Abstract

As children, we lie in fields and stare at the clouds for hours, their form, the shapes they make, and the way they seem to play with the light in the sky are fascinating to us. However, despite of our early fascination with clouds, we know surprisingly little about clouds and cloud processes. This paper outlines a technique developed to look at the formation of ice in clouds as part of a larger effort to understand more about clouds and their role in the atmosphere. This work is still in the early stages of development, however preliminary results obtained thus far seem to reflect our current understanding of ice formation in clouds accurately.

Introduction

The attempt to understand climate and create General Circulation Models (GCMs) as a tool to predicting future states of the climate has generated a need for a better and more thorough understanding of many atmospheric processes. Within this context, the formation of ice crystals in clouds has proven to be one of the most difficult problems to surmount. Generally speaking, modeling the formation of ice in clouds has been difficult because the process by which ice forms in clouds is sensitive to temporal and spatial variations by a factor of 100 or 1000\(^1\), making cloud ice formation difficult to generalize and difficult to model in the lab.

A large portion of what makes understanding ice formation difficult is that the commonly held notion that pure freezes as 0\(^\circ\) C is incomplete. Thermodynamics reveals that because it is energetically unfavorable for ice crystals to form at warmer temperatures, homogenous (pure) water will not freeze until it reaches ~-40\(^\circ\)C. However, in heterogeneous (impure) systems, which we more commonly observe, non-H\(_2\)O materials can provide a substrate, or nucleus, on which water molecules can collect to form an ice-like aggregate. When these aggregates come into contact with any water that is below 0\(^\circ\) C, they can act as a seed which causes the water to freeze.

Because water is brought into clouds as a vapor, it is exceptionally homogenous. However, if the ice in clouds were homogenous, we would only expect to only find ice at temperatures around -40\(^\circ\) C. However, experiments looking at the actual concentration of ice in clouds with respect to temperature

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indicate that ice readily forms in clouds at temperatures warmer than -15° C. This observation demonstrates that ice formation in clouds must include a heterogeneous component involving ice nuclei.

Our current understanding of ice nucleation in clouds includes four possible processes for the formation of ice. 1) Water vapor can form ice directly on deposition nuclei. 2) Water can condense on condensation nuclei and then freeze. 3) Pure water droplets can form and then be hit by contact nuclei. 4) Immersion nuclei can enter into a droplet and subsequently cause freezing. These four processes are coming to be understood, however their function in the atmosphere still contains a great deal of uncertainty and complexity. Figure 1 shows findings on ice nucleation with respect to temperature from Vali (1976), Deshler (1982), Rogers (1982), and Cooper (1980).

Figure 1

These findings illustrate the general trend that concentration of ice nuclei increases with decreasing temperature. However, the distribution of their data also shows that there is still a great deal of uncertainty in the formation of ice in clouds. In addition to their findings about the general relationship between temperature and ice nucleation, all four investigations revealed that at -15°C, for instance, that the concentrations of immersion and deposition nuclei were approximately 10 to 20 times lower than those of condensation nuclei.

A number of different types of experiments have been done in an attempt to gain a greater understanding of the ice nucleation process. Two of the main means through which investigations into ice nucleation have proceeded are: looking at the freezing process in cloud chambers and other methods in which particles are suspended in air, and studying the water freezing process on filters on which ice

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2 Beard, Kenneth  
3 Rogers and Yao  
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nuclei have been collected. Both these experimental techniques have advantages and shortcomings. Cloud chambers can be an effective way of looking at ice nucleation because the basic notion of a cloud as water droplets suspended in air can be recreated. However, cloud chamber investigations are hindered by the difficulty of inserting and controlling ice nuclei found in the actual atmosphere. On the other hand, collecting ice nuclei on filters and then observing the freezing properties of water on the filters provides a convenient way of modeling freezing processes associated with actual ice nuclei. However, resolving the contribution of the filters themselves to the nucleation process and separating it from contributions of actual nuclei is a challenging puzzle. As a result, part of the work on ice nucleation currently underway involves improving experimental techniques with the hope of gaining a more accurate picture of the ice nucleation process and incorporating it more effectively into GCMs.

Experimental Methods

The basic process we developed to look at filters that had been used to take atmospheric samples involved four steps, placing the filter on a cold stage, cooling it down to the testing temperature, making water droplets with humid air, and observing the filter for ice nucleations.

First, the filter was placed on the cold stage to where it would be cooled down. The cold stage consisted of a thick aluminum plate with holes drilled through the core and a thin copper plate on top to help cool the sample evenly. The stage was connected to a cooling bath which continuously circulated liquid silicone cooled to ~−35°C through the cavity in the aluminum plate as the cooling mechanism. Additionally, an adjustable heater with feedback was installed, which enabled us to set the temperature of the stage as required to test ice nucleation at varying temperatures. To prevent the copper from possibly acting as an ice nucleating mechanism, a piece of saran wrap was placed on top of the copper plate. Finally, a layer of microscope slide oil was place on top of the saran wrap, which soaked through the filter, helping to minimize the effects of the filter on the ice nucleation process.

Once the filter was placed on the cold stage and had become saturated with oil, a ring of Styrofoam with six holes cut in it was placed around the filter and a Buchner funnel was placed over the ring. The funnel and ring system created a chamber into which air, humid, dry, or semi-humid, could flow. A Buchner funnel was used because the built-in fritted glass disk disrupted inflowing air creating a situation, which did not favor one area of the filter for the condensation of water droplets. As the filter was cooled, air, dried to ~8% relative humidity at ~23°C flowed into the chamber at a rate of 1 liter per minute preventing the formation of water droplets until the target temperature was reached.

Once the filter was cooled to the test temperature, the dry air flowing on the filter was replaced with humid (several hundred percent humidity relative humidity at the test temperature) air flowing at a rate of 1 liter per minute for 2–4 minutes, in order to form water droplets. This process was chosen in an
attempt to replicate the process of formation and freezing of water droplets in clouds as closely as possible. The greatest difficulty of creating water droplets with humid air was found to be that at different temperatures, because relative humidity varies with temperature, water droplets formed at different rates. As a result, filters at different temperatures were exposed to humid air for different periods of time based on a qualitative analysis of droplet size.

Finally, after droplets had formed on the filter, the humid air was replaced with a mix of humid and dry air, ~15% relative humidity at ~23°C, which flowed onto the filter at a rate of ~.2 liters per minute. The mixed air created a balanced situation within the chamber humid enough to prevent water droplet evaporation and dry enough to minimize the deposition of ice on already formed ice nuclei. At the end of three minutes under the mixed airflow, the number of ice nucleations was counted visually and the filter was disposed of.

**Results and Analysis**

Developing an effective technique for analyzing atmospheric samples of ice nuclei taken on filters in a manner closely mimicking the formation of ice in clouds was the primary focus of this experiment. In attempting to achieve that goal, real filter samples, taken at ground level, were used from the area of Boulder Colorado to generate data reflecting the number of ice nuclei in the atmosphere. In as much as these data are very preliminary findings based on this technique, they do seem to correlate with other data taken from around the world by other research groups.

In analyzing the data found from the experimental procedure, the number of nucleations counted for a blank filter was subtracted from the number of nucleations counted for a test filter at a given temperature. This number of net nucleations was then divided by the number of liters of air sucked through the test filter to yield nucleations per liter. Figure 2 shows a summary plot of the data obtained from this process.

![Nucleations per Liter Air vs Temp](image)

*Figure 2*
The two sample groups, taken at the east and west ends of Boulder seem to generally represent some kind of exponential growth in the number of ice nucleations per liter of air as temperature was decreased. Figure 3 shows these same data sets with the number of nucleations converted to a logarithmic scale.

Comparing Figure 3 to Figure 1 reveals a general correlation of the data taken with the technique developed in this experiment and data sets previously contributing to our understanding of the ice nucleation process.

**Conclusions**

Generally speaking the process developed in this experiment yielded results consistent with our knowledge of ice nucleation in clouds. However, the process could be developed considerably from its current evolution. There are a number of aspects of the process, which have not yet been standardized; for example, the time that humid air is pumped into the test chamber varies from temperature to temperature on a qualitative basis. Additionally, more data is needed to help establish the repeatability of the process and the statistical uncertainty associated with each measurement of the ice nuclei concentration for a given temperature. Finally, in developing the process, an unforeseen effect was noticed the number of ice nucleations at a given temperature based on the testing technique. Generally speaking, water droplets created at 0°C and cooled to a low temperature froze less often than water droplets created when the filter was already fully cooled down.

Hopefully, the work from this investigation will be helpful to understanding of the function ice and ice nuclei in the atmosphere and our modeling attempts through GCMs. Currently, ice nucleation is one of the aspects of climate modeling that we understand least well. In January of 2001, the Summary...
for Policy Makers from Working Group I of the Intergovernmental Panel on Climate Change summarized our understanding of the science behind climate in a single figure. Figure 4 shows that our understanding of the Earth’s climate system is relatively complete in some areas, such as the contribution from greenhouse gases, however in other cases, such as mineral dust (a primary ice nuclei), we cannot tell if their contribution to radiative forcing will be positive or negative. A better understanding of ice nucleation could help us to come to more certain conclusions about the contributions of mineral dust to climate change, and as a result, help us to make better climate related decisions.

Figure 4\(^5\)

\(^5\) IPCC, Working Group I, Summary for Policy Makers
References


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