processes are in other temperature ranges. Likewise, the observed formation of ice crystal concentrations exceeding 100 L^{-1} in cumulus clouds by Hobbs and Rangno (1990) and Rangno and Hobbs (2001) suggests that the rime-splinter process is not fast enough to account for these observations. Also, it is not known how important ice multiplication is in continental cumulus. Over the decades of measurements of ice particle size distributions that span a wide range of cloud temperatures, a consistent result shows a preponderance of small ice crystals relative to large ones. This observation often occurs under conditions in strong sublimation zones or where the small ice particles should grow rapidly to larger sizes. This could be a major ice multiplication process that has not been explained and could be operative under far-reaching conditions, or it could be an instrumentation artifact due to crystal breakup on the probe inlets (Field et al. 2003, 2006; Korolev and Isaac 2005, Heymsfield et al. 2008). This question can be addressed by using newer probes that generate fewer artifacts, or older probes fitted with new inlets.

Secondary processes have been found to be quite important in some types of cumulus clouds (e.g., Blyth and Latham, 1993; Rangno and Hobbs, 1991; Huang et al. 2008) and in winter California orographic storms (Gordon and Marwitz, 1986, Marwitz, 1987), but less important in orographic storms with colder cloud base temperatures (e.g., Cooper and Saunders 1980; Rauber and Grant, 1987). Secondary processes are thought to be especially important in tropical maritime clouds (Hallett et al., 1978), although the effects may be modulated by the strength of the updraft (e.g. Lopez et al., 1985).

2.3 Dependence on Droplet Spectrum

The formation of ice by both primary and secondary nucleation is likely to be strongly influenced by the size spectrum of cloud droplets, which is determined by CCN spectra, updraft velocity and entrainment. Similar to findings in warm clouds (Hudson et al. 2009a; Hudson and Noble 2009), Hudson et al. (2009b) found similar correlations between CCN and cloud droplet concentrations for various liquid water content thresholds for most of the supercooled clouds of ICE-L. However, these correlations were disrupted in some mixed phase wave clouds where the ice seemed to reduce droplet concentrations relative to CCN concentrations.

For typical ice nucleus concentrations in warm based cumuli, initial ice concentrations may be produced by the impaction of giant (> 1 μm diameter) ice nuclei with drizzle or raindrops larger than about 200 μm (Beard, 1992) or by immersion freezing of these same drops. Once frozen, these drops can grow by accretion rapidly to produce graupel and initiate secondary ice formation through the Hallett-Mossop process (e.g. Hallett et al., 1978). This process may explain why tropical clouds are observed to glaciate more rapidly than mid-latitude continental clouds; in tropical clouds, drizzle drops are found at warmer sub-freezing temperatures than in mid-latitude clouds.

The speed of glaciation of a cloud is also highly dependent upon the prior history of warm-rain processes. Several modeling studies (Cotton, 1972a,b; Koenig and Murray, 1976; Scott and Hobbs, 1977) have shown that the coexistence of large, supercooled raindrops and small ice crystals nucleated by deposition, sorption, or Brownian contact nucleation favors the rapid conversion of a cloud from the liquid phase to the ice phase. In the absence of supercooled raindrops, small ice crystals first grow by vapor deposition until they become large enough to commence riming or accreting small cloud droplets. The riming process proceeds relatively slowly until the ice has grown to millimeter-sized graupel particles, after which time the conversion of cloud water to ice can proceed relatively quickly. However, if supercooled raindrops are present, the slow-growth period (depositional growth of the ice to sizes large enough to rime efficiently) can be circumvented. The large raindrops then quickly collide with small ice crystals, they immediately freeze, and are large enough to rapidly collect small supercooled cloud droplets, enhancing the rate of conversion of a cloud to the ice phase. Secondary ice-crystal production by the rime-splinter mechanism, if active, further accelerates the glaciation rate of the cloud. Several modeling studies (Chisnell and Latham, 1976; Koenig, 1977; Lamb et al., 1981; Huang et al. 2008) have shown that the presence of supercooled raindrops accelerates the cloud into a mature riming stage wherein large quantities of secondary ice crystals can be produced in the temperature range -3°C to -8°C. The small secondary ice crystals collide with any remaining supercooled raindrops, causing them to freeze and further accelerate the glaciation process.

However, as noted by Sax and Keller (1980), in broad, sustained rapid-updraft regions, even when the criteria for rime-splinter secondary production are met, the secondary crystals and graupel can be swept upward and removed from the generation zone. Until the updraft weakens and graupel particles settle back into the generation zone, the positive-feedback aspect of the multiplication mechanism is broken. Therefore, the opportunities are greatest for rapid and complete glaciation of a single steady updraft, if the updraft velocity is relatively weak. In contrast, Sax and Keller (1980) observed high concentrations of ice particles in the active updraft portion of a pulsating convective tower. They postulated that the graupel particles swept aloft in the first bubble of a pulsating convective tower settled downward into the secondary ice-particle production zone (-3 to -8°C), wherein they became incorporated into a new convective bubble and contributed to a prolific production of secondary ice crystals by the rime-splinter mechanism. This demonstrates that there exists a very intimate, nonlinear interaction among buoyancy production by glaciation of a cloud, the evolution of the microstructure of the cloud, and the evolving cloud motions.

2.4 Evaporation Effects on Ice Formation

Several studies suggest that evaporating cloud droplets may be highly effective ice nuclei (Kassender, et al. 1957; Rosinski, 1995). Beard (1992) postulated that during evaporation, an organic shell forms and promotes hydrogen bonding and sulfate absorption sites that lead to freezing. He also suggested that residues of evaporated cloud droplets carrying high electric charges might act as ice nuclei through "electro-freezing." Cooper (1995) speculated that changes in mass and thermal accommodation coefficients during evaporation can lead to stronger cooling than would be predicted for a simple wet-bulb process, causing the activation of freezing nuclei.

In addition, laboratory studies by Oraltay and Hallett (1989) suggest that evaporating graupel particles produce copious numbers of "ice bits", which if entrained into an ice-supersaturated region of a cloud, could contribute to enhanced ice particle concentrations associated with an evaporated region of the cloud. This process, however, is a secondary production mechanism, not primary.

Recent laboratory observations of surface-enhanced ice nucleation (Shaw et al. 2005) have led to a new hypothesis for evaporation freezing. As a droplet containing an insoluble particle evaporates, eventually the surface of the droplet will come into contact with the particle. The feasibility of evaporation freezing been tested in the laboratory (Durant and Shaw 2005), and the data suggest that collision with the inner drop surface increases the temperature at which the particle initiates droplet freezing by several °C, in accord with the similar impact of classic contact freezing nucleation with the outer drop surface. Therefore, it is plausible that the number density of active ice nuclei will increase in a region of evaporating cloud, relative to a region of non-evaporating cloud.

Evidence from field studies that show evaporation enhances IN activity is mostly indirect or inferential. This evidence, at the moment, is perhaps more intriguing than it is compelling. Some field studies have related unusually high ice nuclei numbers or unusual increases in ice crystal numbers to circumstances in which clouds were evaporating. Langer et al. (1979) found IN enhanced in thunderstorm outflow regions compared to surrounding regions of the atmosphere. Some of the observations of ice crystal number enhancement versus expected IN number in the comprehensive cloud studies of Hobbs and Rangno (1985; 1990) and Rangno and Hobbs (1994) were also observed to originate in close proximity to regions of cloud evaporation. Nevertheless, these authors focused attention on the relation between cloud droplet diameter and high ice crystal concentration. Stith et al. (1994) followed the development of ice in a cumulus turret near its top at -18°C. During the updraft stages, low ice concentrations were observed in the turret (similar to what would be expected from primary ice nucleation), but during the downdraft stages (where evaporation was prevalent), the ice concentrations increased by an order of magnitude. This observation cannot be explained by rime splintering.

Observations in some orographic wave clouds prior to ICE-L provided a strong argument for some type of ice nucleation process associated with droplet evaporation. Cooper (1995) observed the onset of up to hundred-fold increase in ice crystal concentrations in the evaporation region of orographic wave clouds. The largest ice enhancements in the Cooper study were observed in clouds with temperatures approaching the onset temperature for homogeneous freezing. Smaller enhancements were found in warmer clouds and no enhancements were found at temperatures greater than about -20°C. Neither Cooper (1995) nor Rogers and DeMott (2002) found ice crystal concentrations to progressively increase in wave cloud trains, as might be expected if ice nucleating particles were being created by cloud cycling. Further evidence of the possible role of evaporation nucleation has been presented by Field et al. (2001), Cotton and Field (2002) and Baker and Lawson (2005). Field et al. (2001) and Baker and Lawson (2005) show observational evidence from repeated wave cloud penetrations suggesting that high concentrations (several per cm^{-3}) of ice had to form close to where the supercooled liquid evaporated. The concentration of ice produced in the evaporation regions is typically much greater than that produced initially, near the leading edge of the wave cloud. Enhanced ice formation in evaporation regions in ICE-L clouds were only found associated with ice generated via homogeneous ice nucleation that fell into these layers from above (Field et al., manuscript in preparation).

2.5 Pre-Existing Ice

Pre-existing ice, resulting from remnants of decayed convection, fallout from higher anvil or cirrus clouds or incomplete sublimation of ice crystals, may effectively seed clouds. If these particles are small or present in low concentrations, they may not be visible. During ICE-L, lidar and radar observations from the C130 aircraft showed snow from Elk Mountain that lofted into the leading (upwind) edge of a wave cloud under study (Figs. 2.4, 2.5). Lidar and radar detected ice at the leading edge of the wave cloud. For trajectories higher up that did not entrain these ice particles, radar did not detect ice until ice generated in the cloud grew to a radar-detectable size. The pre-existing ice was not visually observed upwind of the cloud.

Because shallow maritime convective clouds are typically short-lived, they can potentially provide a source of ambient ice. Such pre-existing ices may effectively seed future

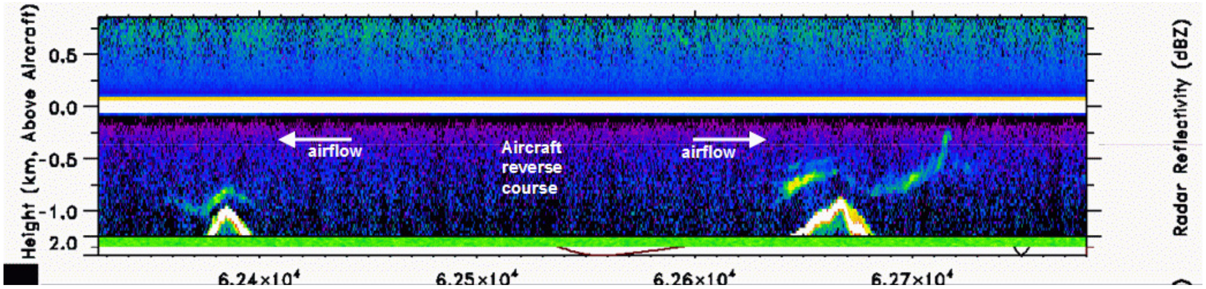


Figure 2.4 Wyoming Cloud Radar Data (ordinate; height above and below aircraft level; abscissa: time) obtained from a pass from (left to right) upwind of the cloud under investigation (RF11, ICE-L), a turn, then a return pass (beginning at about 6.255x104 sec) from upwind to downwind. Elk Mountain and a cloud overhead are indicated in this plot. Plot courtesy of Sam Haimov

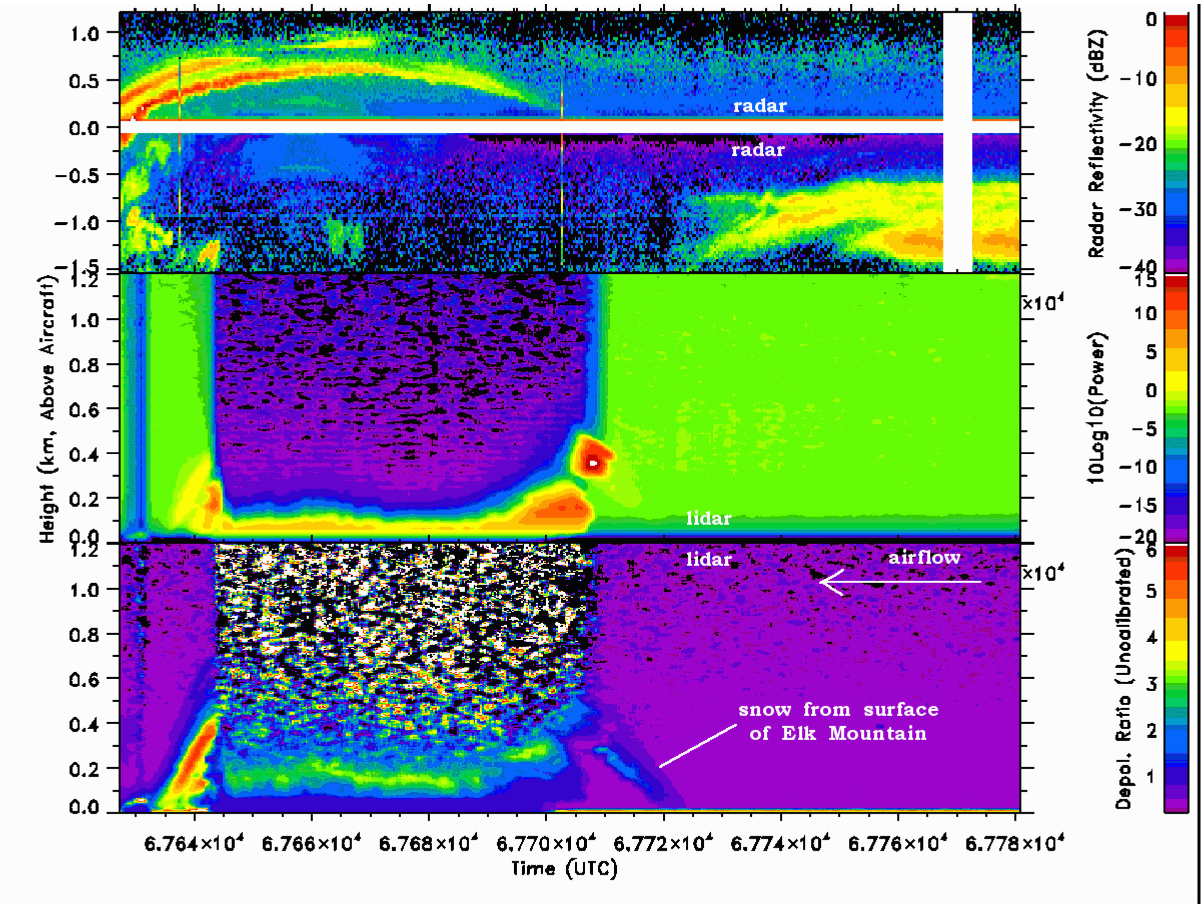


Figure 2.5 Cloud sampled during RF11, with penetration from downwind to upwind. The wind blows from right to left in the figure. Top panel: upward viewing Wyoming Cloud Radar Observations. Center panel: Relative lidar backscattered power. Bottom panel: Lidar depolarization ratio. Ice feeding into the cloud layer is seen to extend from 6.770x104 to 6.772x104 sec and from the aircraft level to about 0.4 km. Plot is courtesy of Zhien Wang.

developing cumulus clouds at relatively warm (-10 °C) temperatures, as well as promote secondary ice production. The question is whether there are ice nuclei active in concentrations on the order of 1 L^{-1} (a number which must be confirmed through modeling studies) at -10 °C and above, or whether the ambient ice concentrations originate from processes occurring at colder temperatures. In order to effectively model these systems we need to understand the origin of these particles must be determined.

2.6 Air Parcel History and Cloud Dynamics

The microphysical evolution of the cloud must be considered within the overall dynamical evolution. The thermodynamic history of the air reaching a given temperature and the amount and location of entrainment of dry air are likely to be major factors in determining the formation of ice, and depend intimately on the dynamics of the cloud and its overall lifetime. For example, the height (temperature) of the cloud base plays a major role, together with the stability of the air (i.e., buoyancy and updraft velocity), in determining the age of the cloudy air arriving at a given temperature level. Thus air parcels rising in clouds with low, warm, bases have longer residence times and are more likely to produce precipitation by warm-rain processes before the air in the updraft reaches the freezing level. The rate of glaciation and secondary ice production as just described are likely to be similar to what is observed in tropical clouds, given similar aerosol profiles and stability. Most continental locations (such as the US Midwest) have higher cloud bases and a tendency towards cold (ice) precipitation formation. The concentrations of CCN and giant CCN may play a major role, given their effect on droplet size distributions.

The contribution of ice nuclei from the cloud base relative to those entrained into the cloud is not known, but could have substantial implications for the initiation and subsequent spread of cloud ice. The strength and continuity of the updrafts clearly influence ice entrainment and recirculation. An active warm rain process would remove potential ice nuclei.

Thus knowledge of the thermodynamic history of the air moving through the cloud, and the timescales associated with the residence of the air within different thermodynamic and microphysical regimes, are likely quite important in determining ice nucleation by primary or secondary processes. This time history of air parcel characteristics is difficult to quantify with observations alone, but progress can be made on these issues by collecting data to constrain Lagrangian microphysical calculations conducted within detailed 3D numerical models of the clouds. In such a manner, ice formation and its transport within the cloud can be considered, and even used to aid in the interpretation of the in-situ observations.

2.7 Dust as Ice Nuclei

"Dust" aerosol consists of mineral particles of surface crustal origin, composed primarily of silicon, aluminum, potassium and calcium. Large scale events of dust production have been detected from satellites and have been tracked across large distances. Studies of dust deposition indicate potential distribution on global scales (Husar et al., 2001; Prospero, 1996; 1999). Large scale sources include African Sahara and eastern Asia (Huebert et al., 2003). Dust is ubiquitous at most times, locations, and altitudes, at least in the Northern Hemisphere. A preponderance of evidence of a relationship between dust and ice nuclei on a global scale comes from several different types of studies, some of which involve direct examinations of IN particles, and others that are based on inferential evidence.

Some inferential evidence of a connection between dust and ice nuclei comes from analysis of weather modification studies in Israel (Gabriel and Rosenfeld 1990). The Israeli analyses suggested that cloud seeding increased precipitation on days with low natural IN concentration, but decreased precipitation when IN concentration was high. Ice nucleating aerosols were measured with membrane filters. Higher natural IN concentrations were associated with days having greater amounts of desert dust, as determined by meteorological trajectories, rain water chemistry and total suspended particulate analyses. An earlier study by Gagin (1965) reported that desert dust, especially *loess*, produces large quantities of ice nuclei.

Early direct evidence for mineral dust association with ice nuclei came from identifying the apparent nucleating particles in fresh snowfall. Kumai (1951) and Isono et al. (1959) made Formvar casts of precipitating snow crystals. They found particles at the crystal centers and analyzed them using ion microprobe and electron microscope (EM) techniques. The chemical composition indicated up to 85% of particles were clay materials, including illite, kaolinite, halloysite and other minerals, as well as particles containing sodium chloride. While this evidence is suggestive that the central particles were the ice nucleating particles, the approach has inherent uncertainties: (1) although the location at the crystal centers suggests the particles were the nuclei, they may have been collected by processes other than nucleation scavenging; (2) when several particles are near the center, there is an inherent bias towards the larger ones (Mossop 1963); (3) the analysis identifies elements, weighted by mass fraction – the nucleating structure may be a minor component; (4) structure and chemical bonding are not characterized; and (5) nucleation occurs on the surface at a particular site, the properties of which are not characterized. Nevertheless, some of these same limitations apply to most methods that identify ice nuclei by the major composition of particles associated with ice formation.

Similar studies to capture ice particles from clouds aloft and determine the compositions of their residual nuclei have used counter-flow virtual impactor (CVI) inlets. Heintzenberg et al. (1996) evaporated cirrus crystals collected with a CVI and impacted the residual particles onto transmission EM grids for single particle analysis. The dominant particles were identified as minerals, containing silicon and iron. Likewise, Targino et al. (2005) describe results from an airborne study of wave clouds in which CVI ice particle residues showed a high occurrence of elements associated with mineral dust. These studies may have contained some influence of scavenging on ice crystal residual properties. These possible limitations motivated Cziczo et al. (2004) to sample only the smallest ice crystals (< 25 μm) from in-situ and anvil cirrus using a CVI and to measure their compositions in real-time with a single particle laser ablation mass spectrometer (PALMS). These measurements occurred over Florida and surrounding oceanic regions during the NASA CRYSTAL-FACE project (July 2002). Appreciable quantities of African dust are transported over large areas of the Atlantic Ocean to the Caribbean region during much of the year, peaking from June through August (Prospero and Lamb, 2003). Mineral dust particles larger than about 0.2 μm were one of the major residual particle classes identified in cirrus by Cziczo et al. (2004) during the project as a whole and predominated during the presence of lower altitude Saharan dust layers. Twohy and Poellot (2005) reached a similar conclusion using EM analyses regarding the predominant composition of residues of CVI-collected anvil cirrus crystals found at temperatures warmer than -36°C during the same project. . Very high concentrations (30 to

70%) of Saharan dust were found in residual particles from tropical cirrus anvils in the eastern Pacific (Twohy, personal communication). We note here that since the CVI collects all particles above a certain equivalent aerodynamic diameter, the utility for specifically identifying ice nuclei in warmer clouds may be restricted to regions that are fully glaciated. For example, Pratt et al. (2009) identified unreacted mineral dusts as predominant compositions in ice crystal residuals in one glaciated cloud case during ICE-L.

The most direct evidence for dust as atmospheric ice nuclei comes from measurements of ice nuclei within transported dust plumes. Airborne ice nuclei and ground-based polarization lidar measurements in Florida during CRYSTAL-FACE provide direct evidence of a connection between dust of Saharan origin (identified by lidar, satellite data, and trajectory analyses)and very high concentrations of heterogeneous ice nuclei (Sassen et al., 2003; DeMott et al., 2003a). Prenni et al. (2007) also identified the highest concentration of ice nuclei re-processed in a CFDC from CVI residuals collected in cumulus anvil clouds in the most dust-influenced clouds during CRYSTAL-FACE. Similar association of high ice nuclei concentrations with Asian dust plumes have been inferred in the depolarization lidar studies of Sassen (2002; 2005) over the Western U.S. and Alaska in Spring, where rapid glaciation of clouds forming in regions of lidar-detected dust layers was noted. Recently, direct measurements of ice nuclei, aerosol size distribution, and aerosol model forecasts over the Pacific Basin during the Pacific Dust Experiment have confirmed the association of Asian dust plumes with elevated ice nuclei concentrations (Stith et al. 2009). Use of aerosol trajectory forecasting and remote sensing, studies such as those proposed for ICE-T can target airborne sampling of dust plumes to more fully explore the impact on a wide range of cloud temperature regimes.

Direct evidence for dust as a major contributor to atmospheric IN also comes from measurements of the composition of aerosol particles processed as ice nuclei; the small ice crystals formed in ice nuclei instruments leave little time for additional scavenging. Chen et al. (1998) and Rogers et al. (2001) processed aerosols in a continuous flow diffusion chamber in the vicinity of cirrus and Arctic stratus clouds respectively and found silicates and other crustal materials to represent about half on average of residual ice nucleating particles based on EM studies. Phillips et al. (2008) summarize TEM measurements from several such studies, indicating mineral dust-like particles as the dominant IN composition about 55% of the time. Real-time mass spectrometric measurements of IN were made by DeMott et al. (2003b) at a high altitude continental U.S. site and similarly identified mineral dust and metallic (possibly oxides that compose dust) as two-thirds of all ice nuclei under remote aerosol conditions. These estimates are only modestly lower than estimates of mineral dust IN contributions from early analyses of the inferred nuclei of collected snow crystals. We note though that most of the more recent studies have emphasized ice nuclei active at temperatures below -20°C.

The primary ice nucleation efficiency of dust has now been examined and validated in numerous laboratory studies focusing on freely suspended or flowing mineral dust aerosols from major source deserts and their major components. These studies have indicated that at temperatures warmer than -35°C and colder than about -10°C the onset of ice forming on some desert dust aerosols occurs at the same time as the activation of liquid water droplets through a condensation or immersion process (e.g. Field et al. 2001; DeMott et al. 2008; Welti et al. 2009). Only at colder temperatures does the deposition mode become apparent (e.g. Mohler et al. 2006; Koehler et al. 2007). Up to 10% of the dust was found to act as ice nuclei, but fractions activating are strongly dependent on temperature, ice relative humidity, and to some extent water supersaturation (DeMott et al. 2008; Welti et al 2009). Less than a few % of all dust particles are typically found to be active in the mixed phase cloud regime warmer than -30°C in most recent studies. There remain needs to validate earlier work indicating the proclivity of mineral dusts to be subject to preactivation effects (e.g., Roberts and Hallett, 1968) due to prior cloud ice activation and to determine the impacts of atmospheric chemical and cloud processing on the IN activity of mineral dusts.

A significant question that has arisen from laboratory studies and atmospheric lidar studies of dust interacting with supercooled clouds regards the warm temperature limit of dust influences on ice formation. This issue is extremely important as regards the source IN for ice formation in modestly supercooled maritime cumuli and the essential IN that act as the primary trigger for the Hallett-Mossop process. The laboratory studies indicate an ever decreasing efficiency of natural dust particles at sizes below 1 micron, presumably those that travel the longest distances, reaching immeasurable ice formation warmer than about -15°C. Nevertheless, a few studies of IN activation from dusts up to sizes of 10 μm diameter suggest that the first onset of ice formation may occur as warm as -9 to -13°C (Kanji and Abbatt, 2006; Knopf and Koop, 2006). These varying estimates span the range of the selected lidar studies, with Sassen et al. (2003) indicating that Saharan dust may stimulate ice formation in clouds as warm as -6 to -9°C, while the studies of numerous Saharan dust-cloud interactions over Africa by Ansmann et al. (2008) suggest that glaciations never ensues warmer than about -15°C. It is possible that discrepancies reflect differences in dust size distributions and/or source regions, but more direct measurements of IN in dust plumes and documentation of dust interactions with clouds at warmer supercooled temperatures are clearly warranted.

ICE-T will aim to study the role of dust on ice nucleation. Figure 2.5 shows the climatology (1965-2004) of dust concentration at Barbados (J. Prospero priv. comm.). Dust loadings increase from April through the July coinciding with the increase in precipitation and deeper summertime convection in the Caribbean region. Dust events occur in discrete outbreaks providing the opportunity of investigating the evolution of cumulus in both the presence and absence of dust.

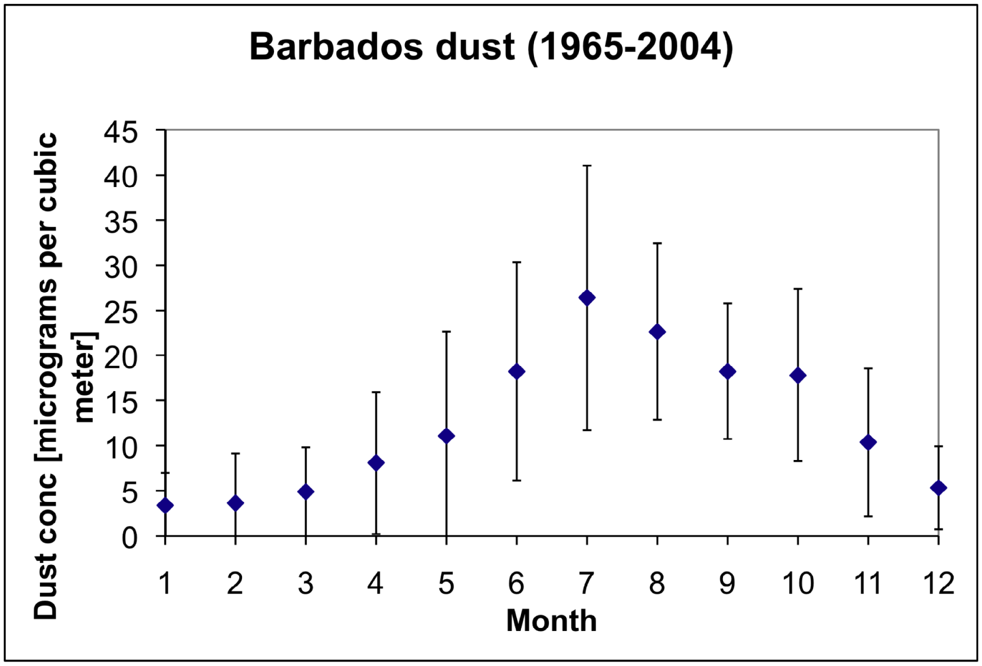


Figure 2.6 Climatology of dust concentration at Barbados averaged over the period 1965-2004 (J. Prospero priv. comm.).

2.8 Biological Particles as Ice Nuclei

Biological particles, including certain bacteria, fungi, pollen, and decayed organic material have been identified as ice nuclei of potential importance for clouds and precipitation since the 1960's (e.g., see reviews by Cantrell and Heymsfield, 2005; Möhler et al. 2007; Morris et al., 2004; Szyrmer and Zawadzki, 1997). Biological ice nuclei could represent an alternate (to mineral dusts) or an additional major source of atmospheric IN in modestly supercooled clouds (Möhler et al., 2007). A key unknown is their number concentrations in the atmosphere. Christner et al. (2008a) demonstrated the ubiquity of biological IN in fresh snow from mid- to high-latitude locations, but quantified only the number of such IN active at -7˚C per volume of snow water. Bowers et al. (2009) estimated numbers of ice nucleation active (INA) bacteria per unit volume of air at one of the same remote continental sites based on the assumption that drops formed from collected particles (also examined for their microbial-diversity and the presence of INA bacteria) that froze warmer than -10°C were from biological nucleators. Number concentrations varied from 1 to 50 m^{-3}, increasing with relative humidity. These are similar to maximum IN number concentrations at -15ºC that were attributed to bacteria in the Arctic (Bigg and Leck, 2001) based on processing particles collected on filters in a static thermal diffusion chamber. Phillips et al. (2008) use other considerations to roughly estimate this same concentration of bacteria active at -30˚C in "background-troposphere" conditions. Nevertheless, there is tremendous uncertainty in such estimates because little is known about the spatial and seasonal distributions of biological IN, the indirect manner of calculating atmospheric concentrations of individual bacterial IN from studies of large frozen drops of melted snow or rainwater, and the many possible quantitative errors in determining IN number concentration in air from filter collections processed in static diffusion chambers (Bigg, 1990). Also, since biological particles may be transported long distances at high altitudes (Pratt et al. 2009) biological IN activity must be determined at temperatures characterizing the full range of mixed-phase clouds. This is especially important if other bacterial species show more modest ice nucleation activity in comparison to known INA bacteria such as *Pseudomonas syringae*, as indicated by the recent measurements of Mortazavi et al. (2009). Another critical issue to resolve regarding biological ice nuclei, especially with regard to the objectives of ICE-T, is if sources are primarily terrestrial and from plant matter (Christner et al. 2008b), or if significant oceanic sources exist as suggested by Rosinski et al. (1986; 1987) who measured highly active organic IN that they ascribed to biological activity over certain currents and upwelling regions of the Pacific Ocean. These particles could be collected on slides from the C130 and processed in the laboratory.

2.9 Aircraft-Produced Ice Particles (APIPS)

In the early 1980s, Rangno and Hobbs directed attention to a phenomenon called "APIPs" (aircraft-produced ice particles). During research flights that measured cloud properties, the production of ice crystals from the passage of propeller aircraft at temperatures as warm as –8°C was documented (Rangno and Hobbs 1983, 1984). Ice particle concentrations were more than 10^4 times greater than the expected concentrations of ice nuclei at this temperature. The APIPs were contained in a relatively narrow cylindrical region of cloud; the diameter of the cylinder increased with time, at a rate of about 1 m/s (Rangno and Hobbs, 1983, their Fig. 22).

Rangno and Hobbs (1983) suggested that the Mossop et al. (1968) observation of high ice particle concentrations in a cumulus with a cloud top temperature of -4°C was the result of APIPS. The Mossop et al. measurements were made in a cloud that was penetrated five times by a DC-8 aircraft and three times by a Constellation. Given the horsepower of these propeller aircraft, it is conceivable that they did influence the ice production in that cloud based on the Woodley et al (2003) study. This possibility was vigorously rejected by Mossop (1984). Determining if such observations of the glaciation of cumulus can be made, and if they are due to primary and secondary ice nucleation or an artifact resulting from the aircraft that were used to sample them, is a question to be addressed during the ICE-T field campaign.

2.10 Climate Implications

It is well accepted that CCN can have an important impact on large scale climate because changes in CCN directly affect cloud droplet size distributions, hence cloud albedo and the earth's radiation budget (Twomey 1977; Penner et al. 1994). Rogers (1994) argued that since a large fraction of the earth's clouds can consist at least partly, if not entirely, in ice or mixed phase, changes in aerosols acting as ice nuclei may have a strong effect on clouds and their impacts. There is potential for a strong effect attributable to changes of IN aerosols. Support for this assertion can be found in GCM simulations by Fowler and Randall (1996). They performed an assessment of the CSU GCM model sensitivity to ice phase and mixed-phase clouds. Significant changes in cloud optical depth and cloud fraction resulted from altering the partitioning between cloud ice and supercooled cloud water; this is the essential function of IN. These changes produced significant variations in longwave and shortwave cloud radiative forcing. Penner et al. (2001, IPCC) reported an experiment with the ECHAM climate model. In one experiment, all clouds between 0 and –40°C were assumed to be liquid. In the second experiment, all clouds were assumed to be ice. The difference in cloud forcing between the two experiments resulted in a total forcing of +13 W m^{-2} by clouds in this region if they are ice. Thus, there is a large potential for climate forcing due to changing microphysical properties associated with ice nuclei in this range of temperatures.

2.11 Modeling Studies Overview

The fundamental influence of ice on cloud properties is strong motivation for representing ice processes in a realistic way, if the modeled cloud properties are to be believed. Recent papers point to microphysical parameterizations as a significant uncertainty in models at many different scales (Grabowski, 2003; Randall et. al., 2003, Gilmore et al., 2004; van den Heever and Cotton, 2004). This uncertainty brings higher relevance to fundamental research; more realistic descriptions of ice processes within numerical models must build on a better foundation of understanding. Laboratory studies provide unique insight under controlled conditions, but are unable to simulate all the changes in ice nuclei chemistry, effects of evaporative cooling, dynamical transport, water vapor competition, feedbacks between ice processes and storm dynamics, and processes with time scales longer than ~10 minutes such as secondary ice production processes.

Recent model sensitivity studies using bulk microphysical parameterizations (Thompson et al, 2004; Colle et al, 2005; Garvert et al, 2005; van den Heever et al., 2006; Carrio et al., 2007) reveal that more observations are needed to characterize and model the following aspects: ice initiation and subsequent number concentration, the activation and role of both CCN and IN on ice processes, production and depletion of supercooled liquid water, evolution of snow and graupel size distributions, accurate representations of ice fallspeeds, and transition from rimed snow to graupel. Secondary ice generation is often ignored or treated inappropriately (Connolly et al., 2004) despite current thinking regarding its overwhelming importance in some cloud types.

2.12 Summary

From the discussion above, it is clear that fundamental knowledge is lacking about the nature of ice formation in clouds that can be addressed by coupling modeling and laboratory studies with new instrumentation deployed in a carefully designed field experiment:

- Measurements of ice nuclei need to be compared against measurements of ice concentrations in natural clouds under conditions that are well-defined. These measurements of ice concentrations need to be made with the newest instrumentation that can better resolve small ice particles than in the past, and are less affected by shattering artifacts.
- Numerous field and laboratory studies have suggested droplet evaporation can enhance ice nucleation, and although such an enhancement was not observed in the layer clouds studied during the recent ICE-L field campaign, new measurements with improved instrumentation are needed to determine if such ice nucleation could be important in convective clouds.
- The effects of variations in the chemical composition of aerosol and ice nuclei, as well as aqueous chemical changes in droplets, upon the ice initiation process are not known. New

studies can explore this question quantitatively by combining instruments for ice nuclei measurements, cloud particle separators (CVI), and single-particle mass spectrometry.

- The role of dust to act as ice nuclei in cumulus at temperatures below -15°C has been studied extensively, but its ability to act as CCN and immersion nuclei at warmer temperatures has not been suitably determined.
- The ice-nucleating efficiency of biological material at temperatures warmer than -15°C may be important in some clouds, and the influence of these types of particles needs to be characterized.
- Although ample laboratory evidence has characterized one secondary ice initiation process (*Hallett-Mossop*) that operates in a restricted temperature range and only in the presence of large particles undergoing riming, the mechanism for this process is still undetermined. Field observations suggest that other ice multiplication processes occur, but they have not been identified outside of this temperature range, or in the absence of liquid water. Future lab and field studies with more sensitive instruments are required to explain the *Hallet-Mossop* mechanism, and to identify other secondary ice nucleation processes.
- For cumulus, the location and processes responsible for initial ice formation are not known. The roles of the cloud dynamics, thermodynamic environment, secondary processes, and the relative importance of ice nuclei lofted from cloud base versus those entrained laterally or from cloud top, are important questions. These must be addressed in order to advance basic knowledge and to improve the modeling of cold cloud systems.
- Past observations of subtropical and tropical maritime cumulus clouds with top temperatures of -10 °C and below may have been influenced by pre-existing ice from prior convection, or by the research aircraft making the measurements. New observations in such clouds using newer particle probes and remote sensors (lidar and radar) are necessary to document if primary or secondary ice formation processes are active in these clouds, at these temperatures.

3. Scientific Direction

The **Ice in Clouds Experiment-Tropical (ICE-T)** field campaign, to be conducted in concert with laboratory measurements and numerical simulations, focuses on primary, heterogeneous ice nucleation and secondary processes in maritime cumulus clouds. It is proposed for Summer 2011 based out of St. Croix, Virgin Islands. This location was selected because it offers an opportunity to make significant progress in our fundamental understanding of ice formation processes, and allows synergy with other ongoing ground-based programs in the area (e.g. Barbados, Puerto Rico). Towering cumulus clouds are of particular interest because they offer simple dynamics (relative to deeper convective systems), both liquid and ice phase growth processes and their interaction, and high frequency of occurrence in the region. The Caribbean area is subject to episodic intrusions of African dust. Airborne experiments in these clouds can use guidance from satellite imagery and trajectory forecast models in order to time research flights during periods where long range transport of African dust is likely to affect the Caribbean.

Towering cumulus clouds have the desirable feature that the formation and evolution of water droplets, rain drops and ice particles can be characterized as a function of time with combinations of remote sensing, airborne sampling and modeling. These clouds provide the opportunity to characterize ice formation processes, the influence of a strong warm rain process upon different ice formation processes, to quantify the influence of dust and to derive ice nucleus activity and composition as constraints on ice initiation processes. We can also test and evaluate the influence of the research aircraft making the measurements on the production of ice in the clouds under study (i.e., APIPs) in order to limit such effects on the data set.

3.1 Science Objectives and Goals

This section describes field studies, laboratory experiments and model simulations that focus on ICE-T objectives. The studies are designed to:

1. Establish which primary heterogeneous ice nucleation modes are active and important by measuring ice formation in tropical maritime cumulus clouds:
   a. Detecting the initial formation of ice particles, with a combination of in-situ microphysical measurements and remote sensing observations. Microphysical properties will be measured over short spatial scales (~100 m) with the latest generation of particle probes with proper care to reduce the influence of artifacts on the measurements from APIPS and shattering.
   b. Defining the cloud and environmental conditions (temperature, pressure, water vapor and kinematics) when and where the first ice is observed. Measure the vertical velocity structure with in-situ probes and airborne Doppler radar.
   c. Determining whether preexisting ice particles are present in the environment, and whether they are ingested into the cumulus clouds under study, using remote sensors (airborne lidar, radar).
   d. Comparing ice production in dust-free and dust-laden conditions.

2. Identify ice nuclei by:
   a. Directly measuring the activation properties of ice nuclei in air ingested by the clouds from the boundary layer and surrounding cloud levels, for use in numerical modeling studies to predict ice initiation.
   b. Characterizing the physical and chemical properties of ice nuclei collected within the air ingested by the clouds.
   c. Characterizing the physical and chemical properties of the residuals of ice particles sublimated during passage through the Counterflow Virtual Impactor (CVI) probe. Use a large cut size for the CVI to reduce the collection of cloud droplets during some periods in liquid and mixed-phase regimes. Focus on glaciated regions in more mature clouds.
   d. Measuring the cloud activation properties of droplet and ice crystal CVI residual nuclei for comparison to ambient CCN and ice nuclei.
   e. Developing methods to improve the statistical sampling of ice nuclei at the warmer supercooled temperatures (e. g., bag samples, aerosol concentrators).

3. Characterize secondary ice production processes:

   a. In regions with first ice (from radar observations), measure ice particle size distributions and ice particle shapes as a function of time and with high spatial resolution.
   b. Characterize the nuclei of particles within the region of secondary ice production.
   c. Quantify the drop size distributions (including drizzle) created by the warm rain process at lower altitudes in the clouds, ascending above the freezing level and compare with ice formation in mixed-phase regions.

4. Determine the likely influence of aircraft produced ice on the ICE-T measurements.
   a. By making carefully designed flight patterns and with tracers of combustion and entrainment, measure the size distributions, shapes and concentrations of ice particles as a function of temperature.
   b. Characterize the nuclei of the APIP particles.
   c. Using in-situ and remote sensing observations, characterize the spread of these particles through the cloud volume.

5. Conduct laboratory experiments:
   a. That characterize the properties of the nuclei of particles generated by the Hallett Mossop (1974) process, to provide a benchmark for what is to be expected in clouds.
   b. That characterize the nuclei of APIP particles, using dry ice as a proxy for particles generated in the expanding air behind the blades of a propeller-driven aircraft.

6. Improve the prediction of ice concentrations within numerical models by:
   a. Determining the thermodynamic history of air parcels in which ice nucleation occurs in smaller, developing mixed-phase cumulus clouds.
   b. Performing numerical experiments that demonstrate the importance to ice formation on dynamical processes that drive the thermodynamics, such as updrafts, downdrafts, turbulence, entrainment and cloud-edge mixing events.
   c. Testing the sensitivity of numerical modeling results to secondary ice nucleation production rates and mechanisms.
   d. Simulating the overall development of the clouds sampled during ICE-T, ranging from simpler to more complex cases.

3.2 Scientific questions

A. Can we document the observed evolution of ice formation in maritime cumulus with top temperatures warmer than -10°C, and if this is dependent upon the generation of ice by propeller aircraft sampling the clouds?

B. Which types of aerosol act as CCN and IN and how do they depend on temperature, size and aging?

C. How do the warm rain and primary and secondary ice processes vary as a function of cloud lifecycle and with changing environmental conditions, particularly ambient dust? What is the fraction of vapor flowing into cloud base (the cloud base mixing ratio) that arrives at the 0°C, -5°C and -10°C temperature levels in the form of vapor, supercooled liquid water and ice (see A. Heymsfield et al, 2009)? How does dust affect these fractions? How does this depend on the cloud life-cycle?

D. Does primary nucleation, specifically the number concentrations of ice nuclei, explain the onset and glaciation of cumuli?

E. Are secondary ice formation processes critical to the glaciation process in these clouds, what concentration of primary IN are sufficient to trigger them and how does the process work?

F. Does mid-level entrainment play a role in feeding CCN and IN, particularly dust, into convective clouds? There will be a proposal submitted to NSF as part of ICE-T to use water isotopes to characterize the originating temperature of the entrained air, as in Webster and Heymsfield (2003). Furthermore, vertical profiles of ozone and CO show nearly zero variability below the trade inversion, and thus that the difference in CO and ozone concentrations below the trade cumuli and their environment is nearly zero. Thus, conserved tracer studies are not likely to be successful below the trade inversion. However, the air above the trade inversion may be vastly different, and it is thus possible that CO and ozone may be excellent tracers of both source and amounts entrained in the upper parts of the ICE-T clouds.

G. Can numerical models which treat primary and secondary ice production processes adequately treat the spread of ice and precipitation initiation in tropical maritime clouds?

3.3 New Measurement Capabilities

To address the science objectives outlined above, a complete set of aerosol and microphysical probes, remote sensors and bulk probes that use the latest technology are requested (Table 2). Listed alongside each instrument are the particular science questions requiring data from each of these instruments to be addressed (Table 2A). With a few exceptions, each of these probes flew on the C-130 during the ICE-L field campaign. The software necessary to process the data from most of the probes has already been developed and used for ICE-L field analyses. There is redundancy in some categories, especially for the microphysical probes, because each probe offers somewhat different measurement capabilities. Improvements to some of the probes are anticipated (e g., installation of Korolev-type probe tips to reduce shattering of large ice on the tips of the 2D probes). *Proposals will be submitted to NSF to operate some of the user-supplied probes and/or to process and analyze the data.*

There has been significant improvement in the suite of probes used to measure the microphysical properties of ice particles in all size ranges. Particle size distributions and shape information can now be obtained for sizes as small as a few microns and extending across the full range of particle sizes expected to be observed in maritime cumulus clouds (several millimeters). New probe tips and open path inlets (to reduce shattering of large ice on the inlets), higher resolution and smaller detection sizes, faster electronics, and improved data analysis software will help produce an unprecedented data set of the formation of ice in the tropical maritime cumulus clouds sampled during ICE-T.

Ice nuclei and cloud condensation nuclei measurements, including chemical composition and size distributions, provided a wealth of information on the properties of the aerosols during ICE-L and are also ideally suited for use in ICE-T. Airborne cloud condensation nuclei (CCN) instruments can measure activation spectra (e.g., Hudson 1989). The continuous flow diffusion chamber (CFDC, Rogers et al., 2001; Eidhammer et al. 2009) has been used

Table 2: Proposed Instrument Complement

Cloud and precipitation microphysics

Probe/Source	*PI Objectives/questions (from Section 3.1)*
SID IIH# (EOL/MMM)	1a, 1c, 3a, 4a, 4c, A, C, D, E
HOLODEC > 5 µm drops/crystals (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
CDP 1 – 47 µm (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
FSSP (tubeless) 1 – 47 µm (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
Fast 2D-C (10 µm res.)#* 15 – 640+ µm (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
Fast 2D-C (25 µm res.)#* 25 – 1600+ µm (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
2D-P (200 µm res.) 200 - 6400+ µm (EOL)	1a, 1c, 3a, 4a, 4c, A, C, D, E
3V-CPI (2.5 & 10 µm res.)# 2.5 µm – 1000+ µm (EOL/SPEC)	1a, 1c, 3a, 4a, 4c, A, C, D, E
CIP with depolarization# 15 – 1000+ µm (DMT)	1a, 1c, 3a, 4a, 4c, A, C, D, E

Aerosol and ice nuclei probe

Probe	*PI Objectives/questions*
CDFC with TEM (CSU and others)	1e, 2b, 3b, 4b, 5a, 5b, B, D, E, F, G
ATOF-MS (UCSD)	2a, 3b, 4a, 4b, A, C, D, E, F
CVI with TEM and SEM (OSU, ASU)	2a, 2b, 3b, 4b, B, C, D, E, F
CN >10 nm (EOL)	
RDMA 10 – 100 nm (EOL)	2a, 3b, 4a, B, C, F
UHSAS 0.1 – 1.5 µm (EOL)	2a, 3b, 4a, B, C, F
SP2 (DMT)	2a, 3b, 4a, A, B, D
CCN (DMT)	2a, 3b, 4a, B, C, F
CCN (DRI)	2a, 3b, 4a, B, C, F
GNI (giant aerosol) (EOL)	1e, C, F

Bulk probes

Probe	*PI Objectives/questions*
Water isotopes (CU)	1d, 4a, F
King LWC (EOL)	1b, 6a

Remote Sensing

Probe	*PI Objectives/questions*
Up/down 94 GHz radar (UWyo.)	1a, 1b, 1c, 1d, 3a, 4c, A, C, E, F
Up/down lidar (UWyo)	1a, 1b, 1c, 1d, 3a, 4c, A, C, E, F
Ophir temperature (EOL)	1b, 6a
Remote sky/surface temp (EOL.)	
Digital video (EOL.)	

Trace Gases

Probe	*PI Objectives/questions*
Fast ozone (ACD)	4a, C, F
CO (ACD)	4a, C, F
CO_2# (ACD or Toohey (CU) instrument)	4a, C, F
TDL & VCSEL hygrometers (EOL)	1b, 1d, 4a, 6a, C, F
Lyman-alpha hygrometer (EOL)	4a, C, F

Notes # New and/or improved probe * It is highly desirable that the probe be modified to have tips to reduce shattering successfully to measure ice nucleus concentrations of aerosols in the size range ~50 nm to 1 μm. Improvements in phase discrimination detection could potentially extend ice nuclei detection to larger aerosol sizes. The *counterflow virtual impactor* (CVI) separates particulate residues from evaporated cloud particles. CVI-derived aerosols can be fed to the CFDC to examine their ice nucleating properties, to a single particle mass spectrometer and to a non-refractory aerosol mass spectrometer to measure size and chemical composition, or to CCN, EM grids (for TEM analysis) and membrane filters (for SEM analysis), and other aerosol characterization devices. Similarly, residual particles from ice crystals that nucleate and grow in the CFDC can potentially feed a single particle mass spectrometer (DeMott et al., 2003b), a method that has not been exercised to date on aircraft. Aerosol particle mass spectrometers have been adapted for airborne use (Pratt et al., 2009) and provide mass and chemical composition of single particles larger than ~50 nm. Similar instruments that use particle collections are sensitive to ~5 nm; these are not adapted for airborne use yet, although work is ongoing to achieve this capability.

Cloud profiling with millimeter radar provides information on cloud structure and dynamics, water phase and turbulence, and the location of first ice (e. g., Fig. 2.5). The addition of a downward-viewing lidar provides the capability of evaluating whether the towering cumulus clouds under study (Section 3.3) are developing upward into preexisting ice or dust, or are entraining ice or dust from the adjacent environment.

Chemical tracers (ozone and CO will be used in an attempt to determine the amount and altitudes of entrained air in the upper part of the cumuli. The isotopic composition of water vapor and cloud particles constrain the history of cloud nucleation and air mass origin. The two heavy isotopologues of water ($H_2{}^{18}O$ and HDO) combined give a measure of the supersaturation conditions under which ice particles were formed because the rate of diffusion growth differs for the two species. On the other hand, supercooled liquid drops are close to being in thermodynamic equilibrium with vapor (and linked to a vapor pressure dependent isotopic fractionation). As such, upon being frozen the particles resulting reflect the particle formation process linked to having been formed as supercooled drops. So too, riming of ice crystals formed from diffusional growth will have a mixture of the isotopic signature. The disequilibrium between cloud particles and vapor can be provided directly by measuring both the isotopic composition of vapor and liquid and further insight can be made by profiling through the depths of the cumuli.

3.4 Towering Cumulus Clouds

Towering maritime cumulus clouds offer an excellent opportunity for characterizing warm rain and ice microphysical processes in an environment with fewer hazards to aircraft than deeper convection (Fig. 3.1). These processes can be quantified using remote sensing—airborne and ground-based radar and lidar, and in-situ aircraft observations. In such clouds, the measurements needed to address the scientific questions listed earlier in this section can be made using a single aircraft that has the ability to sample the aerosol and cloud properties in-situ.

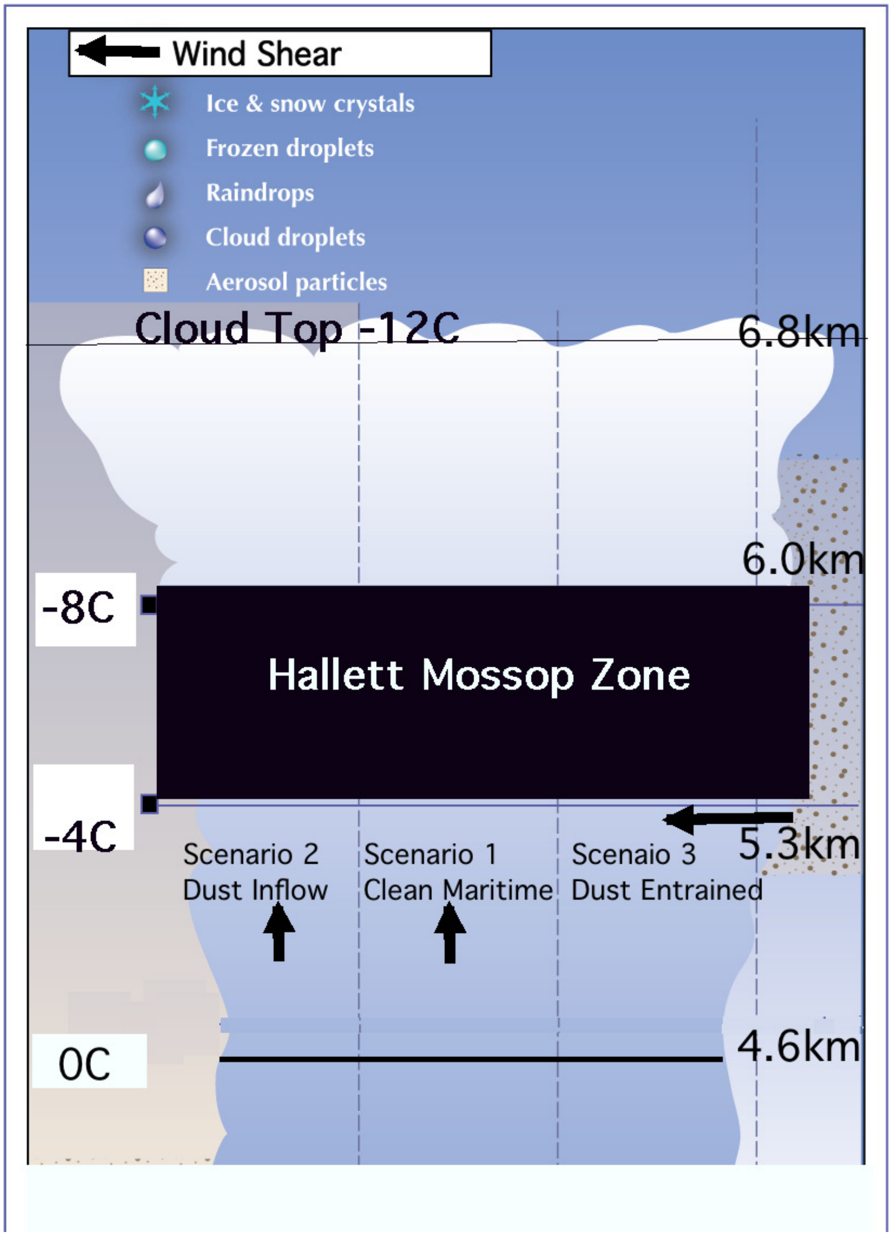


Figure 3.1. Maritime towering cumulus cloud with three scenarios: clean maritime air, maritime air with dust in the inflow, and mid-level entrainment of dust. Significant temperatures and heights and direction of vertical wind shear are shown.

3.5 Climatology of Cumulus Clouds and Dust in the Caribbean During Proposed Project (June/July) Timeframe

Satellite active remote sensors and surface observations now provide a wealth of information available to develop a cumulus cloud climatology in the vicinity of St. Croix (17.70N 64.80W) during the June/July timeframe. For later reference to the satellite observations, July soundings from Luis Muñoz Marín International Airport in San Juan, Puerto Rico and from

Grantley Adams Airport in Barbados, indicate that the 0, -10 and -15 °C levels are near 4.6, 6.6, and 7.4 km, respectively (Fig. 3.2) and fall within the C130's flight envelope.

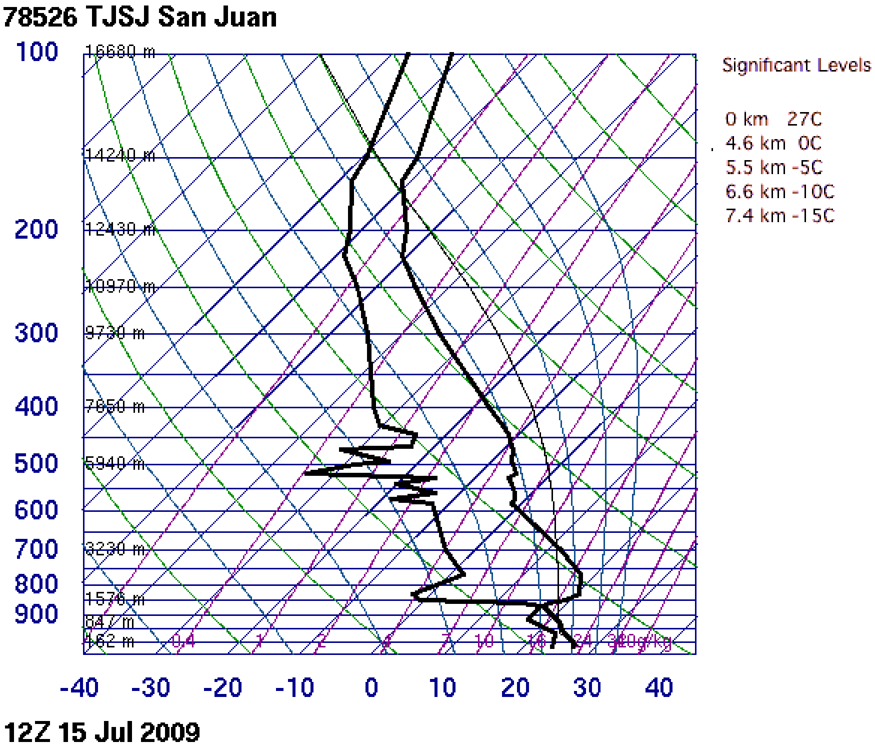


Figure 3.2 Typical July sounding from San Juan, Puerto Rico.

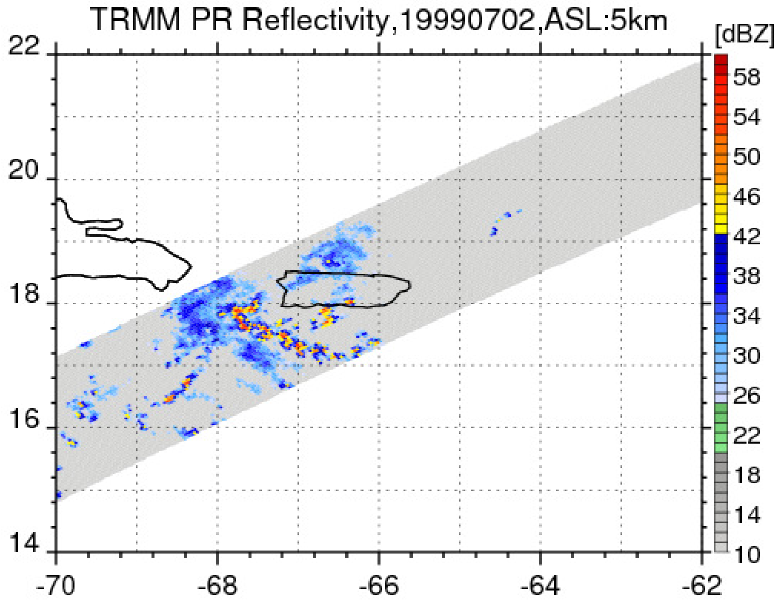
Using the revised Hahn and Warren ship-based climatology that uses 44 years of surface observations (http://www.atmos.washington.edu/~ignatius/CloudMap/), statistics on the daytime frequency of occurrence of cumulus clouds indicate that in the July timeframe, cumulus cloud coverage of the sky is about 23%, while cumulonimbus cover an average of 9%. Stratus coverage is an average of 2%. Average cloud base heights are about 670 m.

Since 1997, the TRMM Precipitation Radar (PR) has provided information on the locations and coverage and heights of precipitating regions, where reflectivity exceeds about 18 dBZ. The footprint of the TRMM PR at the surface is about 4x4 km and 5x5 km after the orbit boost. These data are valuable for developing an ICE-T cloud climatology because we are interested in studying those clouds with an active secondary ice production process where presumably rimed particles and frozen drops are present.

From the TRMM historical data base, we have developed a climatology of TRMM radar echo locations for each of the months June, July and August in a rectangular domain that is 8 degrees wide in longitude and latitude centered on St. Croix (Figs. 3.3, 3.4). All locations within this domain are within about one hour ferry time for the C130. Using the 12 years of TRMM radar observations, we have developed a climatology of the frequency of occurrence of the tops of precipitation regions, which we define as the top of a vertically contiguous radar echo that extends from the surface to height H. These would be precipitating cumulus clouds. Over the 12 years of TRMM PR radar data, there are approximately 500 overpasses (or 42 per year) within this domain for each month (Fig. 3.4).

For the height intervals of relevance to ICE-T, we can expect up to 3% of the area of the domain to be covered by precipitating convective clouds during the month of July. Within the domain and assuming that the convective cells are 5 km wide corresponding to the TRMM footprint size, we should have hundreds of opportunities to sample cumulus clouds at any one time during the month of July (e. g., Fig. 3.3). Note also that for each of the three months, the probability distribution function (PDF) of echo top frequency peak a few km above the average cloud base height (from the Hahn and Warren study), presumably due to the minimum depth required for the warm rain process to develop. The frequency distributions of occurrence of deep precipitating convective clouds and the distribution with height are slightly lower for June than July, therefore, July evidently has somewhat more numerous and deeper convection (Fig. 3.4). August is the start of the Atlantic hurricane season and will be avoided if at all possible.

The normalized probability of a TRMM PR radar echo at any given height H, without the constraint of a vertically continuous radar echo structure, is shown in Fig. 3.5. There is a rapid drop-off in the probability of “precipitation” in clouds with tops from 4.5 km (about 0°C) to 6.0 km (-10°C). Thus, warm rain processes probably dominate convection in the area near St. Croix but also argue for making this region exceptional to study the interaction of warm rain and ice processes.



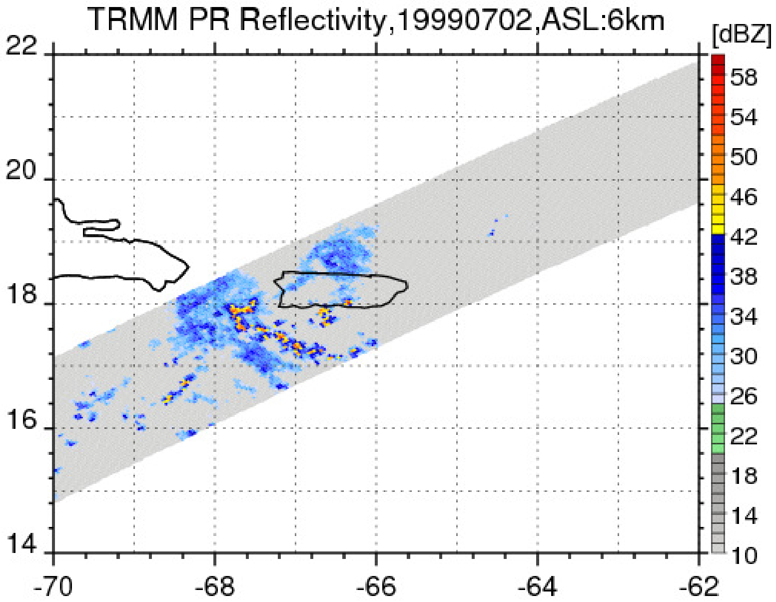
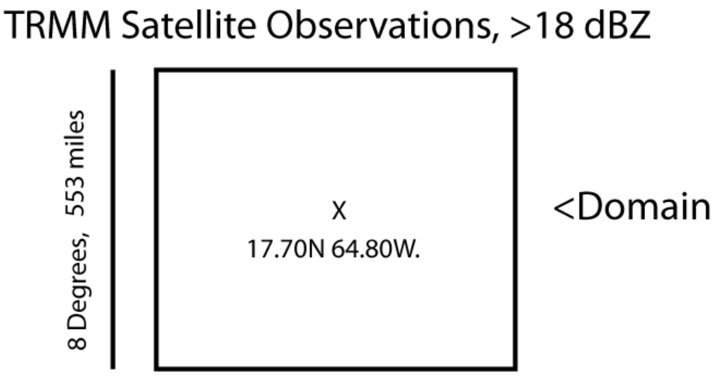


Figure 3.3 TRMM precipitation radar observations (above a minimum detectable reflectivity of 18 dBZ during one overpass during the month of July, for (top) 5 km, and (bottom) 6 km levels.



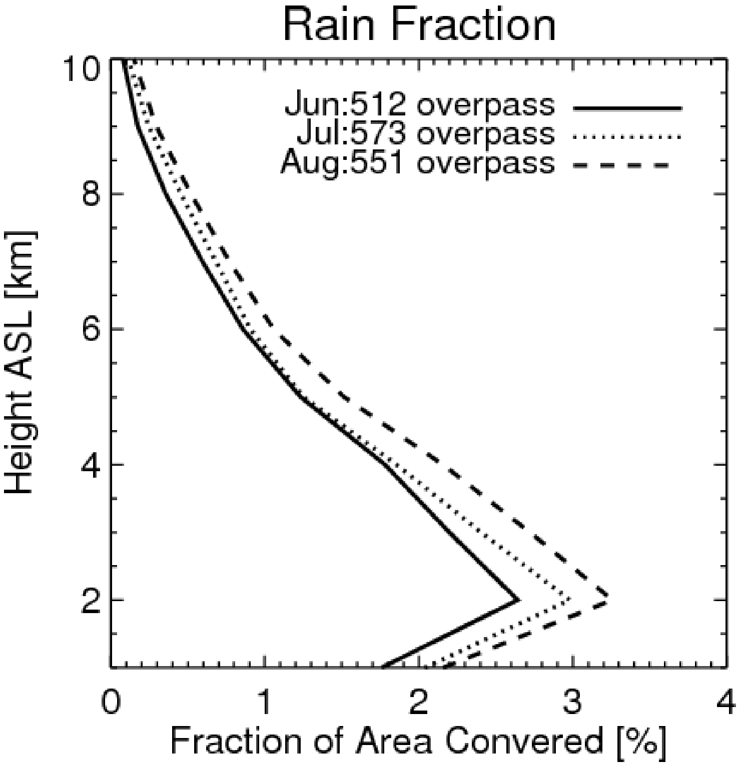


Figure 3.4 Fraction of TRMM radar echo occurrence over the domain shown in the top of the figure. Whenever there is a TRMM PR overpass over this study box, the AREA of > 18dBZ based on the radar swath size is calculated at each 1km in the vertical from the surface to the 10 km level. The fraction of area covered is derived from the total area of > 18dBZ in all the overpasses divided by the total AREA of PR sampling during these overpasses. Data kindly compiled by Weixin Xu and Edward Zipser, University of Utah.

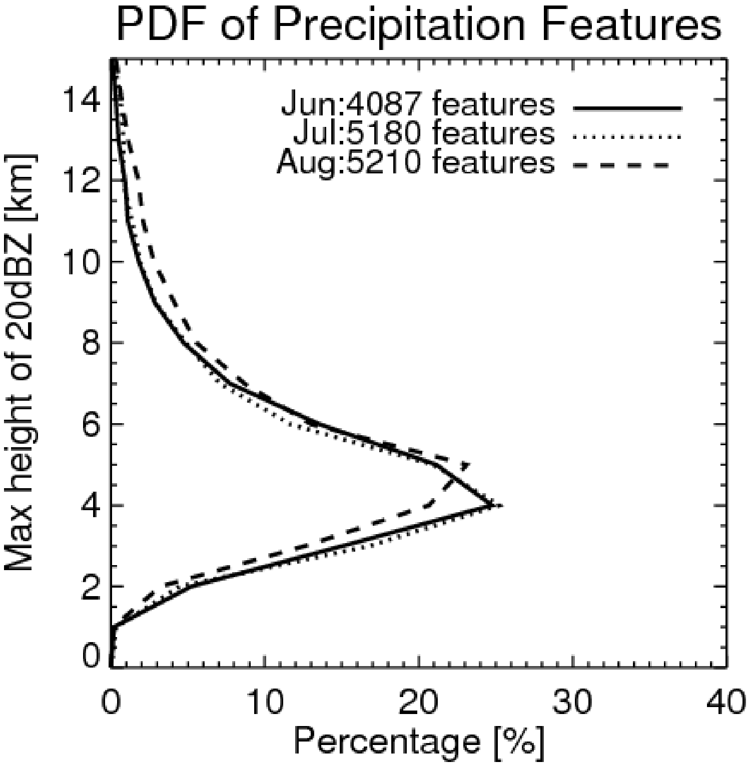


Figure 3.5 Probability Distribution Function of cloud top heights. Data kindly compiled by Weixin Xu and Edward Zipser, University of Utah.

CloudSat, with a 94 GHz Cloud Profiling Radar (CPR), provides more detailed information on cloud height occurrence due to its improved detection sensitivity but with fewer overpasses of the domain, because it has only been in operation since 2006 and the CPR only provides vertical cross-sections under the satellite (Fig. 3.6). The CPR signals are averaged along the track to provide a profile every 1.1 km with an effective footprint size of 1.4 km cross-track × 1.8 km along-track. We can use GOES imagery but CloudSat provides us with information on the vertical distribution of cloud occurrence. At approximately the same longitude as St. Croix, CloudSat crosses the same latitude while heading toward the northwest in daylight hours (St. Croix is -4 hours UTC) and toward the northeast at night. The averaged cloud layer number detected under this narrow CloudSat path—discrete tracks across the domain in Fig. 3.6, is presented in Fig. 3.7. There appears to be diurnal cloud variability, with some deeper cloud encountered.

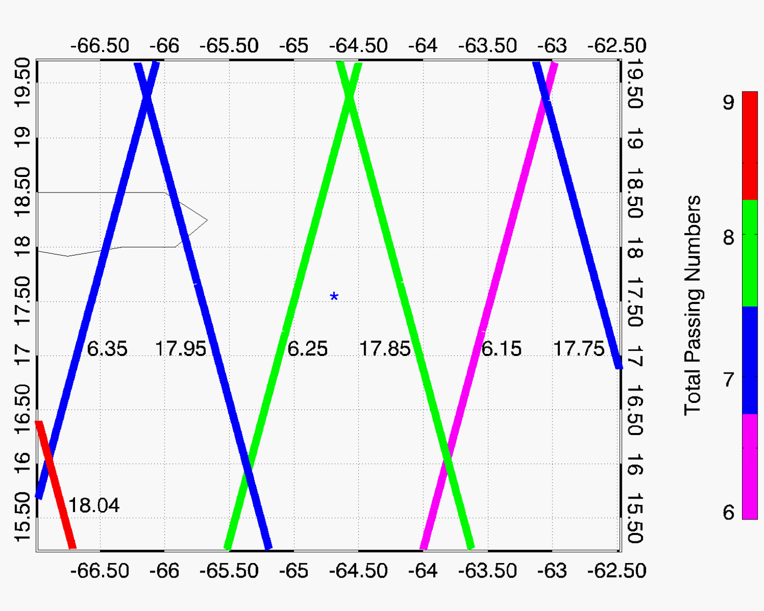


Figure 3.6 CloudSat tracks around St. Croix and number of overpasses used for cloud statistics. The numbers near the track indicate approximate overpass time in UTC for each track.

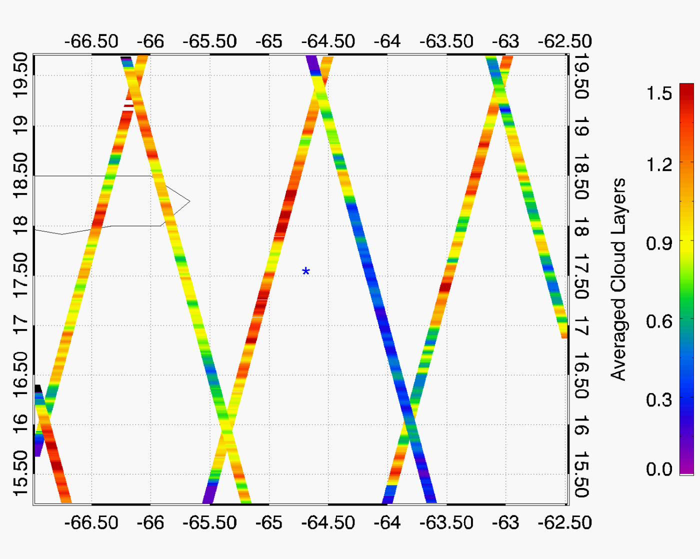


Figure 3.7 As in Fig. 3.6, but for mean cloud layer detected by radar at any height in the atmosphere. The scale ranging from 0 to 1.5 is the average number of discrete clouds encountered across the swath as a function of the position along the swath.

Along the **CloudSat** tracks, we have examined the length scales of continuous cloud occurrence. The minimum distance of a radar "pixel" is 1.1 km and we have derived PDF's for more than 4 pixels to reduce noise. The highest frequency of occurrence of continuous radar reflectivity at heights of 4-6 km is about 5 km in length, with the occurrence dropping quite rapidly for longer scales (Fig. 3.8). Along the 500 km CloudSat track in the area of interest, we can expect to encounter clouds with 5 km in length about 0.3% of the time.

Assuming clouds have the same size along and cross track, there are very roughly 500/5*0.3 or a total of about 30 clouds with diameter ~5 km within the 500 km long domain. Counting clouds with size large than 5 km given in Fig. 3.8, there are more than 50 cloud systems extending up to 4-6 km heights at a given time within 500 × 500 km domain, i.e., over 120 cloud systems in the 800 × 800 km domain used in the TRMM data analyses.

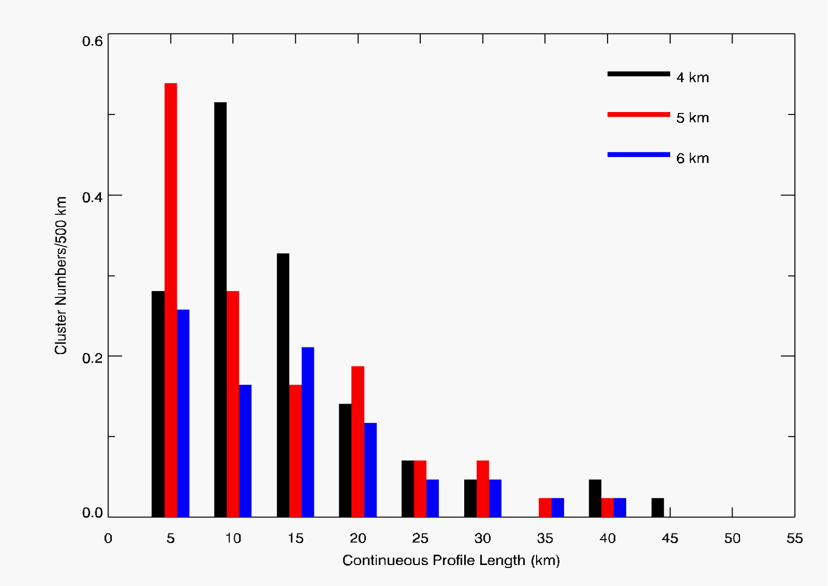


Figure 3.8 Frequency distribution of continuous horizontal lengths of along-track radar echoes from CloudSat CPR observed from south to north across the 500 km domain within transit range of C130. Data are shown according to the altitude, so, for example, for an altitude of 4 km, there will be 0.28 cloud clusters of diameter 5 km.

The distribution of mean CloudSat radar top heights for cloud clusters observed under the CloudSat track (Fig.3.9) shows a dominance of cloud tops below 4 km height, but throughout the range 4 to 7 km we can expect numerous clouds reaching the levels of our interest.

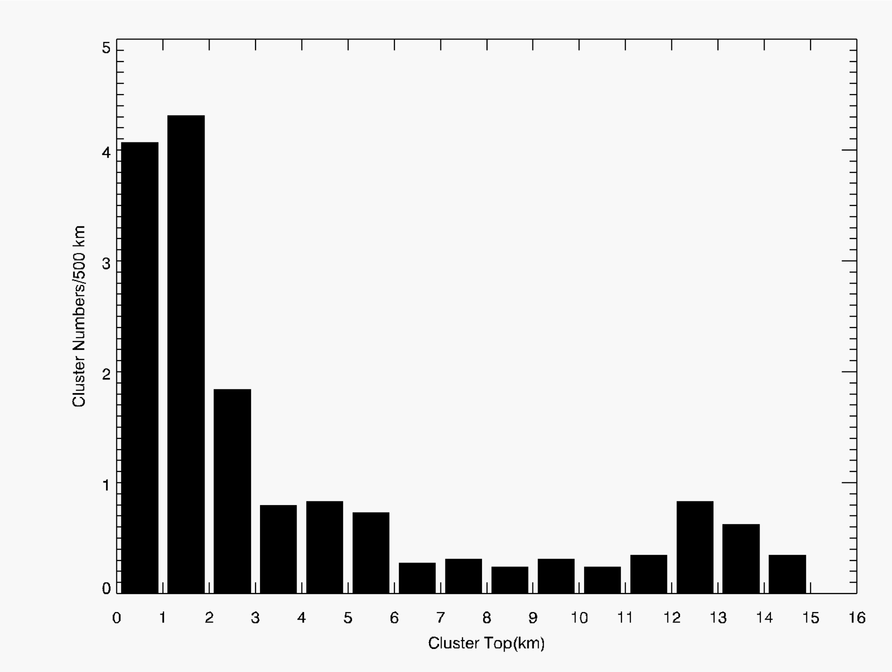


Figure 3.9 CloudSat radar top height distribution for the month of July in the 500 x 500 km^2 domain within range of the C130's base at St. Croix.

In summary, soundings indicate that the NCAR C-130 is capable of sampling convection in the Caribbean area—specifically the area around St. Croix, Virgin Islands to temperatures of about -20°C during the month of July. During daytime hours, we can expect 100 cumulus of diameter 4 km and larger daily in a domain within a one hour ferry time of the C130 based in St. Croix.

The influence of dust on the properties of the supercooled regions of maritime cumulus clouds is a primary objective of ICE-T. Observations of boundary layer dust concentrations have been derived on a daily basis since 1973 by Prospero and coworkers. Daily data for July1973-2009, kindly furnished by Joe Prospero, suggests that perhaps 50% of the days during July will have enhanced concentrations of dust (Fig. 3.10) relative to the background dust levels during the winter (see Fig. 2.5) at Barbados, not far from St. Croix. Relatively high concentrations, above 50 ug/m^3, might be present 20% of the time and with transits to dust enhanced areas by the C130 aircraft, a greater percentage of cases can be attained (Fig. 3.10).

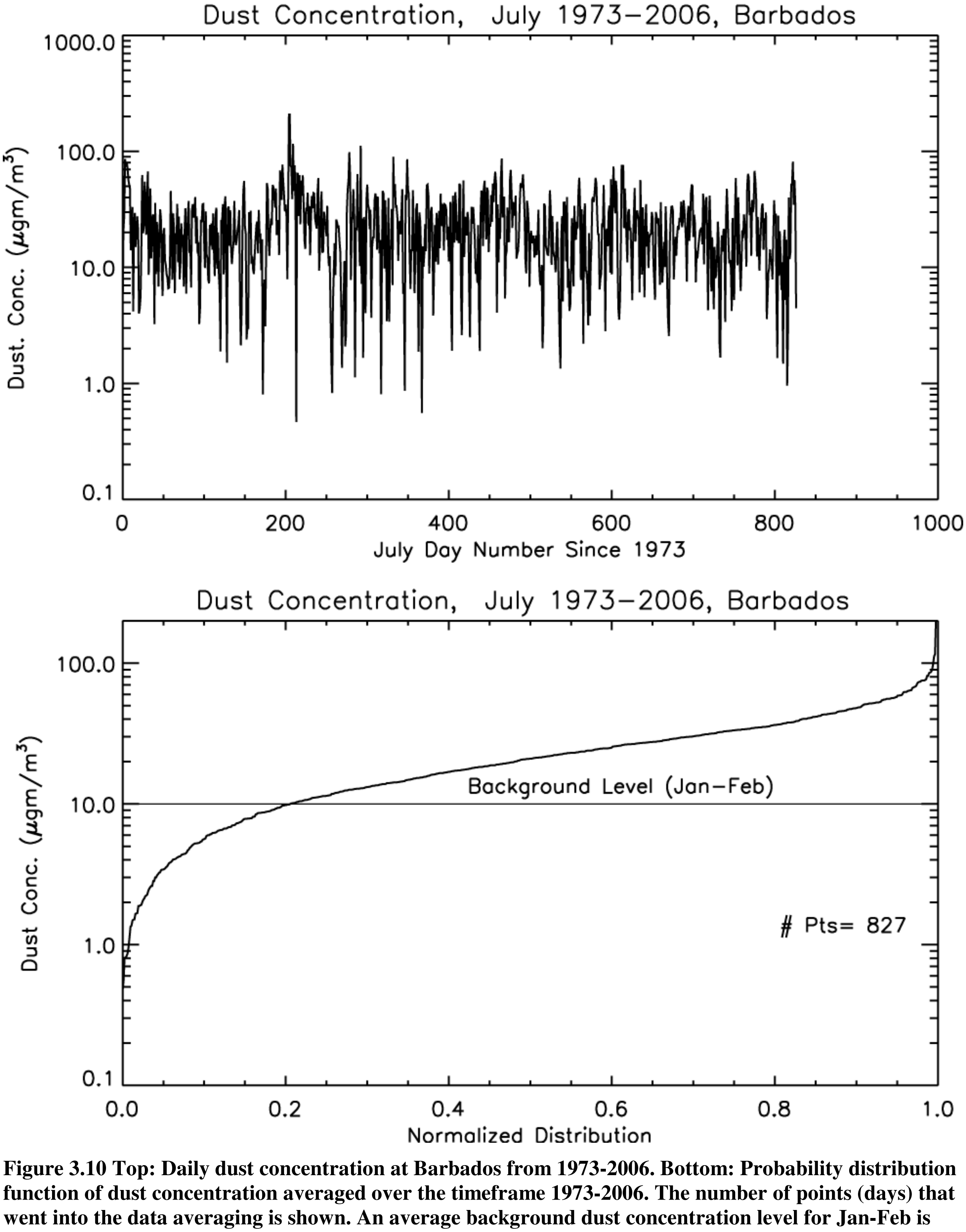


Figure 3.10 Top: Daily dust concentration at Barbados from 1973-2006. Bottom: Probability distribution function of dust concentration averaged over the timeframe 1973-2006. The number of points (days) that went into the data averaging is shown. An average background dust concentration level for Jan-Feb is plotted as a horizontal line. Data courtesy of Joe Prospero. It is quite possible that the dust concentrations surrounding the upper parts of the cumuli may be significantly different from these surface measurements.

Detailed flights patterns designed to investigate ice initiation in cumulus clouds are presented in Section 4. The instrumentation set (Section 3.2) will characterize ice nuclei and differentiate water droplets and ice particles with far greater sensitivity than previous projects. Opportunities to characterize ice nucleation with different aerosol types (e. g., dust) will be given emphasis. African dust influences on primary ice concentrations will be investigated as part of ICE-T. With measurements in pristine and dust-laden cloud, this research can address the central question, if it is possible to predict primary and second ice concentrations from measurements of ice nuclei and ice particle measurements.

3.6 Synergy with Other Programs

In an NSF-funded study, "Impact of African Dust on Clouds and Precipitation in a Caribbean Tropical Montane Cloud Forest" (2009-2012), PI Mayol-Bracero and Co-PI Prather will combine field and modeling investigations to address how the physico-chemical properties of long range transported African dust aerosols influence Caribbean cloud properties and precipitation levels in a Puerto Rican tropical montane cloud forest. The distance to one of the field sites, Luquillo Experimental Forest (18.27N 65.75 W), is only 110 miles from St. Croix. Their intensive field campaign coincides with the proposed ICE-T time period. They will continuously measure aerosol properties using an ATOFMS, Nephelometer, PSAP, Particle Counter, and a GRIMM Optical Counter. Coordination with this program will allow us to better quantify the aerosol properties in the region of ICE-T. See more details of the proposed collaboration with this ongoing study in the Appendix.

The NASA micropulse lidar network (MPLNET) includes a site on Barbados, a 470 mile transit from St. Croix. NASA also makes continuous spectral radiometer measurements at Barbados as part of their AERONET (AErosol RObotic NETwork) network. The Aerosol-cloud-precipitation-climate interactions (ACPC) instruments are operated at Barbados by the Max Planck Institute under the direction of Bjorn Stevens. They include:

- Daily Soundings
- DIAL and Raman Lidar
- Scanning S and K-band Doppler polarization radars

These observations will be made during the proposed ICE-T time frame. Such data may also be useful for characterizing general aspects of the aerosol and clouds in the ICE-T region.

3.7 Modeling Studies

In the past, simple modeling approaches to ice in clouds have been used in part because of limitations in our basic understanding of the underlying microphysical processes, as well as inability to resolve the key scales at which microphysical-dynamical processes operate because of the coarse resolution of climate and numerical weather prediction models. With increasing computer power and hence improved representation of small-scale dynamic and thermodynamic variability through better model resolution (including the "superparameterization" approach, e.g., Grabowski et al. 2003) and/or assumed-PDF methods (e.g., Golaz et al., 2002), combined with model tests that indicate important sensitivities to details of ice evolution, it is becoming clear that a more realistic approach to ice evolution—a more cause-and-effect approach—is both possible and warranted. The recent development of two-moment bulk ice microphysics schemes (Meyers et al., 1997; Liu et al. 2007; Morrison and Gettelman 2008; Lohmann and Roeckner 1996), combined with improved representation of sub-grid scale dynamics and thermodynamics, provide a framework for representing detailed ice microphysical processes in climate and weather models. For example, Kärcher and Lohmann (2002; 2003) developed a physically-based parameterization, that has been validated using parcel model results, to treat homogeneous and heterogeneous ice nucleation, respectively, in global models (Lohmann and Kärcher, 2002; Lohmann et al., 2004). Liu and Penner (2005) developed a parameterization that treats the combined homogeneous and heterogeneous nucleation that is based on a mechanistic description of ice nucleation. These parameterizations need to be tested through application to specific field situations in order to bring higher relevance to fundamental research. Most representations of ice generation prediction still use a single parameter dependence (e.g., temperature or ice super saturation) of nucleated ice concentrations such as the Fletcher curve (Fletcher, 1962) or Meyers parameterization (Meyers (1992) to represent primary ice nucleation, despite the near universal opinion among cloud physicists that it is unjustified because it is inconsistent with recent evidence (Rangno and Hobbs, 1994; see above ice nuclei measurement figure), and an oversimplification of the ice production process. Its appeal is its simplicity. Numerical cloud models usually do not include sources and transport of ice nuclei (Lin et al. 2002, Tao et al. 2003), and geographic and altitude dependences are seldom represented (Bigg 1976, Oishi 1994). Modeling the sources of these aerosols is thus still a problem.

Some models such as RAMS have attempted to address these issues. RAMS has initial vertical and horizontal variability of aerosols that can serve as CCN, GCCN, ice nuclei, and activation and sinks of those aerosol are explicitly modeled (Cotton et al., 2003; Saleeby and Cotton, 2004; Carrio et al., 2005; van den Heever et al., 2006; Prenni et al. 2007). This model has also recently been interfaced with a dust model (Smith, 2007) in order to represent aerosol sources associated with dust generation. RAMS simulations of convective storm development under the influence of dust observed during the CRYSTAL-FACE field campaign demonstrated the impacts of varying concentrations of dust acting as IN on the surface precipitation, the partitioning between liquid and ice species, the dynamical structure of the storm, and the optical and microphysical characteristics of the anvil produced by the deep convection (van den Heever et al., 2006; Carrio et al., 2007). These simulations also showed the importance of the interactions between CCN and IN on storm characteristics, and highlighted the fact that the roles played by IN and GCCN were just as important as those of CCN. Finally, the influence of IN and the associated ice processes on the updraft strength were found to be important, with subsequent influences on the anvil longevity and depths. Obtaining measurements of updraft strengths associated with varying IN concentrations would therefore be useful to enhance our understanding of the feedbacks between storm dynamics and IN in convective storms.

With the parameterizations described above (Lohmann and Kärcher, 2002; Liu and Penner, 2005; Barahona and Nenes, 2009), it is critical to develop the means to treat ice nucleation in coupled aerosol/cloud microphysics schemes. These parameterizations would need to be extended to treat heterogeneous nucleation at temperatures warmer than –40°C. A few heterogeneous ice nucleation parameterizations that link aerosols to ice nucleation have been suggested, such as the empirical parameterization by Phillips et al (2008) and the classical theoretical nucleation parameterization by Khvorostyanov and Curry 2004. Still, this area of development is perhaps the most critical need, given the estimates of possible climate forcing by ice clouds in this region (Penner et al., 2001). A key point is that most global modeling systems now include detailed representation of aerosol, often with links to two-moment cloud microphysics schemes (Ming et al. 2007; Liu et al. 2007; Morrison and Gettelman 2008; Lohmann 2002). Other modeling systems, including the "superparameterization" or "multi-scale modeling framework" (e.g., Randall et al. 2003), will be able to link the embedded cloud model into the global aerosol fields; such efforts are currently underway. However, it remains a challenge both to further improve the representation of sub-grid variability in the large-scale, conventional GCMs, as well as improving the detailed treatment of ice microphysical processes, including nucleation, in models of all scales.

3.8 Education and Outreach

The ICE-T field campaign will provide an excellent education opportunity for a broad range of future scientists and to provide an opportunity for the public to view the C130. On two prearranged days, the C130 will have a hard down day for the airport to reside at the St. Croix airport. On a second day, the C130 will spend most of the day at San Juan Airport for students and the public to view the aircraft.

Graduate students: Over 10 universities are expected to participate in the ICE-T field campaign, introducing significant numbers of graduate students to field work, including mission planning and execution, and data quality control and analysis. Time spent at the operations center talking with the ICE-T PIs can be quite valuable to students: observing the data collection strategies makes later analysis of the data far easier, and such an opportunity also helps the students build networks in and outside their fields, and potentially gain new mentors that can serve them far later into their careers. The data collected during ICE-T will also assist the laboratory and numerical modeling efforts of additional graduate students at the participating institutions, as they conduct research on ice processes while progressing toward the attainment of their degrees.

Strong collaboration with an ongoing NSF-funded project that is studying the effects of dust on precipitation over Puerto Rico is anticipated (see letter from Profs. Mayol-Bracero and co-I Prather in Appendix). It is anticipated that graduate and undergraduate students associated with this program will be involved either directly or indirectly in ICE-T.

Undergraduate students: The University of the Virgin Islands- St. Croix (UVI) is a participant in the White House Initiative on Historically Black Colleges and Universities. The proposed timing of the project (June/July) coincides with the summer break at UVI. This institution conducts an Emerging Caribbean Scientists (ECS) Program that hosts multiple opportunities for summer scientific research for their undergraduates. Two particular events under the ECS program in which ICE-T might participate include SURE (Summer Undergraduate Research Experience—a quality REU program) and the SSRI (Summer Sophomore Research Institute—a program to provide research opportunities for sophomores and rising juniors, to foster confidence in the academic abilities, and foster skills in science). If ICE-T is approved, we will contact appropriate personnel at UVI to participate in the mentoring of some of these students during the project, and for the rest of the summer (remotely).

4. Flight Patterns

Flight planning will be conducted on a daily basis, aided by forecast and real-time radar products from the NWS in Puerto Rico, which were valuable for flight planning during the RICO project. Real-time radar imagery that covers the St. Croix area is available (http://radar.weather.gov/radar.php?rid=jua). Upper-air soundings are available from San Juan as well (station number 78526). Satellite imagery is available in real-time. Dropsondes will provide periodic vertical profiles of the thermodynamics and windfields.

A set of flight patterns was developed by the investigators to address the science goals outlined in Section 3.1. There are several other issues that were considered in the design of these patterns, including:

1. The transient nature of the updrafts of tropical maritime cumulus clouds.
2. Potential for APIP production (*aircraft-produced ice particles)*
3. Safety considerations, including aircraft icing and turbulence penetration.

Periodic outbreaks of dust can be expected in the study area during the ICE-T timeframe (Sections 2 and 3). As shown in Fig 3.10, 50% of the days during July might be expected to have enhanced concentrations of dust relative to the background dust levels during the winter at Barbados, not far from St. Croix. Relatively high concentrations, above 50 ug/m3, might be present 20% of the time, and with transits to dust-enhanced areas yielding a greater percentage of cases. Dust outbreaks can be tracked from satellite imagery and from models run by the Navy Postgraduate School in Monterey (see Sections 2 and 3). With lidar and aircraft profiling from near the surface to mid-levels, the flights will characterize the vertical distribution of dust layers. Cloud sampling will be conducted using the patterns described in this section. The presence and amount of dust in cloud particles will be derived from several instruments, including the CVI and mass spectrometers and the activity of the dust as ice nuclei from the CFDC.

4.1 Clear Air Sampling

Routine measurements will be made below the bases of cumulus clouds to determine the region where the updrafts are active and to characterize the inflow thermodynamic conditions, as well as the properties of the aerosols, cloud-active nuclei and trace gases flowing into the clouds. This sub-cloud base sampling will be made during the transit to the cloud sampling area and on the return to the C130's base airport. Aircraft soundings in the clear air in the vicinity of the clouds will also be conducted to characterize the local thermodynamic environment.

4.2 Stair-step Patterns

Two patterns are designed to provide data on the temporal evolution of the ice phase in developing cumulus.

1. Quasi-Lagrangian Ascent

An ideal experiment will be to sample an isolated growing region of a cloud, so that the history of the upper cloud regions could be documented as the rising top encounters the 0°C temperature level and above (Fig. 4.1). Sampling near the ocean surface is conducted during climbout and descent and at other times as needed. The C130 first penetrates cloud 100 m above the base to characterize the aerosol, then climbs to the 0°C level and looks for a target cloud either visually and/or with the aid of the downward looking WCR Doppler measurements. Details of the penetrations include:

- A penetration 100 m above cloud base is made upon entry into the area of cloud sampling to characterize the aerosol (from the CVI residual particles).
- The second cloud penetration is made in a developing cloud at about the 0°C (4.5 km) level to document dynamical and thermodynamic characteristics of the updraft, as well as particle size distributions produced by the warm rain process.
- The growing cloud top is subsequently sampled during its ascent to document the appearance of the first ice.
- The flight strategy should be oriented relative to the direction of shear whenever significant shear is present, where shear is defined here as vertical shear of the horizontal wind. When shear is present (visually and/or in soundings), then the strategy will be to make the first penetration perpendicular to the shear, and subsequent passes up-shear and down-shear. This provides a measurement of the shear-induced structure by using both in-situ and remote sensing.
- The specific climb rates of the C130 for the various horizontal legs can be determined following the first transit through the cloud. (Typical vertical velocities in developing maritime cumulus are of order 4 m/s or less, G. Heymsfield et al. 2009).
- The cloud penetrations should be done as expeditiously as possible, in order to minimize cloud evolution.
- CO and SP2 measurements can identify whether the C130 is re-intercepting its own exhaust in the subsequent cloud penetrations. Microphysics data will identify re-interceptions are associated with ice crystals produced by the C130 (APIPs).

The details of the cloud environment can be documented by leveling off or holding a heading for a few minutes after exiting cloud to improve the wind data. Although some aerosol information is obtained in this way, dedicated aerosol sampling (as well as documentation of the thermodynamic conditions and winds in the cloud environment) can be conducted at several altitudes during a descending spiral, leveling off at various altitudes for aerosol sampling, following the cloud sampling or on the return ferry back to aircraft base.

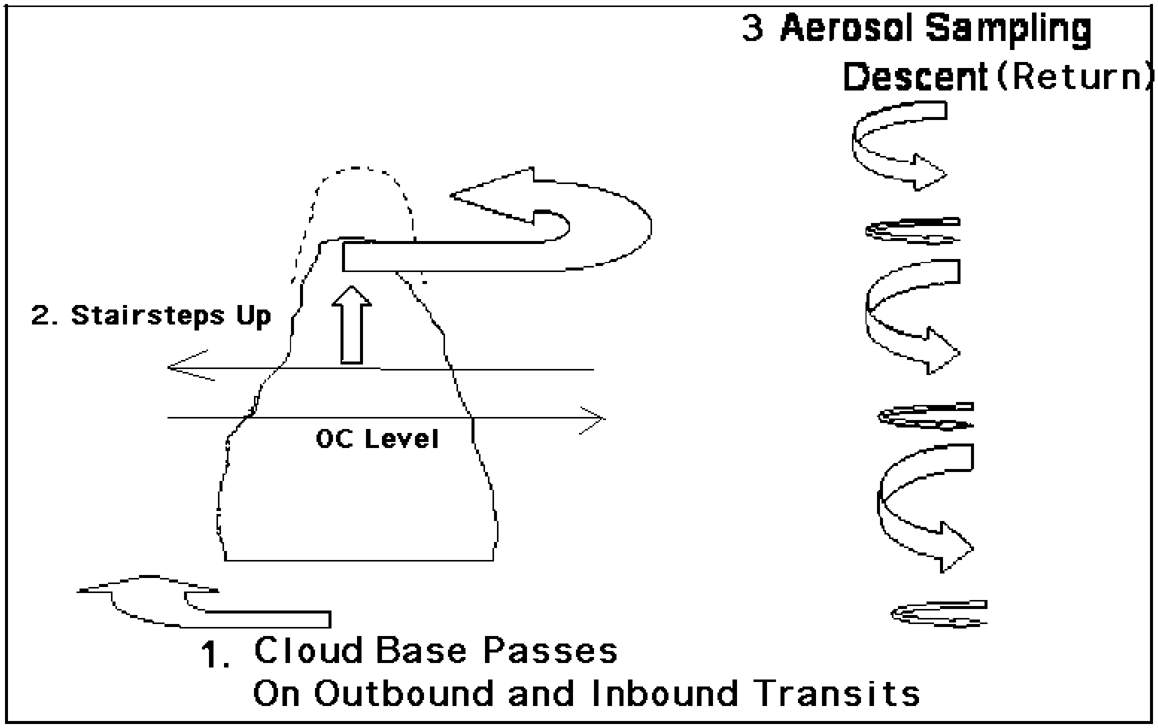


Figure 4.1. Single cloud or cloud turret flight sampling plan for tropical maritime cumulus clouds.

2. Descending Pattern

This pattern is designed to characterize primarily the tops of individual or several targeted maritime cumulus clouds, and to minimize the potential impact of APIPS on the dataset. If the updrafts remain active, the C130 will descend through cloud in a stair-stepped pattern.

First, the C130 makes a penetration through a cluster of cumulus clouds at a temperature near 0°C (4.5 km) and/or to avoid APIP production, climbs to a height from 2000 to 5000 feet above the tops of a developing, candidate cloud or cluster of clouds (Fig. 4.2). An over flight provides a radar and lidar overview at the beginning of a cloud sampling sequence. This remote sensing information indicates the stage of development of the liquid phase, vertical velocity structure, and the location and status of ice phase development.

Thereafter, the tops of candidate cumulus clouds will be penetrated, with preference given to clouds that have a dome-like top, indicating that the updrafts are likely to be active. With wind shear near cloud tops, cloud is carried down-shear. If the sheared portion is in the temperature range of interest, the evolution of the droplet to ice transition can be characterized in passes along the direction of the shear vector. This pattern might give us information on secondary ice production—the Hallett-Mossop process, for example.

Following the sampling near cloud top, profiling will be conducted primarily from cloud top downwards (to reduce the chance of sampling APIPS in the updrafts). Penetrations are made as rapidly as possible. Our expectation is that most maritime cumulus clouds that with maximum tops at about the -10°C level are not likely to present any flight hazards from airframe icing, turbulence, or lightning. (Vertical velocities are weak, see G. Heymsfield et al., 2009. Many studies have shown that lighting production is minimal in maritime cumulus). Data for the CVI and CFDC instruments will be obtained during extended horizontal legs during transits from cloud to cloud and also during levels runs in the boundary layer as noted earlier.

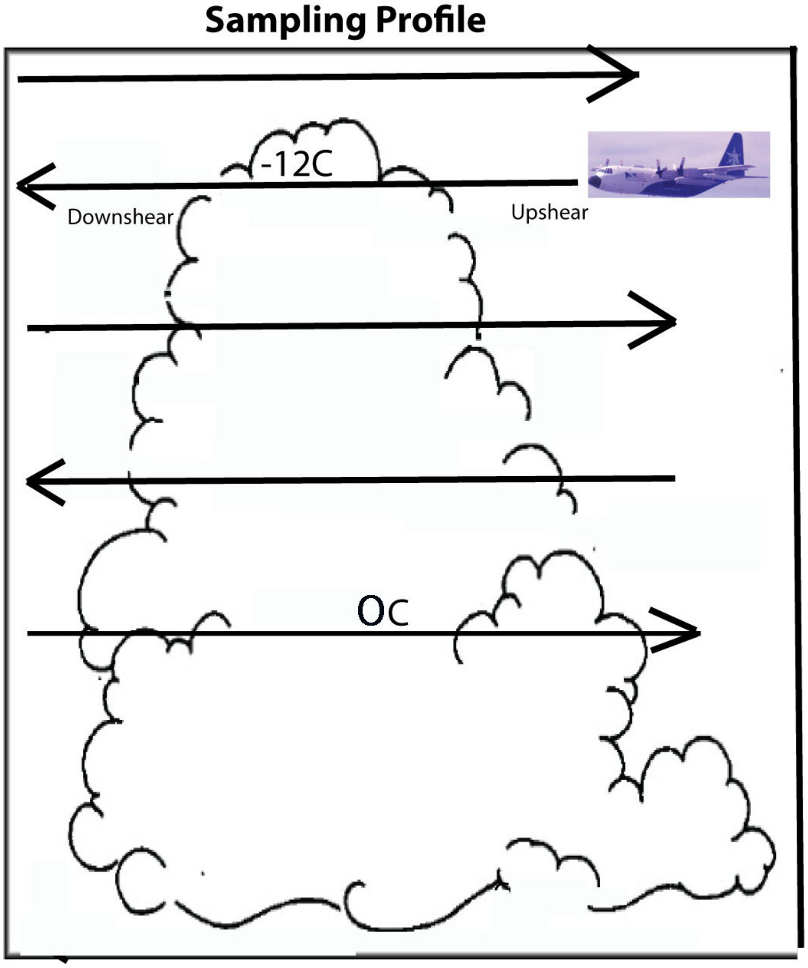


Figure 4.2 Stair-stepped descending flight pattern

4.3 Statistical Sampling

When the numbers of clouds reaching cloud top temperatures of -10°C are plentiful, statistical sampling will be conducted, similar to the RICO field campaign (Rauber et al. 2007). Flight tracks are proposed wherein penetrations are made through the upper parts of the cumulus clouds, without regard to their stage of development (Fig. 4.3). This type of pattern will provide a large sample of data from which we can characterize ice concentrations as a function of cloud top temperature, and when dust is present, can be used to determine how dust affects these concentrations. APIP production will not be of concern because re-penetration of the same cloud is unlikely.

This sampling approach requires a field of numerous cumulus clouds that can be penetrated in turn to provide a statistical view of an ensemble of cumulus. Assuming cloud top temperatures of ~ -12°C, the general procedure is:

1. Transit to operating area

2. Profile descent to below cloud to provide an environmental profile.

3. Fly straight and level legs 100 m above the ocean surface to characterize below-cloud aerosol and chemistry

4. Fly at least one leg through cloud at 100 m above cloud base to characterize the cloud droplet spectrum and to characterize the properties of the aerosols as derived from the CVI residuals. The height of the cloud base, the aerosol concentration just below base, and the cloud spectra just above the base are critical parameters for understanding the microphysics of these clouds and the role of dust.

5. Ascend to -10°C level and perform series of runs through different cumulus clouds.

6. Descend to -5°C level and perform a series of runs through different cumulus clouds.

7. If cloud top is well defined then characterize aerosol and chemistry just above. Perform downward looking remote sensing runs.

If sampling is conducted along the direction of wind shear, we may be able to characterize secondary ice production and how it is influenced through the role of dust.

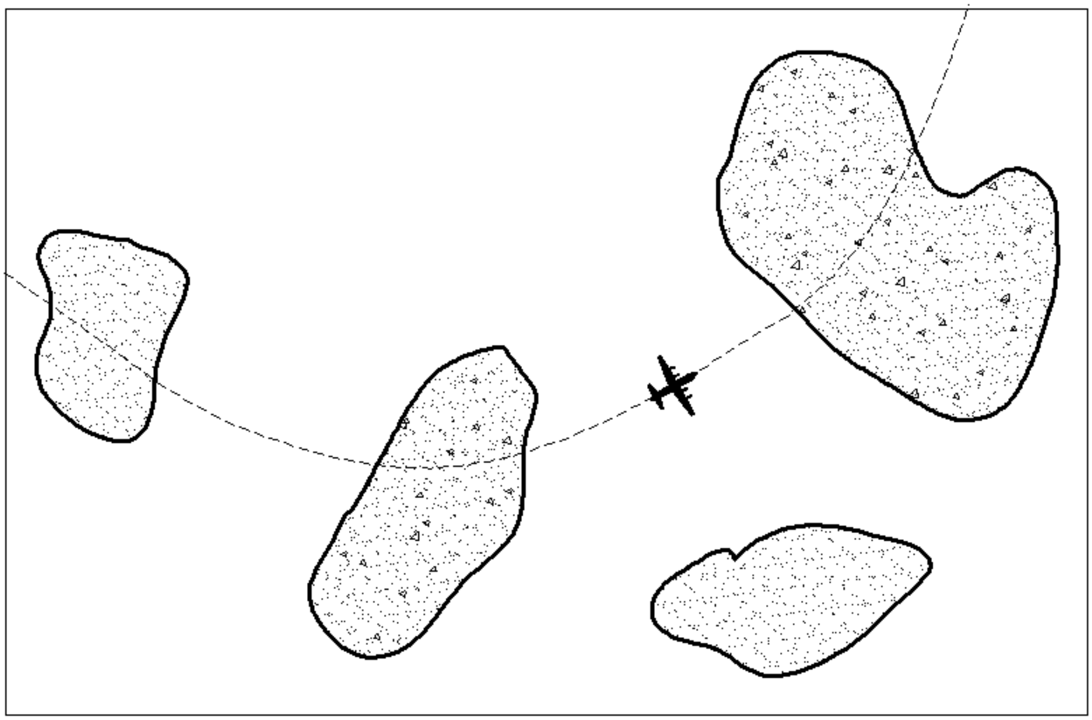


Figure 4.3 Statistical sampling flight pattern

4.4 APIPs

One of our goals is to determine whether glaciation of warm-topped cumulus clouds found in the Mossop et al. (1968) study was due to natural causes, e. g., the Hallet-Mossop effect, or whether it may have been due to APIPS. The study of Woodley et. al (2003) provides some insight into when the C130 will produce APIPS. Those researchers found that the onset of APIP production decreased with the number of propeller blades, for a given engine power and blade area. With 3000 kw of power per engine, the C130 has more power than any of the airplanes studied by Woodley et al., but the four propeller blades to distribute the power reduces the onset temperature of APIP production. Currently (2009), the C130 has 4-bladed propellers, and the prop disk diameter is 4.1 m. In comparison with the aircraft in Woodley's experiment, the prop disk area is very large, so that expansion and adiabatic cooling behind the propeller blades is further mitigated. NCAR may upgrade the propellers to 6-blade versions, which are more efficient and quieter. Our expectation is that this modification will further reduce the likelihood of APIP generation. Flying the C130 at relatively low power through the target clouds will further mitigate the chances of APIP production. Thus, the C130 is unlikely to produce APIPS at temperatures warmer than -10°C (see Woodley article).

Nonetheless, we propose a flight pattern whereby the C130 penetrates a cumulus cloud, turns 90 degrees and then tries to cross the initial track (Fig. A5). A number of instruments onboard the C130, including the Soot Photometer (SP2), condensation nuclei concentrations, and chemistry (CO), should be good tracers of prior penetration by the C-130. If APIPs are identified, we will evaluate the spread of the ice with repeated penetrations of the cloud at different heights or out-of-cloud with radar and lidar.

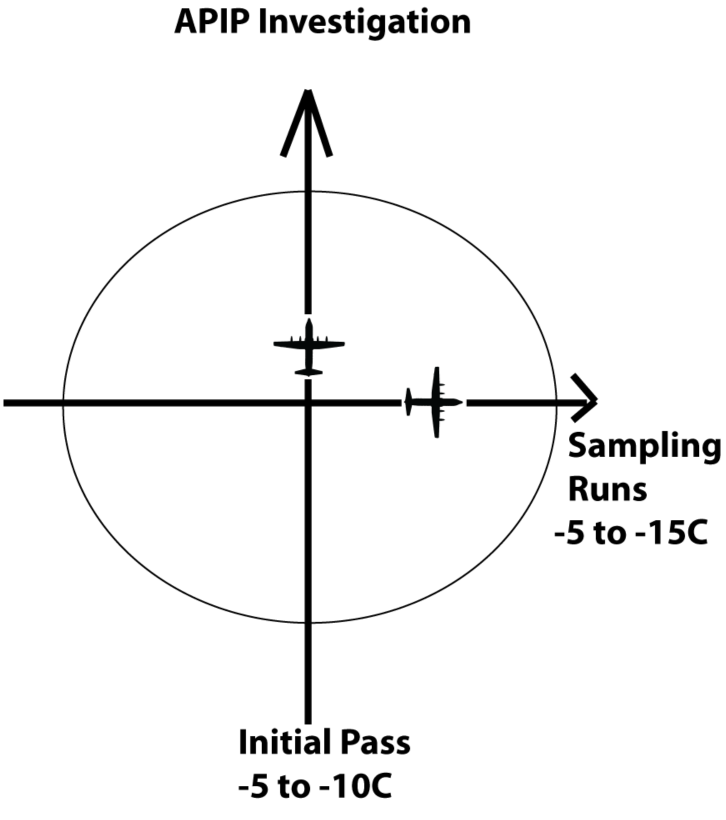


Figure 4.4 Flight pattern designed to identify APIP production in the -5 to -10°C temperature range.

5. References

Ackerman, Thomas P., Liou, Kuo-Nan, Valero, Francisco P.J., Pfister, Leonhard. 1988: Heating Rates in Tropical Anvils. *J. Atmos. Sci.*, **45**, 1606 – 1628

Ackerman, TP, 1988: Cirrus microphysics and infrared radiative transfer: a case study. *Annalen der Meteorologie, Offenbach a/M.*, n.s., 1(25), 133-134.

Ansmann, A., I. Mattis, D. Müller, U. Wandinger, M. Radlach, and D. Althausen, 2005: Ice formation in Saharan dust over central Europe observed with temperature/humidity/aerosol Raman lidar. *J. Geophys. Res.*, **110**, D18S12, doi:10.1029/2004JD005000.

Ansmann, A. , M. Tesche, D. Althausen, D. Müller, P. Seifert, V. Freudenthaler, B. Heese, M. Wiegner, G. Pisani, P. Knippertz, and O. Dubovik (2008), Influence of Saharan dust on cloud glaciation in southern Morocco during the Saharan Mineral Dust Experiment, *J. Geophys. Res.*, **113**, D04210, doi:10.1029/2007JD008785.

Arakawa, Akio. 2004: The cumulus parameterization problem: Past, present, and future. *J. Climate*, **17**, 2493–2525.

Archuleta, C.M., P.J. DeMott, S.M. Kreidenweis, 2005: Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures. *Atmospheric Chemistry and Physics Discussions*, 5, 3391-3436.

Baker, B., 1991a: On the role of phoresis in cloud ice initiation. J. Atmos. Sci., 48, 1545-1548.

Baker, B., 1991b: On the nucleation of ice in highly supersaturated regions of clouds. *J. Atmos. Sci.*, **48**, 1904-1907.

Baker, B.A., and R.P. Lawson, 2005: In situ observations of the microphysical properties of wave, cirrus and anvil clouds. Part 1: Wave clouds. *J. Atmos. Sci.*, (in press).

Barahona, D. and A. Nenes, 2009: Parameterizing the competition between homogeneous and heterogeneous freezing in cirrus cloud formation – monodisperse ice nuclei, *Atmos. Chem. Phys.,* **9**, 369–381.

Beard, K., 1992: Ice Initiation in warm-base convective clouds: An assessment of microphysical mechanisms. *Atmos. Res.*, **28**, 125-152.

Bergeron, T., 1935: On the physics of cloud and precipitation. *Proc. 5th Assembly U.G.G.I. Lisbon,* 2, 156.

Bigg, E.K, 1976: Size distributions of stratospheric aerosols and their variations with altitude and time. *J.*

Bigg, E. K. (1990), Measurement of concentrations of natural ice nuclei. *Atmos. Res.*, **25**, 397-408.

Bigg, E. K. and C. Leck (2001). Cloud-active particles over the central Arctic Ocean. *J. Geophys. Res.*, **106**, 32,155-32,166.

Bowers, R. M., C. L. Lauber, C. Wiedinmyer, M. Hamady, A. G. Hallar, R. Fall, R. Knight, and N. Fierer (2009), Characterization of Airborne Microbial Communities at a High-Elevation Site and Their Potential To Act as Atmospheric Ice Nuclei, Appl. Environ. Microbiol., 75, 5121–5130

Blyth, A.M. and J. Latham, 1993: Development of ice and precipitation in New Mexican summertime cumulus clouds. *Q. Jl. Roy. Meteor. Soc.*, **119**, 91-120.

Cantrell, W., and A.J. Heymsfield, 2005: Production of ice in tropospheric clouds: A review. *Bull. Amer. Meteor. Soc.*, **86**, 795-807.

Carrio, G.G., H. Jiang, and W.R. Cotton, 2005: Impact of aerosol intrusions on the Arctic boundary layer and on sea-ice multing rates.Part I: May 4, 1998 case. *J. Atmos. Sci.*, **62**, 3082–3093.

Carrio, G.G., S.C. van den Heever, and W.R. Cotton, 2007: Impacts of nucleating aerosol on anvil-cirrus clouds: A modeling study. *Atmos. Res.*, 111-131.

Chen, Y., S.M. Kreidenweis, L.M. McInnes, D.C. Rogers and P.J. DeMott, 1998: Single particle analyses of ice nucleating particles in the upper troposphere and lower stratosphere, *Geophys. Res. Lett.*, **25**, 1391-1394.

Chisnell, R.F and J. Latham, 1976: Ice particle multiplication in cumulus clouds. *Q. Jl. Roy. Meteor. Soc.*, **102**, 133-156.

Christner, B., C. E. Morris, C. M. Foreman, C. Rongman, and D. C. Sands, 2008a: Ubiquity of biological ice nucleators in snowfall. Science, 319, 1214-1220.

Christner, B., R. Cai, C. E. Morris, K. S. McCarter, C. M. Foremand, M. L. Skidmore, S. N. Montross, and D. C. Sands (2008b), Geographic, seasonal, and precipitation chemistry influence on the abundance and activity of biological ice nucleators in rain and snow, Proc. Natnl. Acad. Sci., 105, No. 48, 18854–18859.

Colle, B. A., J. B., Wolfe, W. J. Steenburgh, D. E. Kingsmill, J. A. Cox., and J. C. Shafer, 2005: High resolution simulations and microphysical validation of an orographic precipitation event over the Wasatch Mountains during IPEX IOP3. Submitted to *Mon. Wea. Rev.*

Connolly, P.J., T.W. Choularton, M.W. Gallagher, K.N. Bower, M.J. Flynn, and J.W. Whiteway, 2004: Observations and modeling of cirrus outflow from HECTOR during the EMERALD-II campaign. *Proc. 14th Intl. Conf. Clouds and Precip.*, Bologna, WMO, 747-750.

Cooper, W.A., 1991: Research in cloud and precipitation physics: Review of U.S. theoretical and observational studies, 1987-1990. *Rev. Geophys. Suppl.*, 69-79.

Cooper, W.A., 1995: Ice formation in wave clouds: Observed enhancement during evaporation. *Proc. Conf. Cloud Physics*, Dallas, Amer. Meteor. Soc., 147-152.

Cooper, W.A. and C.P.R. Saunders, 1980: Winter storms over the San Juan Mountains. Part II: Microphysical processes. *J. Appl. Meteor.*, **19**, 927-941.

Cooper, W. A., 1986: Ice initiation in natural clouds. Precipitation Enhancement-A scientific challenge. *Meteor. Monogr.*, **21**, 29-32.

Cotton, W.R., 1972a: Numerical simulation of precipitation development in supercooled cumuli—Part I. *Monthly Weather Review*, **100**, 757–763.

Cotton, W.R., 1972b: Numerical simulation of precipitation development in supercooled cumuli—Part II. *Monthly Weather Review*, **100**, 764–784.

Cotton, R.J. and Field P.R., 2002: Ice nucleation characteristics of an isolated wave cloud. *Q. J. Roy. Meteor. Soc.*, **128**, 2417-2437.

Cotton, W.R., R.A. Pielke, Sr., R.L. Walko, G.E. Liston, C.J. Tremback, H. Jiang, R.L. McAnelly, J.Y. Harrington, M.E. Nicholls, G.G. Carrio, J.P. McFadden, 2003: RAMS 2001: Current status and future directions. *Meteor. Atmos. Phys*., **82**, 5-29.

Crutzen, P.J., M.G. Lawrence, U. Poeschl, 1999: On the background photochemistry of tropospheric ozone. *Tellus*. **51A-B**, 123-146.

Cziczo, D.J., D.M. Murphy, P.K. Hudson, and D.S. Thomson, 2004: Single particle measurements of the chemical composition of cirrus ice residue during CRYSTAL-FACE, *J. Geophys. Res.-Atmos.*, ***109*** (D4), D04201.

DeMott P. J., K. Sassen, M. R. Poellot, D. Baumgardner, D. C. Rogers, S. D. Brooks, A. J. Prenni, S. M. Kreidenweis, 2003a: African dust aerosols as atmospheric ice nuclei, *Geophys. Res. Lett*., **30**, 1732, doi:10.1029/2003GL017410.

DeMott, P.J., D.J. Cziczo, A.J. Prenni, D.M Murphy, S.M. Kreidenweis, D.S. Thomson, R. Borys and D.C Rogers, 2003b: Measurements of the concentration and composition of nuclei for cirrus formation. *Proc. Nat'l Acad. Sci*., **100**, 14655-14660.

DeMott et al., 2008: The fourth international ice nucleation workshop (ICIS-2007): Objectives and preliminary results, *Proc. 15th International Conference on Clouds and Precipitation*, Cancun, Mexico (on CD-ROM).

DeMott, P.J., A. J. Prenni, X. Liu, M. D. Petters, C H. Twohy, M. S. Richardson, T. Eidhammer, S. M. Kreidenweis, and D. C. Rogers, 2009: Predicting global atmospheric ice nuclei distributions and their impacts on climate, Submitted to Proc. Natnl. Acad. Sci.

Durant, A. J. and R. A. Shaw, 2005: Evaporation freezing by contact nucleation inside-out. *Geophys. Res. Lett.*

Eidhammer and coauthors, 2009: Ice initiation by aerosol particles: Measured and predicted ice nuclei concentrations versus measured ice crystal concentrations in an orographic wave cloud. J. Atmos. Sci., ICE-L Special Issue, under revision.

Fan, j., M. Ovtchinnikov, J. M. Comstock, S. A. McFarlane, and A. Khain (2009), Ice formation in Arctic mixed-phase clouds: Insights from a 3-D cloud-resolving model with size-resolved aerosol and cloud microphysics. J. Geophys. Res. 114, D04205, doi:10.1029/2008JD010782

Field, P.R., RJ Cotton, K Noone, P Glantz, PH Kaye, E Hirst, RS Greenaway, C Jost, R Gabriel, T Reiner, M Andreae, CPR Saunders, A Archer, T Choularton, 2001: Ice nucleation in orographic wave clouds: Measurements made during INTACC. *Q. J. Roy. Meteor. Soc*., **127**, 575, A, 1493-1512.

Field, P.R., R.J. Hogan, P.R.A Brown, A.J. Illingworth, T.W. Choularton, P.H. Kaye, E. Hirst and R. Greenaway, 2004: Simultaneous radar and aircraft observations of mixed-phase cloud at the 100-m scale. *Q. J. Roy. Meteor. Soc.*, **130** (600) 1877-1904.

Field, P. R.,R. Wood, P. R. A. Brown, P. H. Kaye, E. Hirst, R. Greenaway, and J. A. Smith, 2003: Ice particle interarrival times measured with a fast FSSP. *J. Atmos. Ocean. Tech.*, **20**, 249-261.

Field, P. R., A. J. Heymsfield, and A. Bansemer, 2006: Shattering and particle interarrival times measured by optical array probes in ice clouds. *J. Atmos. Ocean. Tech.*, **23**, 1357–1371.

Findeisen, W., 1938: Die kolloidmeteorologischen Vorgange bei der Niederschlagsbildung (*Colloidal meteorological processes inthe formation of precipitation). Met. Z.,* 55, 121.

Fletcher, N. H. 1962: *The physics of rain clouds*. Cambridge University Press, Cambridge, UK

Fowler L.D., and D. A. Randall, 1996: Liquid and ice cloud microphysics in the CSU general circulation model .2. Impact on cloudiness, the earth's radiation budget, and the general circulation of the atmosphere. *J. Climate*, 9, 530-560.

Fowler, Laura D., D. A. Randall. 2002: Interactions between Cloud Microphysics and Cumulus Convection in a General Circulation Model. *J. Atmos. Sci.*, **59**, 3074–3098

Fridlind, A. M., A. S. Ackerman, G. McFarquhar, G. Zhang, M. R. Poellot, P. J. DeMott, A. J. Prenni, and A. J. Heymsfield (2007), Ice properties of single-layer stratocumulus during the Mixed-Phase Arctic Cloud Experiment: 2. Model results, *J. Geophys. Res.*, **112**, D24202, doi:10.1029/2007JD008646.

Gabriel, K.R. and D. Rosenfeld, 1990: The Second Israeli Stimulation Experiment: Analysis of precipitation on both targets. *J. Appl. Meteor.*, **29**, 1055-1067.

Gagin, A., 1965: Ice nuclei, their physical characteristics and possible effect on precipitation initiation. *Proc. Intl. Conf. Cloud Physics*, Tokyo-Sapporo, p.155

Garvert, M. F., B. A. Colle, and C. F. Mass, 2005: Synoptic and mesoscale evolution of the 13-14 December 2001 IMPROVE II storm system and comparison with a mesoscale model simulation. *J. Atmos. Sci.*, in press.

Gierens, K, U. Schumann, M. Helten, H., Smit, A. Marenco: 1999. A distribution law for relative humidity in the upper troposphere and lower stratosphere derived from three years of MOZAIC measurements. *Annales Geophysicae. Atmospheres, Hydrospheres and Space Sci.*, Berlin, Germany. 17, 1218-1226.

Gilmore, M.S., J.M. Straka, and E.N. Rasmussen, 2004: Precipitation uncertainty due to variations in precipitation particle parameters within a simple microphysics scheme. *Mon. Wea. Rev.*, **132**, 2610-2627.

Golaz, J.-C, V. E Larson and W. R. Cotton, 2002: A PDF-Based Model for Boundary Layer Clouds. Part I: Method and Model Description. *J. Atmos. Sci.*, **59**, 3540-3551. 2002

Gordon, G.L. and J.D. Marwitz, 1986: Hydrometeor evolution in rainbands over the California Valley. *J. Atmos. Sci.*, **43**, 1087-1100.

Grabowski, Wojciech W. 2003: Impact of cloud microphysics on convective-radiative quasi equilibrium revealed by cloud-resolving convection parameterization. *J. Climate*, **16**, 3463 - 3475.

Griggs, D. J., T. W. Choularton, 1983: Freezing modes of riming droplets with application to ice splinter production. *Q. Jl. Roy. Meteor. Soc.*, **109**, 243-253.

Gultepe, I., G. A. Isaac, and S. G. Cober, 2001: Ice crystal number concentration versus temperature. *International J. Climate*, **21**, 1281-1302

Harris-Hobbs, R. L., and W. A. Cooper, 1978: Field evidence supporting quantitative predictions of secondary ice production rates. *J. Atmos. Sci.*, **44**, 1071-1082.

Hallett, J. and Mossop, S.C., 1974: Production of secondary ice particles during the riming process. *Nature*, **249**, 26-28.

Hallett J., Sax R.I., Lamb D., Murty A.S.R., 1978. Aircraft measurements of ice in Florida Cumuli. *Q. Jl. Royal Meteor. Soc.*, **104**, 631-651

Harris-Hobbs, Raymond L., Cooper, William A. 1987: Field Evidence Supporting Quantitative Predictions of Secondary Ice Production Rates. *J. Atmos. Sci*, **44**, 1071–1082

Heintzenberg, J., K. Okada and J. Strom, 1996: On the composition of non-volatile material in upper tropospheric aerosols and cirrus crystals. *Atmos. Res.*, **41**, 81-88.

Heusel-Waltrop, WA, K. Diehl, SK Mitra, HR Pruppacher, 2003: A Laboratory and Theoretical Study on the Uptake of SO sub(2) Gas by Large and Small Water Drops Containing Heavy Metal Ions. *J. Atmos. Chem.*, **44**, 211-223.

Heymsfield, A.J., LM Miloshevich, C. Twohy, G Sachse, S. Oltmans, 1998: Upper-tropospheric relative humidity observations and implications for cirrus ice nucleation. *Geophys. Res. Lett.*, **25**, 1343-1346.

Heymsfield, A. J., P. R. Field and A. Bansemer, 2008: Exponential size distributions for snow. J.Atmos. Sci., 65, 4017–4031.

Heymsfield, A. J., Bansemer, G. Heymsfield, and A. O. Fierro, 2009: Microphysics of maritime tropical convective updrafts at temperatures from -20 to -60°C. *J. Atmos. Sci.*, in press.

Hobbs, P. V. and A. L. Rangno, 1985: Ice Particle Concentrations in Clouds. *J. Atmos. Sci.*, **42**, 2523-2549.

Hobbs, P.V. and A.L. Rangno, 1990: Rapid development of high ice particle concentrations in small polar maritime cumuliform clouds. *J Atmos. Sc*i., **47**, 2710-2722.

Huang and coauthors, 2008: Four-dimensional variational data assimilation for WRF: Formulation and preliminary results. *Mon. Wea. Rev.,* **137**, 299–314.

Hudson, J.G., 1989: An instantaneous CCN spectrometer. *J. Atmos. Ocean. Techn.*, 6, 1055-1065.

Hudson, J.G. and S. Noble, 2009: CCN and cloud droplet concentrations at a remote ocean site. *Geophys. Res. Let.,* **36,** L13812, doi:10.1029/2009GL038465.

Hudson, J. G., S. Noble, V. Jha, and S. Mishra, 2009a: Correlations of small cumuli droplet and drizzle drop concentrations with cloud condensation nuclei concentrations, *J. Geophys. Res.*, **114**, D05201, doi:10.1029/2008JD010581.

Hudson, J. G., S. Noble, and V. Jha, 2009b: Comparisons of CCN with supercooled clouds. Submitted to *J. Atmos. Sci.*

Huebert, B.J., T. Bates, P.B. Russell, G. Shi, Y. Kim, K. Kawamura, G. Carmichael, and T. Nakajima, 2003:An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts. *J. Geophys. Res.*, 108 (D23), 8633, doi:10.1029/2003JD003550

Husar RB, Tratt DM, Schichtel BA, et al., 2001: Asian dust events of April 1998. *J. Geophys. Res*.**, 106** (D16), 18317-18330.

Isono, K., M. Komabayasi, and A. Ono, 1959: The nature and origin of ice nuclei in the atmosphere, *J. Meteorol. Soc. Japan*, **37**, 211– 233.

Kanji, Z. A., and J. P. D. Abbatt (2006), Laboratory studies of ice formation via deposition mode nucleation onto mineral dust and n-hexane soot samples, *J. Geophys. Res*., **111**, D16204, doi:10.1029/2005JD006766.

Kärcher, B. and U. Lohmann, 2002: A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols,. *J. Geophys. Res.* **107**, doi: 10.1029/2001JD000470.

Kärcher, B. and U. Lohmann, 2003: A parameterization of cirrus cloud formation: Heterogeneous freezing. *J. Geophys. Res.* **108**, doi: 10.1029/2002JD003220.

Kassander, A.R., L.L. Sims and J.E. McDonald, 1957: Observations of freezing nuclei over the southwestern U.S., In: *Artificial Stimulation of Rain*. Pergamon, New York, 392-403.

Khain, A., M. Ovtchinnikov, M. Pinsky, A. Pokrovsky, H. Krugliak, 2000: Notes on the state-of-the-art numerical modeling of cloud microsphysics. *Review Atmospheric Research*, Amsterdam, The Netherlands. Vol. 55, no. 3-4, pp. 159-224.

Khvorostyanov, V. I., and J. A. Curry, 2004: The theory of ice nucleation by heterogeneous freezing of deliquescent mixed CCN. Part I: Critical radius, energy, and nucleation rate. *J. Atmos. Sci*., **61**, 2676-2691.

Knopf, D. A., and T. Koop, 2006: Heterogeneous nucleation of ice on surrogates of mineral dust, *J. Geophys. Res.*, **111** (D12), D12201.

Koenig, L. R., 1977: The rime-splintering hypothesis of cumulus glaciation examined using a field-of-flow cloud model. *Q. Jl. Roy. Meteor. Soc*., **103**, 585-606.

Koenig, L. R., and F. W. Murray, 1976: Ice-bearing cumulus cloud evolution: numerical simulation and general comparison against observations. *J. Appl. Meteor.*, **15**, 747–762.

Koehler, K. A., S. M. Kreidenweis, P. J. DeMott, A. J. Prenni, and M. D. Petters, 2007: Potential impact of Owens (dry) Lake dust on warm and cold cloud formation, J. Geophys. Res., 112, D12210, doi:10.1029/2007JD008413.

Korolev, A., and G. A. Isaac, 2005: Shattering during sampling by OAPs and HVPS. Part I: Snow particles. J. Atmos. Ocean. Tech., 22, 528–542.

Kumai, Motoi. 1951: Electron-microscope study of snow-crystal nuclei. *J. Atmos. Sci.*, **8**, 151–156.

Lamb, D., J. Hallett, and R.I. Sax, 1981: Mechanistic limitations to the release of latent heat during the natural and artificial glaciation of deep convective clouds. *Q. Jl. Roy. Meteor. Soc.*, **107**, 935-954.

Langer, G., G. Morgan, C.T. Nagamoto, M. Solak and J. Rosinski, 1979: Generation of ice nuclei in the surface outflow of thunderstorms in northeast Colorado. *J. Atmos. Sci.*, **36**, 2484-2494.

Lawson, R. P., B.A. Baker, C.G. Schmitt, and T.L. Jensen, 2001: An overview of microphysical properties of Arctic clouds observed in May and July 1998 during FIRE ACE. *J. Geophys. Res.*, 106 , D14 , 14989-15014.

Lin, R., D.O. Starr, P.J. DeMott, R. Cotton, K. Sassen, E. Jensen, B. Kaercher, X. Liu, 2002: Cirrus parcel model comparison project. Phase 1: The critical components to simulate cirrus initiation explicitly. *J. Atmos. Sci*., **59**, 2305-2329.

Liu, X. H., and J. E. Penner, 2005: Ice nucleation parameterization for global models, *Meteorol. Z.*, **14**(4), 499–514.

Liu, G. S., and J. A. Curry, 1999: Tropical ice water amount and its relations to other atmospheric hydrological parameters as inferred from satellite data. *J. Appl. Meteor.*, 38, 1182-1194.

Liu, H. , P.K. Wang and R.E. Schlesinger, 2003: A Numerical Study of Cirrus Clouds. Part II: Effects of Ambient Temperature, Stability, Radiation, Ice Microphysics, and Microdynamics on Cirrus Evolution. *J. Atmos. Sc*i., **60**, 1097-1119.

Liu, X., and J.E. Penner. 2005: Ice nucleation parameterization for global models. *Meteorologische Zeitschrift, **14,*** 499-514.

Liu, X., J. E. Penner, S. J. Ghan, and M. Wang, 2007: Inclusion of ice microphysics in the NCAR Community Atmosphere Model Version 3 (CAM3). *J. Climate*, **20**, 4526-4547.

Lohmann, U. 2002: Possible aerosol effects on ice clouds via contact nucleation, *J. Atmos. Sci.,* **59**, 647–656.

Lohmann, U., and E. Roeckner, 1996: Design and performance of a new cloud microphysics scheme developed for the ECHAM general circulation model. *Clim. Dyn*., **12**, 557-572.

Lohmann U., and B. Kärcher, 2002: First interactive simulations of cirrus clouds formed by homogeneous freezing in the ECHAM general circulation model. *J. Geophys. Res.-Atmospheres*, **107** (D10): Art. No. 4105

Lohmann, U., B. Kärcher, and J. Hendricks, 2004: Sensitivity studies of cirrus clouds formed by heterogeneous freezing in the ECHAM GCM. *J. Geophys. Res.*, **109,** doi: 10.1029/2003JD004443.

Lopez, R.E., R.F. Reinking, J. Hallett, and D. Rosenfeld, 1985: 5-cm radar echoes and their microphysical significance in Florida cumuli. *J. Geophys. Res.*, **90**, 10667-10673

Lord, S. J., and J. M. Lord, 1988: Vertical velocity structure in an axisymmetric, nonhydrostatic tropical cyclone model. *J. Atmos. Sci*., **45**, 1453--1461.

Martin, ST, HM Hung, A. Malinowski, 2001: Chemistry of cirrus cloud formation. *J. Aerosol Sci*., **32**, suppl. 1, pp. S925-S926.

Marwitz, J.D., 1987: Deep orographic storms over the Sierra Nevada. Part II: The precipitation process. *J. Atmos. Sci*., **44**, 174-185.

Mason, B., 1998: The production of high ice-crystal concentrations in stratiform clouds, *Q. Jl. Roy. Meteor. Soc*., **124**, 353-356.

McCumber, Michale, W. –K. Tao, J. Simpson, R. Penc, S. -T Soong, 1991: Comparison of Ice-Phase Microphysical Parameterization Schemes Using Numerical Simulations of Tropical Convection. *J. Appl. Meteor*., **30**, 985–1004.

Meyers, M.P., P. J. DeMott, W. R. Cotton, 1992: New Primary Ice-Nucleation Parameterizations in an Explicit Cloud Model. *J Appl. Meteor*, **31**, 708-721.

Meyers, M.P., R.L. Walko, J.Y. Harrington and W.R. Cotton, 1997: New RAMS cloud microphysics parameterization. Part II: The two-moment scheme. *Atmos. Res*., **45**, 3-39.

Möhler, O., and coauthors, 2005: Effect of sulfuric acid coating on heterogeneous ice nucleation by soot aerosol particles. *J. Geophy. Res*., **110**, D11210: 0148-0227.

Möhler, O., P.J. DeMott, G. Vali, and Z. Levin (2007), Microbiology and atmospheric processes: The role of biological particles in cloud physics, *Biogeosciences,* **4**, 1059–1071.

Morris, C. E., D. G. Georgakopoulos, and D. C. Sands, (2004), Ice nucleation active bacteria and their potential role in precipitation, J. Phys. IV, 121, 87–103.

Morrison, H., and A. Gettelman, 2008: A new two-moment bulk stratiform cloud microphysics scheme in the community atmosphere model, version 3 (CAM3), Part I: Description. *J. Climate*, **21**, 3642-3659.

Ming, Y., V. Ramaswamy, l. J. Donner, V. T. J. Phillips, S. A. Klein, P. A. Ginoux, and l. W. Horowitz, 2007: Modeling the interactions between aerosols and liquid water clouds with a self-consistent cloud scheme in a general circulation model. *J. Atmos. Sci.,* **64**, 1189-1209.

Mortazavi , R., C. T. Hayes and P. A. Ariya (2009), Ice nucleation activity of bacteria isolated from snow compared with organic and inorganic substrates, Environ. Chem., 5, 373–381.

Mossop, S. C., 1968: Comparison between concentration of ice crystals in cloud and the concentration of ice nuclei. *J. Rech. Atmos.,* **3**, 119-124.

Mossop, S. C., 1984: Comments on "Production of ice particles in clouds due to aircraft penetrations". *J. Appl. Meteor.*, **23**, 345–345.

Mossop, S.C., Ono, A. and Wishart, E.R., 1970: Ice particle in maritime near Tasmania. *Q. J. Roy. Meteorol. Soc.*, 25, 889-508

Mossop S.C. 1985: The origin and concentration of ice crystals in clouds. Bull. Amer. Meteorol. Soc., 66(3), 264-273.

Oishi, S., E. Nakakita, H. Itsuji, S. Ikebuchi, 1994: Numerical approach about the effect of updraft on local rainfall. *Eos*, **75**, no. 25, suppl., 29 p.

Oraltay, R.G. and J. Hallett, 1989: Evaporation and melting of ice crystals: A laboratory study. *Atmos. Res.*, **24**, 169-189.

Penner, J.E., R.J. Charlson, J.M. Hales, N.S. Laulainen, R. Lefier, T. Novakov, J. Ogren, L.F. Radke, S.E. Schwartz and L. Travis, 1994: Quantifying and minimizing uncertainty of climate forcing by anthropogenic aerosols. *Bull. Amer. Meteor. Soc.*, 75, 375-400.

Penner, J. E. et al. Aerosols, their Direct and Indirect Effects. in *Climate Change 2001: The Scientific Basis* (eds Houghton, J. T. et al.) Cambridge Univ. Press, 289-348.

Phillips, V.T.J., T. W. Choularton, A. J. Illingworth, R. J. Hogan, P. R. Field, 2003: Simulations of the glaciation of a frontal mixed-phase cloud with Explicit Microphysics Model, *Q. Jl. Roy. Meteor. Soc.*, 129, 1351-1371.

Phillips, V. T. J., P. J. DeMott, and C. Andronache, 2008: An empirical parameterization of heterogeneous ice nucleation for multiple chemical species of aerosol, J. Atmos. Sci., 65, 2757 – 2783, doi:10.1175/2007JAS2546.1.

Pratt, K.A., C.H. Twohy, S.M. Murphy, R.C. Moffet, A.J. Heymsfield, C.J. Gaston, P.J. DeMott, P.R. Field, D.C. Rogers, T.R. Henn, M.K. Gilles, J.H. Seinfeld and K.A. Prather. Observation of playa salts as nuclei of orographic wave clouds. Journal of Geophysical Research, submitted.

Pratt, K.A., S.M. Murphy, R. Subramanian, P.J. DeMott, G.L. Kok, D.C. Rogers, A.J. Heymsfield, J.H. Seinfeld and K.A. Prather. Flight-based chemical characterization of biomass burning aerosols within two prescribed burn smoke plumes. Atmospheric Chemistry & Physics. In Preparation.

Prenni, A.J., P.J. DeMott, C. Twohy, D.C. Rogers, S.D. Brooks, S.M. Kreidenweis, A.J. Heymsfield and M.R. Poellot, 2007: Examinations of ice formation processes in Florida cumuli using ice nuclei measurements of anvil ice crystal particle residues. *J. Geophys. Res.*, **112**, D10221, doi:10.1029/2006JD007549.

Prospero, J.M., 1996: Saharan dust transport over the North Atlantic Ocean and Mediterranean: an overview, in *The Impact of Desert Dust Across the Mediterranean*, eds. S. Guerzoni and R. Chester, Kluwer, Dordrecht.

Prospero, J. M., 1999: Longterm measurements of the transport of African mineral dust to the Southestern United States: Implications for regional air quality, *J. Geophys. Res.*, **104**, 15,917-15,927.

Prospero, J.M., and J.P. Lamb, 2003: African droughts and dust transport to the Caribbean: Climate change and implications, *Science*, **302,** 1024-1027.

Randall, David, M. Khairoutdinov, A. Arakawa, W. Grabowski, 2003: Breaking the Cloud Parameterization Deadlock. *Bull. Amer. Meteor. Soc***., 84**, 1547-1564.

Rangno, A. L. and P. V. Hobbs, 1983: Production of ice particles in clouds due to aircraft penetrations. *J. Cli. Appl. Meteor.*, **22**, 214–232.

Rangno, A. L and P. V. Hobbs, 1984: Further observations of the production of ice particles in clouds by aircraft. *J. Cli. Appl. Meteor.,* **23**, 985–987.

Rangno A.L. and P. V. Hobbs, 1991: Ice particle concentrations and precipitation development in small polar maritime cumuliform clouds. *Q. J. Roy. Meteorol. Soc.*, **117**, 207-241.

Rangno, A., and P. Hobbs, 1994: Ice particle concentrations and precipitation development in small continental cumuliform clouds. *Q. J. R. Meteorol. Soc.,* **120**, 573-601.

Rangno A.L. and Hobbs P.V. 2001: Ice particles in stratiform clouds in the Arctic and possible mechanisms for the production of high ice concentrations. *J. Geophys. Res.*, **106**(D14), 15065-15075.

Rasmussen, R.M., 1995: A review of theoretical and observational studies in cloud and precipitation physics: 1991-1994. *Rev. Geophys. Suppl.*, 795-809.

Rasmussen, R., M. Dixon, F. Hage, J. Cole, C. Wade, J. Tuttle, S. McGettigan, T. Carty, L. Stevenson, W. Fellner, S. Knight, E. Karplus, N. Rehak, 2001: Weather Support to Deicing Decision Making (WSDDM): A Winter Weather Nowcasting System. *Bull. Amer. Meteor. Soc***. 82**, 579-596.

Rasmussen, Roy, Politovich, Marcia, Marwitz, John, Sand, Wayne, McGinley, John, Smart, John, Pielke, Roger, Rutledge, Steve, Wesley, Doug, Stossmeister, Greg, Bernstein, Ben, Elmore, Kim, Powell, Nick, Westwater, Ed, Stankov, B. Boba, Burrows, Don. 1992: Winter Icing and Storms Project (WISP). *Bull. Amer. Meteor. Soc*. **73**, 951–976.

Rauber, R.M. and L.O. Grant, 1987: Characteristics of cloud ice and precipitation during wintertime storms over the mountains of northern Colorado. *J. Climate Appl. Meteor.*, 26(4), 488-524.

Roberts, G., and A. Nenes, 2005: A continuous-flow streamwise thermal-gradient CCN chamber for airborne measurements, *Aerosol Science and Technology,* **39**, 206-221.

Roberts, P. and J. Hallett. A laboratory study of the ice nucleating properties of some mineral particulates. *Q. Jl. Roy. Meteor. Soc*., **94**, 25-34, 1968.

Rogers, D.C., 1994: Detecting ice nuclei with a continuous flow diffusion chamber—some exploratory tests of instrument response. *J. Atmos. Ocean. Techn*., **11**, 1042-1047.

Rogers, D.C., P.J. DeMott, S.M. Kreidenweis and Y. Chen, 2001: A continuous-flow diffusion chamber for airborne measurements of ice nuclei, *J. Atmos. Oceanic Techn*., **18**, 725-741.

Rogers, D.C., P.J. DeMott and S.M. Kreidenweis, 2001: Airborne measurements of ice nucleating aerosol particles in the Arctic spring, *J. Geophys. Res.*, **106**, D14, 15053-15063.

Rosinski, J., P. L. Haaagenson, C. T. Nagamoto, and F. Parungo (1986), Ice forming nuclei of maritime origin, J Aerosol Sci, **17**, No. l, 23-46.

Rosinski, J., P. L. Haaagenson, C. T. Nagamoto, and F. Parungo (1987), Nature of ice forming nuclei in marine air masses, J Aerosol Sci, **18**, No. 3, 291-309..

Rosinski, J., 1995: Cloud condensation nuclei as a real source of ice forming nuclei in continental and marine air masses. *Atmos. Res.*, **38**, 351-359.

Saleeby, S.M. and Cotton, W.R. 2004: A large-droplet mode and prognostic number concentration of cloud droplets in the Colorado State University Regional Atmospheric Modeling System (RAMS). Part I: Module descriptions and supercell test simulations. *J. Appl. Meteor.*, 43 (1), 182-195.

Sassen, K. 2002: Indirect climate forcing over the western US from Asian dust storms. *Geophys. Res. Lett.,* **29**, 10.1029

Sassen, K., 2005: Dusty ice clouds over Alaska, Nature, 434, 456.

Sassen K., P. J. DeMott, J. M. Prospero, M. R. Poellot, 2003: Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results. *Geophys. Res. Lett*., 30 (12), 1633, doi:10.1029/2003GL017371

Sax, R. I., J. Thomas, M.Bonebrake, and J. Hallett, 1979: Ice evolution within seeded and nonseeded Florida cumuli. *J. Appl. Meteor.*, **18**, 203-214.

Sax, R.I., and V.W. Keller. 1980: Water-ice and water-updraft relationships near –10°C within populations of Florida cumuli. *J. Appl. Meteor*., **19**, 505–514.

Schiller, C, A. Afchine, N. Eicke, C. Feigl, H. Fischer, A. Giez, P. Konopka, H. Schalger, F. Tuetjer, FG Wienhold, M. Zoeger, 1999: Ice particle formation and sedimentation in the tropopause region: a case study based on in situ measurements of total water during POLSTAR 1997. *Geophys. Res. Lett*., 26, 14, 2219-2222.

Scott, B.C. and P.V. Hobbs, 1977: A theoretical study of the evolution of mixed-phase cumulus clouds. *J. Atmos. Sci.*, **34**, 812-826

Shaw, R. A., A. J. Durant, and Y. Mi, 2005: Heterogeneous surface crystallization observed in undercooled water. *J. Phys. Chem. B*, **109**, 9865-9868.

Simpson, Joanne, Brier, Glenn W., Simpson, R.H.. 1967: Stormfury Cumulus Seeding Experiment 1965: Statistical Analysis and Main Results. *J. Atmos. Sci.*, **24**, 508–521.

Smith, M.A., 2007: Evaluation of mesoscale simulations of dust sources, sinks and transport over the Middle East. M.S. Thesis, Colorado State University, 126 pp. [Available from Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523.]

Stith, J. L., D. A. Burrows, P. J. DeMott, 1994: Initiation of ice: comparison of numerical model results with observations of ice development in a cumulus cloud. *Atmos. Environ*. **32**, 13-30.

Stith, J. L., V. Ramanathan, W. A. Cooper, G. Roberts, P. J. DeMott, G. Carmichael, C.D. Hatch, B. Adhikary, C. H. Twohy, D. C. Rogers, D. Baumgardner, A. J. Prenni, T. Campos, R. S. Gao, J. Anderson, and Y. Feng, 2009: An overview of aircraft observations from the Pacific Dust Experiment campaign. J. Geophys. Res., 114, D05207, doi:10.1029/2008JD010924.

Stoelinga, Mark T., Hobbs, Peter V., Mass, Clifford F., Locatelli, John D., Colle, Brian A., Houze, Robert A., Rangno, Arthur L., Bond, Nicholas A., Smull, Bradley F., Rasmussen, Roy M., Thompson, Gregory, Colman, Bradley R. 2003: Improvement of Microphysical Parameterization through Observational Verification Experiment. *Bull. Amer. Meteor. Soc.*, **84**, 1807–1826.

Sun, A., H.-Y. Chun, J.-J. Baik, M. Yan, 2002: Influence of electrification on microphysical and dynamical processes in a numerically simulated thunderstorm. *J. Appl. Meteor.*, **41**, 1112-1127.

Szyrmer, W. and I. Zawadzki (1997), Biogenic and anthropogenic sources of ice-forming nuclei: A review, Bull. Amer. Meteor. Soc., **78**, 209–228.

Tao, W. -K., and J. Simpson, 1993: The Goddard Cumulus Ensemble Model. Part I: Model descripton. *Terr., Atmos. Oceanic Sci.*, **4**, 35-72.

Tao, W-K., 2003: Goddard Cumulus Ensemble (GCE) Model: Application for Understanding Precipitation Processes. *Meteorological Monographs*, 29, No. 51, pp. 107–107.

Targino, A.C., R. Krejci, K.J. Noone, and P. Glantz, 2005: Single particle analysis of ice crystal residuals observed in orographic wave clouds over Scandinavia during INTACC experiment, *Atmospheric Chemistry and Physics Discussion*, 5, 8055-8090,

Thompson, G., R. Rasmussen, B. Bernstein, 2002: Detailed comparisons of aircraft icing environments and Model-Predicted Microphysics. *Proc. 10th AMS Conf. Aviation, Range, Aerospace Meteorology*, 10.

Thompson, G., R.M. Rasmussen and K. Manning, 2004: Explicit forecasts of winter precipitation using an improved bulk microphysics scheme. Part I: Description and sensitivity analysis. *Monthly Weather Review*, **132**, 519-542.

Tinsley, B., and R. Heelis, 1993: Correlations of atmospheric dynamics with solar activity: Evidence for a connection via the solar wind, atmospheric electricity, and cloud microphysics. *J. Geophys. Res*., **98,** 10375-10384.

Tinsley, B.A., R.P. Rohrbaugh, and M. Hei, 2001: Electroscavenging in clouds with broad droplet size distributions and weak electrification. *Atmos. Res.*, **59-60**, 115-135.

Toon, O.B., C.P. Mckay, T.P. Ackerman, K. Santhanam, 1989: Rapid calculation of radiative heating rates and photodissociation rates in inhomogeneous multiple scattering atmospheres. *J. Geophys. Res.*, **94**, D13, 16287-16301.

Twohy, C. H., and M. R. Poellot (2005), Chemical characteristics of ice residual nuclei in anvil cirrus clouds:evidence for homogeneous and heterogeneous ice formation, Atmos. Chem. Phys. 5, 2289–2297.

Twohy, C. H., P. J. DeMott, K. A. Pratt, R. Subramanian, G. L. Kok, S. M. Murphy, T. Lersch, K. A. Prather, J. H. Seinfeld, A. J. Heymsfield and Z. Wang, 2009: Relationships of biomass burning aerosols to ice in orographic wave clouds. *J. Atmos. Sci.*. Submitted.

Twomey, S., 1977: The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.*, **34**, 1149-1152.

van den Heever, S.C., and W.R. Cotton, 2004: The impact of hail size on simulated supercell storms. *J. Atmos. Sci.*, 61, 1596-1609.

van den Heever, S.C., G.G. Carrio, W.R. Cotton, P.J. DeMott, and A.J. Prenni, 2006: Impacts of nucleating aerosol on Florida storms. Part I: Mesoscale simulations. *J. Atmos. Sci.*, 1752-1775.

Van Diedenhoven, B., A.M. Fridlind, A.S. Ackerman, E.W. Eloranta, and G.M. McFarquhar, 2009: An evaluation of ice formation in large-eddy simulations of supercooled Arctic stratocumulus using ground-based lidar and cloud radar. *J. Geophys. Res.*, **114**, D10203, doi:10.1029/2008JD011198.

Vardiman, L, 1978: The generation of secondary ice particles in clouds by crystal - crystal collision. *J. Atmos. Sci.*, **35**, 2168 - 2180.

Welti, A., F. Lüönd, O. Stetzer, and U. Lohmann (2009), Influence of particle size on the ice nucleating ability of mineral dusts, Atmos. Chem. Phys. Discuss., 9, 6929–6955.

Willoughby, H.E., Jorgensen, D.P., Black, R.A., Rosenthal, S.L.. 1985: Project STORMFURY: A Scientific Chronicle 1962–1983. *Bull. Amer. Meteor. Soc.*, **66**, 505–514

Woodley, W. L.and coathors, 2003: Aircraft-produced ice particles (APIPs): Additional results and further insights. *J. Appl. Meteor.*, **42**, 640–651.

Zuberi, B., A. K. Bertram, C. A. Cassa, L. T. Molina, and M. J. Molina, 2002: Heterogeneous nucleation of ice in (NH4)2SO4-H2O particles with mineral dust immersions, *Geophys. Res. Lett.*, **29**, doi:10.1029/2001GL014289

Zurovac-Jevti , Dan e, G. J. Zhang, 2003: Development and Test of a Cirrus Parameterization Scheme Using NCAR CCM3. *J. Atmos. Sci.*, **60**, 1325–1344.

6. Appendix – Letter of Support for Educational Outreach

1 March 2010

Andrew Heymsfield
Senior Scientist
MMM Division
NCAR
3450 Mitchell Lane
Boulder, Co 80301

Dear Dr. Heymsfield:

Collaboration with ICE-T would be a great opportunity for our research group since **it augments our recently funded NSF proposal** titled *"Impact of African Dust on Clouds and Precipitation in a Caribbean Tropical Montane Cloud Forest"*, whose CoPIs are Kimberly Prather (Scripps and UCSD) and Elizabeth Andrews (University of Colorado, Boulder). The ICE-T program would also benefit from the proximity of the University of Puerto Rico-Rio Piedras (UPR-RP) just 80 air miles from your St. Croix, USVI base for ICE-T. The Arecibo Observatory is close, with potentially useful LIDAR capacity. Having the C130 conduct some research flights in the vicinity of San Juan would support our proposed studies, strengthen both ICE-T and our research program in atmospheric chemistry, as well as provide students new opportunities for research.

As per our discussions, we envisage at least two students being directly involved in the field operations on St. Croix. I understand that Dr. Heymsfield has submitted a proposal to NCAR for funds for the students to visit NCAR and to participate in the flight operations in July, 2011. The interaction of these and other students with collaborators during any time the PI's could spend in San Juan following a mission (e.g., a mini-conference/science meeting) may inspire them to pursue careers as higher-level scientists within the field of aerosols, clouds, and climate studies. The PI noted that a prearranged day will be called when the C130 spends most of the data at the airport in San Juan to allow students and the public to view the aircraft.

Puerto Rico is an ideal location for coordinating some ICE-T flights with our NSF funded project, as this is feasible. The island has good exposure to the easterly trades, and is free of major land masses upwind, minimizing the effects of anthropogenic aerosol sources. Also, in Puerto Rico, our group has an aerosol sampling station (Cape San Juan, CSJ) at the northeast coast (18° 23' N, 65° 37' W) that operates since the 90s, and a more recently added cloud/rain/aerosol sampling station, downwind from CSJ, operating since 2004 (Pico del Este, PE, in the Luquillo Experimental Forest (18° 16' N, 65° 45' W). CSJ measurements are supported by NOAA ESRL, the station is part of the NASA AErosol RObotic NETwork (AERONET), and it recently became one of the regional stations of the Global Atmosphere Watch (GAW) program. PE's elevation (at 1051 m amsl) sits above the cloud condensation level thus facilitating the study of clouds without the need for aircraft and the related complexity and costs. Mean annual precipitation at PE is > 5000 mm/yr.

Both stations (CSJ and PE) have the major advantage of having a large university research center, UPR-RP, within an hours' drive. University staff, faculty, and facilities facilitate the research activities (e.g., minimizing concerns about sample handling and preservation, and providing highly reliable data). The close proximity of UPR-RP to the sampling site also permits a continuous sampling program that is essential to monitoring the variations in aerosol and cloud properties that occur due to seasonal changes.

The measurements that we have been performing in Puerto Rico together with the ones that will be performed as part of our recently funded project and the ones that you propose for ICE-T would definitely strengthen both of our projects. It would be great to be able to join efforts in these projects. We look forward to collaborating with you.

Best regards,

Olga L. Mayol-Bracero

Associate Professor and PI of NSF funded "Impact of

African Dust on Clouds and Precipitation in a Caribbean

Tropical Montane Cloud Forest"

Institute for Tropical Ecosystem Studies

University of Puerto Rico