2.1 ICE NUCLEI VARIABILITY, RELATION TO AMBIENT AEROSOL PROPERTIES, AND IMPACTS ON MIXED-PHASE CLOUDS

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1. INTRODUCTION

First ice formation in mixed-phase clouds is thought to result from special particles known as ice nuclei (IN). IN from natural and possibly manmade sources thereby impact issues ranging from precipitation prediction to cloud radiative forcing (ice indirect effects). After a consideration of our capabilities for detecting IN, we review evidence supporting that IN measurements predict first ice formation in clouds and present results regarding the role of aerosol size and mineral dust transports on impacting the spatial, temporal, and seasonal variability of IN. We conclude with a few notes on the implications of these results and research needs.

2. MEASURING ICE NUCLEI (IN)

Ice nuclei typically represent only a small fraction of the population of all particles. They possess the capability to heterogeneously activate ice formation and may do so by multiple pathways that can depend on temperature, relative humidity, particle size and composition, and time (Figure 1). The Colorado State University Continuous flow diffusion chamber (CFDC) processes particles for specific, but variable, conditions of temperature and relative humidity (RH) via the confinement of a cylindrical aerosol particle lamina within a vapor diffusion field created between cylindrical ice-coated walls held at different temperatures. Readers are referred to Rogers et al. (2001a) and references therein for a complete description of the technique. Exposing particles to constant conditions for several seconds residence time does not specifically limit the ice formation mechanisms examined, although CFDC conditions can be selected to favor contributions from one or more mechanisms and it is indeed the total contributions of all mechanisms at a given temperature which one often desires to know for direct comparisons to cloud ice formation.

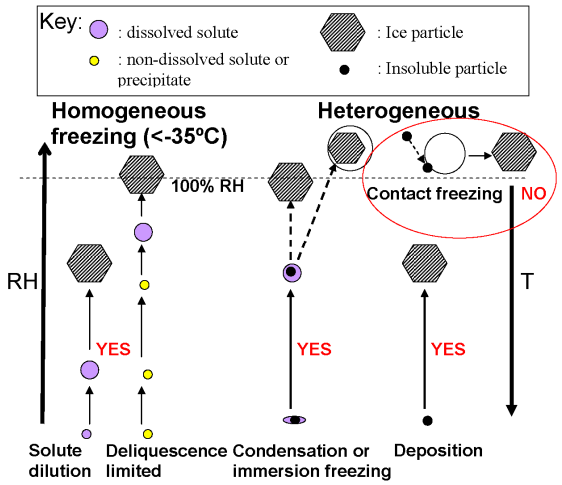

Figure 1. A conceptual representation of ice formation mechanisms indicating (in red) those that are directly measured by the CFDC technique. See the text for further description.

No existing portable ice nucleus measuring system is capable of detecting ice formation by all known mechanisms. Figure 1 indicates the mechanisms for ice formation that are sensed using the CFDC technique. The CFDC has previously been used for measurements of homogeneous freezing of aqueous solution particles (left side of Figure 1) (DeMott et al. 2003a). Typically, heterogeneous ice nucleation is limited to deposition nucleation (vapor to ice formation on a particle surface) within the RH regime below water saturation. This process is observed in laboratory CFDC studies of mineral dust type particles (Archuleta et al. 2005). Condensation freezing, specifically condensation immediately followed by freezing, can occur in this same RH regime for solute-concentrated haze

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particles, but usually at much lower temperatures (e.g., below -30ºC). It is distinguished from immersion freezing by the fact that immersion freezing nuclei are those present within a liquid droplet (due to nucleation or other scavenging) that undergoes cooling prior to achieving a freezing temperature condition. The effectiveness of both freezing mechanisms depends not only on temperature but on the peak RH achieved in a parcel, since this can determine both the water content of particles and the numbers activated as CCN in the water supersaturated regime. We generally assume that the CFDC technique detects both freezing mechanisms, but it will not effectively detect these nuclei if the freezing process is strongly stochastic. The rate of contact freezing nucleation is dependent on the collision rate between nuclei and liquid droplets in clouds. The CFDC does not have sufficient residence time to effectively detect contact freezing nuclei. Nevertheless, based on the studies of Shaw et al. (2005) suggesting that an immersion freezing nucleus will act as a contact freezing nucleus some 4 to 7ºC warmer, one might constrain the numbers of contact freezing nuclei by operating a CFDC at relatively high water supersaturations (DeMott et al. 1998) to encapsulate all particles within drops and measure the immersion freezing nuclei temperature spectrum. This suggestion would be invalid if the population of potential contact freezing nuclei is much larger because a larger proportion of all particles possess enhanced efficiency for freezing on contact for transient periods during certain cloud processing, as suggested by Beard (1992).

A final important note regarding IN sampling is that it occurs on aircraft via an inlet. The inlet may be a forward-facing aerosol inlet for collecting particles irregardless of clear or cloudy conditions, or it may be a counterflow virtual impactor (CVI) to sample only cloud particles above a certain size. In either case, aerosol particles are exposed to heating, whether the dynamic heating that occurs in standard inlets or the forced heating of cloud particles in a CVI that is required to drive water off the particles prior to reprocessing in instruments such as the CFDC. Heating may alter the ice nucleating properties of atmospheric particles.

3. RESULTS

3.1 *IN relation to ice formation in clouds*

Existing evidence for the correspondence between IN and ice formation in clouds is rather modest. Rogers and DeMott (2002) show correspondence between IN measurements by the CFDC technique and ice concentrations in modestly supercooled orographic wave clouds. In Figure 2 we show some more recent evidence from the Alliance Icing Research Study II (AIRS-II) in Southeast Canada suggesting that the first appearance of ice in clouds is directly linked to the presence of IN feeding the clouds and that the atmosphere may at times be quite inhomogeneous in this regard. CVI cloud particle residual nuclei that were reprocessed by the CFDC showed a lack of IN in a cloud that contained little or no ice, while the nuclei from a nearby cloud containing ice also contained IN.

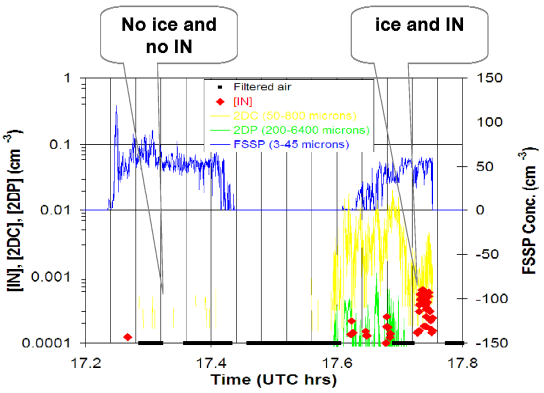

Figure 2. Cloud particle and ice nuclei concentrations (1-min averages) measured from reprocessed cloud particle residuals that were initially sampled via a counterflow virtual impactor inlet during transit through two successive stratocumuli cloud lines during the AIRS-II study. The presence of ice in the clouds, particularly as indicated by larger particles on the 2D probes, was directly associated with the presence of IN. CFDC and cloud temperatures were equivalent at about -16ºC and the CFDC RH was 102%.

Figure 2 indicates that correspondence between IN and ice concentrations was only modestly good in some regions of the second cloud sampled. This situation is duplicated in many other scenarios of sampling IN from cloud particle residuals. An example from a deeper cloud system is shown in Figure 3. While [IN] correlated within a factor of 3 with 2DC probe ice crystal concentrations in the upper ice cloud regions, [IN] were a factor of 50 lower on average in other regions of the cloud. This may have reflected the existence of a secondary ice formation process or may reflect IN sampling issues. Nevertheless, the discrepancy between IN and ice concentrations in some clouds is an issue that has been ever present in the cloud physics

literature for many years and remains unresolved. It is quite possible that there remain unidentified ice formation mechanisms.

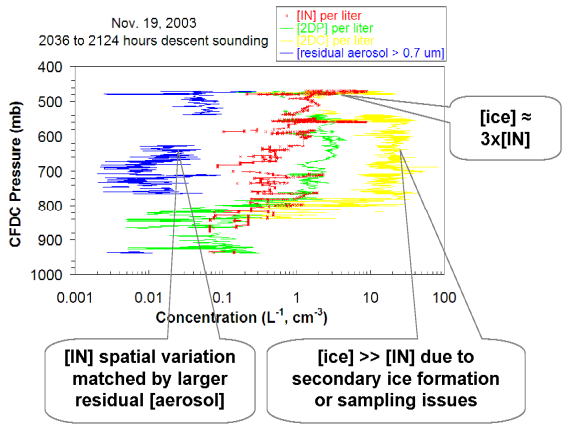

Figure 3. Cloud particle, ice nuclei, and aerosol concentrations measured from reprocessed cloud particle residuals that were initially sampled via a CVI inlet on the NCAR C-130 aircraft during a combined spiral/stepped descent profile through a deeper cloud system during the AIRS-II study. Cloud top temperature (not shown) was near -23ºC and base temperature was 3ºC. The CFDC processed IN at -13ºC and RH = 102%. The cloud was mostly ice in the upper regions, but liquid was present with ice in the lower regions near and above the freezing level.

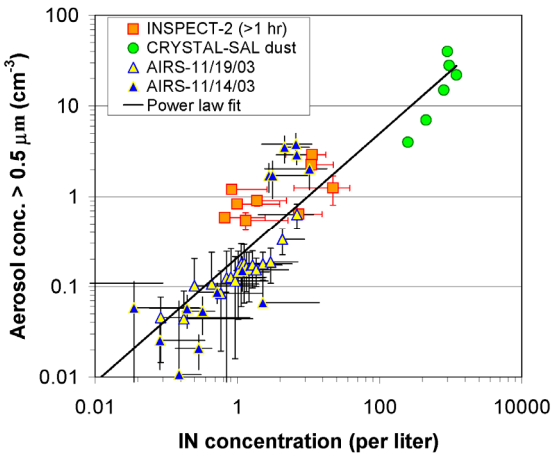

Figure 4. Ice nuclei concentrations related to the concentration of aerosol particles at sizes above 0.5 μm in three projects. The data are from measurements in and around mixed-phase clouds in AIRS-II, in clear air at Storm Peak Laboratory (Colorado) during INSPECT-2 in April-May 2004 (Richardson et al. 2006), and from within the Saharan Aerosol Layer (SAL) during the NASA CRYSTAL-FACE study in 2002 (DeMott et al. 2003b). A power law (aerosol concentration = $0.2084[IN]^{0.6894}$) fits these data with an $r^2 = 0.75$.

3.2 *IN relation to aerosol size, season and locale*

Two additional points evident in Figure 3 are the strong spatial variability of IN and the relation between IN concentrations and the concentrations of all cloud-residual aerosol particles of larger sizes. Residual aerosol particles at sizes above 0.3 μm were measured in AIRS-II using the same optical particle counter (OPC) system that detects IN by their growth as ice crystals to sizes above 2 μm. We are exploring the relation between IN and aerosol particle size distributions in existing CFDC data sets from programs representing a range of locales and seasons. Figure 4 shows a selection of IN concentration data (three projects represented, as indicated in figure caption) correlated with the concentrations of aerosol particles exceeding 0.5 μm. The selection of this aerosol particle size was guided primarily by the availability of aerosol data at these sizes in all projects rather than identification of a specific relation of highest correlation. Further, aerosol concentrations were measured by different instruments in the separate studies, a factor that may contribute to the scatter of data in Figure 4. Nevertheless, data of this type could provide a powerful means for formulating ice initiation parameterizations for use in numerical modeling.

Richardson et al. (2006) show that relations of IN to larger aerosol concentrations are in many cases a direct consequence of the presence or absence of mineral dust particles. This may explain differences in the probability distributions of [IN] measured in studies conducted at different locations and at different times of the year. For example, Figure 5 shows that IN measurements in similar Arctic regions and with similar CFDC processing conditions during spring and fall seasons show the distinctly different [IN] data that may be the signature of cycles of mineral dust transports. Namely, measurements during the spring season FIRE-ACE/SHEBA project in 1998 (Rogers et al. 2001b) reflect, on average, quite high [IN], similar to the dust-affected midlatitude spring data of the INSPECT-2 (Ice Nuclei Spectroscopy Study – 2) project (Richardson et al. 2006). In contrast, average [IN] measured during the Mixed Phase Arctic Cloud Experiment (M-PACE) study (Prenni et al. 2006) during the Arctic fall season, when dust transports to the area are at a minimum, are among the lowest ever measured. Similarly, a large percentage of the IN concentrations measured during the NASA CRYSTAL-FACE study (DeMott et al. 2003b) are

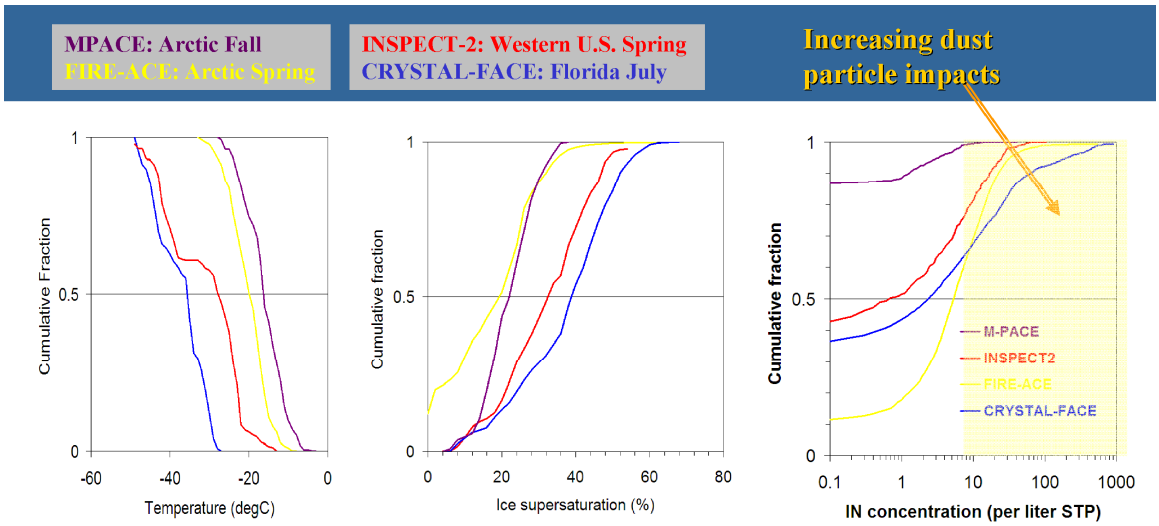

Figure 5. Cumulative fractions of 1 minute observations of CFDC temperature, ice supersaturation, and IN concentration in four research projects. Arctic region data are from the DOE M-PACE (Fall 2004) and NASA FIRE-ACE (Spring 1998) programs, while mid-latitude data are for the NSF INSPECT-2 (Spring 2004) and NASA CRYSTAL-FACE (Summer 2002) programs. Dust aerosol impacts are evident in the region of frequency of IN concentrations exceeding 10 per standard liter.

among the highest ever measured in ambient air and reflect the direct impact of Saharan aerosol transports to the Florida region during July.

4. SUMMARY

Current measurements support that ice nucleating aerosols are critical for the initiation of ice in clouds. The data also strongly support the need for more coordinated studies of ice nuclei and ice formation in clouds in order to resolve and understand the roles of known primary (IN), secondary, and currently unresolved ice formation processes in determining cloud properties.

Mineral dust aerosol particles are well known and now well fairly well documented sources for IN populations. While many other potential IN sources (primary and secondary aerosol contributions from combustion processes and biological processes in particular) remain unconstrained, a consequence of the relation between IN and mineral dust sources is that [IN] relate to the concentrations of larger aerosol particles. Research continues on quantifying this relationship to test for robustness in a variety of locales and seasons as this information could be quite powerful from a numerical modeling perspective. Nevertheless, a great deal of research remains to understand the multivariate sensitivities of [IN] on temperature, humidity and time.

Cloud properties may be particularly sensitive to ice nuclei at certain times and locations, particularly for periods favoring low mineral dust mass loadings and low ice nuclei concentrations. The M-PACE project and fall Arctic season seems to generally fit in this latter category (Prenni et al. 2006), with strong implications for cloud phase and the surface radiation budget, although spatial variations of ice nuclei and impacts on clouds in that study are the subject of continued study.

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