

**Observational evidence for  
anthropogenic effects on Arctic  
boundary layer clouds over Eureka,  
Nunavut**

**by**

**Richard Hildner**

**Master of Science Thesis**

## Abstract

The relationship between air-mass origin and Arctic Boundary Layer Clouds over Eureka, Nunavut is investigated. This paper examines different modes of entry for air masses over the Eureka region using calculated back-trajectories with the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Five-day-long back-trajectories all ending 1.5 km above Eureka were calculated twice daily for a three year interval (2006-2008). Each trajectory was then associated with a cloud type existing at the time and place of the end-point using a ground-based multi-sensor cloud phase classifier. All the trajectories then were grouped into similar pathways using an average-linkage clustering algorithm. Each of these clusters represented a general mode of entry into the boundary layer above Eureka and the distributions of cloud phases associated with each cluster were statistically compared. Consistent with an anthropogenic origin for many ice-forming nuclei (IFN), it was found that the modes of entry which quickly cross the arctic ocean and originate in the industrial regions of Eurasia had a statistically significant difference in their distribution of phases and exhibited the most arctic mixed-phase cloud occurrences. This has important impacts on Arctic climate since Arctic mixed-phase stratus serve as a greenhouse blanket.

## Acknowledgements

Thanks go to my academic advisor, Dr. Gregory J. Tripoli for his support, motivation and his belief in me. Special thanks go to Dr. Edwin Eloranta, my unofficial advisor for his guidance in using the AHRSL data and motivation in researching this topic as well as providing the computing resources to run my code on. A special thanks goes to Gijs de Boer for his ideas and helping me in the day to day process of initiating this study.

Additional thanks go out to Dr. Mathew Shupe for assisting me in the use of his cloud phase classifier. Thanks to Mr. Joe Garcia and Mr. Pete Pokerandt for answering any question I had with computer related issues and fixing any bugs I encountered along the way.

Thanks to NASA for their support and funding of this study. A special thanks to my family.

# **Contents**

**Abstract**

**Acknowledgements**

## **1 Literature Review**

1.1 Clouds and Climate

1.2 Observations of Cloud Nucleation

1.3 Observations of Arctic Cloud Nucleation

1.4 Trajectory Analysis and Clustering

1.5 Mixed Phase Arctic Clouds

1.6 Immersion Freezing

1.7 References

## **2 Article**

2.1 Introduction

2.2 Instrumentation

2.3 Trajectories

2.4 Clustering

2.5 Results

2.6 Discussion

2.7 Conclusion

2.8 References

**Literature Review:****Clouds and Climate:**

The importance of cloud radiative properties has been highlighted by many important studies and even the earliest of these pointed to the importance of Arctic clouds in global-warming scenarios. Wetherald and Manabe 1988 investigated the effect of global-warming by increasing the solar constant in a highly simplified three dimensional global circulation model (GCM). Although they found that the net global radiative forcing due to clouds was by and large balanced in their model, they never-the-less identified changes in cloud distribution with increases in lower tropospheric cloudiness and a decrease in the upper troposphere. This translated to a net increase in cloudiness as one went poleward and a decrease near the equator (Wetherald and Manabe 1988). Mitchell and Ingram 1992 specifically investigated the effects of carbon dioxide warming on clouds using another low resolution GCM and found that the raising of the tropopause across the globe creates increased radiative cloud-top cooling. They suggest that cloud parametrizations at the time of their study remain largely arbitrary and are the largest source of uncertainty in all GCM models. This is the same results Bromwich et. al. 1994 arrived to when they compared simulations on the National Center of Atmospheric Research (NCAR) Community Climate Model Version 1 (CCM1) to European Center for Medium-Range Weather Forecasts (ECMWF). The Arctic climate in particular has the largest deviations between models and observations.

Studies like these motivated a variety of scientific programs such as the Atmospheric Radiation Measurement program (ARM) which specifically singled out the importance of focusing on the physical process which occurred in the Arctic as being crucial to predicting the climate as a whole. The 1990 Intergovernmental Panel on Climate Change (IPCC) as well emphasized the vulnerability of the Arctic region due to the unique physical processes which govern its climate. Namely, arctic sea ice serves the two-fold purpose of increasing the surface albedo and insulating the cold winter air from the relatively warm ocean water underneath. Additionally the flow of Arctic Sea water and its sea ice into the North Atlantic is a fundamental component of the global thermohaline circulation (IPCC 1990).

In an overview of Arctic cloud-radiation characteristics Curry et. al. 1996 identify four unusual cloudy boundary-layer types: 1. summertime boundary-layer with multiple decks of clouds, 2. mixed phase boundary-layer clouds, 3. low level ice crystal clouds and "clear sky" ice precipitation in stable winter-time boundary-layers, and 4. winter-time ice crystal plumes emanating from leads or cracks in the sea ice. These four cloud types create a difficult modeling problem that is still far from being solved. Thus up until today large uncertainties arise from the unsatisfactory handling of micro-physical and sub-gridscale dynamical parameterizations and in addition, observational uncertainty in the arctic is sparse while in situ measurements are rarely coincidental with satellite measurements in order to compare important fine scale variables such as liquid

water path, updraft velocity, and size distribution. (NRC, 2005; IPCC, 2007; Chin et al., 2009).

#### **Observations of Cloud Nucleation:**

Due to the sub-freezing temperatures present throughout most of the year in the Arctic, the nucleation of cloud hydrometeors even in the lower troposphere involves understanding both liquid drop nucleation about cloud condensation nuclei (CCN) and ice deposition nucleation on ice-forming nuclei (IN). Laboratory studies dealing with ice nucleation on aerosol particles have been undertaken since the 1950's to determine the ice-forming capabilities of different types of aerosols. Isono et. al. 1959 established that Asian silicate or clay dust particles advected from the Gobe desert were especially active IN (Isono et. al. 1959). Similar types of experiments employing deposition and condensation chambers have been carried out since then but with the advent of depolarization LIDAR and back-trajectory modeling it has been possible to observe condensation events in nature.

Sassen et. al. 2003 used the Facility for Atmospheric Remote Sensing(FARS)Polarization Diversity Lidar (PDL) along with a continuous flow diffusion chamber (CFDC) on-board an aircraft to simultaneously observe a nucleation event in southern Florida from the ground and sample the CCN content of the air-mass during a Saharan dust outbreak. It was concluded that Saharan dust aerosol particles can be just as active IN as Asian dust particles and



Saharan dust plumes may be responsible for seeding a variety of clouds (Sassen et. al. 2003).

Similarly, in Leipzig, Germany , Ansmann et. al. 2005 used a Raman lidar to observe the nucleation event (Fig.1) of a gravity-wave induced alto cumulus ice cloud and used the FLEXTRA langrangian integrator to calculate the back-trajectory of the over-head air-mass (Fig.2). The origin was determined to be the Sahara desert as well (Ansmann et. al. 2005).

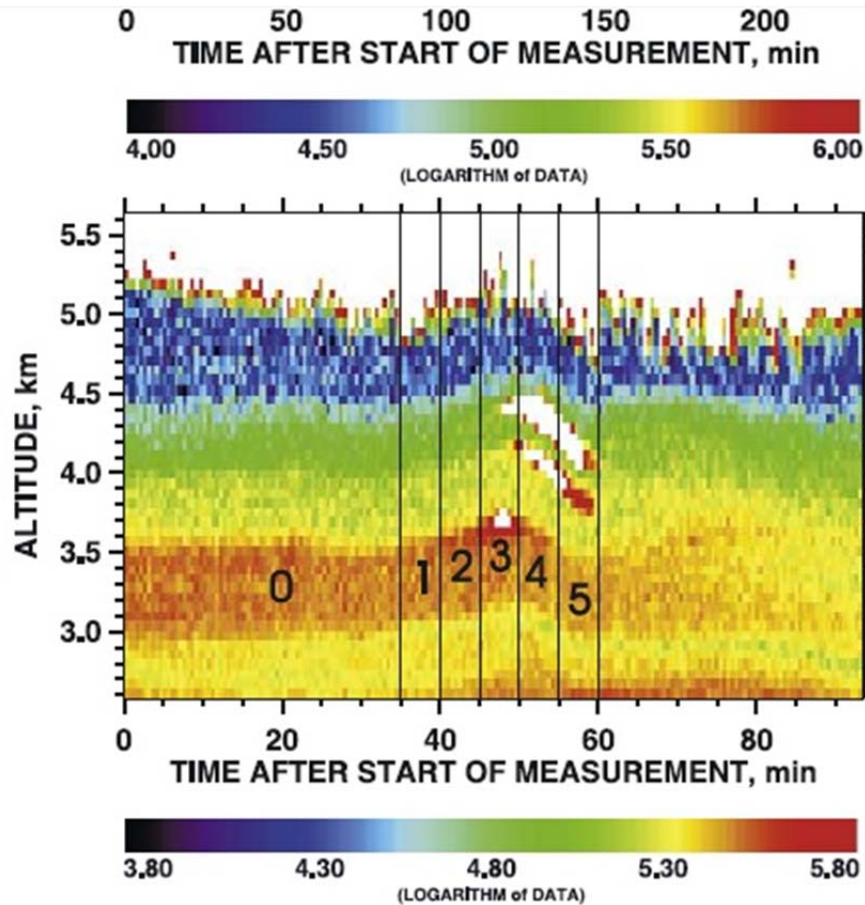
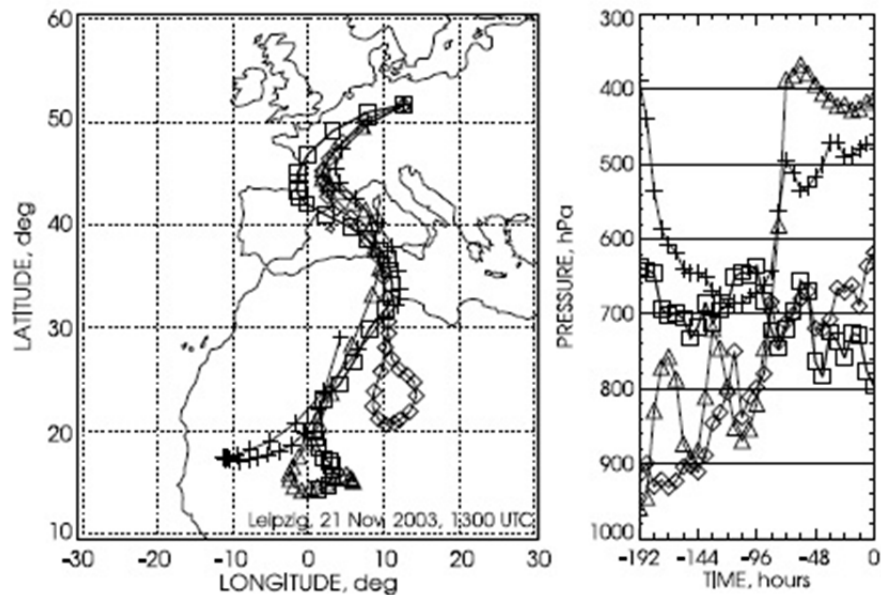


Figure 1: Raman lidar image of a nucleating gravity wave induced cloud (Ansmann et. al. 2005)



**Figure 2:** Calculated back-trajectories tracing dust plume to Saharan Desert (Ansman et al. 2005)

In addition to employing ground-based lidar, the advent of the CALIOP Lidar aboard the CALIPSO satellite has provided a top-down view of clouds which has also proved invaluable to subsequent studies. Yumimoto et. al. 2009 is a prime example of a study which illustrates in vivid detail the interaction between dust-plumes and cloud nucleation. Following an intense dust storm which occurred in May 2007 over the Taklimakan desert in northwestern China, the SPRINTARS global aerosol transport model was used to reconstruct the trajectory of the resulting dust plume and images from CALIPSO overpasses were used to confirm this trajectory (Fig. 3).

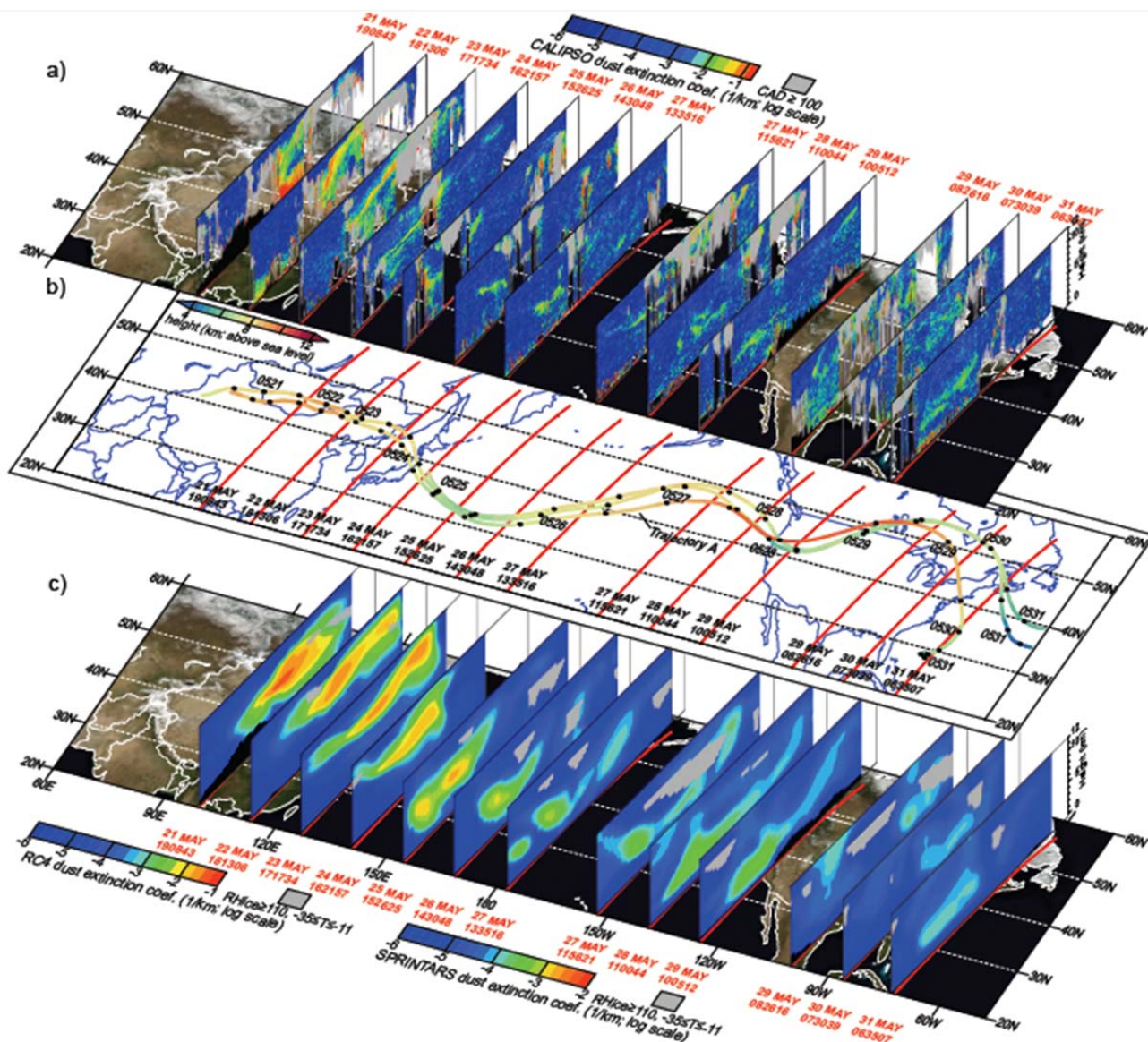


Figure 2: Correspondence between SPRINTARS calculated trajectory and CALIPSO's images of the plume (Yumimoto et al. 2009)

### Observations of Aerosols and Arctic Cloud Nucleation:

The nucleation event studies presented thus far have all focused mostly on non-boundary-layer clouds in the mid-latitudes. Due to the far greater concentration of CCN in the mid-latitudes compared to the



arctic along with a far greater number of observations leads to less studies showing arctic-boundary layer cloud nucleation events.

De Boer et. al. 2011 shows several specific cases using the Arctic High Spectral Resolution Lidar and other instruments located at Eureka, Nunavut and the world's northern-most manned weather station. In Fig.4 they show a thin persistent aerosol plume at 2km in both sub-saturated and super-saturated conditions with respect to ice.

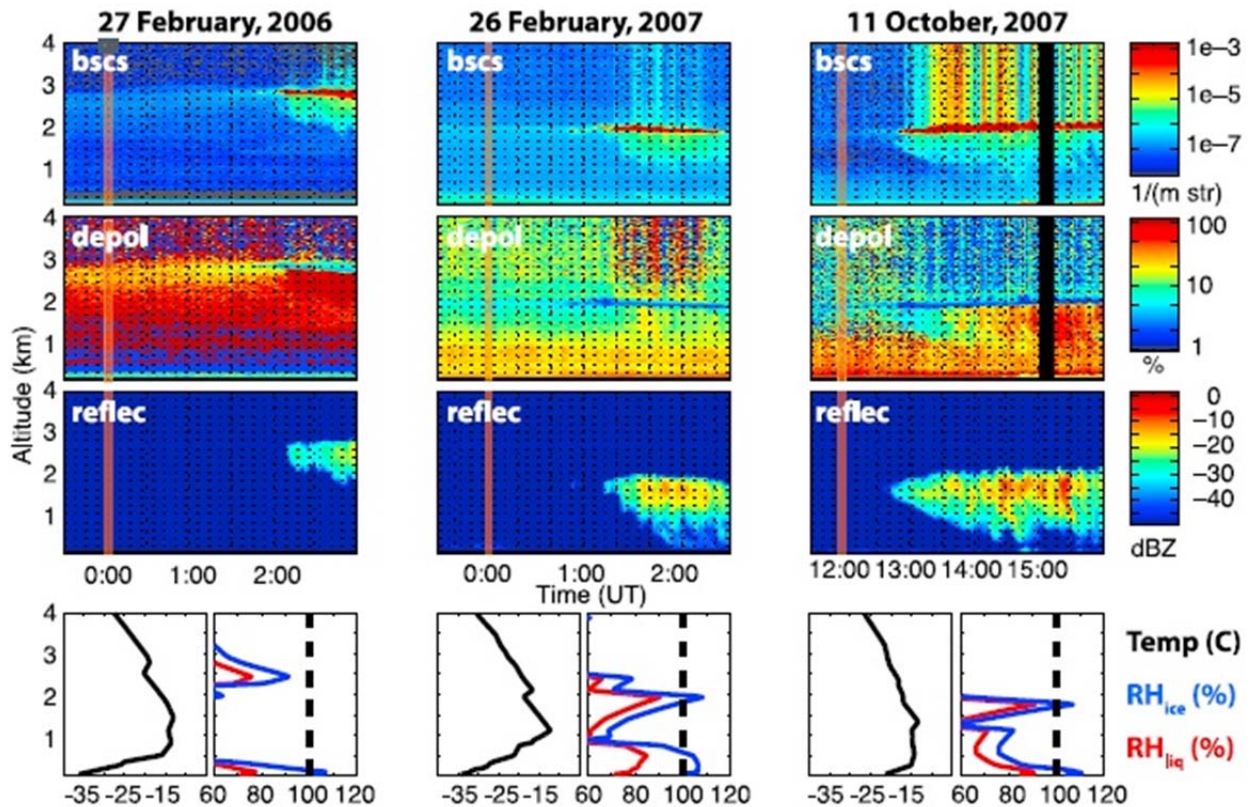


Figure 3: Three different Arctic mixed phase cloud nucleation events (de Boer et. al. 2011).

The plume eventually nucleates into a mixed-phase cloud which clearly contains liquid droplets in conditions which are below freezing and super-saturated with respect to ice (de Boer et. al. 2011). In

conjunction with similar data from two other Arctic locations they conduct statistical analysis and find strong evidence that in the range of -30C to -20C, liquid dependent ice formation in the lower troposphere is the most frequent mechanism of nucleation.

### **Trajectory Analysis and Clustering:**

Analyzing back-trajectories of air-parcels to determine their source of origin has been a mainstay in the study of atmospheric chemistry and pollutant transport. For example, Jickells et. al. 1982, Church et. al. 1992 and Galloway et. al. 1993 all used a subjective manual classification of different calculated back-trajectories to study the composition of precipitation in Bermuda.

Not until Moody and Galloway 1988 was cluster analysis used to group back-trajectories together in order to obtain a classification of chemical composition of an air-parcel. Cluster analysis is an algorithm used to group similar trajectories together and this in turn is used to create categories of air masses based on region of origin. This study related ion and anion concentrations to different source regions for air masses advected into Bermuda and it was found that the greatest deposition of these ions occurred with transport on trajectories from the west and northwest, off the east coast of US, indicating an anthropogenic source (Moody and Galloway 1988).

### **Mixed Phase Arctic Clouds:**

The first heuristic numerical model of a mixed phase Arctic stratus

clouds was developed by Herman and Goody 1976 which simulated some key aspects of Arctic strati such as a near constant height of cloud base and the splitting of cloud layers. They ran two cases, the first one was a stable case where warmer air was advected over a colder surface, and the second one was an unstable case where cold air was advected over a warmer surface. Both cases produced the same results except that in unstable case the onset of cloud formation was earlier.

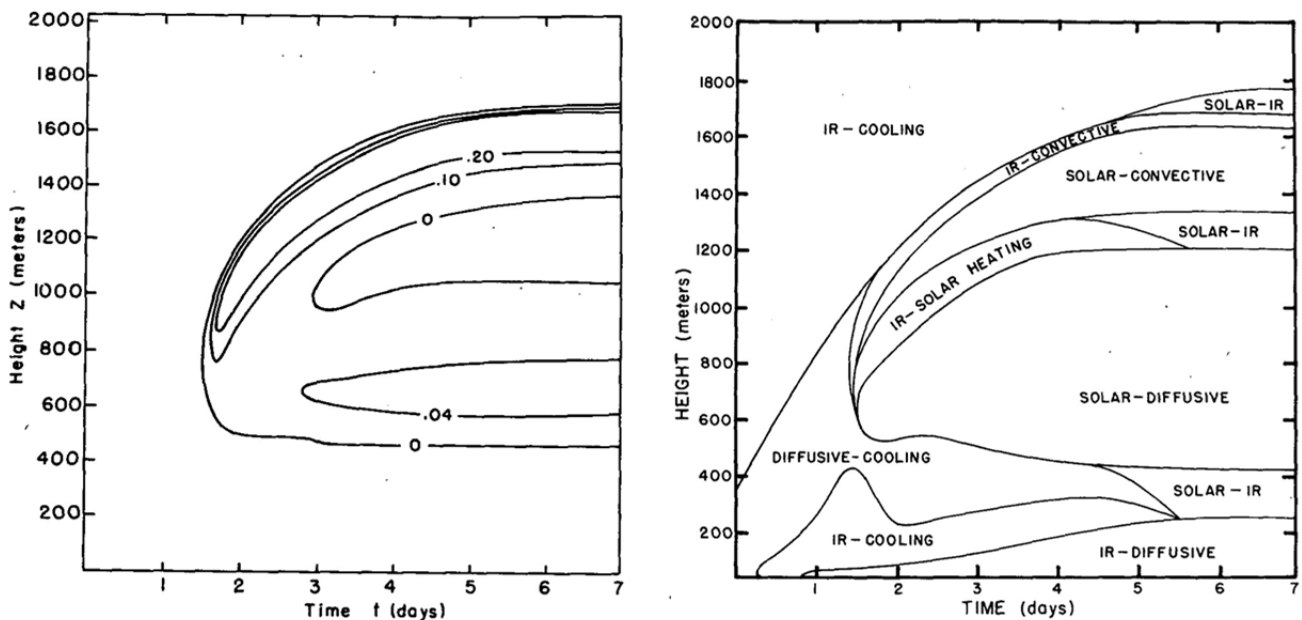


Figure 4: Heuristic numerical model capturing basic features of mixed phase clouds(Herman and Goody 1976).

Conceptually, they describe the process of cloud formation in three main steps: BL cooling to saturation, cloud formation with cloud-top radiative LW cooling, and then solar "green-house" heating within the cloud evaporating the middle, producing layers. Initially the BL cools though turbulent heat-flux into the surface and LW emission by water vapor into space, while LW absorption by other

atmospheric gases merely delays cloud formation but does not prevent it. Once a given level cools to the point of saturation, condensation dramatically alters the radiative regime and increased LW cooling at cloud top is turbulently distributed throughout the cloud.

Herman & Goody then proceed to alter various parameters and analyze the results. For example, when they make the cloud droplets radiatively inactive, only a tenuous haze is formed with no layering, or when they make the eddy diffusivity constant with height, the cloud base starts at 75 meters. Although this simple model captures some key features of Arctic strati, the crucial aspect which is still not well understood is how the nucleation of ice effects the various cloud properties (Herman and Goody 1976).

### **Immersion Freezing:**

In order for the liquid phase to not glaciate immediately based on the Bergeron-Findeissen process (Pruppacher and Klett 1997), there must exist a mechanism to separate these two phases inside the cloud. 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 formation of large ice-particles is favored and thus they fall

out of the liquid layer more readily. Gijs de Boer 2009 conducted 2-dimensional simulations using Prof. Greg Tripoli's NMS model and Tempei Hashino's Advanced Micro-Physics Scheme (AMPS) which tracks nucleation explicitly. He found that for runs with low IFN, immersion freezing heavily dominated over deposition freezing during much of the life-cycle of the single-layered mixed phase cloud (de Boer 2009).

**Motivations:**

Dr. Matt Shupe's cloud classifier provides an excellent opportunity to combine both the methods from atmospheric chemistry to ground based remote sensing in order to investigate the roll of source region on cloud properties. By applying the method of trajectory cluster analysis to the observables measured by ground based remote sensing instruments we can attempt to illuminate a link between modes of entry into the Eureka region to different cloud regimes. More specifically a link will be sought between mixed phase clouds and potential anthropogenic sources.



**Observational evidence for anthropogenic effects  
on Arctic Boundary Layer clouds over Eureka, Nunavut**

R. Hildner, G. J. Tripoli AND Edwin W. Eloranta

*Department of Atmospheric, Oceanic & Space Science, University of Wisconsin,  
Madison, Wisconsin*

ABSTRACT

The relationship between air-mass origin and Arctic Boundary Layer Clouds over Eureka, Nunavut is investigated. This paper examines different modes of entry for air masses over the Eureka region using calculated back-trajectories with the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) model. Five-day-long back-trajectories all ending 1.5 km above Eureka were calculated twice daily for a three year interval (2006-2008). Each trajectory was then associated with a cloud type existing at the time and place of the end-point using a ground-based multi-sensor cloud phase classifier. All the trajectories then were grouped into similar pathways using an average-linkage clustering algorithm. Each of these clusters represented a general mode of entry into the boundary layer above Eureka and the distributions of cloud phases associated with each cluster were statistically compared. Consistent with an anthropogenic origin for many ice-forming nuclei (IFN), it was found that the modes of entry which quickly cross the arctic ocean and originate in the industrial regions of Eurasia had a statistically significant difference in their distribution of phases and exhibited the most arctic mixed-phase cloud occurrences. This has important impacts on Arctic climate since Arctic mixed-phase stratus serve as a greenhouse blanket.

## **Introduction**

More than 50 years ago pilots flying in the Canadian and Alaskan Arctic observed a strange haze of an unknown origin (Greenaway, 1950; Mitchell, 1956). By the end of the 1970's the anthropogenic sources of these particles were illuminated through "chemical fingerprinting" and spatial gradients showing the surprisingly large extent of these clouds of pollution. A combination of intensive field programs and long-term measurements over the past decades have confirmed these early observations that this haze is anthropogenic in origin due to emissions from Europe and the former Soviet Union (Quinn et al. 2006).

In the same 50 year period, climatic changes in the arctic have been observed on an unprecedented scale. The most dramatic change has been the downward trend in summer minimum sea ice extent and the anthropogenic cause for this trend is being further and further established. Furthermore cloud formation and maintenance are seen as the largest source of uncertainty in future models (IPCC 2007).

Of the unusual cloud types which exist in the Arctic, mixed phase Arctic clouds remain the most persistently difficult to accurately model due mainly to their complex dependence on the composition, morphology and history of the condensation or ice-forming nuclei which seed them. Thus the nucleating properties of these naturally occurring as well as anthropogenic cloud-forming aerosols depend heavily on the aerosol particle's region of origin and mode of entry into the Arctic.

This study investigates this relationship between Arctic cloud-type and air-mass origin over a specific Arctic location. Using a ground-based sensor array stationed at Eureka, Nunavut, three years of daily cloud-type data was linked to the type of trajectory along which that air-mass travelled in order to arrive at our region of interest.

## **Instrumentation**

A multi-sensor approach is necessary in order to classify different cloud types and data from a combination of Arctic high spectral resolution depolarization lidar (AHSRL), millimeter cloud radar, microwave radiometer and radiosondes at Eureka, Nunavut run by the University of Wisconsin Lidar Group. These observations were all integrated into a cloud phase classification algorithm developed by Shupe 2007 in order to determine cloud-type (Shupe 2007).

## **Trajectories**

This study used 2171 five-day-long back-trajectories arriving at Eureka at 0000Z and 1200Z for every day from 2006 to 2008. These times coincided with the twice daily radiosonde launches in order to obtain the best classification of phase. All trajectories were calculated using the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model with ECMWF reanalysis data.

In order to evaluate how realistically model calculated trajectories approximate true parcel trajectories, Stohl et al. 1998 compared constant volume balloon (CVBs) tracks against those calculated from ECMWF reanalysis using a dispersion model similar to HYSPLIT known as FLEXTRA. When matching the height of the model trajectories at each time step to the height of the CVBs in order to focus on the horizontal motion, it was found that largest error was 11% of the trajectory length and the average error being 6% of the trajectory length. Even in cases where the trajectory dipped into the planetary boundary layer the average error could be 15%. Some of these sources of error include small scale convective vertical mixing, unknown radiative cooling rates and of course error in the horizontal wind field (Stohl et al. 1998).

The question then becomes which reanalysis dataset to use and what level of error this would introduce. Harris et al. 2005 show that the difference in calculated trajectories between using different reanalysis data sets can be up to 30%-40% of the trajectory length; however, it was also concluded that no data set could be considered superior to another even though differences exist in the trajectories they result in (Harris et al., 2005). Engström et al. 2009 show through a perturbation method that errors in a single reanalysis data set can lead to errors in trajectories of about this same magnitude (Engström et al. 2009).

If a link was sought between individual trajectories and different cloud regimes, then these errors would be very significant and thus it was preferable to study large groups of trajectories that together form broad modes of entry into the Eureka region in order to “average out” these errors and have a broader acceptable range of accuracy.

## **Clustering**

The present study relies on grouping back-trajectories using the average-linkage clustering method described by R. R. Sokal (Sokal 1958) which yields similar results

to the centroid clustering algorithm employed by S.R. Dorling when clustering back trajectories arriving at Eskdalemuir, south Scotland (Dorling et al. 1992). A FORTRAN programs was coded implementing the average-linkage method because it avoids producing outlying clusters which only have a handful of members as opposed to the centroid method or Ward's method (Kalkstein et al. 1987). When clustering trajectories over Mace Head, Ireland, Cape et. al. 2000 explicitly decide to use average-linkage for its suitability to synoptic variables (Cape et al. 2000).

The longitude-latitude back-trajectories were converted to Euclidean  $x,y$ -coordinates. Let  $X_{ijk}$  be the  $x,y$ -coordinates of the  $i$ th back-trajectory ( $i = 1, 2, \dots, N_k$ ) at the  $j$ th time step ( $j = 1, 2, \dots, J$ ) belonging to the  $k$ th cluster of  $K$  clusters. The centroid of a given cluster is a trajectory formed by averaging over all of the back-

$$X_{@jk} = \frac{1}{N_k} \sum_{i=1}^{N_k} X_{ikj}$$

trajectories in that cluster, i.e.

The @ sign denotes averaging over the index it replaces. To calculate the measure of similarity between two clusters (1 and 2) the squared Euclidean distance ( $D_{pq}^2$ ) between each pair of trajectories is found, one from each cluster, and averaged:

$$L_{1,2} = \frac{1}{N_1} \frac{1}{N_2} \sum_{p=1}^{N_1} \sum_{q=1}^{N_2} D_{pq}^2, \text{ where}$$

$$D_{pq}^2 = \sum_{j=1}^J (X_{pj1} - X_{qj2})^2$$

Starting with a set amount of “seed” clusters, in this case 30 randomly chosen ones. An iterative process is followed, at each step first finding the root mean square deviation of each cluster (RMSD):

$$RMSD_k = \sum_{p=1}^{N_k} \sum_{j=1}^J (X_{pj} - X_{@jk})^2$$

This represents how “broad” the  $k$ th cluster is and then all clusters are averaged over to find the total average RMSD. Next the two clusters which are most similar, i.e. have the smallest  $L_{1,2}$ , are merged into one large cluster bringing the total number of  $K$  clusters down by one and the process is iterated again.

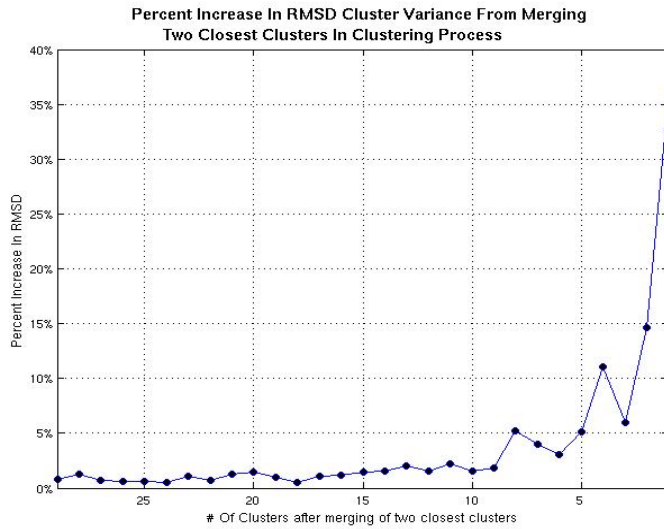
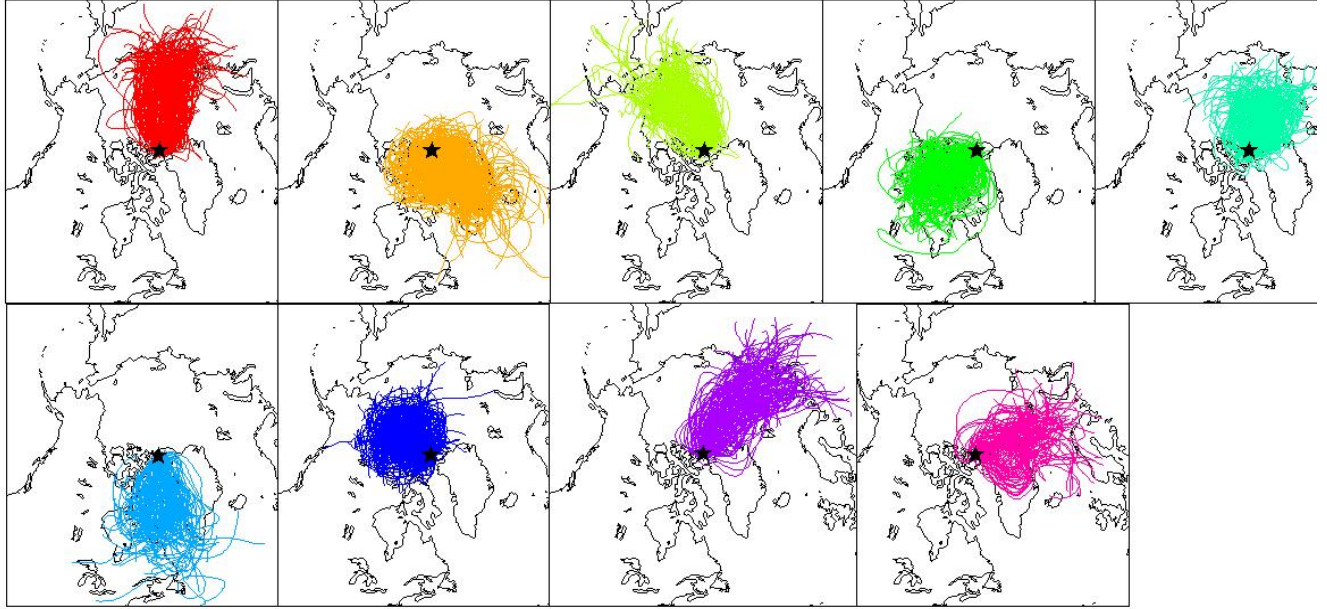


Figure 5: When 9 clusters are merged into 8 clusters the first large percent increase of RMSD is observed.

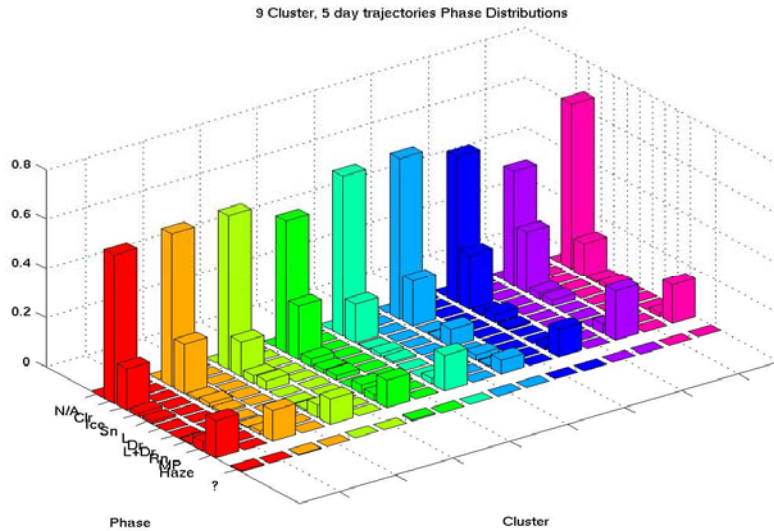
## Results

Fig. 1 illustrates the percent change of RMSD with each iteration and the point where it first changes more than 5% is the point where two clusters which were significantly different from each other were merged. This means that with less than 9 clusters, information about modes of entry is lost, making it an optimal number.

Fig. 2 shows the clusters which were obtained:



**Figure 6: 9 modes of entry into Eureka**



**Figure 7:** Probability distribution of phase classifier observations for each cluster. *N/A = no data, Clr = clear skies, Ice = ice cloud, Sn = falling snow or diamond dust, L = liquid cloud, Dr = drizzle, Dr + L = drizzle and liquid water cloud, Rn = Rain, MP = mixed phase cloud, Haze = Arctic haze and ? = indeterminate.*

Each of these clusters is associated an end point time stamp. For this time stamp the ground-based sensors at Eureka are consulted and analyzed with the cloud classifier. A distribution of cloud types was obtained for each cluster and normalized since different clusters had different numbers of members. Fig. 8 illustrates the probability that any given cloud type, including clear skies, would occur for each cluster.

## Discussion

The unusual cloud types that form in the polar regions include ice crystal clouds that reach the surface, commonly referred to as “diamond dust;” convective plumes associated with leads or polynyas (openings in the sea ice); persistent mixed-phase clouds; and multiple layers of thin cloud decks that occur in the statically stable arctic environment (Curry et al. 2000).

Mixed phase Arctic clouds in particular can be nucleated by Arctic haze sulphate particles. The microphysical composition of these haze particles consists of a soluble component composed of varying mixtures of sulphate, ammonium and nitrate enveloping a solid nucleus composed of particulate organic matter, silicate dust, black carbon and rich in heavy metals which identify the industrial sources (Shaw, 1983).

This particle size is very efficient at scattering visible solar radiation through Mie scattering and also exhibits weak absorption which may have a significant influence on the arctic climate (Shaw and Stamnes, 1980); however, the other mechanism through which arctic haze may play a significant role in affecting the climate is through the aerosol in-direct effect which is the influence Arctic haze exerts on the nucleation of clouds.

In the Arctic, extensive stratiform mixed phase cloud layers may persist in a quasi steady state for up to days and even weeks (Shupe et al. 2006). A better understanding of the microphysical processes of formation, growth and interaction of drops and ice particles in arctic mixed-phase clouds is crucial to improving their representation in general circulation models and our understanding of their impact on Earth's climate (Intergovernmental Panel on Climate Change, 2007). Several studies through cloud-resolving and large eddy simulations have revealed that these cloud systems are very sensitive to even rather modest changes in cloud condensation nuclei (CCN) and ice forming nuclei (IFN) (Olsson and Harrington 2000; Jiang et al. 2000; van Diedenhoven et al. 2009). There also exists observational and model evidence that ice nucleation is dependent on the initial formation of a liquid phase in which Arctic haze particles first serve as CCN and then subsequently as IFN through the process known as immersion freezing. In this process water droplets which initially condense around Arctic haze particles have their freezing point depressed due to the haze particle's soluble component. Eventually the drop grows, diluting the soluble portion and in turn raising the freezing temperature until it becomes large enough to freeze around the solid nucleus. This mechanism arranges for only large drops to freeze and thus fall out of the liquid portion of the cloud avoiding complete glaciation through the Bergeron-Findeisen mechanism (de Boer et al. 2011; Shupe et al. 2007).

In order to determine if the phase observations for each cluster are samplings from distinct probability distributions, a non-parametric Mann-Whitney test was performed between each pair of distributions. This rank-sum test can be conducted between two very different size populations to determine if there exists a statistically significant difference between them.



	Clus 1	Clus 2	Clus 3	Clus 4	Clus 5	Clus 6	Clus 7	Clus 8	Clus 9
Clus 1									
Clus 2								x	
Clus 3									
Clus 4								x	
Clus 5								x	
Clus 6								x	
Clus 7								x	
Clus 8		x		x	x	x	x		x
Clus 9								x	

*Fig 9: Non-parametric Mann-Whitney test between pairs of distributions. An X indicates that there is a statistically significant difference between these two distributions at the 1% level.*

Cluster 8 (Purple) which moves directly across the Arctic Ocean originating in Eurasia is the only cluster which is statistically different from most of the other clusters. The only other modes of entry which it does not show a statistical difference from follow paths which are quite similar and follow more or less direct paths across the Arctic Ocean from the former Soviet Union.

Cluster 8 additionally exhibits the highest probability (20% chance) of containing an Arctic haze event but not the highest probability of a mixed phased cloud event (3.9% chance). It does however show the highest probability out of all the other clusters to contain an ice cloud event (25% chance). This could be due to the fact that mixed phase clouds being in the form of stratus decks are extremely vertically thin and depending on the height of the boundary-layer could be situated above or below the actual end point height (1.5 km over Eureka) of the calculated back-trajectory. At these low levels below the top of the boundary-layer many, if not most of the events which the cloud phase

classifier is categorizing as ice events are due to ice falling out of mixed phase stratus decks. Fig. 5 (g) shows how ice event classifications are associated with mixed phase cloud event classifications. This is consistent with the immersion freezing method in which the larger droplets are the ones to freeze and drop out below the liquid phase. In light of this it is highly probably that Cluster 8 has the phase distribution with highest occurrence of mixed phased clouds in the vicinity of the trajectories' end point.

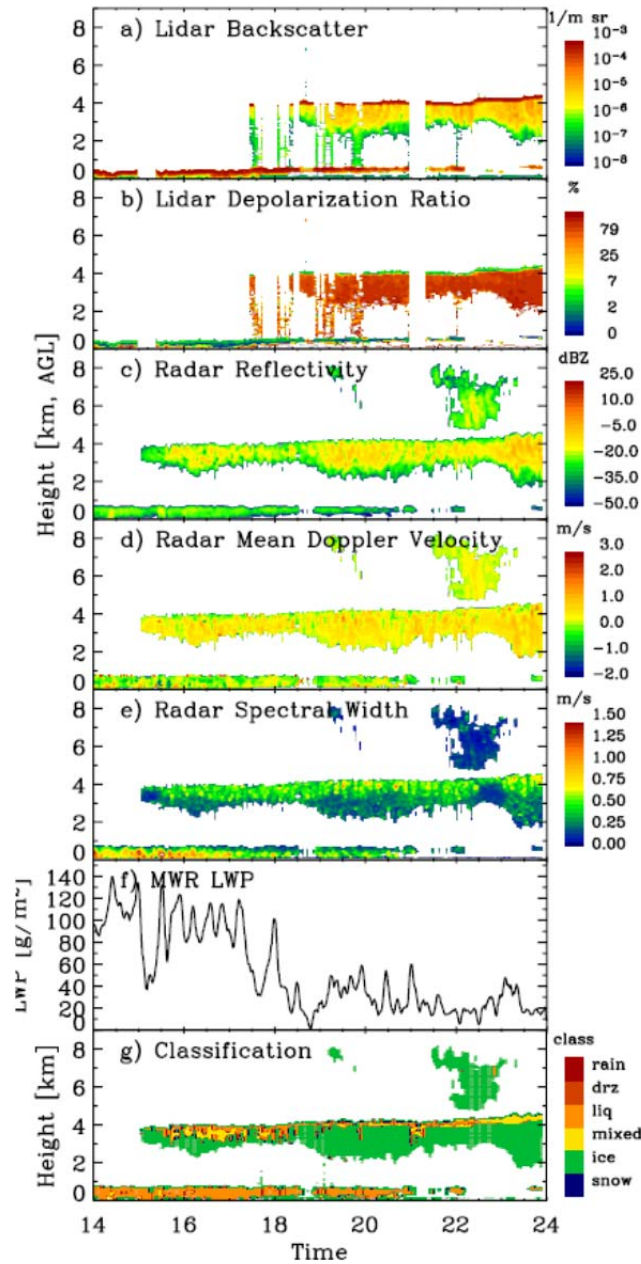


Fig 10: Observations from different ground

## Conclusion

Three years of data from a specific arctic location with ground-based instruments at Eureka, Nunavut was used to link the occurrence of Arctic mixed phase clouds with an air mass's mode of entry into the region of origin. A link between occurrences of Arctic haze events and mixed-phase cloud events was found, with air masses from Eurasia containing higher occurrences of both of these types of events.

For the three years studied, the mode of entry of air masses into Eureka which originate in the northern Eurasian industrialized region of the world and crosses the Arctic Ocean over the course of 5 days shows a statistically significant difference in its distribution of cloud occurrence and phase compared to other modes of entry (Fig. 6). The results of this study show that air masses which originate in Eurasia and cross the Arctic Ocean have a higher probability of producing mixed phase clouds over Eureka. Decades of observations showing the anthropogenic origin of arctic haze via industrial sulfates or agricultural burning in northern Eurasia combined with the results of this study suggest observational evidence for an additional anthropogenic effect on mixed phase cloud formation and life-cycle. This has important implications for the unique role of the arctic in climate change.

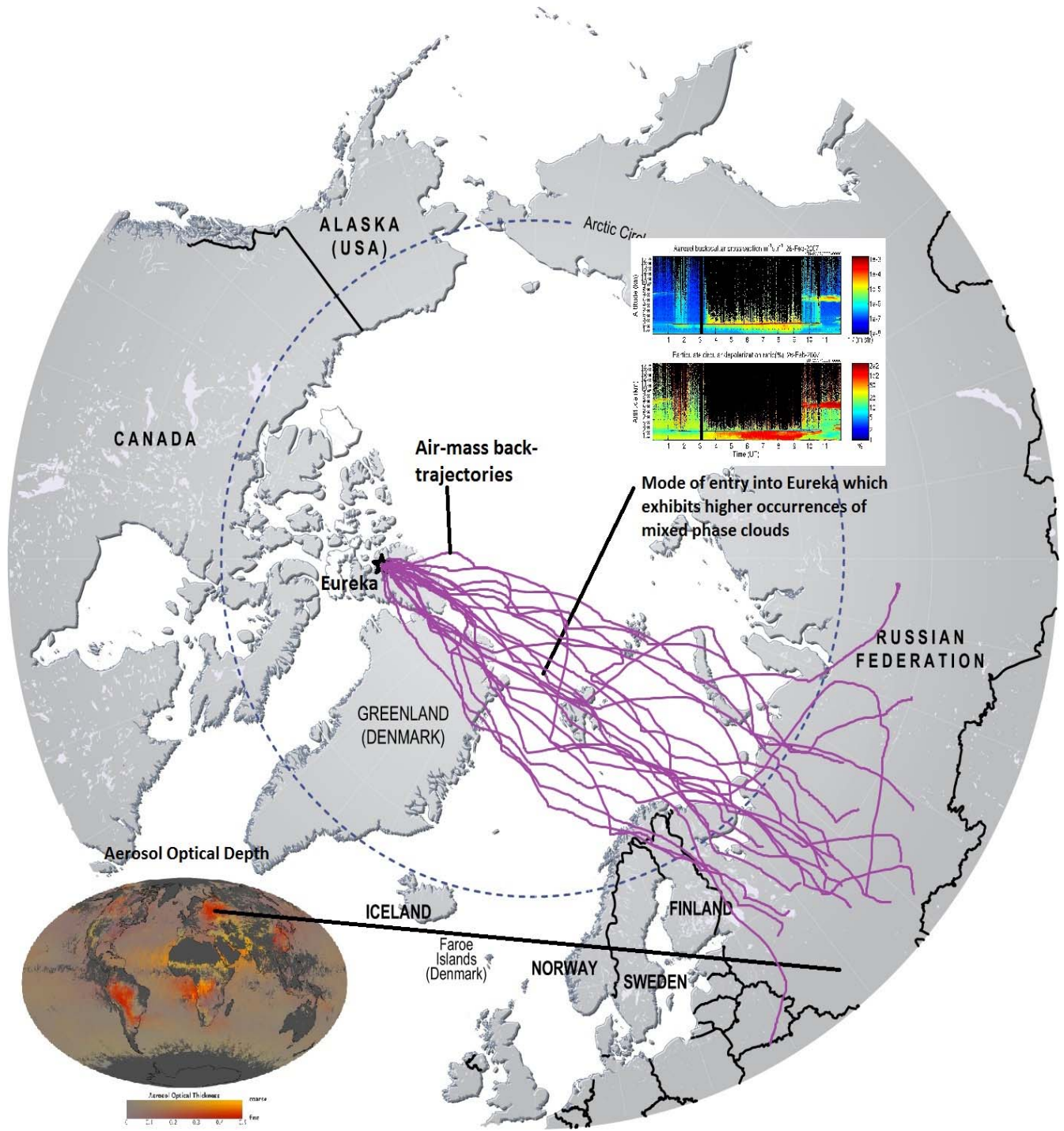
The unprecedented loss of sea ice over the last 50 years is intimately tied to the degree of downward long-wave flux (DLF). The accumulation of contributions to DLF change based on several factors such as cloud fraction, cloud-base height, liquid/ice water path, precipitable water, surface skin temperature, and lower-tropospheric temperature. Francis et al. 2007 found through investigating a variety of satellite derived products that in the Arctic where mixed phase clouds exist, the primary variable which affects DLF appears to be changes in water vapor, i.e. precipitable water. Low Arctic stratus clouds cause increased emission of longwave radiation to the surface that exceeds the increased reflection of incoming solar energy by the clouds back to space. (Francis et al. 2007).

Low lying mixed phase clouds in the Arctic nucleated inside of air masses lofted into the Arctic from regions heavy in anthropogenic Arctic haze would produce a greenhouse blanket which would act as an amplifier in the already present ice-albedo feedback mechanism.

Another, and perhaps opposite effect of increased mixed phase cloud nucleation due to polluted air on the Arctic climate is their dehydrating effect proposed by Blanchet et al. If mixed phase clouds are primarily nucleated through the immersion freezing process, then Arctic haze nucleated clouds favor formation of large ice crystals thus substantially increasing the water flux to the surface. This would result in a greater dehydration of maritime air which could explain the increasing lapse rate over the Arctic Ocean present in general circulation models (Blanchet et al. 1994, Blanchet et al. 1995).

This effect could be presently masking some of the arctic warming produced by the Arctic stratus greenhouse blanket and merely delaying a point-of-no-return tipping point in the ice-albedo feedback cascade.

Thus our study shows observational evidence for an additional mechanism through which anthropogenic activity influences the Arctic climate in addition to large-scale warming.



References

Barrie, L. A. Fisher, D. & Koerner, R. M. *Atmos. Envir.* 19. 2055-2063 (1985).

Blanchet, J.-P. and E. Girard, 1994: Arctic greenhouse cooling., *Nature*, 371, 383.

Blanchet, J.-P. and E. Girard, 1995: Water-vapor temperature feedback in the formation of the continental Arctic air: Implications for climate., *Sci. Total Environ.*, 160/161, 793-802.

Cape, J.N., Methven, J., Hudson, L.E., 2000. The use of trajectory cluster analysis to interpret trace gas measurements at Mace Head, Ireland. *Atmospheric Environment* 34, 3651-3663.

Church, T. M., Galloway, J. N., Jickells, T. D. and Knap, A. H. 1982. The chemistry of western Atlantic precipitation at the mid-Atlantic coast and on Bermuda. *J. Geophys. Res.* 87, 11013-1 1018.

Curry, J.A., P. V. Hobbs, M. D. King, D. A. Randall, P. Minnise, G. A. Isaac, J. O. Pinto, T. Uttal, A. Bucholtz, D. G. Cripe, H. Gerber, C. W. Fairall, T. J. Garrett, J. Hudson, J. M. Intrieri, C. Jakob, T. Jensen, P. Lawson, D. Marcotte, L. Nguyen, P. Pilewskie, A. Rangno, D. C. Rogers, K. B. Strawbridge, F. P. J. Valero, A. G. Williams, D. Wyliep. 2000: FIRE Arctic Clouds Experiment. *Bull. Amer. Meteor. Soc.* 81, No. 1, 5-29.

de Boer, G., "An Improved Understanding of the Lifecycle of Mixed-Phase Stratiform Clouds Through Observations and Simulation". PhD dissertation, The University of Wisconsin - Madison, 2009.

de Boer, G., H. Morrison, M. D. Shupe, R. Hildner. (2011) Evidence of liquid dependent ice nucleation in high-latitude stratiform clouds from surface remote sensors. *Geophysical Research Letters* 38:1.

Dorling, S.R., Davies, T.D., Pierce, C.E., 1992. Cluster analysis: a technique for estimating the synoptic meteorological controls on air and precipitation chemistry - method and applications. *Atmospheric Environment* 26A, 2575-2581.

Engström, A. and Magnusson, L.: Estimating trajectory uncertainties due to flow dependent errors in the atmospheric analysis, *Atmos. Chem. Phys.*, 9, 8857-8867, doi:10.5194/acp-9-8857-2009, 2009.

Francis, J.A. and E. Hunter, 2007: Changes in the fabric of the Arctic's greenhouse blanket, *Environ. Res. Lett.*, 2, doi:10.1088/1748-9326/2/4/045011.

Galloway, J. N., Knap, A. H. and Church, T. M. 1983. The composition of western Atlantic precipitation using shipboard collectors. *J. Geophys. Res.* 88.

Greenaway, K. R. 1950. Experiences with Arctic flying weather, Royal Meteorological Society Canadian Branch (Nov. 30, 1950), Toronto, Ontario.

Harris, J. M., Drexler, R. R., and Oltmans, S. J.: Trajectory model

- sensitivity to differences in input data and vertical transport method, *J. Geophys. Res.*, 110, D14109, doi:10.1029/2004JD005750, 2005.
- IPCC, 1990: *Climate Change: The IPCC Scientific Assessment*. J. T. Houghton, G. J. Jenkins, and J. J. Ephraums, Eds., Cambridge University Press, pp. 365.
- IPCC: *Climate Change 2007: The Physical Scientific Basis*, edited by: Solomon, S., Qin, D., Manning, M., et al., Cambridge Univ. Press, New York, USA, 2007.
- 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.
- Jiang, H., W. R. Cotton, J. O. Pinto, J. A. Curry, and M. J. Weissbluth, 2000: Cloud resolving simulations of mixed-phase Arctic stratus observed during BASE: Sensitivity to concentration of ice crystals and large-scale heat and moisture advection. *J. Atmos. Sci.*, 57, 2105-2117.
- Kalkstein, L.S., Tan, G., Skindlov, J.A., 1987. An evaluation of three clustering procedures for use in synoptic climatological classification. *Journal of Climate and Applied Meteorology* 26, 717-730.
- Mitchell, J. F. B., W. J. Ingram, 1992: Carbon Dioxide and Climate: Mechanisms of Changes in Cloud. *J. Climate*, 5, 5-21.
- Mitchell, M. 1956. Visual range in the polar regions with particular reference to the Alaskan Arctic. *J. Atmos. Terrestrial Phys.*, Special Suppl.: 195-211.
- Moody J. L. and Galloway J. N. (1988) Quantifying the relationship between atmospheric transport and the chemical composition of precipitation on Bermuda. *Tellus* 40B, 463-479.
- Olsson, P. Q., and J. Y. Harrington, 2000: Dynamics and energetics of the cloudy boundary layer in simulations of off-ice flow in the marginal ice zone. *J. Geophys. Res.*, 105D, 11 889-11 899.
- Quinn PK, Shaw G, Andrews A, Dutton EG, Ruoho-Airola T, Gong SL (2007). Arctic haze: current trends and knowledge gaps. *Tellus* 59B: 99 - 114.
- Sassen, K. , P. J. DeMott, J. M. Prospero, and M. R. Poellot (2003), Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results, *Geophys. Res. Lett.*, 30(12), 1633.
- Schwarzenboeck, A. Duroure, C. Gayet, J. F. Herber, A. Krecji, R. Lefevre, R. Minikin, A. Neuber, R. Shcherbakov, V. Stroem, J. Aerosol-cloud interaction during the transition time period of Arctic haze to clean summer conditions. *Journal of Aerosol Science*. 2004, VOL 35; SUPP/2, pages 727-728.
- Seibert, P., Helga Kromp-kolb, Anne Kasper, Michael Kalina, Hans Puxbaum, Dieter T Jost, Margit Schwikowski, Urs Baltensperger, Transport of polluted boundary layer air from the Po Valley to high-alpine sites, *Atmospheric Environment*, Volume 32, Issue 23, 1 December 1998, Pages 3953-3965.

Shaw, G. E. 1983. Evidence for a central Eurasian source area of Arctic haze in Alaska. *Nature* 299, 815-818.

Shaw, G. E. and Stamnes, K. 1980. Arctic haze: perturbation of the Polar radiation budget. *Am. N.Y. Acad. Sci* 338, 533-539.

Shupe, M. D., Kollias, P., Persson, P. O. G., and McFarquhar, G. M.: Vertical motions in Arctic mixed-phase stratiform clouds. *J. Atmos. Sci.*, 65, 1304-1322, 2008b.

Shupe, M. D.: A ground-based multiple remote-sensor cloud phase classifier, *Geophys. Res. Lett.*, 34, L2209, <http://dx.doi.org/10.1029/2007GL031008>doi:10.1029/2007GL031008, 2007.

Shupe, M.: Clouds at Arctic atmospheric observatories. Part 2: Thermodynamic phase characteristics. *J. Appl. Meteorol.* 50, 645-661 (2011).

Shupe, M.D., S. Y. Matrosov, and T. Uttal, 2006: Arctic mixed-phase cloud properties derived from surface-based sensors at SHEBA. *J. Atmos. Sci.*, 63, 697-711.

Sokal R R, Michener C D, 1958 A Statistical Method For Evaluating Systematic Relationships. *Univ Kans Sci Bull* 38:1409-1438.

Stohl, A. & Koffi, N.E. Evaluation of trajectories calculated from ECMWF data against constant volume balloon flights during ETEX. *Atmospheric Environment* 32, 4151-4156 (1998).

Tietze, K., Riedi, J., Stohl, A. & Garrett, T.J. Space-based evaluation of interactions between pollution plumes and low-level Arctic clouds during the spring and summer of 2008. *Atmospheric Chemistry and Physics Discussions* 10, 29113-29152 (2010).

Tsushima, Y., S. Emori, T. Ogura, M. Kimoto, M. J., Webb, K. D. Williams, M. A. Ringer, B. J. Soden, B. Li, N. Andronova 2006: **Importance of the mixed-phase cloud distribution in the control climate for assessing the response of clouds to a carbon dioxide increase**, *Climate Dynamics*, 27, 113-126.

Van Diedenhoven, B., A.M. Fridlind, A.S. Ackerman, E.W. Eloranta, and G.M. McFarquhar, 2009: An evaluation of ice formation in large-eddy simulations of supercooled Arctic stratocumulus using ground-based lidar and cloud radar. *J. Geophys. Res.*, 114, D10203, doi:10.1029/2008JD011

Wetherald, Richard T., and Syukuro Manabe, 1988: Cloud feedback processes in a general circulation model. *Journal of the Atmospheric Sciences*, 45(8), 1397-1415.



