# Investigation of the Impact of CALIPSO Measured Aerosol Variations on Arctic Stratus Detected by CloudSAT and SEARCH Measurements During the International Polar Year

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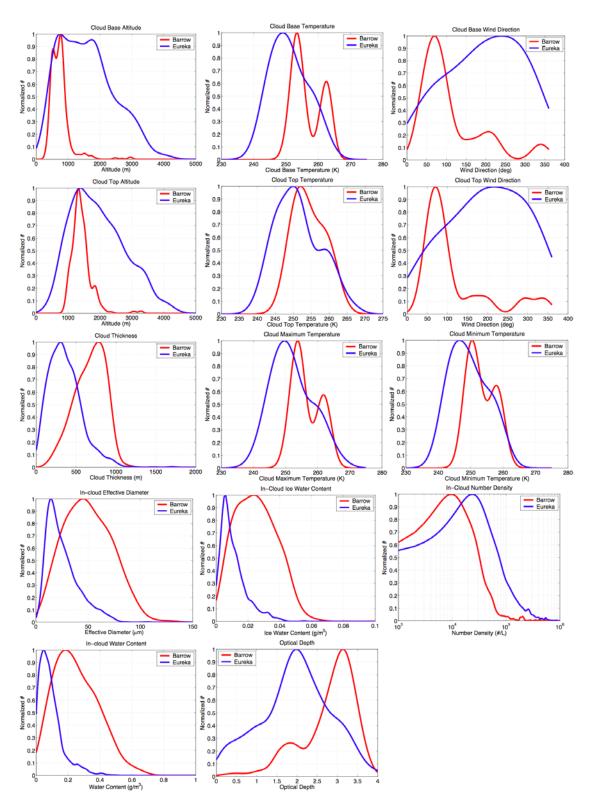
#### 1. Introduction

This report outlines findings from NASA grant number NNX07AQ81G. This project was to increase our understanding of ice nucleation processes in Arctic Stratus clouds through the combined use of lidar and cloud radar sensors looking down from space (CALIPSO, CLOUDSAT) and looking up from the ground (UW AHSRL and NOAA MMCR). Specifically, we strived to understand the ice-aerosol interactions leading to heterogeneous nucleation of ice within the mixed-phase cloud layer. These processes have not been captured well by models, due to oversimplified nucleation schemes, and a lack of accurate aerosol handling. Since the formation of ice in these clouds has a large influence on the extended survival of cloud liquid, improper handling of ice nucleation can lead to unrealistic lifetimes, often too short, for the cloud structure. The following sections outline the results and resulting publications of this project.

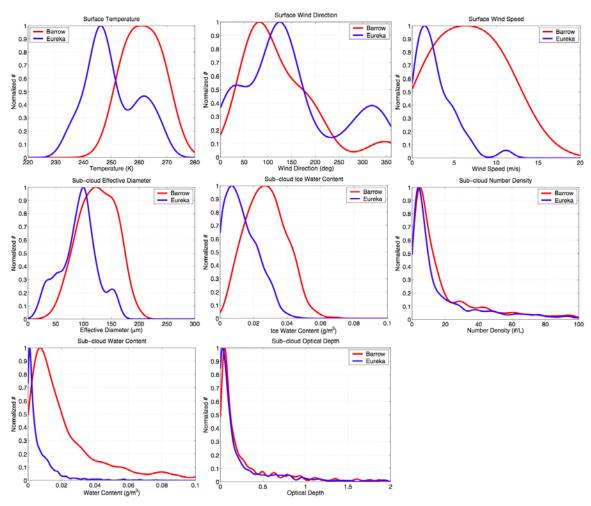
## 2. Overview of Work Completed

#### 2.1. Collection/Evaluation of data from satellite an ground-based sensors

In order to gain a more thorough understanding of characteristics shared by these clouds, we utilized data collected by the University of Wisconsin Arctic High Spectral Resolution Lidar (AHSRL), NOAA and CANDAC Millimeter Cloud Radars (MMCR), and the University of Idaho Polar Aeri (PAERI) to establish a multi-year dataset of cloud properties. To accomplish this, all measurements from the previous two and a half years in Eureka were manually reviewed for mixed-phase stratus cases. All cases were divided into half hour segments. From these half hour sections, average values of cloud base height, cloud top height, cloud thickness, cloud optical depth, and precipitation optical depth were collected. Additionally, estimates of average cloud and precipitation particle number density, particle effective size, and water contents were made using combined lidar-radar retrieval methods (Donovan and Van Lammeren, 2001). Distributions of average values for each variable over half hour periods are shown in figures 1 and 2. Some highlights of this work include validation of a large temperature range over which these clouds exist (240-270 K), a characterization of some of the main microphysical properties of the clouds and precipitation, and discovery of significant differences between clouds observed at Eureka and those observed in Barrow. Plus, distributions of surface and in-cloud wind directions show distinct peaks, hinting at the sources of moisture and possibly aerosols. More detailed results from this work, as well as an overview of methods used will be included in an article currently in preparation (de Boer, 2008b).



**Figure 1:** Distributions of mixed-phase stratus cloud properties from Eureka (blue lines) as compared with Barrow (red lines).



**Figure 2:** Distributions of precipitation and surface properties for time periods with mixed-phase stratus clouds from Eureka (blue lines) and Barrow (red lines).

In addition to the resulting information about mixed-phase cloud properties, this work also helped lead to important findings about the ground-based methods used to observe them, and our current state of observational competence. In a collaborative effort with scientists from several other institutions, it was determined that by far the most accurate observations we can achieve are those of cloud macrophysical properties such as cloud heights and dimensions. Microphysical properties were found to be significantly more difficult to accurately portray, particularly those pertaining to the liquid portion of mixed-phase clouds. Also, measurement of characteristics of aerosols associated with the formation of these clouds was shown to be severely lacking. Further information will be available in the Shupe et al. (2008) article to be published in the Bulletin of the American Meteorological Society.

Ground-based information was also utilized to do a preliminary evaluation of some of the CloudSAT products to be used in analysis of these clouds. Because of the difficulties associated with observing mixed-phase clouds combined with the newness of the CloudSAT instrument and data, this evaluation was necessary to better understand our capabilities in using this data. CloudSAT algorithms readily detected clouds in all of the cases evaluated, though they were not always recognized to be stratus or stratocumulus. Although some microphysical estimates from these products showed agreement with ground-based values, there were also some major discrepancies that came out of this comparison, particularly for the liquid retrievals (figure 3). In the end, this work revealed that some significant improvements are needed in the CloudSAT data products in order to efficiently utilize these measurements for mixed-phase cloud research. A more detailed explanation of these results is given in de Boer (2008a).

In order to make the comparison between the ground-based and A-Train derived measurements, a web tool was developed to easily provide dates and times where the A-Train passes within a given distance from the Eureka site. Not only does this interactive webpage provide the dates and times, but it also provides quick-look type images of the data from ground based sensors, so that investigators looking for specific types of events (such as mixed-phase stratus cases) can easily identify these cases for dates that have coincident ground and space born measurements. This tool is available for use by the general science community at <a href="http://lidar/cgi-bin/processeddata/retrievedata.cgi">http://lidar/cgi-bin/processeddata/retrievedata.cgi</a> by toggling the "file mode" parameter to "satellite".

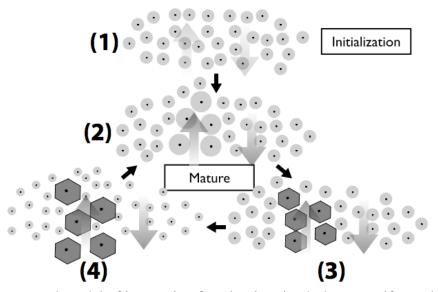


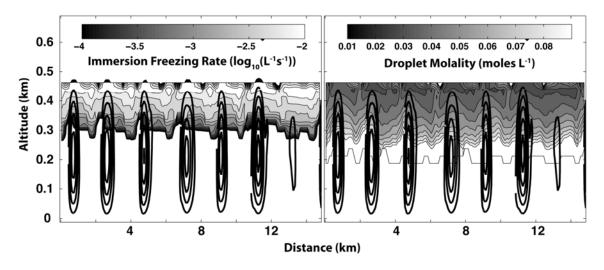
Figure 3: A conceptual model of immersion freezing in mixed-phase stratiform clouds.

#### 2.2. Analysis of Observations

This study was published in the Journal of Atmospheric Science (de Boer et al., 2009a). From this study and others, it is clear that Arctic Mixed-Phase clouds occur frequently. In addition, an assessment of CloudSAT's ability to detect these clouds was completed in this work, and it was determined that for the current location, CloudSAT would have difficulties detecting approximately 10% of all observed cases due to their altitude (cloud top < 1000m), and approximately 8% due to their low maximum reflectivity (Z < -29 dBZ). Based on observational evidence from these studies, as well as in-situ observations, a theory focusing on immersion freezing as a dominant nucleation mechanism within mixed-phase stratiform clouds was developed (Figure 3). An article describing this theory is currently in review for publication in Atmospheric Research (de Boer et al., 2009b).

#### 2.3. High-resolution simulation of local cloud processes

Because of instrument limitations, one of the most powerful tools available for improving our understanding of cloud processes and testing the above theory on immersion freezing is cloud-resolving simulation. A series of simulations of a mixed-phase cloud layer observed during the Surface Heat Budget of the Arctic (SHEBA) campaign were completed as a part of the SHEBA Global Cloud System Study (GCSS) model inter-comparison. These simulations are in addition to those completed for an earlier inter-comparison based on observations from Barrow, Alaska. Results from the later study appeared in the Quarterly Journal of the Royal Meteorological Society (Klein et al., 2009).



**Figure 4:** Immersion freezing rate (left) and droplet molality (right) from simulations of mixed-phase stratiform clouds. Upward atmospheric vertical motion is contoured in black lines.

Comparison of simulated ice production rates through multiple modes of nucleation showed that immersion freezing produced a large fraction of ice as compared to deposition/condensation freezing. Based on the results of these simulations, it is certain that rates of immersion freezing and deposition/condensation freezing are at the very least comparable.

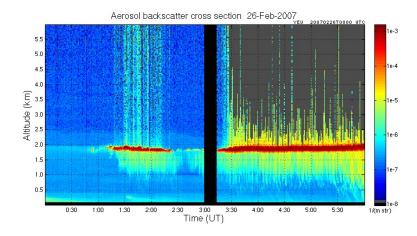
A modeling analysis of the effects of CCN properties on cloud lifetime was also completed. Aerosol soluble mass fraction was found to influence the initiation of freezing via the immersion mode by requiring droplet growth to larger sizes when soluble mass fraction was increased (Figure 4). These larger droplets were found to form near cloud top in simulations completed with the immersion freezing mode active. Originally, these droplets were hypothesized to grow through expansion within updrafts. This hypothesis was shown to correctly predict the location of elevated immersion freezing rates. However, completed simulations also revealed that particles were nucleated via immersion freezing over downdrafts. The downdraft nucleation regions were shown to be the result of isobaric radiative cooling near the cloud top, which resulted in a colder and more humid environment. Despite ice particles nucleating throughout the upper portions of cloud layers, simulated radar reflectivity features maximum values in regions of ascent, similar to observations. This occurs because of the large volume of the particles growing in and falling out of the updrafts, many of which have been rimed during their extended lifetime within the super cooled liquid layer.

In addition to soluble mass fraction, CCN insoluble mass type was also found to have a large influence on freezing via the immersion mode. Droplets forming on aerosol particles containing insoluble fractions with high freezing efficiencies (e.g. illite, montmorillonite) froze at smaller sizes than those containing particles with lower freezing efficiencies (e.g. kaolinite, soot). This effect was found to be more influential than that imposed by the aerosol soluble mass fraction. An article describing the study of aerosol effects was published in the Journal of Geophysical Research later this summer (de Boer et al., 2009c).

# 2.4. Collection, evaluation and processing of SEARCH, CALIPSO and CloudSAT data for specific case study cases

Analysis of particular dates for use as a case study continues to be an evolving process. Currently, cases are being sought in which aerosol layers evolve into mixed phase cloud layers. An example of an interesting case is the morning of 26 February, 2007 (Figure 5, top). Here, a liquid layer forms from an aerosol layer. The 00Z sounding indicates that the atmosphere is

saturated with respect to ice at the altitude where an aerosol layer is found. Since ice does not nucleate at all until after a liquid layer forms, this implies that there is something about the aerosol type that is not conducive to direct ice formation. This would support the immersion freezing hypothesis alluded to in the previous section. To further investigate these interactions, we are analyzing links between aerosol plumes and ice nucleation using measurements by AHSRL and CALIOP along with twice daily temperature and dewpoint profiles obtained from Eureka soundings. Figure 5 (bottom) illustrates the frequency of occurrence of low lidar backscatter cross-sections (indicating cloud and hydrometeor free air) under different levels of relative humidity with respect to ice. A database of cases such as this is being collected and analyzed to gain a better understanding of how commonly cases with aerosols not readily nucleating ice occur. At this time, statistical relationships have been found between backscatter coefficient, depolarization and relative humidity. It is clear that clouds do not necessarily form when saturation with respect to ice is reached.



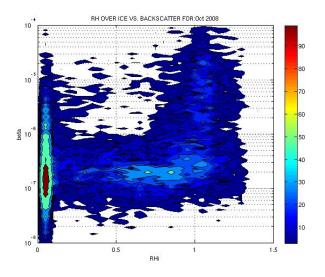
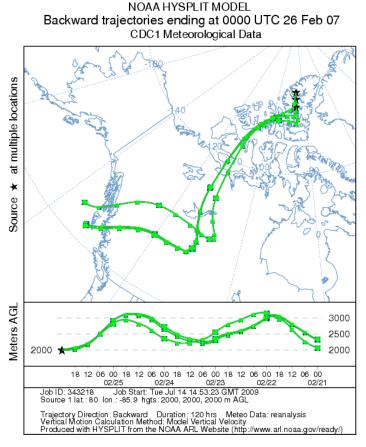


Figure 5: An example of liquid cloud layer forming from an aerosol layer that has been exposed to ice-supersaturated conditions (top) as well as a histogram of the frequency of occurrence of lidar backscatter cross-section versus relative humidity over ice (bottom).

Simulation of regional aerosol transport for specific case study dates to determine sources and types. The NOAA HYSPLIT model is being utilized to evaluate aerosol source regions and transport for the case study dates in the above dataset. An example is provided in Figure 6. Information on where the aerosol particles have been and have come from could provide clues in what they may consist of. Since aerosol composition was shown to significantly influence simulated cloud structures, having some information on sources and composition may help in explaining some of the characteristics of observed cloud structures. Patterns between ice nucleation (or lack thereof) and aerosol source region are being tracked.



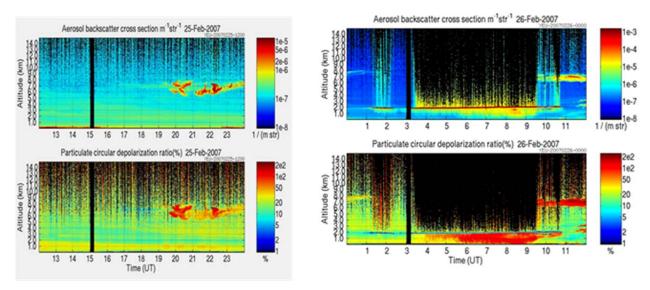
**Figure 6:** A back trajectory analysis of the aerosol layer depicted in Figure 3.

#### 2.5. Origin of Mixed Phase clouds observed in the Arctic

The intent of this study was to investigate the relationship between aerosol origin and the mixed phase arctic clouds which are observed over Eureka. Aerosol origin is an indirect way of studying aerosol composition which plays a crucial role in the formation, growth and life-time of water droplets and ice particles by serving as cloud condensation nuclei (CCN) or ice-forming nuclei (IN). The morphological and chemical properties of aerosols heavily influence the phase,

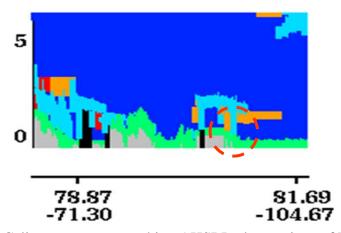
particle number concentration, effective radius, and liquid water path (LWP) of a cloud. Mixed phased clouds in particular are sensitive to this composition. Due to the meta-stable nature of super-cooled water, it seems that as soon as there could be a mechanism to produce ice inside a cloud, the entire cloud would quickly glaciate through the Bergeron-Findeisen process. A mechanism which is hypothesized to exaggerate this separation of phases is nucleation through immersion freezing. In this process the soluble salt portion of IFN/CCN suppress the freezing temperature of a liquid water droplet and as the droplet grows in an updraft, this salt content is diluted and the freezing temperature is raised until the point where the drop gets big enough to freeze. This would mean that the formation of large ice-particles is favored and thus they fall out of the liquid layer more readily. For this reason it is believed arctic haze, which tends to be comprised of such a soluble salt portion surrounding a silicate nucleus would have a positive effect on the formation of mixed phase arctic clouds.

A specific example of a mixed phase cloud nucleation event seen both by the downward viewing CALIOP and the upward looking Arctic High Spectral Resolution Lidar (AHRSL) was found. A thin layer of arctic haze can be seen at 2 km (Figure 7). This is readily identifiable from the higher backscatter/ low circular depolarization indicating a spherical aerosol, yet the environment is not saturated with respect to water. This layer then goes on to nucleate into a mixed phased cloud as water saturation is reached.



**Figure 7:** AHSRL observations taken on 25-26 February 2007. Aerosol backscatter shown (top) and Particulate Depolarization ratio (bottom).

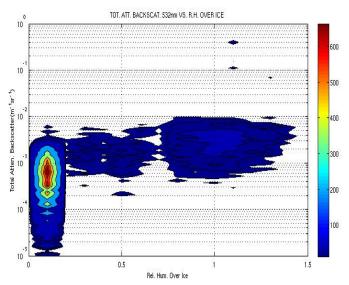
An overpass of CALIPSO (Figure 8) within about 10 km from the AHRSL at Eureka (80°N, -86°W) was found for this same event only a few hours before the nucleation event occurred.



**Figure 8:** Calipso overpass matching AHSRL observations of Figure 7.

The CALIOP level 2 feature mask shows an aerosol layer (orange) and nucleated cloud (aqua blue). Due to the large areal extent of the aerosol layer and the stratus cloud deck we believe that CALIOP is also observing a similar if not the same nucleation event as it passes over the region.

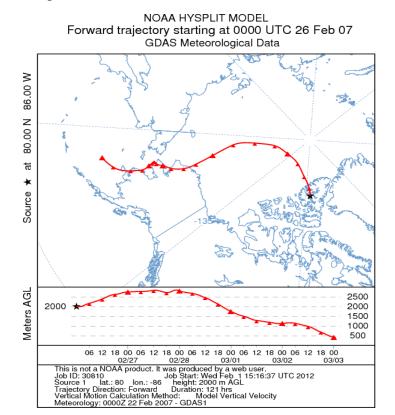
Focusing on the Eureka coordinates (80°N, -86°W) at the time of the overpass, the attenuated backscatter was compared to the reanalysis vertical temperature and humidity profile included in the level 1 product. A histogram (Figure 9) was obtained using every data point in the whole vertical column above Eureka. The coloring indicates the number of data points falling within a given bin.



**Figure 9:** Comparison of backscatter to reanalysis of temperature.

The typical backscatter of atmospheric aerosols is a little bit above 10<sup>-3</sup> so we see a peak corresponding to the signature of the aerosol layer and the environment is supersaturated with respect to ice. The regions on the histogram of lower backscatter but supersaturation with respect to ice would correspond to the arctic haze layer which eventually nucleates into a cloud.

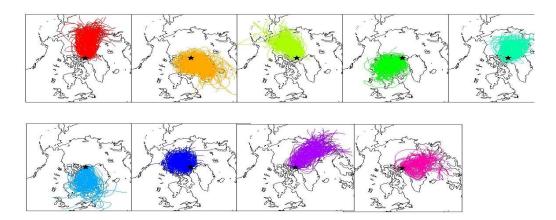
In order to link examples such as these to source regions of aerosols, back-trajectories were calculated using the HYSPLIT\_4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) model available from NOAA's Air Resources Laboratory. The model uses gridded meteorological data to integrate in reverse the wind field and obtain the Lagrangian trajectory. An individual back trajectory ending on February 26th, 2007 0000Z, 2 km above Eureka shows an example of this.



**Figure 10:** Sample Hysplit analysis of particle trajectories showing the origin of particles moving over the Eureka observations, with trajectories beginning at 00UTC 25<sup>th</sup> of February.

The individual 5 day trajectory shows the airmass arching over the Arctic Ocean but originating in a flow off of East Asia. Due to the errors inherent in the reanalysis data used to do the backward Lagrangian integration, an individual trajectory does not necessarily tell us reliable information about airmass origin. To make a more robust comparison between airmass origin and cloud phase, a whole ensemble of trajectories were calculated twice a day for all the days from 2006 to 2008. Each day corresponds to two different trajectories which show a possible

origin for that airmass five days earlier. This whole ensemble of trajectories was then grouped into different clusters, each cluster representing a "mode of entry" into Eureka. This was achieved by coding a recursive clustering algorithm and the most optimal result took the form of 9 different "modes of entry."



**Figure 11:** Cluster analysis depicting source of trajectories grouped into 9 different "modes of entry".

The endpoint of each trajectory was then associated with a different type of hydrometeor determined by a phase classifier developed by Mathew Shupe which had the categories of clear sky, ice, snow, liquid cloud drops, liquid cloud drops & drizzle, rain, mixed phase cloud and arctic haze (Shupe, 2007).

Each cluster then produced a different distribution of phases.

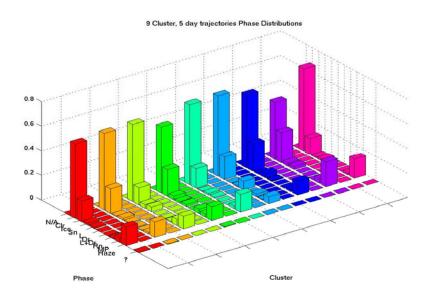
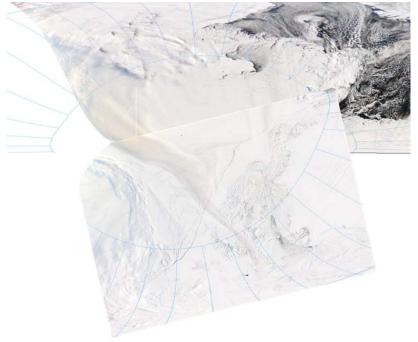
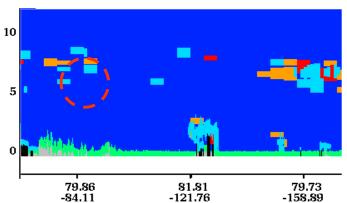


Figure 12: For each cluster, the distribution of ice types was shown with a histogram, depicting a relationship between air parcel origin and the characteristics of ice that form.

Using a Mann-Whitney U statistical significance test it was found that to a 1% level of confidence that the 8th cluster (purple) was sampled from a different distribution from all the other clusters except the 1st (red) and 3rd (pea green) clusters. These three unique clusters correspond to fast moving airmasses originating in Eurasia. Cluster 8 also had the highest raw number of mixed phase cloud days and arctic haze days in it. This is consistent with the fact that arctic haze is thought mainly originate from industry of European and former Soviet countries (Quinn, et. al. 2007).

A concrete example of one such event can be seen with CALIPSO on August 12, 2008 (Saha et. al., 2009).





**Figure 13:** Satellite and Calipso observation for trajectories associated with a particular case (12 August 2008).

Despite the elevated altitude of this plume, it's mode of entry into the Eureka region would fit well into clusters which cross the Arctic Ocean from Eurasia.

#### 2.6. Mixed Phase microphysics in the Tropical Tropopause Layer

A preliminary investigation of mixed phase microphysics occurring within the Tropical Tropopause Layer (TTL) was performed by Mr. Daniel Henz with a small amount of support from this project. The TTL exhibits many of the properties of arctic stratus, particularly the large influence of acidic solutions on the ice nucleation process. It was anticipated that this could result from the injection of pollutants into the tropical upper troposphere by convective plumes, possibly containing aerosols linked to the slash burning or industrial sources. We anticipated that the evoling characteristics of the TTL could be observed by Calipso. Figure 16 depicts one such simulated result from Henz (2010).

The preliminary study of Henz (2010) was to determine if a 2D cloudd model could produce the observed humidity and thermal characteristics of the TTL and crude microphysical structures to qualify it to be used as a laboratory for highly detailed microphysical studies as discussed by de Boer, and supported by Calipso observations.

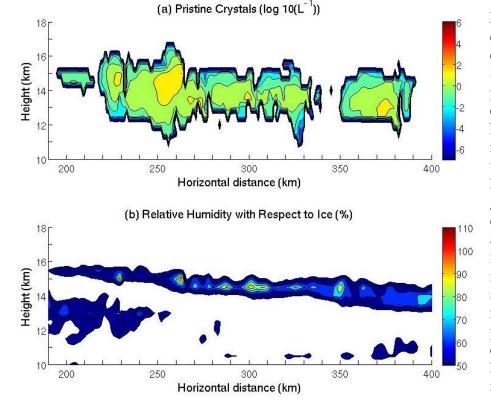


Figure 15: TTL cirrus from control experiment at 8694000s. a) Plot of pristine ice crystal concentration vs. height. b) Plot of relative humidity with respect to ice vs. height. TTL cirrus generated from detrained convective anvil located in the model domain. There is a small region of supersaturation with respect to ice where pristine ice crystals can continue to grow homogeneous nucleation.

Henz's results did indeed confirm that the 2d idealized framework could produce the TTL basic structural characteristics and therefore be used as a laboratory to study ice nucleation

on a fine scale using very detailed explicit microphysics. This study is currently expanding in scope and will generate published Journal papers.

#### 3. Educational

This project provided support for 3 students and one post doc during is funding period. The students included:

- 1. Gijs de Boer, PhD (2010)
- 2. Richard Hildner, MS (2012)
- 3. Daniel Henz (MS 2010)

# Post Docs supported:

1. Tempei Hashino (2009-2010), now at University of Tokyo

#### 4. Summary

This project resulted in a new understanding of what appears to be one of the most fundamental and under-appreciated forms of ice nucleation processes in the atmosphere. This nucleation process was shown to be tied directly to aerosol chemistry, forever closely mixed phase microphysical processes to the aerosol chemistry giving rise to these processes. It was shown through the HSRL observations that these processes could be observed, but were below the minimum detection level of Calipso during the important nucleation phases.

Far from demonstrating any failure, this project has shown that the Calipso mission was spot on the needs for future assessment and prediction of global microphysical processes. It forever merges our understanding of saturation based microphysical nucleation and growth processes with sub-saturated Kohler curves and aerosol deliquescence, and brings the cloud itself into the realm of atmospheric chemistry.

The prediction of cloud microphysics has long been known to be highly dependent on the assumptions made about nucleation. This study has clearly shown the nucleation and so the entire cloud is dependent on this underlying chemistry and that is dependent on the origins of aerosols and the dry and moist chemical evolution of the aerosols along those trajectories. These evolutions must be observed by remote sensing platforms if true cloud prediction and therefore albedo prediction and precipitation prediction are to improve. The role of space-borne lidar and cloud radar cannot be underestimated in this pursuit.

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- -----, E.W. Eloranta, T. Hashino, and G.J. Tripoli **A Potential Role for Immersion Freezing in Arctic Mixed-Phase Stratus.** *ARM Science Team Meeting* March 10-14, 2008, Norfolk, VA
- -----, E.W. Eloranta, I.A. Razenkov, J.P. Hedrick, J.P. Garcia, and M.D. Shupe, **Long-Term Lidar and Radar Observations of Arctic Stratus at Two Locations.** *Annual Meeting of the American Meteorological Society* January 20-24, 2008, New Orleans, LA.
- -----, E.W. Eloranta, and G.J. Tripoli. **Understanding Formation and Maintenance of Mixed-Phase Arctic Stratus Through Long-Term Observation at two Arctic Locations** 2007 Fall Meeting of the American Geophysical Union (AGU), December 10-14, 2007, San Francisco, CA.

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