

# The Ice in Clouds Experiment

- Research Plan -

# **Scientific Overview Document**

March 1, 2006



Contributing authors:

Andy Heymsfield<sup>1</sup>, Jeff Stith<sup>1</sup>, Dave Rogers<sup>1</sup>, Paul Field<sup>1</sup>, Paul DeMott<sup>2</sup>, Charles Knight<sup>1</sup>, Joyce Penner<sup>3</sup>, Ken Sassen<sup>4</sup>, Greg Thompson<sup>1</sup>, Bill Cotton<sup>2</sup>, Will Cantrell<sup>5</sup>, Sharon Lewis<sup>6</sup>, Gabor Vali<sup>7</sup>, Al Cooper<sup>1</sup>, Steve Platnick<sup>8</sup>, Jim Dye<sup>1</sup>, George Isaac<sup>9</sup>, Ulrike Lohmann<sup>10</sup>, Ottmar Möhler<sup>11</sup>, Axel Seifert<sup>12</sup>, Dan Cziczo<sup>10</sup>, Raymond Shaw<sup>5</sup>, Brad Baker<sup>13</sup>, Paul Lawson<sup>13</sup>

Affiliations: <sup>1</sup>NCAR, <sup>2</sup>CSU, <sup>3</sup>U. Michigan, <sup>4</sup>U Alaska, <sup>5</sup>Michigan Tech. U., <sup>6</sup>NOAA, <sup>7</sup>U. Wyoming, <sup>8</sup>NASA, <sup>9</sup>Meteorol. Service Canada, <sup>10</sup>ETH Switzerland, <sup>11</sup>Inst. Meteorol. Climate Res. Germany, <sup>12</sup>U. Karlsruhe Germany, <sup>13</sup>SPEC Inc.

1. Exe 2. Intr	cutive Summary	
2.1	Primary (Heterogeneous) Ice Formation	4
2.2	Secondary Ice Formation Processes	6
2.3	Dependence on Droplet Spectrum	7
2.4	Evaporation Effects on Ice Formation	8
2.5	Air Parcel History	10
2.6	Dust as Ice Nuclei	10
2.7	Climate Implications	12
2.8	Modeling Studies	12
2.9	Summary	14
3. Scie	ntific Direction	
3.1	Wave clouds	16
3.1.	<i>Climatology of wave clouds in Colorado and Wyoming</i>	17
3.2	Upslope Clouds	19
3.3	Cumulus clouds	20
3.4	New Instruments and Aircraft for observing ice formation	21
3.4.	<i>Observing ice concentrations and first ice formation</i>	22
3.4.	2 Airborne measurements of ice nuclei	23
3.5	Laboratory Studies	25
3.6	Numerical Modeling	26
3.7	Remote Sensing Studies	27
4. Research Facilities and Field Campaigns		
5. References		
6 1	Experiment Plans	40
0.1		40
0.1.	Layer and wave Clouds	40
6.1.2	2 Altostratus - altocumulus layers	41
6.1.	3 Convective Clouds	42

# 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.

This document outlines a plan for studies of ice processes in clouds. It includes field observations, laboratory experiments, and numerical modeling. Recent advances in observational tools, laboratory cloud simulation chambers, numerical models, and computer hardware are providing new 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 in selected clouds. We also seek improved quantitative understanding of the roles of thermodynamic pathway, location within the cloud, and temporal dependency.

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. We recognize that secondary ice formation processes occur in many clouds, subsequent to the formation of ice by nucleation. The present focus, however, is on heterogeneous nucleation in clouds where secondary processes do not occur or where they can be separated (in time or space) from the primary process.

The first step in this project is to seek cases with a strong aerosol-ice nucleation signal. It will focus on observational studies with high likelihood of showing a strong connection of aerosols to effect ice formation. These cases occur in geographic areas that experience alternatively dust events and dust-free background. The targets are layer clouds: lenticular wave clouds, nimbostratus, and extensive altocumulus and altostratus 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 provide a range of temperature, humidity, and vertical wind conditions in which first ice may form in a laboratory-like setting. Some of the conditions observed in wave clouds can be approximated in laboratory cloud chamber experiments for ice formation studies and for characterizing the performance of airborne ice nuclei instruments.

An especially intriguing and important feature of wave clouds is an "evaporation glaciation signature" that is often observed in wave clouds. Observations from previous field experiments indicate that high concentrations of ice particles are nucleated near the location where supercooled liquid water evaporates in wave clouds. However, in the previous studies, ice nuclei and detailed aerosol measurements were lacking, which leaves out a critical component to understanding the nucleation process.

Besides dust, other episodic aerosol events (forest fires or pollution) also provide opportunities for observing effects of different aerosol types on layer and convective clouds. Likewise, aerosol characteristics that are different in major ways could be targets of study in the future of this project (e.g., maritime air). Key observations and flight strategies are described in detail later in this document.

In order to make progress towards this goal the following objectives must be met

- 1. Establish which heterogeneous ice nucleation modes are active and important.
- 2. Identify ice nucleating aerosols and obtain quantitative measurements of them.
- 3. Predict ice concentrations in clouds by using numerical models.

Initially, ICE will use airborne measurements of clouds, concentrating on the role of heterogeneous nucleation, along with coordinating ground measurements in mountainous locations such as the Front Range of Colorado and Wyoming. Close collaboration between theory, field, lab, and modeling studies will be emphasized by involving participants who specialize in these different approaches.

# 2. Introduction

This document describes fundamental observational, laboratory, and modeling studies that address how ice forms in clouds, and it proposes a research plan to improve the current representations of ice formation in numerical models. Heterogeneous ice nucleation initiates the ice phase in most clouds at temperatures 0 to -35°C, but it is not known where the first ice particles are produced or which aerosol are favored for ice nucleation. The development of the ice phase is important for many applications but is poorly understood, because historic measurements in clouds have lacked the resolution to adequately sample early ice in clouds or the nuclei that influence its formation. Fortunately, new observational tools and simulation techniques are available to study ice formation mechanisms. After briefly reviewing the major issues related to ice formation in clouds, new opportunities for addressing this problem are presented.

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), water redistribution, and others.

Many mid-latitude clouds have extensive supercooled regions, lending themselves to copious ice production. Even deep tropical clouds, which have a strong warm rain process, produce a major fraction of their precipitation 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). We know that ice may appear as a result of drop freezing by immersion or contact nucleation by ice nuclei (Rasmussen et al. 1992, Stoelinga et al. 2003). Additional ice crystals can be produced through secondary processes like ice-splinter production (Hallet and Mossop, 1974, Harris-Hobbs et al. 1987, Griggs and Choularton, 1983, Mason, 1998, Phillips et al., 2003) or fragmentation (Vardiman, 1978). However, we are unable to accurately predict the primary ice concentration that will form in given situations via nucleation (Meyers, et al., 1992), and only in the case of the Hallett-Mossop processes are the required conditions characterized well enough to support quantitative modeling of secondary ice generation rates. This modeling is based on empirical laboratory data; the physical basis of the mechanism remains uncertain. We suspect that there are other significant secondary ice production mechanisms, but they have not been defined and characterized yet.

Identifying where the first ice originates is difficult observationally, because ice concentrations are low and the small sizes and near spherical initial shapes of ice crystals makes them difficult to distinguish from water droplets using currently available instrumentation. Only under some conditions is it possible to determine whether ice particles are produced from ice nuclei (primary ice) or through a process that involves ice crystals but

not ice nuclei (secondary ice). Entrainment of ice nuclei and ice crystals into cloud updrafts further complicates attempts to identify the processes. Laboratory studies, while insightful, are unable to completely simulate the ice nuclei aqueous chemistry, effects of evaporative cooling, water vapor competition, and potential secondary ice production processes. These poorly understood processes of ice initiation in clouds produce large uncertainties in our ability to model precipitation production (Tao et al. 2003, McCumber et al. 1991) and to predict climate changes (Fowler and Randall 2002, Zurovac-Jevtic and Zhang, 2003).

### 2.1 <u>Primary (Heterogeneous) Ice Formation</u>

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 spatial 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.

Cloud observations confirm the role of heterogeneous ice nucleation in ice phase initiation. While homogeneous freezing field and laboratory studies are recommended to continue, this initiative will focus on the area of heterogeneous ice nucleation where much less is known. Some research over the past 15 years has attempted to identify the relation between heterogeneous nucleation on aerosol particles and ice formation in clouds in cases where initial ice formation was presumed traceable. Much of this work involved flights in orographic wave clouds. These studies, while preliminary glimpses, provided optimism for the ability to resolve the relation between IN and ice formation in well defined studies. Some results of such coordinated sampling, done during Winter Icing in Storms Project in Colorado in 1994, are shown in Figure 1. These indicate the apparent correspondence, within a factor of 2, of IN concentrations measured by two methods and ice concentrations measured in the growth region of orographic wave clouds in the same region. The studies attempt to relate IN and ice formation in the same air mass but not within individual clouds. Much of the uncertainty in these types of studies is due to our inability to measure ice concentrations accurately, due to inadequate resolution of small ice particles. New instrumentation discussed in section 3 offers major improvements to detect small ice.

Airborne studies to relate ice nuclei measurements to ice formation in the same region of space were described by Rogers and DeMott (2002). Figure 2 shows an example of IN versus ice concentration, as detected with conventional cloud particle probes in an orographic wave cloud. The IN instrument processed ambient particles at a temperature approximately equal to that in the wave cloud and water supersaturated conditions. Note the general consistency of IN and ice crystal concentrations. These observations do not demonstrate a cause-effect relationship, but they provide optimism that careful experiments of this type, using the best new instrumentation, will offer an opportunity to test the hypothesis that primary ice nucleation predicts ice formation in clouds and may be quantified as a function of known ice

nucleation modes. In this case, the CFD chamber detects deposition and condensationfreezing nucleation, but is not sensitive to other mechanisms.



Figure 1. Ice nuclei concentrations from large bag samples collected by the NCAR Electra aircraft at the inflow to Colorado wave clouds in 1994 versus ice crystal concentrations in the liquid regions (upward wave segments) of clouds at the same times. IN data were collected using the CFDC technique (particles processed at modest water supersaturations) and by simulated adiabatic expansions of collected air to form clouds in a dynamic cloud chamber (no extra CCN added). The solid line is an exponential fit through IN data; dashed line is exponential fit through the cloud ice concentration data. Figure adapted from Rogers and DeMott (1995).



Figure 2. Along-wind measurements through an orographic wave cloud with Wyoming King Air (Rogers and DeMott 2002). (panel 2) Ice crystal nucleation and growth shown by 1D probe concentration (>  $\sim$ 30 µm, dots) and 2D probe (>  $\sim$ 100 µm, circles). Ice nuclei measured with CFD chamber (solid line). IN concentrations are integrated for 10 s sampling intervals.

For glaciated maritime, continental and Arctic Canadian clouds, Gultepe et al. (2001) showed typical concentrations of ice particles were near 1-10 L<sup>-1</sup> (independent of temperature), for particles greater than 125  $\mu$ m, as measured with a PMS 2D-C, and 1-10 cm<sup>-3</sup> as measured with a PMS FSSP. The FSSP measurements are clearly gross estimates because this probe was not designed to count small ice particles. However, Korolev et al., (2001) summarizing measurements made in the former USSR using an extinction probe, found small (approximately 15-20  $\mu$ m) particles in cold clouds near -30C at the same concentrations as in the Gultepe et al. FSSP measurements in Canada. 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.

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 ( $\sim$  -10°C). An explanation they offered involves 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 ( $\sim$ 15%). The later two hypotheses were found quantitatively inadequate on subsequent analysis (Baker 1991a; Baker 1991b). 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%)."

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  $-12^{\circ}$ 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 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.

## 2.2 <u>Secondary Ice Formation Processes</u>

After primary ice has formed in a cloud, the concentration of ice crystals can be increased through secondary production mechanisms if other conditions are met. Secondary 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). With optimal conditions, a 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, such as graupel in the presence of supercooled droplets with certain sizes and concentrations at specific temperatures. 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, but evidence exists 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 in cumulus clouds by Hobbs and Rangno (1990) and Rangno and Hobbs (2000) 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. Is this a major ice multiplication process that we have not explained and could this process be operative under far-reaching conditions, or is it an artifact due to breakup on the probes' inlets? This question can be addressed by using newer probes that don't generate artifacts.

## 2.3 <u>Dependence on Droplet Spectrum</u>

The formation of ice is likely to be strongly influenced by the size spectrum of cloud droplets. For typical ice nucleus concentrations in warm based cumuli, the highest initial ice concentrations are produced by the impaction of giant (> 1  $\mu$ m diameter) ice nuclei with drizzle or raindrops larger than about 200  $\mu$ m (Beard, 1992). 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 warmrain 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. This is because, 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 then proceeds relatively slowly until they have grown to millimeter-sized graupel particles. Thereafter, the conversion of the cloud water to the ice phase can proceed relatively quickly. However, if supercooled raindrops are present, the slow-growth period can be circumvented. The large raindrops then quickly collide with small ice crystals; they immediately freeze to become frozen raindrops. The frozen raindrops can 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 (Hallett and Mossop, 1974; Mossop and Hallett, 1974) further accelerates the glaciation rate of the cloud. Several modeling studies (Chisnell and Latham, 1976; Koenig, 1977; Lamb et al., 1981) 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 -3C to -8C. 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 will 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 coupling between buoyancy production by glaciation of a cloud and the evolution of the microstructure of the cloud, and the evolving cloud motion field.

## 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 may 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. This evaporation freezing mechanism has been observed in the laboratory (Durant and Shaw 2005), and the data suggests that the temperature at which the particle initiates droplet freezing will increase by several °C. 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, the ice concentrations increased by an order of magnitude. This observation cannot be explained by rime splintering.

Observations in orographic wave clouds in recent years make 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 warmer than about -20°C. Cooper also noted that ice crystal concentrations do not 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<sup>-3</sup>) of ice had to form close to where the supercooled liquid evaporated. Figure 3 shows an example. 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. The nucleation mechanism may be primary, and is a high priority of the proposed study. Two plausible hypotheses exist for explaining the correlation of ice nucleation with evaporation of supercooled liquid droplets; these will be tested with the unique data set from the ICE-L field phase.



Figure 3. Example showing the "evaporation glaciation signature" observed in wave clouds (from Baker and Lawson, 2005). Ice crystal concentration as measured by a 2D-C probe (light blue trace, lower panel) jumps up near the location where supercooled liquid water (shown as blue fill under the green LWC curve, upper panel) evaporates, FSSP concentration (purple trace lower panel) decreases and mean size (red) increases. The black line (upper panel) is potential temperature and the yellow vertical line is location of first ice observed by either the CPI or the 2D-C probe. The LWC and TWC (blue trace, upper panel) are from Nevzorov probe.

One of the hypotheses is relatively new, i.e., extending the laboratory observations mentioned earlier, viz., contact nucleation from inside the drop as well as from outside (Shaw et al., 2005). Those data suggests droplet freezing temperatures via contact are ~4 to 5°C warmer than via immersion-freezing. The hypothesis is that ice nucleation events are enhanced in regions of cloud evaporation. ICE-L will include airborne measurements of ice nuclei extending to temperatures at least 5°C colder than those in cloud. The concentration of immersion-freezing nuclei at these colder temperatures should be comparable to the observed ice concentrations if the hypothesized process is effective.

The other hypothesis is new in the sense that it has not been widely considered in the literature and for example is not found in basic cloud physics text books. The concept is based on laboratory results that are not particularly recent. Hallett (1968) showed that when one component of a supersaturated eutectic solution nucleates, it can initiate nucleation of the other phase. The concept for clouds is that as a supercooled solution droplet evaporates, the point of homogeneous nucleation of the salt will be reached thus initiating rapid precipitation of the salt which could initiate freezing of the liquid in a local region of the droplet where the solute effect is suddenly drastically reduced. Laboratory modeling of this process is underway at DRI, but to evaluate its effectiveness in natural clouds, accurate observations of the composition of the aerosol on which ice has nucleated in evaporating regions of natural clouds is also necessary. These observations are a priority for ICE-L.

# 2.5 <u>Air Parcel History</u>

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. 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 in these types of clouds 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, with higher concentrations of CCN decreasing the rate of initial ice formation and the concentration of giant CCN likely increasing it.

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.

## 2.6 <u>Dust as Ice Nuclei</u>

"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). Evidence of a relationship between dust and ice nuclei comes from several different types of

studies, some of which involve direct examinations of IN particles, and others are based on inferential evidence.

One type of direct evidence comes from identifying the nucleating particles in 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 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 indicate 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; (5) the technique requires particles larger than ~0.02  $\mu$ m; and (6) nucleation occurs on the surface at a particular site, the properties of which are not characterized.

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. Targino et al. (2005) describe results from an airborne study of wave clouds in which their CVI ice particle residues showed a high occurrence of elements associated with mineral dust. Again, it is tempting to assert that these are the nucleating particles, but they could have been scavenged in ways other than ice nucleation. It should be noted that the effect of aerosol scavenging by ice crystals is reduced in wave clouds due to the small elapsed time between ice nucleation and CVI sampling, typically a few hundred seconds or less.

Appreciable quantities of African dust are transported over large areas of the Atlantic Ocean to the Caribbean during much of the year (Prospero and Lamb, 2003). Recent airborne studies and ground-based polarization lidar in Florida during the NASA CRYSTAL-FACE project (July 2002) provide direct evidence of a connection between dust and ice nuclei (Sassen et al., 2003; DeMott et al., 2003a). Measurements were made over Florida in air masses with sources in the Sahara Desert. The dust layer was detected by satellite and ground-based lidar. Trajectory analyses indicated it was of Saharan origin. The observations support the potential importance of mineral dust aerosols to enhance greatly the heterogeneous nucleation of ice at temperatures warmer than -37°C. Using aerosol trajectory forecasting and remote sensing, future studies could target airborne sampling of dust plumes to explore the impact on a wide range of cloud temperature regimes. Cziczo et al. (2004) sampled cirrus (in-situ and anvil) crystals with a CVI during the NASA CRYSTAL-FACE project and measured their compositions in real-time with single particle laser ablation mass spectrometry (PALMS). Mineral dust particles larger than about 0.2 µm were one of the major residual particle classes during the project as a whole and predominated during the presence of lower altitude Saharan dust layers.

Direct evidence for dust as a major contributor to atmospheric IN also comes from measurements of the aerosol particles that are processed as ice nuclei to form small ice crystals, hence leaving little opportunity for additional scavenging, and in some cases the subsequent collection of IN residuals for either EM analyses (Chen et al., 1998) or single particle mass spectrometry analyses (Cziczo et al., 2003). 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 large signals of silicates and other crustal materials in residual ice nucleating particles. Real-time mass spectrometric measurements of IN by the technique of Cziczo et al. (2003) were made by DeMott et al. (2003b) at a high altitude continental U.S. site and they likewise identified mineral dust-like particles as the major contributors during sampling under "background" or remote aerosol conditions. Additional evidence from direct IN measurements has been obtained during airborne measurements made directly in dust layers during the NASA CRYSTAL-FACE experiment.

Inferential evidence of a connection between dust and ice nuclei comes from analysis of weather modification in Israel (Gabriel and Rosenfeld 1990) and from U.S. lidar studies of dust layer interactions with clouds (Sassen, 2002; Sassen et al., 2003). 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 nucleating behavior associated with dust in air (Roberts and Hallett, 1968; Zuberi et al., 2002) and many of the metal oxide components of desert dust (Hung et al., 2003). The Sassen et al. (2002; 2003) studies reported the rapid glaciation of clouds forming in regions of enhanced dust layers.

## 2.7 <u>Climate Implications</u>

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 be ice or mixed phase, 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  $+13 \text{ Wm}^{-2}$  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.

## 2.8 <u>Modeling Studies</u>

The fundamental influence of ice on cloud properties is the strongest evidence that ice generation should be represented 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). This uncertainty brings higher relevance to fundamental research; more realistic descriptions of ice processes must build on a better basic understanding of them. Laboratory studies provide unique insight under controlled conditions, but are unable to simulate all the changes in ice nuclei chemistry, effects of evaporative cooling, water vapor competition, 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) reveal that more observations are needed to characterize and model the following aspects: ice initiation and subsequent number concentration, production and depletion of supercooled liquid water, evolution of snow and graupel size distributions, and transition from rimed snow to graupel.

In the past, simple modeling approaches to ice in clouds have been used in part because of limitations of computer power. With the increase of computer power and 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. 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. A more realistic inclusion of ice processes requires a better basic understanding of them.

Most representations of ice generation use some variation of the Fletcher curve (Fletcher, 1962) to represent primary ice nucleation, despite the near universal opinion among cloud physicists that it is unjustified because it is inconsistent with recent evidence (Meyers et al., 1992, Rangno and Hobbs, 1994), a misinterpretation of the original intent of the formula, and an oversimplification of the ice production process. Its appeal is computational 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). Some models such as RAMS attempt to address these issues. RAMS has initial vertical and horizontal variability of 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). Modeling the sources of these aerosols, however, is still a problem.

Secondary ice generation is often ignored or treated inappropriately (Connolly et al., 2004) despite its overwhelming importance in many cloud types. With the parameterizations described above (Lohmann and Kärcher, 2002; Liu and Penner, 2005), it is critical to develop the means to treat ice nucleation in coupled aerosol/cloud models. These parameterizations would need to be extended to treat heterogenous nucleation at temperatures warmer than – 40°C. 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).

### 2.9 <u>Summary</u>

From the discussion above, it is clear that there exist fundamental uncertainties about the nature of ice formation in clouds that can be addressed by coupling modeling and laboratory studies with new instrumentation deployed in carefully designed field and lab experiments:

- Measurements of ice nuclei need to be compared against measurements of ice concentrations in natural clouds under conditions that are well defined (such as wave clouds). These measurements of ice concentrations need to be done with modern instrumentation that can better resolve small ice particles.
- Field observations suggest that droplet evaporation enhances ice nucleating activity in some circumstances, but the controlling factors are not identified or understood. Several studies have observed highly repeatable and abrupt ice nucleation occurring nearly coincidently with the evaporation of supercooled liquid in orographic wave clouds. Measurements in such clouds, with new instrumentation, can shed light on a process that might extend to convective and other cloud types.
- The effects on the ice initiation process of variations in the chemical composition of aerosol and ice nuclei and aqueous chemical changes in droplets are not known. New studies can explore this question quantitatively by combining instruments for ice nuclei, cloud particle separators (CVI), and single-particle mass spectrometry.
- Although ample laboratory evidence has characterized one secondary ice initiation process (*Hallett-Mossop*) that operates in a certain 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 may help to explain the *Hallet-Mossop* mechanism and to identify other processes.
- For more complex clouds, such as cumulus, the location and processes responsible for initial ice formation are not known. The roles of the 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.

## 3. Scientific Direction

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

- 1. Establish which heterogeneous ice nucleation modes are active and important by:
  - Detecting the initial formation of ice particles in clouds (e.g., is ice forming before droplets, immediately after, or later, and in the evaporating regions of liquid clouds?)
  - Precisely measuring environmental state properties (temperature, pressure, water vapor) and kinematics
  - Measuring ice particle size, concentration and bulk condensed water properties over short spatial scales (~100 m)
- 2. Identify ice nuclei by:
  - Determining which aerosol particles are active as ice nuclei
  - Characterizing the physical and chemical properties of the ice nucleating particles
  - Define the environmental conditions at the point of ice nucleation.
- 3. Predict ice concentrations with numerical models by:
  - Determining the thermodynamic history of air parcels in which ice nucleation occurs in simple clouds such as wave clouds, so that ice nucleation parameterizations can be tested in numerical models.
  - Doing numerical experiments that demonstrate the importance to ice formation of dynamical processes that drive the thermodynamics, such as updrafts, downdrafts, turbulence, entrainment and cloud-edge mixing events.

An initial field campaign is described in this section to study ice initiation processes, focusing on primary, heterogeneous nucleation although recognizing that secondary ice processes may be active and compete with primary nucleation under certain circumstances. Follow-on field programs are being considered, but are not discussed here. The initial campaign is called the **Ice in Clouds Experiment-Layer, ICE-L**. It is proposed for late winter-early springtime clouds along the High Plains at the foot of the Rocky Mountains in the United States. This location was selected because it offers an opportunity to make fundamental progress in understanding ice formation processes, because the High Plains region has a high frequency of layer clouds, including orographic wave clouds and "upslope" clouds. The layer clouds are of particular interest because they offer relatively simple airflow characteristics and are less susceptible to secondary ice production mechanisms. In the early spring, convective clouds, amenable to airborne sampling, also occur intermittently. The High Plains area is subject to episodic intrusions of Asian dust. Airborne experiments in layer clouds can use guidance from satellite imagery and trajectory forecast models in order to time research flights during periods where long range transport of Asian dust is likely to affect the western U.S.

Layer clouds include those formed by upslope winds (e.g., orographic) and by more gradual ascent (e.g., altostratus). Lenticular wave clouds have the desirable feature that the formation and evolution of water droplets and ice particles can be characterized as a function of time as air parcels advect through a mountain wave. Ice particle evolution in this cloud type is not representative of all clouds, but wave clouds provide the opportunity to characterize ice formation processes and to evaluate measurements by ice nucleus instruments. Clouds formed through more widespread gradual ascent (Rasmussen et al., 1995) can be sampled

over extended periods of time. In particular, altostratus and altocumulus clouds have relatively simple air motions and can persist for extended periods; for these mid-tropospheric clouds, the source regions for ice nuclei are at mid-levels rather than near the boundary layer.

## 3.1 <u>Wave clouds</u>

Lenticular wave clouds offer one of the best opportunities for making direct comparisons between atmospheric observations of cloud microphysics and laboratory experiments. The cloud forming process is the forced ascent of stably stratified air, and cloud dissipation occurs as the air descends. Mixing is strongly suppressed, and the airflow is relatively smooth, quasi-steady, primarily horizontal and isentropic. Gradients of scalar atmospheric properties (temperature, humidity, aerosols, etc.) tend to be small along the isentropes but may be large across isentropes, resulting in layering. An aircraft making in-situ measurements cuts across isentropes, which can make interpretation ambiguous. Baker and Lawson (2005) studied seventeen wave clouds using the SPEC Learjet research aircraft and found that the clouds vary from extremely simple to very complex in structure. Figure 4 shows conceptual representations of the cases of both simple and complex wave clouds These figures exemplify the potential difficulty of interpretation from single penetrations, or even multiple-level but incomplete sampling, of the thicker more complex wave clouds.



Figure 4. (left) Schematic representation of a simple wave cloud derived from a numerical model adapted from Heymsfield and Milosovich (1993) showing the expected regions of supercooled liquid water (SLW), mixed-phase, and ice cloud. (right) schematic drawing showing actual variability often observed in complex wave cloud (from Baker and Lawson 2005). Note the waves within the main wave, the vertical layering, and the multiple sequences of supercooled liquid water (SLW), mixed phase and then glaciated areas of cloud.

A direct laboratory analog exists for the simpler wave clouds: cloud formation in a steady expansion (pressure reduction) and cloud dissipation in smooth compression. The conceptual picture of air continually flowing into the upstream edge of the cloud and out the downstream edge, with air parcels spending a few hundred seconds in cloud is idealized and suggests that parcel theory will give a reasonably accurate representation of the microphysical processes occurring in the simpler clouds and parts of the more complex clouds. To the extent that the important variables can be identified and accurately simulated, laboratory experiments will reproduce the fundamental processes of droplet and ice formation and growth. Because the thermodynamic history can be reconstructed to first order from temperature and water vapor measurements, assembled from penetrations at multiple levels of simpler wave clouds, it is possible to identify correlations between various growth stages and the ice formation process.

The measurements needed could be made using a single aircraft that has the ability to sample both the aerosol and cloud properties if simple enough wave clouds are found at the appropriate temperatures. The field project will be stronger by including millimeter radar observations and adding another aircraft capable of reaching all the altitudes of the wave cloud. With such additions, the complexity of the clouds can be derived from observations of the whole cloud system. These also make it possible to detect contamination of the target cloud layer by snow particles precipitating from higher layers. Baker and Lawson (2005) show that their large data set could be segregated to allow for natural laboratory type studies using an appropriate subset of the *in-situ* data.

Given that the thermodynamic structure and temporal evolution of the microphysical properties can be reconstructed from measurements along and against the airflow at multiple levels and the availability of improved water vapor and microphysical probes, one focus of this effort will be to identify where and when evaporation freezing is occurring and attempt to identify the mechanism(s) responsible (e.g., Ansmann et al. 2005; Baker and Lawson 2005; Cooper 1995; Field et al. 2001). The observations will be compared with parallel computational and laboratory studies (e.g., Cotton and Field 2002; Durant and Shaw 2005).

#### 3.1.1 Climatology of wave clouds in Colorado and Wyoming

Wave clouds occur frequently over the mountain ranges in western North America, throughout the late fall into the early spring. A ten-year climatology is shown in Figure 3 for Denver and Cheyenne. It indicates that lenticular clouds occur approximately one day out of three from October through mid-April.



Figure 5. Frequency of occurrence of wave clouds by month as observed by the National Weather Service.

An estimate of the temperature ranges of lenticular clouds was made from NWS standard levels of twice-daily radiosonde observations during March, April and May for Denver, Colorado, and Lander, Wyoming, as shown in Figure 4 for the period 1976 to 1992. Within the range of C-130 maximum altitude ceilings (24,000 to 26,000 feet), the C-130 should be able to reach the -35°C temperature level for approximately one-half of these soundings. March has slightly lower temperatures than April for the Lander soundings and about the

same as that for Denver; May temperatures are considerably warmer. Lander has lower temperatures on average.



Figure 6. Points from NWS standard levels (300, 400 and 500 mb) twice daily soundings from Denver (left panels) and Lander, Wyoming (right panels) for the period 1976 to 1992. Mean temperatures by month are shown by the sloping lines. Reference lines show ceiling of NCAR's C-130 aircraft and -35°C homogeneous ice nucleation threshold (data from Ben Bernstein, NCAR).

Soundings from Denver when wave clouds were observed in Boulder are shown in Figure 5. Boulder is ~15 km from Denver, and the observations were made by John Brown (NOAA meteorologist) for the period March through April over several years. Although the temperatures where the wave clouds were located is not known, the sounding temperature profiles are similar to those in Figure 3, and C-130 maximum altitudes correspond to temperatures -45 to -25°C.

The conclusion from this examination of soundings is that the month of March provides excellent opportunities in the Colorado-Wyoming area to sample wave clouds that span heterogeneous and homogeneous ice nucleation temperatures. Furthermore, the NCAR C-130 is capable of sampling clouds under these conditions. Airborne studies in support of ICE-L will be based from the NCAR Aviation Facility in Broomfield, Colorado, to minimize cost and to capitalize on this climatology. The NCAR C-130 will be proposed to make the measurements we need. A non-facility, upper-level aircraft is desirable and will likely be proposed.



*Figure 7. Denver soundings for days when wave clouds were observed over Boulder, Colorado (from John Brown, NOAA).* 

Detailed flights patterns designed to investigate ice initiation in wave and layers clouds are presented in the Appendix (Section 6.1). The strategy is to fly upwind/downwind flights legs at various temperatures (in wave clouds) and near cloud top (in layer clouds). The instrumentation set (Section 3.4) 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. We plan to conduct the experiment in the March-April timeframe when wave clouds should occur on 1/3 to ½ of the days (Fig. 3). Asian dust influences on primary ice concentrations will be investigated as part of the lenticular wave cloud investigation. Asian dust episodes peak in the springtime and are transported to the Rocky Mountain region, and with height, although the aerosol concentrations diminish, the episodes become more discrete (Jaffe et al., 1999; VanCuren and Cahill, 2002; Huebert et al., 2003; Sassen, 2002). With measurements in clean and dust-laden layers, this research can address the central question, is it possible to predict primary ice concentrations from measurements of ice nuclei?

#### 3.2 Upslope Clouds

In the Front Range region of Colorado, winter and springtime weather systems often produce a shallow upslope cloud. This cloud occurs within cold air masses and can persist for a day or more (Whitman, 1973; Weickman, 1981; Boatman and Reinking, 1984). Whitman (1973) showed that shallow upslope cloud systems with cloud thickness greater than 1 km occurred from 2.5 to 6 times per year in the High Plains using a 10-yr dataset. Cloud top temperatures are often warmer than -12°C and snowfall is usually light (Rasmussen et al., 1995; Politovich and Bernstein, 1995). Precipitating drizzle is often present or mixed with snowfall, depending upon the cloud top temperature and whether there is overlying cloud. Ice crystal concentrations greater than 1  $L^{-1}$  have been reported in upslope clouds, but supercooled liquid water can be long-lived even when cloud top temperatures reach -15°C.

Upslope clouds also provide a laboratory-like setting to examine ice initiation processes. Because these clouds are often long-lived and the air trajectories are relatively simple, Lagrangian parcel-following aircraft flight trajectories at multiple levels through the cloud layer can be repeated to investigate ice formation processes. Ice nuclei measurements upwind of the cloud layer and at cloud top can be compared directly with the in-situ measurements of ice concentrations to evaluate whether the ice nuclei measurements are of predictive value. These cloud layers also provide a unique opportunity to study the effects of episodic dust events on ice nucleation and to evaluate the microphysics attributes of cloud and mesoscale models.

In addition recent studies by Givanti and Rosenfeld (2005) and Jirak et al. (2005) suggest that pollution from major urban areas can alter precipitation downwind of those cities relative to that in more pristine neighboring regions. Jirak et al. (2005) found this effect for the Front Range of Colorado downwind of Denver during upslope events relative to similar areas like west of Greeley. Therefore, the proposed study of upslope clouds provides a unique opportunity to determine if CCN and IN concentrations, and the characteristics of the clouds are different downwind of Denver versus corresponding relatively pristine areas during upslope events.

## 3.3 <u>Cumulus clouds</u>

During the springtime in the Front Range of Colorado, the weak to moderately intense convective clouds that form are amenable to aircraft sampling. Although cumulus clouds are more complex than layer clouds, they offer an opportunity to study ice formation in this important class of clouds under different aerosol environments.

For example, in a recent study of ice formation in maritime and continental convection Rangno and Hobbs (1994) found that the initial stages of ice formation required the presence of large cloud droplets which they found near cloud top. In one of their papers they concluded that the presence of large drops was not essential but that the presence of graupel was. A rapid increase in ice concentration was observed when some drops near cloud top exceeded 25  $\mu$ m diameter. These observations were at temperatures colder than those found in the Hallett-Mossop secondary ice production temperature regime (-3 to -8°C). These observations suggest that large droplet regions are preferred locations for ice formation.

It is well known that locations with drizzle-sized droplets are preferred locations for ice formation. Beard (1992) suggested that ice should readily form in warm-based convective clouds at temperatures colder than  $-10^{\circ}$ C, although in low concentrations. In this case ice is formed by the collisions between giant (greater than 1 µm) ice nuclei and these drizzle droplets; however, the size of the drizzle is much larger than the drops required in the Rangno and Hobbs study and the ice concentrations are much less. The physical mechanisms for the Rango and Hobbs observations are not known. Springtime Colorado convective and upslope clouds provide an opportunity to evaluate the Rangno and Hobbs observations in clouds that are amenable to sampling by research aircraft. The target clouds for cumulus sampling are congestus clouds, which typically occur in spring prior to the main convective season.

Satellite observations indicate that dust from Asian storms is often transported across the Pacific Ocean to North America (Huebert et al., 2003). Sassen (2002) used ground-based lidar to detect mid-tropospheric dust layers in Utah during winter. Trajectory analyses indicated that this dust originated in Asia approximately ten days earlier.

#### 3.4 <u>New Instruments and Aircraft for observing ice formation</u>

In the past few years, there have been significant improvements of instruments and combinations of instruments for measuring cloud-active aerosol particles. The HIAPER and other research aircraft offer compelling capabilities to support new programs for studying ice formation. New instruments are being developed to characterize ice nucleus properties and concentrations of small ice and complement existing and recently available probes. These aircraft can carry a complete set of ice-related instrumentation and have enough performance to sample growing clouds throughout the temperature regions of interest.

The CSU continuous flow diffusion chamber (CFDC, DeMott et al., 1998) measures ice nucleus concentrations of aerosols in the size range ~50 nm to 1 um. The *counter-flow 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 or to a single particle mass spectrometer to measure the size and chemical composition. Similarly, residual particles from ice crystals that nucleate and grow in the CFDC can feed a particle mass spectrometer (DeMott et al., 2003b).

Airborne cloud condensation nuclei (CCN) instruments can measure an activation spectrum (e.g., Hudson 1989), fast-response at a single supersaturation (Roberts and Nenes, 2005). Though not fast response, static-chamber type CCN instruments are small, autonomous, and cover a wide range of supersaturations.

Aerosol particle mass spectrometers have been adapted for airborne use (Cziczo et al., 2004) and provide mass and chemical composition of single particles larger than  $\sim$ 50 nm. Similar instruments that use particle collections are sensitive to  $\sim$ 5 nm; these are not adapted for airborne use yet, although work is on-going to achieve this capability.

All of these instruments have been used recently in both laboratory and airborne studies.

Cloud profiling with millimeter radar provides information on cloud structure, mixing state, and the location of first ice. Particle size distributions and shape information are obtained from a small ice particle detector probe (SID-2, Hirst et al., 2001; Field et al., 2004). One-and two-dimensional Cloud Particle Imager Probes (CPI, Lawson et al., 2001; 2D-S, Lawson et al., 2005) yield detailed information on early ice formation in clouds and the size distributions of coexisting small ice and water droplets (more information in section 3.4.1).

Laboratories offer highly controllable experimental environments (Mohler et al., 2003), new instrumentation, and precise control of aerosol and ice nuclei concentrations, compositions, and size distributions. They can perform experiments to evaluate ice nucleus activity under conditions comparable to those found in the atmosphere. Advancements in computer model simulation have progressed to the point where ice formation mechanisms can be tested and evaluated. Models can also provide near real-time guidance for field experiments (Seifert et al. 2003a, Seifert et al. 2003b, Gierens, 2002, Grabowski, 2003, Randall et. al. 2003).

The important need to extend knowledge about ice processes and the promise of new research tools and methods are forceful arguments for organizing studies of ice formation now. The timing is appropriate to conduct fundamental *in situ* cloud measurements to study ice formation in natural clouds, together with laboratory and modeling efforts to improve the understanding of ice initiation and the parameterization of these processes for cloud and climate models. The following sections describe plans for this research.

#### 3.4.1 Observing ice concentrations and first ice formation

In layer cloud studies over the past 30 years or so, instrument limitations (principally lack of instruments) that can accurately sample ice particles  $<100 \ \mu m$ ) and to reliably measure relative humidity made it difficult to detect and discriminate ice particles at the earliest stages of formation and to characterize the ambient conditions when they form. These limitations occur primarily near the cloud edge (upwind or downwind), outside of liquid water regions or in evaporation regions and can be significant in at least two different cases where ice particle growth is limited. The first case is for relatively small layer clouds having correspondingly small air parcel residence times, and the second case is for any layer clouds having high crystal concentrations. In situations where ice crystals continue to grow larger than ~100 µm, they can be detected with good accuracy, and knowledge of the cloud kinematic setting and air parcel thermodynamic history can be used with modeling in order to infer the spatial and temporal origin of the ice. The challenge is to identify and demonstrate a correspondence between aerosol properties, thermodynamic forcing, and ice initiation.

Heterogeneous freezing in wave clouds was evident in studies by Kelly (1978), Cooper and Vali (1981), and Rogers and Vali (1987). Aircraft passes were made through layer clouds along the direction of the wind. Cloud temperatures were generally in the range -10°C to -30°C, so that homogeneous nucleation processes were not active. The crystal observations were based on PMS 2D-C instruments and typically showed ice concentrations rising quickly from ~zero at the upwind liquid edge to a plateau value within ~100 s parcel time. Liquid water was present throughout the clouds, suggesting large ice supersaturations existed. In these circumstances, small crystals grow quickly (~1  $\mu$ m s<sup>-1</sup>) to sizes readily detectable by the 2D-C probe ( $\sim 100 \text{ }\mu\text{m}$ ). The observations showed crystal size distributions had a single broad mode, and the mode increased uniformly downwind. Backward integrations of crystal diffusion growth indicated that a major fraction of the crystals nucleated in the vicinity of the upstream liquid edge of the cloud. In these circumstances, it seems unlikely that measurements of ice concentrations were seriously underestimated. However, more recent studies in wave clouds (e.g., Field et al. 2001; Baker and Lawson 2005) show that an abrupt, strong increase in the concentration of small ice particles with a bimodal size distribution appears as the droplets evaporate. This phenomenon is observed near the middle of cloud, and is discussed in more detail in Section 2.4.

Wave clouds have little mixing along with relatively simple air parcel trajectories and therefore offer a unique opportunity for airborne studies. Because most of the important components can now be measured, explanations for microphysical processes can be merged into numerical models, and our knowledge of the first stages of ice formation can be tested.

The small ice size-sensitive limitation and shape/phase determination has been improved significantly with the recent development of the SID-2 and 2D-S probes. The SID-2 uses the asymmetry of light scattering from non-spherical particles to discriminate small ice crystals. Airborne and laboratory studies suggest its sensitivity extends down to about 1  $\mu$ m. The 2D-S (stereo) probe provides a shadow image of cloud particles. It has extremely fast electro-optics and uses two orthogonal laser beams that improve definition of the sample volume to detect particles as small as 10  $\mu$ m. Baker and Lawson (2005) show an example of measurements from the SPEC Learjet where the 2D-S probe detects first ice in a wave cloud approximately 1 km upwind of where the 2D-C probe detects ice.

Accurate measurements of relative humidity can be obtained with tunable-diode laser technology at a rate of 10 Hz or greater. Studies of the connection between first ice, in-cloud

thermodynamics and droplet evaporation in the descending branch of a wave cloud can now be pursued. These problems were beyond the reach of earlier technology. Given the possible importance of evaporation nucleation in all clouds, not just lenticular wave clouds, a central thrust of the ICE-L experiment will be to investigate the relationship of first ice formation to evaporation, using a wider complement of instruments and higher precision instruments than those used in previous experiments.

In colder layer clouds, where homogeneous freezing nucleation is the dominant ice formation process, or where there are other causes of high crystal concentration, it is more problematic to discriminate water and ice particles because ice particles probably begin as frozen haze droplets and because ice growth rates are slow. In these cold regions of wave clouds, relatively high concentrations of ice particles  $\sim 10 \mu m$  can be detected with traditional *cloud droplet* probes. However, care must be taken to distinguish regions where there are no particles larger than a few hundreds of microns that could shatter on probe inlets and be counted as small particles (e.g., Field et al. 2003; Korolev et al. 2005). The particle phase can be inferred from humidity measurements, the Rosemount supercooled water detector (Mazin et al., 2001) and verified by new cloud particle instruments such as the CPI, 2D-S, and SID-2. These instruments present significant advantages over earlier studies and will be key factors in addressing first ice formation.

A high altitude aircraft will be needed in ICE-L if we are to make measurements in the homogeneous nucleation layer of wave clouds (see section 6.1).

#### 3.4.2 Airborne measurements of ice nuclei

This section identifies the ice nuclei measurements needed for airborne observations of ice formation in association with layer and convective cloud studies. Current instrumentation is identified, and recommendations are made for developing the necessary measurement capabilities where none exist.

For studying the ice formation processes, there is a distinction between clouds with dominant heterogeneous freezing processes and those with primarily homogeneous freezing. Earlier measurements in layer clouds showed that ice onset occurred near the visible leading edge, that is, within a region a few hundred meters wide. Air parcel residence times in this region are a few to tens of seconds; research aircraft flying along the streamlines will traverse this region in a few seconds. For clouds warmer than about -40C, the dominant heterogeneous ice processes are either condensation-freezing or contact-freezing by very small aerosol particles. Serious questions remain about the deposition mode (formation of ice below water saturation), viz., does it exist and if so, what contribution to the total ice can be attributed to deposition? The challenge in detecting deposition-formed crystals is that they are initially small, and the concentrations are usually so low (<  $\sim$ 0.1 L<sup>-1</sup>) that sampling statistics are typically poor. New instruments may overcome these obstacles, for example with smaller size detection of ice and airborne polarization lidar.

Ice nucleation occurs in response to two primary and independent thermodynamic factors, temperature and humidity. In addition, time and the presence of supercooled drops are important factors. With these factors in mind, airborne IN detectors should have the following capabilities:

- Fast response (~1 second) for accurate temporal and spatial resolution. If the inflow air is steady and has uniform properties, this requirement can be relaxed to allow prolonged sampling in the inflow air upwind of the cloud. This situation is typical for wave clouds.
- Ice nuclei activation spectra: measurement of IN response to both temperature and supersaturation within the ranges observed during aircraft penetrations of the clouds.
- Able to discriminate nucleation mechanisms. IN measurements should be sensitive to the dominant nucleation mode(s), to include condensation freezing, deposition (upwind of water cloud), contact freezing, and immersion freezing. Homogeneous freezing is a distinctly separate mode; its measurement is needed for clouds colder than about -35C and should be closely coupled to simultaneous measurements of CCN activity and chemistry.
- Sensitivity to a hypothesized evaporation ice nuclei (EIN) process, if this process is observed in the field studies.
- Sample particles over a wide size range, from fine particles (<0.1 um) to giant aerosols (>10 um).
- Characterize the physico-chemical properties of IN, in order to help identify source regions and transport processes. This has been done recently by combining IN instruments with a single-particle mass spectrometer (Cziczo et al., 2003).
- Capture cloud crystals in-situ for physico-chemical studies of non-volatile components (including ice nuclei). Airborne counter virtual flow impactor (CVI) instruments provide this capability, and recent studies have combined CVI and IN instruments (DeMott et al., 2003a).

At this time, there is no single IN instrument capable of providing all these measurements at the same time, but there are instruments with strengths in several areas, and combinations of instruments that can meet most of these needs.

A wide variety of different ice nuclei measurement techniques have been used; each one usually favors one or two nucleation mechanisms at the expense of being unable to detect others. Thus, mixing chambers (Langer 1973) are sensitive to contact nucleation because they produce very high concentrations of supercooled water droplets; they have, however, strong and uncharacterized transient supersaturations which can be of primary importance for nucleation, and their time response is  $\sim 20$  to 60 s. Membrane filters (Bigg 1996) sample large volumes of air (few hundred liters). The filters can be processed in different ways to simulate all four nucleation modes, but temporal resolution is usually no better than 20-30 minutes. Complicating factors include possible chemical and diffusion interference from the filter substrate and high concentrations of hygroscopic particles that are captured along with the few ice nuclei. While the continuous flow diffusion (CFD) chamber technique has shown promise in recent field campaigns, it has well recognized limitations: no sensitivity to contact nucleation; sample rate of only about 1 liter per minute; and sample residence time in the chamber of only a few seconds. The CSU version (Rogers 1988) does not sample aerosol particles larger than 2 µm diameter, and it measures at one temperature and supersaturation at a time.

Recent advancements have been made in some of the following areas and augur well for better measurements in the ICE initiative experiments. Additional attention is still needed, and is encouraged as part of this research:

- Greater air sample rates, in order to improve temporal resolution and sampling statistics. An effective approach might put an inertial aerosol concentrator upstream of the CFDC flow instrument.
- New approaches to measuring IN that emphasize or isolate particular nucleation mechanisms. For example, perhaps the time element of contact freezing could be overcome by accelerating droplet-particle interactions by electrostatic charging or an acoustic field. Another example would be segregating particles and testing a subset with the IN instrument, e.g., droplets from CVI, CCN, non-CCN, etc. Recent studies have successfully coupled CVI residual particles into a CFDC ice nuclei instrument.
- An assessment of the effects that warming and drying the sample air before measurement with the IN instrument. Is it important to maintain the sample at ambient conditions?
- Reference standard IN aerosol particles or procedures, in order to compare different measurement methods.
- Comparison of IN measurement techniques, probably in a laboratory-based intercomparison workshop. For example, a large expansion type cloud chamber can provide the most accurate simulation of the natural processes and should be part of the laboratory study.

The ultimate IN detectors are the clouds themselves, and they are the ultimate judge of how ice forms. If it were possible to build an instrument that would sample air and subject it to the same thermodynamic forcing as the cloud environment, then water drops and ice crystals would form and dissipate in the same manner as in the real cloud. However, even if we could build such an instrument, its use would only imitate nature; it would not improve our knowledge of how nucleation processes work or how to describe them in terms of physical and chemical properties of the aerosol. An ice nuclei detector that only imitates the natural process is not sufficient for these studies. Therefore, we argue that a multifaceted approach that emphasizes *mechanistic* studies and new capabilities is not only proper, but has high likelihood for achieving the goals of this research.

## 3.5 Laboratory Studies

A variety of laboratory experiments can be performed to examine the details of the fundamental processes of ice formation. The ultimate goal of these laboratory experiments is testing the basic scientific understanding of the microphysics in cloud forming processes. In many cases, there are close analogs between natural cloud processes and laboratory experiments. There are techniques for generating aerosol particles that closely resemble their natural counterparts, for example, desert dust, soot from biomass burning, and sea-salt.

The unique strength of lab studies is that the initial conditions and thermodynamic histories are well-defined. Often, the same instrumentation is used on research aircraft and in laboratory experiments, helping to eliminate instrumental biases.

A range of lab experiments can be done to examine ice processes in the atmosphere. Where feasible, mechanistic studies will be emphasized. Issues that are especially amenable to laboratory investigation include:

- electrical charge effects on scavenging and nucleation
- ice nucleating activity of specific aerosol such as mineral dust
- evolutionary studies, such as secondary ice production; deposition growth rates and habits of crystals; aggregation sticking efficiency
- the role and effects of organic compounds on ice formation, growth, evaporation

Laboratory experiments are ideal for testing and developing airborne microphysics probes and ice nuclei instrumentation. They also provide support for finding new ways to combine airborne instruments, as has been done recently with CVI feeding CFDC and with CFDC feeding PALMS (Cziczo et al., 2003).

Relevant laboratory experiments that have recently been conducted in cloud chambers and smaller scale experiments include the characterization of the ice nucleation abilities of desert dust samples, and soot particles with both sulfuric acid and sulfate coatings (e.g., Möhler et al., 2005; DeMott et al., 1999; Zuberi et al. 2002; Archuleta et al. 2005). Planned experiments for the AIDA cloud chamber include the investigation of the competition between heterogeneous and homogeneous ice nucleation modes and the effect of coatings on desert dust particles.

Laboratory studies are particularly helpful for investigating nucleation mechanisms. For example, it has been observed that contact-freezing has enhanced activity compared to both deposition and immersion-freezing mechanisms. Cooper (1974) analyzed data from laboratory studies and offered an explanation for contact's enhanced activity relative to *deposition*. It was based on conventional nucleation theory and asserted that the size of the critical nucleus was smaller for a contact process. However, an explanation for the enhancement of contact-freezing compared to *immersion-freezing* has been elusive. Recent laboratory studies, however, have allowed several competing hypotheses for contact nucleation to be eliminated (Durant and Shaw, 2005) and, instead, imply that the enhancement is related to the thermodynamics and kinetics of water-air interface (Shaw et al. 2005). Further study of this phenomenon and its possible role in cloud ice formation can benefit from laboratory studies carried out in parallel with field studies in wave clouds (see Sec. 3.1). For example, the cloud droplet activation-growth-evaporation process can be simulated over a range of realistic wave-cloud conditions, and the resulting ice formation can be observed and compared to field data.

Some lab studies can address differences between lab and aircraft measurement techniques. For example, airborne instruments and sampling methods for measuring cloud particles can be inherently different from those used in the lab. The differences can produce systematic biases that confound lab - aircraft comparisons. Such differences should be characterized.

## 3.6 <u>Numerical Modeling</u>

Numerical models are used to predict ice concentrations from measurements of cloud-active aerosols (CCN and IN), cloud dynamics, and thermodynamics. Detailed microphysical modeling is a crucial component of the planned research. Simulation studies (Numerical Weather Prediction models, Cloud Resolving Models, and parcel models) will support both the field observations and the laboratory experiments. The modeling studies are important for planning the laboratory experiments, for identifying the important measurements, linking

the airborne and laboratory observations, and for developing and testing fundamental understanding and functional descriptions of ice forming processes.

In concert with an ice initiation field project, it is proposed that tests of one or more bulk microphysics parameterizations in real-time simulations be carried out. Results of these simulations will likely become guidance and initial 12 to 48-hour planning tools for field and aircraft operations. This task is well-suited for upslope and wave cloud studies since the national-scale models have coarse grid spacing and often do not resolve/predict shallow boundary-layer (including upslope) clouds or relatively thin wave clouds. The WRF model would be run at high resolution (horizontally and vertically) to resolve small-scale orographic features and relatively shallow clouds at all levels. Simulations will include explicit prediction of five or more hydrometeor species: cloud water, cloud ice, snow, graupel, and rain. Given that proposed aircraft may be susceptible to hazardous in-flight icing, attention will be placed on prediction of supercooled liquid water as a potential threat. The WRF and RAMS models would be run at high resolution (horizontally and vertically) to resolve smallscale orographic features and relatively shallow clouds at all levels. Whereas few current models contain a predictive variable for aerosol particles and/or ice nuclei, we propose to add one or more species to WRF and explicitly predict their movements and ice-initiation effects. In a similar manner, we will have the framework to test the role of snowpack in suppressing ice nuclei sources as well as an ability to add various sources of desert dust (whether from Asia, Africa or Southwest US). Ideally, we desire to ingest real-time data sources to capture dust transport and predict their microphysical effects in the aforementioned real-time simulations.

In addition to the regional-scale modeling using a bulk microphysics scheme in WRF, we propose to test the far more sophisticated detailed/bin microphysics scheme of Geresdi (1998). Recently, this bin scheme has been incorporated into WRF and is currently being tested in two-dimensional simulations. The scheme includes specific deposition and contact nucleation methods that can be tested against data collected during the field and/or laboratory campaign. Secondary ice production mechanisms can also be tested using this framework. Results of this scheme will then be used to improve the bulk parameterizations.

## 3.7 <u>Remote Sensing Studies</u>

Cloud observations from the University of Wyoming Cloud Radar and from lidar at the University of Utah FARS site have shown the potential for detecting ice initiation and spread using remote sensors (Fig. 8). Similarly, satellite remote sensing of cloud properties is relevant to the experiment objectives because it provides large-scale context for understanding the small-scale *in situ* observations. Cloud remote sensing studies are important for the Ice in Clouds Experiment for several reasons:

• Microphysical and optical property validation.

Validation of satellite cloud retrievals in general, and ice clouds in particular, are notoriously difficult. This is due to the differences in spatial scales (1 km satellite versus localized *in-situ* observations) coupled with cloud temporal evolution on the scale of minutes, difficulties in obtaining accurate and representative microphysical measurements, and the lack of field campaign data conducted on a variety of ice cloud types. The use of wave clouds as a validation target is especially advantageous because of their relatively large spatial extent and slow temporal evolution. Cloud parameters retrieved from passive optical imagery that

require validation from in-situ observations include particle effective radius, cloud optical thickness, and ice water path.

• Algorithm improvement.

To date, microphysical models (habit mixtures and size distributions) used in operational ice cloud retrievals (e.g., MODIS) rely on a somewhat limited set of in situ observations. Retrieval uncertainty associated with the variety of habits as a function of size that occur in nature, and as a function of cloud type, is difficult to assess without further in situ observations in a variety of cloud types.



Fig. 8. Wyoming cloud radar (WCR, 94 GHz) observations of the detection of a wave cloud downwind of ice initiation. Air flow is left to right. The liquid cloud at the leading edge of the wave is not detectible but this would be known from the in-situ (C130) m measurements. The minimum detectible signal for the WCR for ICE-L is expected to be -32 to -34 dBZe at 0.5 km (typical range of interest for wave clouds) with useable Doppler velocities for signals greater than approximately -26 dBZe. The WCR will be able to look upward and downward simultaneously for ICE-L -- a capability that will be used to document the shape/structure of the clouds during penetrations through the cloud layers with a gap of ~200 m surrounding the aircraft. Thus, we expect to be able to detect cloud top (hence to infer cloud-top temperature) for penetrations through or below the cloud layer.

• Vertical air motions

The average air motions and variance (turbulence) can be derived from airborne Doppler radar. It is very important to characterize the kinematic structure along the flight path and through the depth of layer clouds.

• Retrieval interpretation

Operational MODIS ice cloud retrievals as well as higher spatial resolution MODIS Airborne Simulator observations show a high correlation between convective ice clouds (large optical thickness) and effective particle radius, where the optically thicker cloud are predominantly associated with a relatively narrow range of retrieved radii (~20-30  $\mu$ m). It is important to understand the physical implication(s) of such results, e.g., nucleation mechanisms and particle evolution, strength of convection, dynamic history, etc.

Modern cloud field research programs are rooted in a combination of in situ and ground- and airborne-remote sensing platforms. This approach evolved from attempts to characterize clouds in terms of their cloud seeding potential in the early 1980s, when most of the modern

remote sensors useful for cloud probing became available. Especially in mountainous terrain where low-level aircraft operations were restricted, millimeter-wave Doppler radars, polarization lidars, and dual-channel microwave radiometers began to be deployed in cloud seeding experiments to supplement in situ data collection (e.g., Sassen 1984). There are two main advantages of this multiple remote sensor approach. The first is that each type of remote sensor has its distinct advantages, and disadvantages, when it comes to sensing various cloud properties, such as discriminating between water and ice particles, while the combined measurements are clearly synergistic. (This has lead to the concept of multiple remote sensor field stations like the ARM sites, and to the development of powerful cloud property retrieval algorithms using two or more types of sensors.) The second advantage is that time records of remote sensing data provide important high-resolution information on the temporal changes in cloud properties from the ground, or on spatial cloud changes from airborne platforms. In both cases, the remote sensing observations provide the context for the improved understanding of in situ data collected essentially along a line at the aircraft height.

Airborne lidar and cloud radar are likely remote sensing instruments for studying ice formation and the kinematic structure of clouds. Airborne lidar is ideally suited for mapping out cloud edges and for thinner clouds the two and three-dimensional structure. Cloud radar can provide information on where the first ice occurs, its spread, and the two and three-dimensional structure of ice and precipitation. The Wyoming 95 GHz cloud radar has been used to investigate ice particle formation in orographic wave clouds and is ideally suited for observing ice initiation and spread for ICE.

Remote sensing observations from the ground have contributed strongly in past studies and offer potential for contributing to the new ICE campaign, especially when airborne studies are conducted over a small geographic area. For example, ground-based sensors could be based near the airport at Laramie, Wyoming. This area has frequent wave and layer clouds and is sufficiently removed from Denver that air-traffic routing is usually not a problem. The most useful remote sensors for the study of orographic or mixed-phase layer clouds are scanning millimeter-wave Doppler radar and scanning polarization lidar. Since the data will be collected in the vicinity of the project aircraft, eye-safe lidars would be far more useful, especially for scanning operations. The radar probing will primarily detect the ice crystal targets in and below the clouds, whereas the lidar data will identify ice virga and the cloud base region of the supercooled liquid source cloud (up to a total optical depth of ~3.0). Dual-channel microwave radiometers to measure liquid water depths would also be useful, but many of the thin liquid water clouds likely to be studied here may not generate usable signals.

## 4. Research Facilities and Field Campaigns

Support for several facilities will be requested or included as a part of the basic ICE research package for an initial field campaign in the Colorado Front Range that is planned for the spring of 2007 (ICE-L). 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 most geographic areas of interest and temperatures low enough to measure zones where heterogeneous nucleation predominates. It and can transport a multitude of instruments and investigators. Because the C-130 is inadequate to sample the upper regions of many wave clouds, an upper level aircraft would be desirable to fully characterize the microphysical properties of wave clouds throughout their vertical depth.

- Millimeter cloud radars, in particular, the Wyoming Cloud Radar (WCR). An airborne version would be employed on the NCAR C130, viewing both upwards and downwards simultaneously. The WCR was used on the C-130 during the DYCOMS-2 (Stevens et al., 12003). It was installed on the belly ramp and viewed only downwards. However, engineering design and evaluations to support upward and downwards viewing are underway, with expectations for implementation in time for ICE-L and other field campaigns. The ability to view upwards will allow the C-130 to monitor the existence of any upper level cloud, above the C-130 ceiling, that might affect the clouds being sampled.
- Airborne polarization lidar, such as described in section 3.7.
- *In situ* instruments as described above for high resolution measurement of small ice and other hydrometeors (SID-2) and for high resolution imagery of particles in the 10-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 the CSU 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 sampling, a CCN sampling instrument, and aerosol size measurements that cover a wide size range (~10nm to 1 µm, electrical mobility and optical particle sizing instruments, CN).
- In-situ seeding with well-characterized nuclei (e.g., Arizona test dust) to benchmark our observations of primary nucleation. In addition, a five-year winter cloud seeding project is starting in Wyoming, with operational and research aspects. It includes plans to use the Wyoming King Air for some research flights. It may be possible to merge some ICE-L missions with those King Air flights to gain the advantages of multiple aircraft observations.
- A focused set of models, including the modified WRF model described above operating at both Numerical Weather Prediction (10 km) and Cloud Resolving (< 1km) resolution, CSU's RAMS, and detailed microphysical parcel models. There is also potential synergy with aircraft icing forecast studies in NCAR's *Research Applications Laboratory* (Politovich, 2003).
- Laboratory studies including the AIDA chamber (Germany), extended range CFDC at CSU, electro-dynamic balance freezing chambers (Penn State, University of Washington).

#### 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.
- 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. and R.P. Lawson, 2004: Riming and other characteristics of columns and rosettes observed in wave clouds. *Proc.* 14<sup>th</sup> Intl. Conf. Clouds and Precip., Bologna, WMO, 1887-1890.
- 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).
- 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. Atmos. Sci., 33, 1080-1086.
- Bigg, E.K., 1996: Ice forming nuclei in the high Arctic. Tellus, 48B, 223-233.
- 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.
- Boatman, D. and R. F. Reinking, 1984: Synoptic and mesoscale circulations and precipitation mechanisms in shallow upslope storms over the western High Plains. *Mon. Wea. Rev.*, 112, 1725-1744.
- 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, 2004: Impact of aerosol intrusions on the Arctic boundary layer and on sea-ice multing rates.Part I: May 4, 1998 case. J. Atmos. Sci., accepted.
- 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
- 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.* 14<sup>th</sup> Intl. Conf. Clouds and Precip., Bologna, WMO, 747-750.

- Cooper, W.A., 1974: A possible mechanism for contact nucleation. J. Atmos. Sci., 31, 1832-1837.
- Cooper, W.A., 1986: Ice initiation in natural clouds. in Precipitation enhancement—a scientific challenge), *Meteor. Monographs*, **21**, Amer. Meteor. Soc., Boston, 29-32.
- 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. and Gabor Vali, 1981: The origin of ice in mountain cap clouds. J. Atmos. Sci., 38, 1244-1259.
- 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., DeMott P.J., Brock C., Hudson P.K., Jesse B., Kreidenweis S.M., Prenni A.J., Schreiner J., Thomson D.S., Murphy D.M., 2003: A method for single particle mass spectrometry of ice nuclei. *Aerosol Sci. Techn.*, 37, 460-470.
- 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., Y. Chen, S.M. Kreidenweis, D.C. Rogers and D. Eli Sherman, 1999: Ice formation by black carbon particles, *Geophys. Res. Lett.*, 26, 2429-2432.
- 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, P.J., D.C. Rogers, S. M. Kreidenweis, Y Chen, C. H. Twohy, D. Baumbardner, A. J. Heymsfield, K. R. Chan, 1998: The role of heterogeneous freezing nucleation in upper tropospheric clouds: Inferences from SUCCESS. *Geophys. Res. Lett.*, 25, 1387-1390.
- 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.
- Durant, A. J. and R. A. Shaw, 2005: Evaporation freezing by contact nucleation inside-out. *Geophys. Res. Lett.*, (in review).
- Field, PR, 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.

- 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
- 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.
- Geresdi, I., 1998: Idealized simulation of the Colorado hailstorm case: Comparison of bulk and detailed microphysics. *Atmos. Res.*, **45**, 237-252.
- Gierens, K, 2002: On the transition between heterogeneous and homogeneous freezing. *Atmos Chem. Phys. Discuss.*, **2**, 2343-2371.
- 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.
- Givati, A. and D. Rosenfeld, 2004: Quantifying precipitation suppression due to air pollution. J. Appl. Meteor., **43**, 1038-1056.
- 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
- Hallett, J. 1968: Nucleation and growth of ice crystals in water and biological systems. *Low temperature biology of foodstuff*, Pergamon press Oxford New York, 23-52.
- 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, AJ, 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.

- Hirst E., Kaye P H., Greenaway R S., Field P., and Johnson D W., 2001: Discrimination of micrometre-sized ice and super-cooled droplets in mixed-phase cloud. *Atmos. Environ.*, **35**, 33-47.
- 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. Sci.*, **47**, 2710-2722.
- Hudson, J.G., 1989: An instantaneous CCN spectrometer. J. Atmos. Ocean. Techn., 6, 1055-1065.
- 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
- Hung, H.M., Malinowski, A. and S.T. Martin, 2003: Kinetics of heterogeneous ice nucleation on the surfaces of mineral dust cores inserted into aqueous ammonium sulfate particles. J. Phys. Chem. A, 107, 1296-1306.
- Husar, R.B. et al., 2001: The Asian dust events of April 1998, J. Geophys. Res., 106, 18317-18330.
- 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, The nature and origin of ice nuclei in the atmosphere, *J. Meteorol. Soc. Japan*, 37, 211–233, 1959.
- Jaffe, D, and 12 coauthors, 1999: Transport of Asian air pollution to North America, *Geophys. Res. Lett.*, 26, 711–714, 1999.
- Jirak, I.L., W.R. Cotton, and W.L. Woodley, 2005: Effect of air pollution on precipitation along the Front Range of the Rocky Mountains. 16<sup>th</sup> Conf. Planned Inadvertent Weather Mod., Amer. Meteor. Soc., San Diego.
- 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.
- Kelly, R.D., 1978: Condensation-freezing ice nucleation in wintertime orographic clouds, M. S. Thesis, Dept. Atmospheric Science, Univ. Wyoming, 88pp.
- 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.
- 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.
- Korolev, A.V., G.A. Isaac, I.P Mazin and H. Barker, 2001: Microphysical properties of continental stratiform clouds. Q. Jl. Roy. Meteor. Soc., 127, 2117-2151.
- 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.
- Langer, G., 1973: Evaluation of NCAR ice nucleus counter. Part I: Basic operation. J. Appl. Meteor., 12, 1000-1011.

- 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.
- Lawson, R. P., D. O'Connor, P. Zmarzly, K. Weaver, B. A. Baker, Q. Mo, and H. Jonsson, 2005: The 2D-S (stereo) probe: Design and preliminary tests of a new airborne, high speed, high-resolution particle imaging probe. J. Atmos. Oceanic Technol., (in press).
- 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. and J.F. Penner, 2005: Ice nucleation parameterization for global models. J. Geophys. Res., (accepted)
- 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., Wang, PK, Schlesinger, RE, 2003: A Numerical Study of Cirrus Clouds. Part II: Effects of Ambient Temperature, Stability, Radiation, Ice Microphysics, and Microdynamics on Cirrus Evolution J. Atmos. Sci., 60, 1097-1119.
- Lohmann U, Kärcher B, 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.
- Mazin, I.P., Korolev, A. V., Heymsfield, A., Isaac, G. A., Cober, S. G., 2001: Thermodynamics of icing cylinder for measurements of liquid water content in supercooled clouds. J. Atmos. Ocean. Techn., 18, 543-558.
- 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.
- Möhler, O., O. Stetzer, S. Schaefers, C. Linke, M. Schnaiter, R. Tiede, H. Saathoff, M. Krämer, A. Mangold, P. Budz, P. Zink, J. Schreiner, K. Mauersberger, W. Haag, B. Kärcher, and U. Schurath, 2003: Experimental investigation of homogeneous freezing of sulphuric acid particles in the aerosol chamber AIDA. Atmos. Chem. Phys., 3, 211-223.
- Möhler, O., C. Linke, H. Saathoff, M. Schnaiter, R. Wagner, U. Schurath, A. Mangold, M. Krämer, 2005: Ice nucleation on flame soot aerosol of different organic carbon content. Submitted to *Meteorol. Ziet.*
- Mossop, S.C., 1963: Atmospheric ice nuclei. Z. angew. Math. Phys. 14, 456.
- Mossop, S. C., and J. Hallett, 1974: Ice crystal concentration in cumulus clouds: Influence of the drop spectrum. *Science*, **186**, 632–634.

- 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.
- Politovich, M.K., 2003: Predicting in-flight aircraft icing intensity. J. Aircraft, 40, 639 644.
- Politovich M.K. and B.C. Bernstein, 1995: Production and depletion of supercooled liquid water in a Colorado winter storm. J. Appl. Meteorol., **34**(12), 2631-2648.
- 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 Hobbs P.V. 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., B. Bernstein, M. Murakami, G. Stossmeister, and B. Stankov, 1995: The 1990 Valentine's Day Arctic outbreak. Part I: Mesoscale and microscale structure and evolution of a Colorado Front Range shallow upslope cloud. J. Appl. Meteor., 34, 1481–1511.
- 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., 1988: Development of a continuous flow thermal gradient diffusion chamber for ice nucleation studies. *Atmos. Res.*, 22, 149-181.
- 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. and P.J. DeMott, 1995: Measurements of natural ice nuclei, CCN and CN in winter clouds. *Amer. Meteor. Soc. Preprints, Conf. Cloud Physics*, 15-20 January, Dallas, TX, 139-144.
- 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.
- Rogers, D.C., and P.J. DeMott, 2002: Ice crystal formation in wave clouds, airborne studies -10 to -35°C. *Amer. Meteor. Soc. 11<sup>th</sup> Conf. Cloud Physics*, 3-7 June 2002, Ogden, UT. (CD-Rom) P1.16.
- Rogers, D.C. and G. Vali, 1987: Ice crystal production by mountain surfaces. J. Clim. Appl. Meteor., 26, 1152-1168.
- 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., 1984: Deep orographic cloud structure and composition derived from comprehensive remote sensing measurements. J. Climate Appl. Meteor., 23, 568-583.
- Sassen, K. 2002: Indirect climate forcing over the western US from Asian dust storms. *Geophys. Res. Lett.*, 29, 10.1029
- Sassen, K. and G. Dodd, 1988: Homogeneous nucleation rate for highly supercooled cirrus cloud droplets. J. *Atmos. Sci.*, 45, 1357-1369.
- Sassen, K. and S. Benson, 2000: Ice nucleation in cirrus clouds: a model study of the homogeneous and heterogeneous modes. *Geophys. Res. Lett.*, **27**, 4, 521-524.
- 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., 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
- Seifert, M., J. Ström, R. Krejci, A. Minikin, A. Petzold, J.-F. Gayet, U. Schumann, and J. Olvarlez, 2003a: Insitu observations of aerosol particles remaining from evaporated cirrus crystals: Comparing clean and polluted air masses. *Atmos. Chem. Phys.*, 3, 1037-1049,
- Seifert, M., J. Ström, R. Krejci, A. Minikin, A. Petzold, J.-F. Gayet, H. Schlager, H. Ziereis, U. Schumann, and J. Olvarlez, 2003b: Thermal stability analysis of particles incorporated in cirrus crystals and of nonactivated particles in between the cirrus crystals: Comparing clean and polluted air masses. *Atmos. Chem. Phys.*, 3, 3659-3679.

- 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.
- Stevens, B. et al. (32 more authors), 2003: Dynamics and Chemistry of Marine Stratocumulus DYCOMS-II, *Bull. Amer. Meteor. Soc.*, **84**, 579-593.
- 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.
- 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.
- 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., D. Starr, A. Hou, P. Newman, Y. Sud, 2003: A Cumulus Parameterization Workshop. Bull. Amer. Meteor. Soc., 84, 1055–1062.
- 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.* 10<sup>th</sup> 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.
- Twomey, S., 1977: The influence of pollution on the shortwave albedo of clouds. J. Atmos. Sci., 34, 1149-1152.
- VanCuren, R. A., and T. A. Cahill, 2002: Asian aerosols in North America: Frequency and concentration of fine dust. J. Geophys. Res., 107, doi:10.1029/2002JD002204.
- Vardiman, L, 1978: The generation of secondary ice particles in clouds by crystal crystal collision. *J. Atmos. Sci.*, **35**, 2168 2180.
- Weickman, H., 1981: Mechanism of shallow winter-type stratiform cloud systems. NOAA Tech. Memo, ERL, NTIS PB82-170176, 61pp.
- Whiteman, C.D., 1973: Some climatological characteristics of seedable upslope cloud systems in the High Plains. NOAA Tech. Rep. 268-APCL-27, NTIS-COM-73-50924/2GI, 43pp.
- 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

- 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. Appendices

## 6.1 Experiment Plans

### 6.1.1 Layer and Wave Clouds

A basic pattern for wave cloud flights with one aircraft involves upwind-downwind transects through the cloud from top to base and below. The intent is to sample the microphysical, kinematic, thermodynamic, and aerosol properties throughout a layer approximately 1000 m thick. The highest altitude is slightly above cloud top, and the lowest altitude is slightly below where a parcel reaches ice saturation in adiabatic ascent. Since wave clouds are relatively steady-state, the data from transects at several altitudes can be combined to produce a vertical cross section of the region in which the cloud forms. Within this region, trajectories of air parcels can be constructed and compared with equivalent numerical model and laboratory experiments.



Figure 9. Wave cloud flight plan on vertical projection aligned with wind. Wind streamlines are sinusoidal. The aircraft makes successive passes at different altitudes from cloud top to base and below, including ice supersaturated region (hatched).

Since wave clouds often repeat in wave trains (i.e., a succession of clouds along the same streamlines), there may be opportunities for longer downstream or upstream transects through repeatedly processed air. In this case, the basic flight plan is modified to focus on cloud processing. Initially, the aircraft would fly pattern #1 on the most upstream wave cloud. Subsequent air sampling and cloud penetrations would occur between and through the wave clouds downstream.

For wave clouds with tops higher than the practical ceiling of the C-130 (~23,000 ft), a second (jet) aircraft that can make microphysical measurements in colder regions of cloud is advantageous. Airborne lidar and radar observations could be done with additional instrumentation on the cloud-penetrating aircraft or with a second aircraft flying in coordination at higher or lower altitudes.

#### 6.1.2 Altostratus - altocumulus layers

The sampling strategy for altostratus/altocumulus clouds is represented in Figure 6. First, the vertical thermodynamic and cloud microphysical structure is surveyed with a saw-tooth run that extends vertically from below cloud base to above cloud top. Second, in-cloud straight and level runs through the center of the cloud and very close to cloud top should be performed to further characterize the microphysical structure and attempt to detect ice shortly after formation. Third, straight and level runs in clear air above and below cloud should be performed to sample aerosol that may be ingested into the cloud system. Given sufficient time this sampling strategy should be repeated to observe the evolution of the cloud system.

Simultaneous lidar and radar observations of the cloud system should be made to identify any liquid layers and provide a larger scale context in which to place the aircraft observations. This remote sensing support could be with the same aircraft, a second aircraft, or ground-based instruments.



Figure 10. Vertical cross section showing aircraft flight patterns (arrows) for sampling in mid-level layer clouds.

#### 6.1.3 Convective Clouds

Figure 7 illustrates a typical flight sampling strategy for isolated convective clouds such as cumulus congestus. The ideal experiment will be to sample an isolated growing region, so that the history of the upper cloud regions could be documented as the rising top encounters cold temperatures. The region below the base should be sampled so that the accurate thermodynamic properties of the cloud base can be measured. Second, details of the lower and mid regions can be documented during ascending spirals (or stair-step climbs—leveling off or holding a heading for periods to improve the wind data) that go in and out of cloud. Third, the growing cloud top can be sampled during its ascent. Finally, although some aerosol information is available during the upward spirals, dedicated aerosol sampling at several altitudes will be accomplished during a descending spiral, leveling off at various altitudes for aerosol sampling. As for the layer cloud experiments, airborne remote sensing could be done with the cloud-penetrating aircraft or a second aircraft flying in coordination at higher or lower altitudes.



Figure 11. Single cloud or cloud turret flight sampling plan for cumulus clouds.