Ph.D. Defense Announcement
Ryan Patnaude
Monday, September 25, at 2:00 pm

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Ph.D. Defense

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Defense
ATS Large Classroom (101 ATS) or Teams

Post Defense Meeting
Riehl Conference Room (211 ACRC)

Committee:
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USING LABORATORY AND AIRBORNE MEASUREMENTS TO INVESTIGATE THE ROLE OF ICE NUCLEATING PARTICLES IN ICE AND MIXED-PHASE CLOUDS

Ice may be present in the atmosphere either in cirrus or mixed-phase cloud regions, each with their own distinctly different characteristics and formation mechanisms. The former is characterized by the presence of only ice crystals at temperatures < -38 ºC, while the latter includes the coexistence of both supercooled liquid cloud droplets and ice crystals between temperatures of 0 ºC and -38 ºC. Cirrus clouds represent an important cloud type as they are ubiquitous in the atmosphere and their radiative effects depend upon their microphysical properties. Their formation mechanisms may proceed via homogeneous or heterogeneous nucleation, and whether one or the other or both occur determines the size and number of ice crystals.

The ocean represents one of the largest sources of aerosols into the atmosphere, and sea spray aerosols (SSA), if they are lofted to the upper troposphere, may act as ice nucleating particles (INPs) to initiate heterogeneous nucleation under cirrus conditions. Although a number of previous studies have investigated the ice nucleating behavior of SSA proxies such as sodium chloride (NaCl), or SSA generated from commercially-available artificial seawater products, ice nucleation under cirrus conditions of SSA generated from natural seawater had not been examined at the inception of this research program. Additionally, whether secondary marine aerosols (SMA), which form via the gas-to-particle conversion of ocean-emitted gas-phase species, may act as an INP in cirrus clouds is currently unknown. The first half of this dissertation highlights two laboratory studies that investigated the role and characteristics of SSA and SMA to act as INPs at cirrus cloud temperatures.

The first study compared ice nucleation results for submicron SSA and NaCl particles and examined whether particle size affected the low temperature ice nucleation. Results showed that both SSA and NaCl initiated heterogeneous nucleation strongly at temperatures below 220 K, and that the size of the particles did not affect the ice nucleating ability of SSA. The similarities between the freezing behaviors of SSA and NaCl particles suggested the salt components were controlling heterogeneous ice nucleation. The second study used a more realistic aerosol generation method, utilizing a Marine Aerosol Reference Tank (MART) that was filled with natural seawater, and investigated the effects of atmospheric oxidation on SSA using an oxidation flow reactor (OFR), which was also used to generate SMA from gaseous emissions released in the MART. SMA alone were also examined for their ice nucleation behavior at cirrus temperatures. Results from this study indicated that atmospheric oxidation did not affect low temperature ice nucleation of SSA, and that SMA are not efficient ice nucleating particles at cirrus temperatures, but could participate in homogeneous nucleation. Finally, the similarities between the findings from the two studies indicated that the generation method of SSA, and any impacts on SSA organic aerosol content, did not affect the ice nucleating behavior of SSA at cirrus temperatures.

Ice in mixed-phase clouds (MPCs), on the other hand, forms initially via heterogeneous nucleation at a wide range of temperatures and relative humidity conditions, depending on the abundance and characteristics of available INPs. Secondary ice production (SIP) may follow heterogeneous nucleation in MPCs, where new ice crystals form either during the heterogeneous freezing event, or through subsequent interactions between the pre-existing liquid cloud droplets and ice crystals. SIP may lead to enhanced ice crystal number concentrations via a number of proposed mechanisms, especially in convective environments. Despite decades of study toward developing better understanding of ice formation in MPCs, the freezing pathways of ice crystals over the course of cloud lifetimes, and the conditions that favor the various proposed SIP pathways, are not fully resolved.

The third study in this dissertation reports and interprets observations of INPs during an airborne campaign over the U.S. Central Great Plains during the Secondary Production of Ice in Cumulus Experiment (SPICULE) campaign that primarily sampled cumulus congestus clouds. Coincident measurements of INP and ice crystal number concentrations in cumulus congestus clouds were used to infer the ice formation pathway, either through heterogeneous nucleation or SIP. Warmer cloud base temperatures and faster updrafts were found to facilitate environmental conditions favorable for SIP. Further, the fragmentation of freezing droplets (FFD) SIP mechanism was found to be critical in the enhancement of ice crystal number concentrations during the earliest stages of the cloud lifetime. Numerical model simulations of an idealized, single congestus cloud, designed to mimic the clouds sampled during SPICULE, were conducted with newly-implemented SIP mechanisms, added to the existing Hallet-Mossop rime-splintering mechanism. The model results indicated that without the FFD SIP mechanisms, the model could not accurately resolve ice crystal number concentrations compared to observations. Inclusion of FFD, which commenced following ice initiation through INPs provided the best closure on the sequence of ice formation processes that occurred during the early stages of convective cloud evolution.

The final portion of this dissertation describes airborne observations of INPs during a field campaign along the U.S. Gulf Coast, also aimed at investigating the impacts of various aerosol-cloud interaction mechanisms on development of convective clouds. During this campaign, a widespread and prolonged Saharan Air Layer (SAL) event took place and INP characteristics during this event are reported and contrasted with INP characteristics prior to the arrival of the SAL. The INP concentrations at temperatures below -20 ºC were enhanced by 1–2 orders of magnitude compared to the flights prior to the dust intrusion, and showed good agreement with one previous study of Saharan dust near Barbados, but lower INP concentrations than another study off the coast of western Africa. The INP concentrations in the SAL also generally overlapped with or exceeded INP concentrations during SPICULE, but only for INPs active temperatures < -25 ºC. These observations were the first airborne measurements in nearly two decades tagging INP concentrations to North African dust that had been transported all the way to the United States. Further, they provide the most comprehensive description of these INPs yet recorded, and suggest a common natural INP perturbation in the southeastern U.S. and Gulf regions in early summer, with implications for cloud processes that warrant further study.