Ice Nucleation Measurement and Parameterizations and the Broad Impact of Associated Ocean and Atmospheric Interactions Within the Global Climate

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Honors Capstone Thesis

Abstract

Current climate models attempt to represent the frequency and characteristics of mixed-phase and ice clouds in the atmosphere. Ice crystals have the ability to scatter incoming solar radiation, which may have a net cooling effect on the Earth's energy budget. Ice forms in the atmospheric by either homogenous freezing at temperatures colder than -36°C, or by heterogeneous freezing, which requires an ice nucleating particle (INP) to initiate freezing at temperatures warmer than -36°C. The stochastic nature of ice formation requires more rigorous development and testing of model parameterizations that aim to predict ice formation. In this study, we have measured the immersion freezing temperatures of sea surface microlayer (SML) samples collected in the North Atlantic Ocean and used our results to examine the accuracy of a global ice nucleation parameterization to predict marine INP concentrations. SML samples from the fourth NASA North Atlantic Aerosol and Marine Ecosystem Study (NAAMES4) field campaign were evaluated for freezing temperature on a custom built ice microscope. Primary marine aerosols generated by the SML have been found to freeze from -28.041°C to -21.341°C showing they are moderately effective INP. DeMott et al. 2010 developed a global parameterization to predict the concentration of INP that is dependent on both the temperature and size of aerosol particles. To apply the parameterization, Scanning Electrical Mobility Sizer (SEMS) size distribution measurements collected concurrently with the SML samples during NAAMES4 were used to select aerosol particles larger than 0.5 μm to input into the parameterization. Results of the comparison between observed and predicted number

concentration of INP show that the parameterization does not represent the high degree of variability present in the observed concentrations of the marine INP in this study. Further refinement of this global parameterization may improve its ability to accurately predict number concentrations of INP produced in a marine environment. A detailed overview of the measurements and parametrizations for INP concentrations will be provided. In addition, an examination of the climate implications of this ocean-atmospheric interaction will also be provided.

Introduction

Earth's radiation budget is the balance between incoming shortwave radiation from the sun and outgoing longwave radiation from Earth that ultimately determines the overall temperature of Earth. Clouds alter the budget because of their unique capability to reflect incoming shortwave radiation back to space, and also reflect longwave radiation back to the surface of the Earth. This reflective property depends on the composition of the cloud. Clouds can be composed of liquid water, ice crystals, or a combination of both. Ice crystals are effective at scattering incoming solar radiation, therefor clouds composed of ice crystals are significantly influential in Earth's radiative energy budget (Brooks et al., 2014). Much of the uncertainty in future climate prediction comes from the lack of understanding of the formation of clouds and their influences on Earth's radiation budget (Glen and Brooks, 2014).

The ice crystals found in the atmosphere form through the process of ice nucleation, which can occur in two different ways in two ways. The first process is homogeneous freezing. Homogenous freezing occurs when temperatures in the atmosphere are colder than -36°C and the ice can form on its own (Brooks et al., 2014). If temperatures are warmer than -36°C,

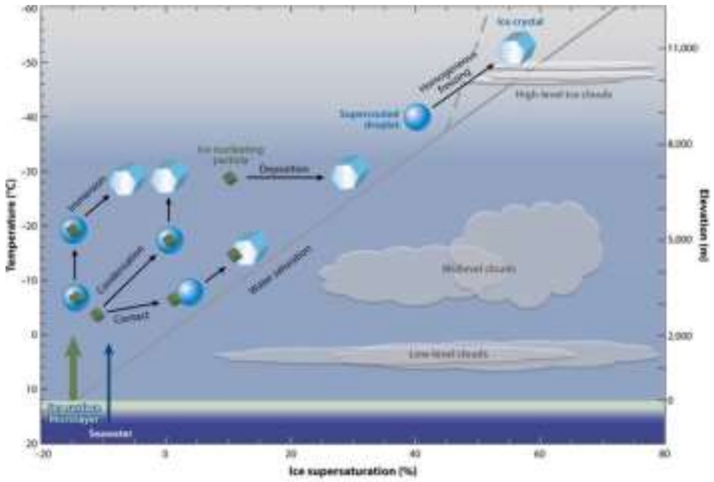

Figure 1: Various pathways ice nucleation occurs using INP from the SML to form low and mid-level clouds above the ocean (Brooks and Thornton 2018).

heterogeneous ice nucleation occurs. Heterogeneous ice nucleation requires an ice nucleating particle (INP) to initiate freezing. Examples of INP are substances like soot, dust and pollen. The process of heterogeneous ice nucleation can occur through four different INP contact pathways. Immersion freezing involves the full INP being surrounded with liquid water within the drop in order to initiate freezing. The second pathway heterogeneous ice nucleation can occur is through condensation where liquid water forms around the INP. The third pathway is contact freezing where the INP is touching the liquid drop along the outer edge in order to initiate freezing. The final pathway is through deposition. This study focuses on the immersions freezing process.

The source of INP that become primary marine aerosols in the samples for this study were taken from Sea Surface Microlayer (SML). This is thought to be a thin film ranging from 1-1,000 micrometers thick (Liss and Duce, 1997). The SML is composed of concentrated organic matter on the surface of the ocean. The organic buildup is caused by the death and subsequent floating of organisms like phytoplankton. The existence and concentration of the SML is

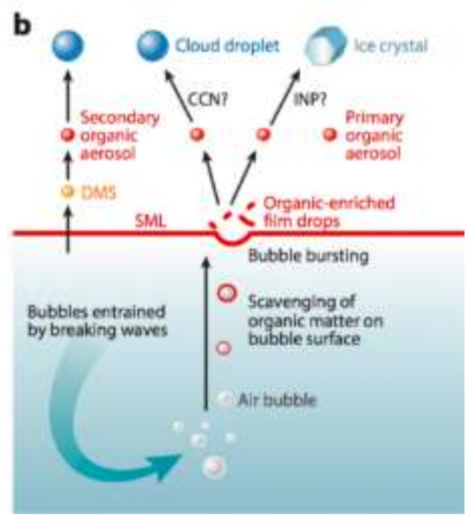

Figure 2: Process of breaking waves causing bubble bursting at the surface, which lofts organic matter from the SML into the atmosphere (Brooks and Thornton 2018).

dependent on many variables like ocean circulation, wave patterns, and weather. The SML is so thin that it is not visible to the human eye, and its existence is predicted based on the current conditions. Research is still being conducted on the characteristics and behavior of the sea surface microlayer. The breakdown of these phytoplankton in the SML have the ability to be lofted through bubble bursting caused by breaking waves at the ocean's surface. The phytoplankton then are able to enter the atmosphere and act as a source of INP to initiate freezing of ice crystals which form low-level clouds.

Experimental Methods

Sample Preparation

The samples used in this study originate from the NASA NAAMES4 cruise deployed over the North Atlantic. This cruise is a part of a five-year study to examine ocean processes, their influences on atmospheric aerosols and clouds, and the overall climate implications for these interactions. The fourth cruise in this series occurred from March 20th, 2018 to April 13th, 2018. The deployment occurred over an accumulating phytoplankton bloom in the North Atlantic. The lab groups who collected the samples that pertain to this research were broadly interested in biological, chemical, and atmospheric interactions that occur in order to prompt cloud formation. Samples and data were recorded with reference to the

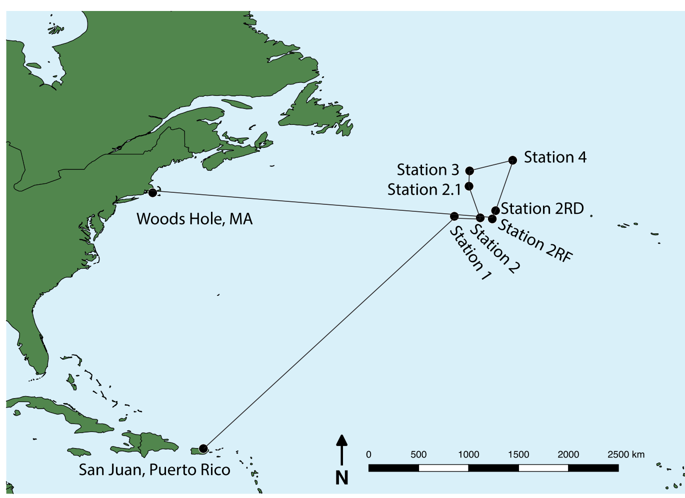

Figure 3: Map of the cruise with stations identified.

station at which they were taken. Station refers to the location and/or stop the ship made during the cruise.

Samples of the SML were taken multiple times throughout the NAAMES4 cruise. If the conditions for a present SML were determined, then a screen was lowered over the side of the ship and immersed in the surface waters. The screen was brought back up onto the ship and the organic material gathered on the screen was collected and stored. These organics and ocean water solutions were then frozen for experimentation in the lab following the cruise. Sampling of the SML occurred about two times per day. The cruise remained at station 2 for multiple days allowing for additional sampling, these stations are referred to as station 2, 2RF, and 2RD. Samples of bulk water were also taken from the same areas the SML samples were taken. Bulk water is the normal ocean water located at a further depth than the SML. The purpose of harvesting bulk water was to have the capability to compare the SML samples with the bulk water samples in the experiment.

Ice Microscope Measurements

The samples of the SML were returned to the lab following the cruise for freezing temperature experiments. The experiments occurred on a custom built ice microscope, and details of this microscope can be found within Fornea et al. 2009. This microscope was built with the purpose to measure the freezing temperatures of liquid drops through the ice

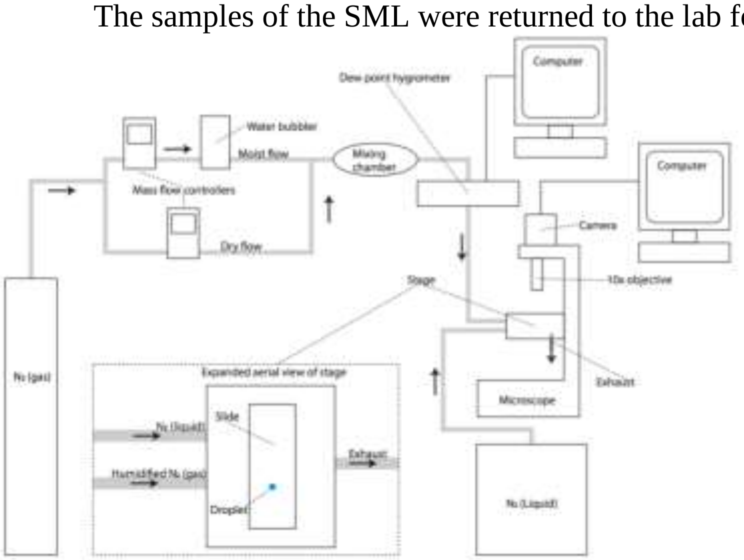

Figure 4: Schematic of ice microscope set up (courtesy of Elise Wilbourn).

nucleation process. The sample is a 2 microliter drop which is placed on to a glass slide and is put on the microscope. Temperature is controlled and monitored through a software package and a liquid nitrogen flow. Condensation and evaporation are controlled within the microscope using mass flow controllers. If the drop appeared to grow because of condensation or shrink because of evaporation the data set was not used. The microscope initiates around twenty-six freeze and thaw cycles on one sample to obtain a significant data set of temperatures that are then averaged. The freezing events are recorded using images taken by a camera every six second throughout the experiment.

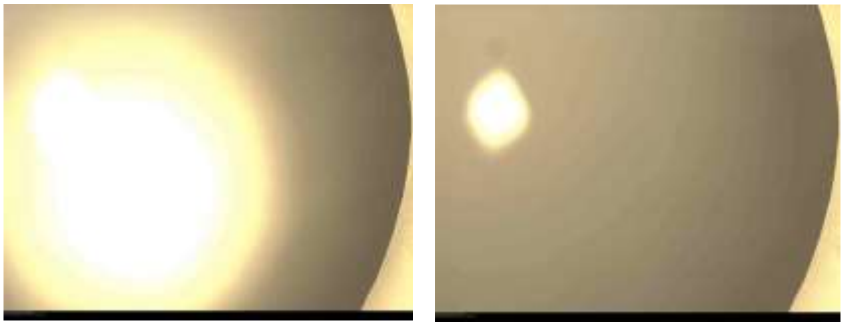

Figure 5: (left) sample image of unfrozen drop from the microscope (right) sample image of frozen drop from the microscope.

Visual evaluation of the images is used to collect the temperature at which the drop froze. Samples of the SML and of bulk water were run to see the freezing temperature difference between the primary marine aerosols from the SML and the deeper ocean water. These temperatures were used to determine the number concentration of INP in the SML. Number concentration refers to the number of INP present that are capable of initiating freezing.

Aerosol Measurements

Also during the NAAME4 cruise, aerosol size distribution measurements were taken using a Scanning Electrical Mobility Spectrometer (SEMS). The SEMS gathers air samples and measures the sizes of particle in the sample based on pre-determined size bins. SEMS data was gathered continuously throughout the cruise, however the data used here was taken simultaneous with the SML samples for a fifteen-minute period, which is the average time the SML sampling took. For this study, sizes of particles counted at 0.5 microns in diameter or larger were used. The use of only aerosols 0.5 microns or larger was determined using previous research, which concluded that aerosols of this size or larger are the most efficient at acting as INP (DeMott et al. 2010). These aerosol size distributions were used as the number concentration of aerosols in the DeMott et al. 2010 parameterization that is describe in the next section.

Parameterization Evaluation

The second component to this study is evaluating an existing global parametrization that attempts to predict the number of INP present that assist in initiating freezing to prompt the formation of clouds. A parametrization is a mathematical attempt to replicate what happens in the physical world in order to improve the ability to model these natural phenomena more precisely. The parametrization used here can be found within DeMott et al. 2010.

$$n_{IN,T_k} = a(273.16 - T_k)^b (n_{aer0.5})^{(c(273.16-T^k)+d)}$$

n_{IN,T_k}= ice nuclei number concentration
T_k= temperature in Kelvin
$n_{aer0.5}$=number concentration of aerosols with diameter >0.5µm
$a = 0.0000594, b = 3.33, \quad c = 0.0264$,
$d = 0.0033$ are best fit coefficients
)

The values for temperature were predetermined to fall between -40°C and 0°C. This is the temperature interval at which freezing of any type occurs in the atmosphere. The values of $n_{aer0.5}$ were determined using the SEMS data collected during the NASA NAAMES4 cruise. The best fit coefficients $a, b, c, and\ d$ remained constant from the DeMott et al. study to this one. This parametrization uses a new approach that not only uses temperature to evaluate INP concentrations, but also considers the diameter of particles to play a key role in properly determining the concentration of INP. Current climate models are not using parametrizations that include a size dependence, and DeMott et al. 2010 argues that their parameterization provides a more accurate representation of the actual number of INP present in the physical world. The DeMott et al. 2010 study utilizes the data resulting from multiple field campaigns to compose the observed INP concentrations. Observed concentrations are then compared to the predicted INP concentrations that were calculated using the parametrization. A strong correlation between the observed INP concentrations and the predicted INP concentration means that the model parameterization is accurately able to predict what is occurring in the physical world. This correlation presents itself on the graph of the observed versus predicted values with a significant amount of data falling on the 1:1 predictive line. DeMott et al. 2010 reports 62% of data fall within a factor of two, shown as the dashed lines, of the 1:1 predictive line, shown as the solid line. DeMott et al. 2010 concludes that this parameterization is more effective at correctly representing the prediction of global INP concentrations than current climate models do.

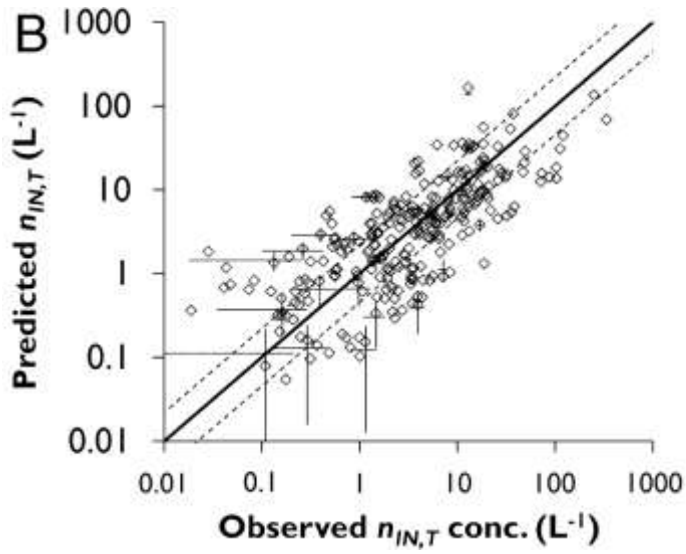

Figure 6: DeMott et al. 2018 results showing the comparison between observed concentrations and predicted concentration.

Results and Discussion

SML Freezing Temperatures

The SML samples taken on the NAAMES4 cruise shown freezing temperatures ranging from -28.041°C to -21.341°C. Certain station samples reveal a significant difference in freezing temperature of the bulk water in comparison to the SML. Other station samples do not show an evident or significant difference between the freezing temperatures of the bulk water and the SML. Exact freezing temperatures of SML and bulk water samples are shown in the figure below. These results show that the primary marine aerosols originating in the SML act as moderately effective INP. This supports subsequent studies that reported marine biological aerosols initiating freezing at high temperatures (DeMott et al 2016, Wilson et al 2015).

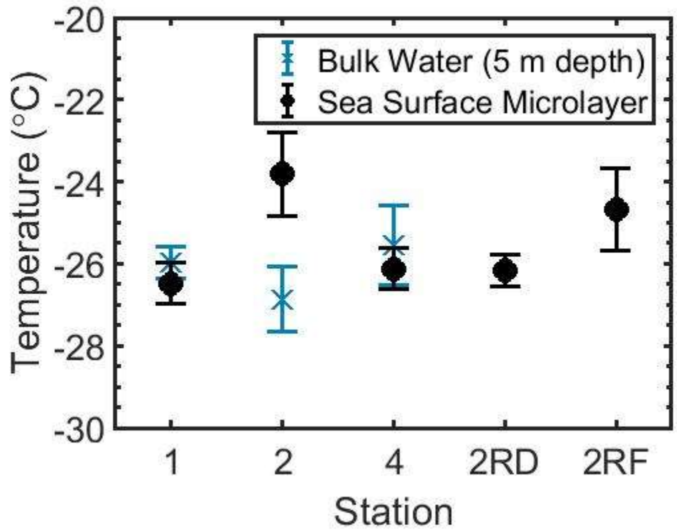

Figure 7: Freezing temperatures obtained from the NAAMES 4 bulk water and SML samples.

The overall implications of the process of phytoplankton decay becoming concentrated in the SML, lofted due to bubble bursting in order to act as moderately effective INP shows the complexity that encompasses oceanic and atmospheric interactions. Primary marine aerosols generated from the SML will add to the list of known INP sources alongside dust, soot, and many other industrial sources (Brooks and Thornton 2018). The identification of the effectiveness of phytoplankton to act as INP, though through a complex process, helps contribute to the knowledge base that is attempting to understand the creation and composition of ice crystals in the atmosphere that play a key role in regulating the climate. More experimentation may be required to continue to identify INP sources that are as effective, if not more effective, than the one of this study.

Parameterization

The results of the parameterization show a significant difference between observed and predicted INP concentrations. A strong correlation along the 1:1 predictive line represents a model that is able to accurately predict INP concentrations as the predicted values in comparison to the observed values that were obtained using the ice microscope measurements of SML samples. Our results show that the DeMott et al. 2010 parameterization was not effective at accurately predicted INP concentrations for our samples of primary marine aerosols.

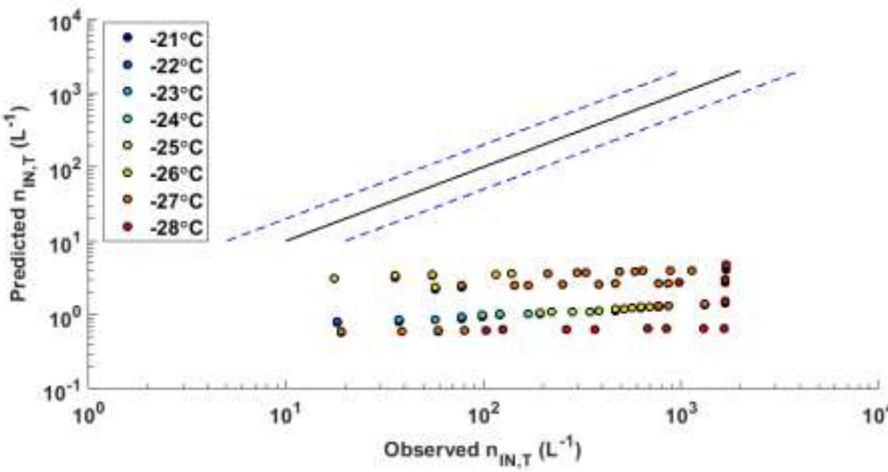

Figure 8: Predicted INP concentration values determined from the DeMott et al. 2010 parameterization compared to the observed INP concentrations obtained from ice microscope measurements.

There are two possible explanations for the parameterization's lack of ability to accurately represent our observed INP concentrations. The first explanation may be that the DeMott et al. 2010 parameterization is not able to encompass the stochastic nature of marine INP along with other sources of INP within a single parameter. If this is the case, more research and refinement of this global parametrization, and ones similar to it, is necessary in order to better represent natural processes in climate models. Though models require a few assumptions about the natural world in order to attempt to recreate it, global parameterizations should be able to replicate both land processes, marine processes, and the interaction of the two generally without the loss of crucial details.

The second possible explanation for the parametrization's inability to accurately represent observed INP concentration is the influences of salt on the samples taken from the SML. Previous studies have concluded that the presence of salt has the capability to widen the range of freezing temperatures to much colder values (Bodnar, 1993). If this is the case, then the actual observed freezing temperatures may be warmer than the results from the ice microscope showed. This explanation would also influence the observed values that are compared to the predicted values calculated by the parameterization, and therefor, the influence of salt may be distorting the observed versus predicted comparison too. Further research is being conducted by co-authors of this paper using dilution techniques in order to understand the influences of salt on freezing temperatures and the associated INP concentrations.

Future Work

Provided that there would be a significant of volume of sample left from the NAAMES4 cruise, additional experimentation could be conducted using samples that have been diluted. This would allow for a comparison to establish the significance of salt on the freezing temperatures of the SML and bulk water samples.

It may also be beneficial to investigate the process of storage and freezing of the samples during the time after they are collected in the field but before they are experimented with in the lab. A concern could be the alterations of the biogenic samples and their associated freezing temperatures because of the initial freezing and storage of the samples after they are collected. However, a few studies have concluded that the initial freezing and storage of seawater samples does not have an effect on the contents of the sample and its ability to act as an effective INP (Irish et al 2017 ;Schnell and Vali, 1975). Controlled lab experiments have recently been performed in mesocosms to initiate the growth of phytoplankton blooms and measure the resulting primary marine aerosols (McCluskey, 2017). This eliminates the need to freeze the samples for a long period of time.

There has also been development in improved parametrizations since the DeMott et al. 2010 parameterization. In Tobo et al. 2013 a small change in the best fit coefficients was made to the original DeMott et al 2010 parametrization in order to better fit the observed data for a forest environment containing fluorescent biological aerosol particles.

Additionally, instrument development is being conducted to create a mobile ice microscope set up that is able to test multiple samples. The Biological Ice Nucleation on the Go (BINGO) is in the development phase. The current in-lab ice microscope would be nearly impossible to take on a field expedition to do in-situ measurements, so the concept behind BINGO is to have a smaller set up that can monitor multiple drops as they freeze and record the respective freezing temperatures.

Climate Implications

The role of clouds in the regulation and prediction of the global climate has been a large source of uncertainty for decades (IPCC 2013). Characteristics of clouds determine their role in Earth's radiative energy budget. Altitude, composition, and longevity of the cloud all influence the clouds capability to reflect incoming solar radiation in order to cool the planet, or reflect longwave radiation back to Earth's surface acting as an insulator to maintain or increase Earth's temperature. The processes previously described in this study have the capability to create low-level clouds that can reflect incoming solar radiation resulting in a net cooling effect of the planet.

In order to create these ice clouds, ample supply of the building blocks of the clouds – INP – are necessary. Since over 70% of the Earth's surface is covered in water, understanding how oceans interact and exchange with the atmosphere is crucial. The importance of biogenic INP sourced from the ocean in remote and high latitude areas have been shown to be essential in the creation of low and mid-level clouds composed of ice (Wilson et al. 2015). Without land influences to create the INP sources of dust, soot, or industrial outputs, remote and high latitude locations rely on the ocean for INP to initiate the creation of clouds.

Understanding the availability of the phytoplankton and the timing of their death and decay is crucial in order to accurately represent their role as INP in the atmosphere. Ocean characteristics such as available nutrients and temperature determine the success of phytoplankton blooms. The effects of climate change have altered the timing of spring onset, and therefor, altered the onset of phytoplankton blooms (Friedland et al. 2016). As ocean temperatures change, the timing and availability of phytoplankton to act as INP will also change. If populations of phytoplankton were to increase or decrease, then the availability of phytoplankton to act as INP could also increase or decrease. This would affect the overall presence and longevity of ice clouds over phytoplankton blooms. Further exploration to understand the temperature dependence of the ocean on phytoplankton blooms would be beneficial to further understand this ocean-atmospheric interaction.

Though the scope of this research project was narrow to encompass a very small part of the ocean-atmospheric interaction that helps determine the climate of the planet, the overall goal to better understand these complicated interactions was achieved. The cloud forcing remains a large uncertainty in modeling the physical world. Further research and refinement to improve the knowledge of INP sources and effectiveness would be beneficial in order to eliminate uncertainty to make more accurate predictions about climate change.

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