CONTRIBUTIONS FROM

THE CLOUD PHYSICS GROUP

REVIEW ARTICLES II

Notes for Nine Lectures on the "Microphysics of Clouds" given by Professor Peter V. Hobbs at the Advanced Study Symposium on the "Microphysics and Dynamics of Convective Clouds" organized by the National Center for Atmospheric Research and held at NCAR, Boulder, Colorado, from July 6 to August 18, 1970.
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I. CONденSATION OF WATER VAPOR IN THE ATMOSPHERE; CLOUD CONденSATION NUCLEI

A. Introduction

Clouds form because of the adiabatic expansion of moist air. During such an expansion, the relative humidity in the air increases to the point of supersaturation (i.e., larger than 100%). At a certain supersaturation, the water vapor condenses to form cloud droplets, the nucleation of which will be briefly reviewed here. At the outset, we shall distinguish two different types of nucleation and discuss them separately.

B. Homogeneous nucleation

This type of nucleation (also known as spontaneous nucleation) occurs in a system of pure water vapor. By raising the relative humidity of such a system (by either cooling it or isothermally compressing it) it can be brought to a state of supersaturation. To attain the state of lowest energy, the vapor should then condense to form small droplets. However, a small droplet is unstable to thermal agitations and will evaporate if the equilibrium vapor pressure over its surface exceeds that in the environment. In other words, condensation does not necessarily occur when the relative humidity exceeds 100%. As the degree of supersaturation increases, small droplets become less unstable. At a certain critical supersaturation, condensation of vapor occurs to form a dense fog. This is homogeneous nucleation.

In experiments with air carefully cleaned of particulates, C.T.R. Wilson found that spontaneous condensation in the form of a dense fog occurred at a supersaturation of about 800%. At this critical supersaturation small embryo droplets can form, by chance aggregation of water molecules, which are large enough to survive and to grow by condensation from the vapor phase.

C. Heterogeneous nucleation

Wilson also observed the formation of a less dense fog at super-
saturations of about 400%. The concentration of droplets in this fog did not increase with increasing supersaturation nor did it decrease by successive expansions. The nuclei responsible for the droplets could not be removed by filtering the air. However, a much denser fog formed when the air was radiated with X-rays. Wilson concluded that this fog was formed by heterogeneous nucleation on ions.

In air containing particulates, condensation occurs at much lower supersaturations than for homogeneous nucleation or heterogeneous nucleation on ions. This is due to the fact that the particulates act as nuclei for the condensation. In the atmosphere the air is never sufficiently clean for either homogeneous nucleation or for nucleation on ions to occur. Instead, condensation occurs at supersaturations of less than 1% on a certain fraction of the solid particles in the air. These are called cloud condensation nuclei (CCN). The efficiency of a particle as a CCN depends on three factors: (a) the size of the particle; (b) the contact angle of water on its surface; and (c) the solubility of the particle in water. These three factors are discussed in turn below.

The larger the size of the particle, the more effective it is as a CCN. This is a direct result of the decrease in the equilibrium vapor pressure over a curved surface with decrease in curvature (Kelvin's equation).

In order for a particle to be an effective CCN, the contact angle $\theta$ of water on the particle must be small. In the limiting case of $\theta = 180^\circ$, the drop merely sits on the surface of the particle and the latter is not effective as a CCN.

The effectiveness of a particle as a CCN increases as its solubility in water increases. This is because the equilibrium vapor pressure above
a solution droplet is less than that above a pure water droplet of the same size. This solubility effect works in the opposite direction to the curvature effect producing the results shown in Fig. 1 (see end of Lecture #1). When the radius of a droplet is small, the solubility effect is dominant. However, as the size of the droplet increases the effect of curvature dominates.

Most particles in the atmosphere appear to consist of an insoluble and a soluble component. For relative humidities less than 70%, the radius of the particle is constant (insoluble part). As the relative humidity increases above 70%, the mixed nucleus changes in size with humidity in much the same way as a wholly soluble nucleus of equivalent size.

D. Sources of condensation nuclei

There are two main processes by which CCN are produced in the atmosphere: dispersion and coagulation.

Dispersion involves the break-up of large particles to form smaller ones which remain suspended in the air. This process generally produces particles greater than 0.1μ in diameter.

Under the title coagulation, we include chemical reactions of gases in the atmosphere to produce particulates and the coagulation of smaller particles by Brownian motion. Particles produced by coagulation are generally less than about 0.1μ in diameter.

It has been thought for some time that salt particles from the ocean may be an important source of CCN. However, measurements made by Radke and Hobbs (1969a) indicate that even in maritime air masses the concentrations of sodium particles are only a small fraction of the concentrations of CCN in the air.

E. Measurement of CCN

The concentration of CCN in the air active at a given supersatur-
ation may be determined by counting the number of water droplets which form in a given volume of air raised to a known supersaturation. The Aitken expansion chamber is not a suitable device for this type of measurement since the supersaturations obtained in expansion chambers are generally several hundred per cent, rather than the fractions of one per cent at which we need to measure the concentrations of CCN. A much better device for measuring CCN is the thermal diffusion chamber in which a small vertical temperature difference is mounted between two horizontal plates which are wetted with water. The water vapor is distributed between the plates by mixing and diffusion. The water vapor pressure $p$ at a given point in the chamber is linearly related to the temperature. However, the saturated vapor pressure $p'$ is not a linear function of the temperature. The result is that at points between the two plates the chamber is supersaturated with respect to water. The maximum supersaturation in the thermal diffusion chamber is determined by the temperature difference between the plates and can be calculated.

An automatic CCN counter has recently been developed by Radke and Hobbs (1969b). In this counter the concentration of water droplets which form in a thermal diffusion chamber is determined by a light scattering technique and the number is recorded digitally (Fig. 2, see end of Lecture #1).

F. Some results of CCN measurements

The results of measurements of CCN in different air masses are shown in Fig. 3 (see end of Lecture #1). It can be seen that the concentrations of CCN in continental air masses are generally greater than those in maritime air masses. Recent measurements at the University of Washington by Hobbs et al. (1968) and Radke (1970) have shown that when clouds evaporate, they can release
high concentrations of CCN. Some results obtained in wave clouds which show this effect are presented in Figs. 4 and 5 (see end of Lecture #1). One possible explanation for this effect is that droplets in clouds collect other particles by various diffusion processes and direct aerodynamic capture, etc. When the droplets evaporate, these captured particles will be released as bigger particles and therefore more efficient CCN. Another possible explanation is that gaseous reactions in cloud droplets may produce compounds (e.g. sulfates) which again will result in efficient CCN when the droplets evaporate.
REFERENCES


Figure Captions

Figure 1 - Equilibrium vapor pressures over droplets of a given radius containing given masses of soluble salt

Figure 2 - An automatic cloud condensation nucleus counter (From Radke and Hobbs, 1968)

Figure 3 - Concentrations of CCN as a function of supersaturation in different air masses (From Radke and Hobbs, 1969)

Figure 4 - Measurements of CCN in wave clouds (From Radke, 1970)

Figure 5 - Measurements of CCN in wave clouds (From Radke, 1970)
Lecture 1, Figure 1

Supersaturation (%) vs. Droplet Radius (μm) for various concentrations of impurities, with 1 ppm/ML being 10^-18 per drop. The graph shows the supersaturation curves for different impurity concentrations and highlights the difference between pure water and impure water at 0°C.

Key:
- Pure Water
- Impurities: 10^-18, 10^-17, 10^-16, 10^-15, 10^-14, 10^-13

Lecture 1, Figure 1
Lecture 1, Figure 2
1700 March 23. Wind W 12 mph.
Snowing. Maritime air.

0900 March 23. Wind SE 3 mph.
Snowing. Modified maritime air.

0900 March 20. Wind S 5 mph.
Clear. Maritime air.


1200 Feb. 15. Wind E 15 mph.
Clear. Continental air.

1330 Feb. 18. Wind SE 15 mph.
Snowing. Transitional maritime-continental air.
Lecture 1, Figure 4

CCN measurements in a lee-wave cloud near Mt. Rainier, Washington

CCN measurements in a "cap" wave cloud over Mt. Rainier, Washington

May 4, 1970
Flight II

Wind 50 KPH
II. GROWTH OF DROPLETS AND PRECIPITATION IN WARM CLOUDS

A. Growth of cloud droplets by condensation

Following our consideration of the initial stages of condensation, we must consider the growth of cloud droplets. The rate of growth of an isolated droplet of radius $r$ by condensation from the vapor phase is given by $\frac{dr}{dt} = \frac{1}{r}$. Therefore, the droplets grow to a uniform size with time but the growth rate decreases (Fig. 1, see end of Lecture #2).

In a cloud we must consider the growth of a large population of droplets in a rising parcel of air. It may be assumed that the direct interactions between droplets is negligible. However, droplets influence each other by their combined influence on their environment. As the parcel of air rises, the saturation ratio increases. Once the saturation point is passed, condensation begins on the most efficient CCN. The supersaturation (SS) continues to rise and CCN increases. The rate of increase of SS is reduced because growing drops consume excess vapor. When the excess vapor is removed as quickly as it is being made available by condensation, the SS falls to zero.

Figure 2 (see end of Lecture #2) shows the growth of droplets by condensation onto various masses of salt for a lift rate of 60.4 cm sec$^{-1}$. We see from these calculations that:

(1) For first 10 sec, when the rate of condensation is negligible, the SS increases linearly with the time. The SS reaches a maximum of 0.35% in 25 sec, and then decreases.

(2) Above certain critical SS, the droplets increase rapidly in size and become uniform.

(3) As SS falls, the smaller droplets which have not been activated fall back to smaller sizes but larger drops continue to grow slowly.
(4) Growth by condensation becomes very slow after a few minutes and drops have only reached a radius about 20μ in 10 min or so.

B. Growth of precipitation particles by coalescence

The relative sizes of cloud and precipitation particles are shown in Fig. 3 (see end of Lecture #2). The volume of a typical cloud droplet (radius 10μ) has to increase by 10^6 in order to produce a precipitable particle of radius 1mm.

The growth of cloud drops from a radius of 10 or 20μ to raindrop size (> 100μ) is an important problem in cloud physics. Condensation alone is too slow; in warm clouds precipitable drops can only form by the collision and coalescence of cloud droplets of different sizes.

We define the collision efficiency E as the fraction of drops lying within the geometrical cross-section of a collector drop which actually collide with the collector.

Then with reference to Fig. 4 (see end of Lecture #2)

\[ E = \frac{y^2}{(a_1 + a_2)^2} \]

or, sometimes

\[ E = \frac{y^2}{\frac{1}{2}a_1^2} \]

where y is the maximum impact parameter (Fig. 4). We may also define a linear collision efficiency as \( y/a_1 \).

Figure 5 (see end of Lecture #2) shows calculated linear collision efficiencies. We see from these theoretical results that:

(1) The larger the collector drop the higher the collision efficiency.

(2) The collision efficiency is very small but still finite when the radius of the collector drop falls below 20μ.
Another question to be answered is: does coalescence always occur when two drops collide? We know from laboratory experiments that under certain conditions (not necessarily representative of those in the atmosphere) droplets can rebound after colliding with a water surface. However, it is generally assumed that in natural clouds the coalescence efficiency is unity.

C. Models for growth by coalescence

1. Continuous model: In this model it is assumed that larger drops grow by continuously capturing smaller drops uniformly distributed in space (Fig. 6, see end of Lecture #2). This model predicts that drops of a given size grow at the same rate.

2. Stochastic model: This is a statistical growth model. Consider those few drops which have made a coalescence collision after a rather short time. They are now in a more favorable position than their fellows to make a further collision because of their larger size. These second collisions are similarly statistically distributed giving a further widening of the spectrum (Fig. 7, see end of Lecture #2). The stochastic model predicts that the droplet spectrum is substantially determined by the first twenty captures. After this the statistical fluctuations are unimportant and the continuous growth equation may be used.

D. Growth by coalescence: The results of some model calculations

Droplets can grow to 20μ by condensation alone and droplets larger than 40μ can grow rapidly by coalescence. But what about the growth of the droplets from about 20μ to 40μ radius?

We will consider the growth from 20μ to 40μ by coalescence based on some model calculations by Bartlett (1966).

Figure 8 (see end of Lecture #2) shows a typical distribution of drop sizes in the early stages of growth of a cumulus cloud. The largest
drops have radius 26µ and are present in concentrations of 3/litre. The tail of the distribution at large sizes is most important for the production of precipitation.

Starting from the distribution shown in Fig. 8 Bartlett computes in a step-wise fashion the development of the spectrum due to growth by coalescence. Resulting drop-size distribution at various times are shown in Fig. 9 (see end of Lecture #2).

Figure 9 shows development of distribution shown in Fig. 8 with time. We see from Fig. 9 that:

1. Large drops develop with time.
2. There is little change in the distribution for drops with radius less than 10µ.
3. The concentration of 40µ drops exceeds 1/litre after 7 min and exceeds 10/litre after 16 min.
4. Most of the liquid water for the growth of the larger drops comes from drops with radius between 11 and 22µ. There must be a good supply of drops in this size range in order for larger drops to grow by coalescence. There is a significant amount of liquid water in drops greater than 40µ which, in this model, are assumed to fall out of the parcel. A convenient way to represent the development of precipitation is to plot this overflow as a fraction of the total liquid-water content. This is shown in Fig. 10 (see end of Lecture #2).

We see that initially there is no significant overflow, but after about 7 min the overflow begins to increase rapidly and after 14 min it increases at an almost constant rate. A rough calculation shows that this overflow can produce a precipitation rate of about 7 mm/hr. This is the right order of magnitude for moderate cumulus shower and indicates that coalescence is capable of producing precipitation.
Figure 11 (see end of Lecture #2) shows assumed initial drop size distributions.

Figure 12 (see end of Lecture #2) shows a table of characteristics of droplet spectra shown in Fig. 11.

A convenient way of comparing the development of precipitation for the different drop size distributions shown in Figs. 11 and 12 is to plot the overflow as a function of time. The results are shown in Fig. 13 (see end of Lecture #2). Figure 13 shows overflow as function of time for different initial distributions. We see from these results that:

(1) In maritime cumulus, if coalescence continued for 15 min, 25% of liquid water would be removed by drops larger than 40μ radius. Hence the coalescence mechanism could produce rain without difficulty in this case.

(2) The continental clouds need more time to get one droplet of 40μ than maritime clouds (since in maritime clouds there are initially some large drops).

(3) Maritime clouds are more efficient than continental clouds in producing precipitable particles by coalescence.

(4) An interesting case is spectrum IV which fails to produce 0.05% overflow in 30 min although it contains a few more large drops than II d. This is because in IV there is a deficiency of drops in the range 14 to 20 microns, which are needed to supply water for large drops.

Bartlett's calculations also show that the minimum collector size drop is the most important single parameter governing the onset of the coalescence mechanism, whereas, any changes which are equivalent to an increase of less than 10% in the collection efficiencies can be ignored.

Notes taken by:
Y. M. Chang
W. H. Mach
REFERENCES


FIGURE CAPTIONS

Figure 1 Growth of a droplet by condensation.

Figure 2 The growth of droplets by condensation onto various masses of salt for a lift rate of 60.4 cm sec\(^{-1}\) (from Howell, 1949).

Figure 3 Relative sizes of cloud and precipitation particles (from McDonald, 1958).

Figure 4 Definition of symbols.

Figure 5 Calculated linear collision efficiencies (from Davis, 1967).

Figure 6 Growth by coalescence according to continuous model.

Figure 7 Growth by coalescence according to stochastic model (from Berry, 1967).

Figure 8 A typical distribution of drop sizes in the early stages of a cumulus cloud. Inset: The same plotted on a logarithmic scale, (from Bartlett, 1966).

Figure 9 Development of distribution shown in Fig. 8 with time (from Bartlett, 1966).

Figure 10 Fraction of total liquid water content in drops with radius greater than 40 microns (from Bartlett, 1966).

Figure 11 Assumed initial drop size distributions (from Bartlett, 1966).

Figure 12 Table of characteristics of droplet spectra shown in Fig. 11 (from Bartlett, 1966).

Figure 13 Overflow as function of time for different initial distributions (from Bartlett, 1966).
Lecture 2, Figure 1

γ

20 μ

TIME

\frac{1}{2} \text{hr}
Conventional borderline between cloud drops and raindrops

Typical condensation nucleus
- $r = 0.1$
- $n = 10^6$
- $V = 0.0001$

Typical cloud drop
- $r = 10$
- $n = 10^6$
- $V = 1$

Typical raindrop
- $r = 1000$
- $n = 1$
- $V = 650$

Key:
- $r =$ radius in microns
- $n =$ number per litre
- $V =$ terminal velocity in cm/sec

Lecture 2, Figure 3
Lecture 2, Figure 4
Lecture 2, Figure 5

\[ \frac{y}{a_i} \]

- \( a_i \) = radius of larger droplet (microns)
- \( a_2 \) = radius of smaller droplet (microns)

\( a_i = 30 \mu \)
\( a_i = 25 \mu \)
\( a_i = 20 \mu \)
\( a_i = 10 \mu \)
Lecture 2, Figure 6

Lecture 2, Figure 7
Lecture 2, Figure 8
Lecture 2, Figure 10
Lecture #2 Dr. Peter V. Hobbs

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Lecture 2, Figure 11

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I TYPICAL OF MARITIME CUMULUS CLOUDS.
II A.B.C.D TYPICAL OF CONTINENTAL CUMULUS CLOUDS,
    CONTAINING DROPS WITH RADIUS UP TO 126,
    24, 22 and 20 μ respectively.
III SAME AS II WITH SMALLER WATER CONTENT.
IV DISTRIBUTION DEFICIENT IN LARGE DROPS.
### Characteristics of Droplet Spectra

<table>
<thead>
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<th>Spectrum</th>
<th>Climatic Type</th>
<th>Liquid-Water Content (g/m²)</th>
<th>Total Droplet Concentration (no./cm²)</th>
<th>Maximum Drop Radius (μm)</th>
<th>Time to Give One 40 μm Drop/Litre (Minutes)</th>
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<td>Maritime</td>
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Lecture 2, Figure 12
Overflow as a function of time for different initial distributions.

Lecture 2, Figure 13
III. NUCLEATION OF ICE AND PROPERTIES OF ICE NUCLEI

A. Homogeneous and heterogeneous nucleation

Clouds frequently extend above the 0°C isotherm and may therefore contain ice particles. Ice can form either directly from the vapor phase or by the freezing of water droplets.

It has been found in the laboratory that very pure water drops, less than a few microns in size, will remain in the supercooled (or undercooled) liquid state to a temperature of about -40°C at which point they freeze spontaneously. This is called homogeneous nucleation. In an environment supersaturated with water vapor, it is also possible, in principle, for ice to be deposited directly from the vapor phase. This process will be in competition with condensation of vapor to liquid water.

Homogeneous nucleation of ice does not occur in the atmosphere because there are foreign particles which permit nucleation to occur at temperatures above -40°C. Nucleation of ice due to the presence of foreign particles is called heterogeneous nucleation. Two types of heterogeneous nucleation must be discussed. One is the direct deposition of ice from the vapor phase and the other is the nucleation of freezing in supercooled drops. For the direct deposition of ice from the vapor phase, the presence of foreign particles reduces the required supersaturation below that needed for homogeneous deposition. Similarly supercooled water containing solid particles will generally freeze at temperatures significantly higher than the homogeneous point.

A macroscopic theory of heterogeneous nucleation has been developed by Fletcher (1958). This approach is based on classical nucleation theory and does not consider in detail the microscopic mechanisms involved in nucleation. The theory consists of writing down an expression
for the change in free energy of a system when nucleation occurs on a substrate, then minimizing this expression to find the size of the critical free energy barrier and size of the critical ice embryo. As the ice embryo forms in the droplet the free energy of the system increases until the embryo reaches a critical radius after which the continued growth of the embryo results in a reduction of the free energy. This implies that if the embryo does grow to its critical radius, the droplet will continue to freeze to lower its free energy.

Figure 1 (see end of Lecture #3) shows the effect of the radius of a spherical particle on the temperature at which it can act as a deposition nucleus according to Fletcher's theory. The surface parameter $m$ is the cosine of the contact angle of ice on the particle. The figure shows that the larger particles are more effective as ice nuclei, particularly for radii less than 1000 Å. However, even for radii greater than 1μ the contact angle must be fairly small (less than 30°) if the substance is to be effective.

Figure 2 (see end of Lecture #3) shows the predictions of the theory when the nucleating particle is immersed in supercooled water. It shows the temperature at which freezing occurs in one second on a spherical particle of radius $R$ in water. The size of the particle becomes important only below a radius of about 0.01μ. Again the surface parameter $m$ has a large influence on the nucleating efficiency.

Since the above treatment is based entirely on thermodynamic considerations, it has a number of serious limitations. The main difficulty is that the chemical bonding and the crystallographic factors that need to be taken into account to calculate the interfacial energies (and hence $m$) are not well-known.
B. **Lattice misfit and entropy effects**

The lattice misfit $\delta$ between a nucleating particle and ice is defined as

$$\delta = \left| \frac{a - a_0}{a_0} \right|$$

where $a$ and $a_0$ are the lattice parameters of the nucleating particle and ice respectively. This misfit causes a certain elastic strain in the ice which increases the energy barrier for nucleation thus lowering the temperature needed to achieve a given nucleation rate.

The value of the surface parameter $m$ also depends upon the way in which the water molecules in ice are bound to the nucleating surface. Since water is polar, this bonding is mainly electrostatic, therefore, ice nucleation should be preferred on materials with intense ionic fields. The energy of the interface will be minimized if the dipole of each water molecule orients parallel to the direction of the electric field in its vicinity. But, this configuration of dipoles may produce a lower entropy and therefore higher free energy in the bulk of the ice well away from the interface. Fletcher (1959b) analyzed the manner in which the relations between interfacial energy and bulk entropy influence the nucleating ability of a material. His conclusion was that any substance which orients the ice dipoles at its surface parallel to one another would be a poor substance for nucleation.

C. **Freezing versus deposition**

Heterogeneous nucleation of ice by direct deposition and by freezing of droplets represent two competing processes. In practice, it may be impossible to distinguish between these two cases since
freezing may not involve a macroscopic droplet but merely a thin layer of water. However, the two processes are quite distinct theoretically. Figure 3 (see end of Lecture #3) demonstrates the situation for a typical nucleating material. The figure shows that for a completely insoluble particle in an environment just saturated with water vapor, condensation cannot occur. Sufficiently large particles (greater than 1\(\mu\)) should act as deposition nuclei at slightly less than water saturation if the temperature is low enough. Figure 3 also shows that in the size range 0.01 to 0.1\(\mu\), particles wetted by water require considerable supersaturation to nucleate drops, but they might act as deposition nuclei at moderate supercoolings and saturation ratios near unity. Condensation should be more rapid than deposition at very small supercoolings and slight supersaturations with respect to water.

D. Experimental studies of heterogeneous nucleation

Studies of heterogeneous nucleation date back to 1724 when Fahrenheit slipped on stairs while carrying a flask of cold water and noticed that the water had become full of flakes of ice. Since then, there have been numerous and often conflicting reports on the means by which ice can be nucleated.

(1) Freezing of water drops

Fig. 4 Freezing temperature of water drops as a function of size (see end of Lecture #3).

From Fig. 4 it can be seen that an order of magnitude increase in the rate of cooling lowers the mean freezing temperature by about 2°C. (Biggs' results for drops >30\(\mu\).) Displacements of the various lines in Fig. 4 is attributed to differing concentrations of ice nuclei in the samples of water used.
(2) Nucleation by particulates

In the belief that good ice-nucleating materials should have crystal properties similar to ice, Vonnegut (1947) searched through X-ray crystallographic data for materials which had cell dimensions and crystal symmetries as close as possible to those of ice. He found that AgI and PbI₂ had small lattice misfits (Fig. 5).

Fig. 5 Crystal structures of AgI, PbI₂ and ice (see end of Lecture #3). AgI and PbI₂ were introduced as powders into supercooled clouds of water droplets at -20°C. In the first trials, AgI had no effect but PbI₂ formed ice crystals. Later, Vonnegut attributed failure of AgI to act as ice nucleus to gross contamination by soluble salts. Cleaner samples of powdered AgI did form ice crystals. Vonnegut also found that electric discharges between Ag electrodes in the presence of iodine vapors produced many thousands of times more ice nuclei than AgI powders. Even more effective were the smokes produced by vaporizing AgI on a hot filament or by dispersing it in a flame. AgI smokes produced by burning an acetone solution of AgI and NH₄I in a hydrogen flame showed that at -20°C over 10⁻¹⁶ ice nuclei were formed per gram of AgI.

Following Vonnegut’s pioneering work, the ice-nucleating ability of a large number of materials was investigated. For example, Fukuta (1958) found 78 inorganic materials to be effective at temperatures above -20°C.

Fig. 6 Some effective artificial ice nuclei (see end of Lecture #3). Because of the widespread use of AgI in cloud seeding, this material has been studied in detail. Reynolds et al. (1951) found that when AgI particles in a container were exposed to strong sunlight, the concentration of active nuclei at -20°C decreased by a factor of about 100 per hour. This decrease
in efficiency is attributed to changes in the surface structure by photodecomposition. Corrin et al. (1967) found that "pure" AgI is less efficient than AgI containing hygroscopic impurities.

Fletcher's theory predicts the conditions under which a particle should act as a freezing nucleus or a deposition nucleus. Edwards and Evans (1960, 1968) determined ice-nucleating activity of a reproducible AgI under controlled temperature and humidity. They found:

(i) Almost all AgI particles of radius 75 Å when exposed to water saturation are inactive as deposition nuclei down to at least -18.5°C. (According to Fletcher's theory, particles of this size should, at water saturation, act as deposition nuclei only below -30°C.)

(ii) AgI particles are much more active as freezing nuclei than as deposition nuclei.

(iii) AgI particles only act as freezing nuclei if the relative humidity with respect to water exceeds 110%. (Since the relative humidity in clouds seldom exceeds 101%, AgI particles should act only as relatively inefficient deposition nuclei unless they collide with cloud droplets.)

Is a particle more efficient as a freezing nucleus when it comes into contact with the surface of a supercooled drop or when it is imbedded in the drop? Gokhale (1965) concluded that nucleation by contact is much more effective than nucleation by particles imbedded in the drop. The difference in the nucleating temperature for these two cases may be 5° to 10°C, depending on the material.

E. Organic nuclei

Recent studies have revealed a large number of organic compounds which can act as ice nuclei at fairly high temperatures. The first organic
ice nucleus to be discovered was phloroglucinol (Bashkirova and Krasikov, 1957), effective at -8°C. In a study of more than 300 organics Fukuta (1963) found metaldehyde to be the most effective; this material nucleated ice at -1°C.

Mention should be made of the unusual ice-nucleating properties of urea. Due to the high endothermic heat of solution (-60.5 calories/gm) and high solubility, this organic produces strong local cooling when it dissolves in water and thus can cause ice nucleation when the air temperature is as high as +6°C.

F. Nucleation of high pressure forms of ice

By using certain organic compounds as nucleators, Evans (1967) has crystallized most of the high pressure forms of ice from the liquid at pressures outside the range of thermodynamic stability. He also used the high pressure forms of ice to investigate the role of lattice misfit in ice nucleation.

Fig. 7 Portion of phase diagram for water substance (see end of Lecture #3).

Aqueous suspensions of AgI were frozen under pressures up to 3000 bars, which includes ice I and ice III phases. Now up to a pressure of 3000 bars, AgI has a hexagonal structure with a mismatch of not more than 2% with ice I. Ice III however has a tetragonal lattice in which none of the spacings correspond to the AgI lattice. For doubly-distilled water (without AgI suspension), ice I formed up to a pressure of 2060 bars while ice III formed above this pressure. Both phases nucleated at about 20°C below their respective melting curves. However, when an aqueous suspension of AgI was cooled, ice I nucleated at a supercooling of 4°C, (i.e. along curve EF) but nucleating temperature of ice III was the same as that for doubly distilled water. Drops containing AgI cooled at a pressure of 2500 bars nucleated at point X, on extrapolation of EF, as
ice I! These observations show that in cases where the nature of chemical bonding is the same, the phase which has the closest lattice fit with the nucleator is preferentially nucleated.

G. **Summary of requirements of an ice nucleus**

We have seen that the small mismatch between the crystal structure of AgI and ice led Vonnegut to test the ice-nucleating ability of AgI which was found to be very effective in this respect. Subsequently, it was widely held that a small mismatch with ice was the sole criterion for an effective ice nucleus. More recently, however, it has become evident that while lattice spacing does play an important role in determining the effectiveness of a material as an ice nucleus, it certainly is not the only factor involved.

AgI is essentially hydrophobic in nature and the surfaces of AgI and ice are probably energetically incompatible. Zettlemoyer et al. (1961) postulated that ice first develops on AgI at a few impurity sites which are hydrophilic in character. This hypothesis is supported by Corrin's work on impure and pure AgI.

To check this idea, Zettlemoyer et al. hydrophobed several types of silicas and determined their nucleating ability. They found that hydrophobing a silica surface significantly increased its ice-nucleating efficiency and some of these materials rivaled AgI. Studies of the absorption properties of these silicas showed that water was absorbed in clusters at high energy sites. A balance between hydrophilic and hydrophobic surface groups and proper distribution of sites were shown to be important for heterogeneous nucleation of ice. The relative inefficiency of pure AgI can be attributed to lack of impurity sites on which ice can form.
While the general requirements for an ice nucleus outlined above appear to be valid, it should not be assumed that the mechanism of heterogeneous nucleation is completely understood. Indeed, different materials probably nucleate ice in different ways.

Notes taken by:
R. M. McGehee
J. R. Miller

REFERENCES


Figure Captions

Figure 1 - Temperature $T$ at which a spherical particle of radius $r$ and surface parameter $m$ will nucleate an ice-crystal in 1 sec by sublimation from an environment at water saturation. The effect of elastic strain is not included (Fletcher, 1958).

Figure 2 - Temperature $T$ at which a spherical particle of radius $r$ and surface parameter $m$ will nucleate an ice-crystal from water in one second by freezing. The effect of elastic strain is not included (Fletcher, 1958).

Figure 3 - Activity surfaces for a typical nucleating substance (Fletcher, 1959a).

Figure 4 - Freezing temperature of water drops as a function of size.

Figure 5 - Models of the crystal structures of ice, lead iodide and silver iodide. In the latter white spheres are iodide ions and black spheres metallic ions (Fletcher, 1959b).

Figure 6 - Some artificial ice nuclei.

Figure 7 - Portion of phase diagram for water substance (Evans, 1967).
Lecture #3

Dr. Peter V. Hobbs

Lecture 3, Figure 1
Lecture 3, Figure 3

Temperatures and particle radii for condensation and sublimation processes.
(a).—Bigg's relationship between the median freezing temperatures and the diameters of water drops containing foreign nuclei (heterogeneous nucleation).

(b).—Median freezing temperatures for groups of droplets of very pure water having diameters $<500\mu$ and the lowest freezing temperatures recorded for drops of $d>500\mu$. 

Lecture 3, Figure 4
Lecture #3

Dr. Peter V. Hobbs

Ice

Lead iodide

Silver iodide

Lecture 3, Figure 5
<table>
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<tr>
<th>Substance</th>
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</thead>
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<td>Mercuric iodide</td>
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<td>- 8</td>
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<td>Silver sulphide</td>
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<td>- 8</td>
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<td>Ammonium fluoride</td>
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<td>- 9</td>
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<tr>
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FULL LINES DENOTE PHASE BOUNDARIES BETWEEN ICE I, ICE III, AND THE LIQUID AS A FUNCTION OF TEMPERATURE AND PRESSURE. DASHED LINES DENOTE NUCLEATION LEVELS. AB, ICE I IN PURE WATER; CD, ICE III IN PURE WATER; EFG, ICE I IN SILVER IODIDE SUSPENSIONS.
IV. PROPERTIES AND DISTRIBUTION OF ICE NUCLEI IN THE ATMOSPHERE

A. Experimental techniques for investigating ice nuclei in the atmosphere

The accurate determination of the numbers and types of ice nuclei that will be activated in a natural cloud is inherently difficult for a variety of reasons. First the conditions under which ice nucleation can occur in the atmosphere are highly variable. The potential nuclei are exposed for various periods of time to different conditions of supersaturation and undercooling, depending on the particular synoptic, mesoscale and cloud conditions. Since nucleation is dependent to some extent on the past history of the particle, it is evident that nuclei which may not be activated in one situation may well be in another. Therefore, in order to use data obtained from measurements one should account for differences between the conditions to which the nuclei were exposed and the situation in which the data are to be applied. For example, the threshold temperature for activation by vapor deposition may differ considerably from that for ice nucleation from the liquid phase by a given nucleating agent.

Ice nuclei exist in the air both as free particles and embedded in cloud and precipitation particles. In the latter case, the particles may be collected and examined for ice nuclei. If the snow crystal is found to have a particle near its center this is usually assumed to be the ice nucleus on which the particle nucleated. The size and nature of the ice nucleus may be investigated by electron microscopy and micro-chemical analysis. The threshold temperature of the most efficient freezing nucleus in a snow crystal may be determined by melting the crystal, cooling it, and noting the temperature at which it nucleates.

Ice nuclei in suspension in the air have been studied using the following techniques:
(1) Isolating a quantity of air, cooling it below its dewpoint, and noting the freezing temperature of the condensation products.

(2) Allowing particles in the air in a cold box to settle onto a supercooled soap film (or sugar solution). The particles which act as freezing or contact nuclei at the temperature of the solution proceed to grow to visible ice crystals which can be counted.

(3) An air sample is introduced into an expansion chamber which is slowly pressurized and then suddenly expanded. The consequent cooling to temperature $T$ activates those ice nuclei effective at temperature $T$ or less. The ice particles which form in the chambers may be counted visually in a collimated light beam or allowed to settle out on a supercooled film.

(4) Using mixing chambers which are similar to the cold box technique with the major difference being found in the method of sampling the environmental air. The cold box uses a passive sampling system, in the sense that the box is open to the environment. The mixing chamber is a closed system into which the sample is drawn in, humidified and allowed to equilibrate. Nucleation detection is the same as those mentioned above.

(5) A known volume of air may be drawn through a millipore filter. The filter may subsequently be exposed to a known supersaturation and cooled to a given temperature and the number of ice crystals which form on the filter counted.

There are problems associated with each method of detection. For example, in the cold box, it is not known what proportion of the nucleating events occur as contact nucleation at the surface of the solution, and what proportion occur as vapor deposition nucleation in the air above the solution. The expansion chamber exposes the particles to the threshold temperature for only a short period; it is not known how long a nucleus should be exposed
before concluding that activation will not occur. The filter technique introduces the unknown influence of the filter base on the nucleation threshold.

B. Distribution and sources of ice nuclei in the atmosphere

The concentrations of natural ice nuclei show great variability in time and space. The nuclei count at a given temperature may vary by an order of magnitude during a period of one hour. Over longer periods of about a day the fluctuations in concentrations may be several orders of magnitude. During so-called ice nucleus "storms", ice nucleus concentrations an order of magnitude greater than normal may persist for many days.

On the average the local nucleus concentration can usually be fitted by a curve of the form \( \ln N = a(T_1 - T) \) where \( N \) is the number of ice nuclei per liter of air activated at temperature \( T \) and "\( a \)" is a constant which varies between 0.3 and 0.8. \( T_1 \) is the temperature at which the concentration of ice nuclei is one per liter. Although \( T_1 \) varies with conditions, \(-20^\circ C\) is a representative figure. Since there is normally a total of about \( 10^8 \) particles per liter in the air, only about one in \( 10^8 \) particles acts as an ice nucleus at \(-20^\circ C\). If "\( a \)" is given the value 0.6, the concentration of ice nuclei increases by an order of magnitude for each \( 4^\circ C \) decrease in temperature.

Measurements of the concentrations of ice nuclei in the atmosphere in the vertical have given rather inconclusive results. Some show an increase with height, while others indicate no particular change with height. Results from measurements made in Australia indicate that the stratosphere acts as a source of ice nuclei, which observation, together with apparent correlations between periods of rainfall peaks and preceding meteor showers, led Bowen (1953) to suggest that meteoritic material might be an important source of ice nuclei in the earth's atmosphere.
Figure 1 (see end of Lecture #4) shows some measured ice nuclei concentrations against temperature for the northern and southern hemispheres using several different measuring techniques. There appears to be a real difference in the average concentration of nuclei in the two higher concentrations being observed in the northern hemisphere. The reason for this difference is probably the greater area of land in the northern hemisphere which is the most obvious source for ice nuclei in the atmosphere. The ice nucleating ability of various minerals have been tested in the laboratory. Some of the results of these measurements are shown in Figure 2 (see end of Lecture #4). It can be seen from these results that some silica and clay minerals are quite effective as ice nuclei and may be the primary sources of natural ice nuclei in the air. This conclusion receives support from the few observations which have been made on the composition of the central nuclei in snow crystals. In crystals collected in Michigan, Kumai (1961) found that 87% had clay mineral particles as the central nucleus. In Greenland 85% of the snow crystals had a clay particle as the central nucleus and one-half of these were kaolin. The diameters of the central nuclei ranged from 0.3 to 8 microns with a mean of 3 microns.

Figure 3 (see end of Lecture #4) shows the influence of wind direction on the concentration of ice nuclei at a site on the Pacific coast of Washington. The results support the hypothesis that the land is the major source of ice nuclei.

Some industries have been observed to emit ice nuclei into the atmosphere. Figure 4 (see end of Lecture #4) shows a wind rose of ice nucleus concentrations for Seattle, Washington, and two locations remote from urban industrial areas. The concentrations of ice nuclei in the city were much greater than those in the non-urban areas and reached maximum values when the wind was from the city center (SW of sampling site).
C. Effects of synoptic conditions on ice nuclei

The concentrations of ice nuclei in the air can vary rapidly with changing synoptic conditions. Figure 5 (see end of Lecture #4) shows an increase in ice nucleus concentrations by a factor of 50 during a rain shower. Figure 6 (see end of Lecture #4) shows increase in ice nucleus concentrations during the evaporation of fogs. The increase in ice nucleus count in a rain shower might be due to the release of ice nuclei by evaporating drops. In order for fogs to increase the concentrations of ice nuclei in the air during evaporation they would have to act as a sink for ice nuclei during formation.

As previously mentioned, increases in the concentrations of ice nuclei which exist for several days are termed ice nucleus "storms". An example of ice nucleus storms in Alaska and Washington are shown in Fig. 7 (see end of Lecture #4). Analysis of air trajectories coincident with this event showed that the particles were carried along in the air mass from Alaska to Washington. Significantly, the Japanese investigation of materials in these storms showed increases of nuclei of earth materials (Isono, et al., 1970). One such storm was traced back to a dust storm in the Mongolian desert. It is hypothesized the particles were carried on strong winds from Asia across the Pacific Ocean to the west coast of the United States.

D. Comparison of concentrations of ice nuclei and ice particles in natural clouds

Following Bergeron's suggestion on the role of ice crystals in producing precipitation in clouds, it was generally accepted that each ice particle in a cloud originates on an ice nucleus. However, recently several field observations have indicated that this is not always the case. Figure 8 (see end of Lecture #4) shows measurements of the concentrations of ice
particles and ice nuclei in natural clouds. It can be seen that there are many more ice crystals than ice nuclei for cloud top temperatures higher than about -20°C. However, as the temperature of cloud top decreases, the ratio of ice crystals to ice nuclei approaches unity. In regions of clouds where this ratio is greater than unity ice multiplication mechanisms may be occurring. One possible mechanism by which the number of ice particles in a cloud may increase without the action of ice nuclei is by the explosion or fragmentation of droplets during freezing. Such fragmentation produce a number of tiny ice splinters, each of which is capable of nucleating freezing in another droplet which can subsequently shatter, and so on. Recent experiments (Hobbs and Alkezweeny, 1968) suggest that a small fraction of cloud droplets might fragment during freezing. Other mechanisms of propagating ice in clouds are the mechanical breaking-up of ice crystals due to collisions and thermal shock when supercooled drops collide with and are nucleated by delicate ice crystals. It has also been suggested that cloud drops might shatter or fragment during freezing after colliding with ice particles in clouds. However, this seems unlikely since in this case freezing occurs from the ice substrate outwards and the stresses necessary for explosion and fragmentation are unlikely to build up.
REFERENCES


Figure Captions

Figure 1 - World-wide measurements of ice nuclei made during 1956-1960.
Figure 2 - Threshold temperatures for ice nucleation of some earth minerals.
Figure 3 - Wind rose of ice nucleus concentrations measured at a site on the Pacific Coast of Washington (from Hobbs and Locatelli, 1970).
Figure 4 - Wind rose of ice nucleus concentrations measured in a city (Seattle) and two non-urban sites (from Hobbs and Locattelli, 1970).
Figure 5 - Variations in ice nucleus concentrations with weather conditions (from Hobbs and Locattelli, 1970).
Figure 6 - Variations in ice nucleus concentrations during the evaporations of fogs (from Hobbs and Locattelli, 1970).
Figure 7 - Ice nucleus storms in Alaska and Washington (from Hobbs et al., 1970).
Figure 8 - Measurements of the concentrations of ice particles and ice nuclei in natural clouds (from Hobbs, 1969).
AVERAGE ICE NUCLEUS CONCENTRATIONS IN THE NORTHERN AND SOUTHERN HEMISPHERES (FROM MOSSOP, 1963).  
A SOUTHERN HEMISPHERE, EXPANSION CHAMBER, BIGG (1960).  
B SOUTHERN HEMISPHERE, MIXING CHAMBER, BIGG (1960).  
C NORTHERN HEMISPHERE, EXPANSION CHAMBER, BIGG (1960).  
D NORTHERN HEMISPHERE, MIXING CHAMBER, BIGG (1960).  

Lecture 4, Figure 1
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Lecture 4, Figure 2
Quillayute (Pacific Coast)

Stampede Pass (Cascade Mountains)

University of Washington (Seattle)
Lecture 4, Figure 5

Wind Direction
Light Showers
Air Temperature
Dew Point
Wind Direction
Calm
- S
- E
- N
- W
- S

Concentration of ice nuclei per 300 liters
of air active at -21°C

29 Oct. 1200 2400
30 Oct. 0000 1200 2400
Lecture 4, Figure 6

Concentration of ice nuclei per 300 liters of air at -21°C

Wind direction

Dew point

Air temperature

Visibility, miles

November 23, 1968

October 14, 1968
Lecture #4, Figure 8

- Concentrations of ice particles
- Concentration of ice nuclei effective at estimated temperature of cloud top (cm$^{-3}$)
- Ratio of concentration of ice particles to concentration of ice nuclei

Ratio of ice particles to ice nuclei

Ice particles

Ice nuclei

Estimated temperature of cloud top (°C)
V. GROWTH OF ICE CRYSTALS

A. Role of ice crystals in clouds

One of the most important aspects of cloud physics that needs detailed study is the physical processes that produce precipitation. We now consider the suggestion first put forward by Franklin (1789), and developed by Wegener (1911) and Bergeron (1935), that most of the precipitation reaching the ground originates as ice crystals in supercooled clouds. This process is probably dominant in the formation of precipitation in temperate latitudes and in the interior of continents where the growth of precipitable particles by coalescence may be limited (see Lecture 2).

To understand the Wegener-Bergeron mechanism, it is necessary to study in detail the germination and the growth of ice crystals in clouds. In the previous lecture we discussed the origin of the ice crystals; in this lecture we will consider the mechanisms by which they may increase in size.

Ice crystals can grow by the direct condensation of water from the vapor phase, by aggregation, and by the accretion of water droplets. Each of these processes is considered in turn below.

B. Growth of ice crystals by condensation from the vapor phase

We may assume that ice crystals in subfreezing clouds are initially formed by condensation and freezing (or by deposition) on to active ice nuclei, or as somewhat larger crystals by the freezing of cloud droplets. At temperatures just below 0°C the concentrations of ice crystals in clouds is comparatively low; also the difference between the saturation vapor pressure over ice and liquid water is small so that the growth rate of an ice crystal at these temperatures is little different from that of a cloud droplet. However, as the temperature is lowered the number of ice particles in a cloud
will increase and they will grow more rapidly than the cloud droplets since the saturation vapor pressure over ice is less than that over liquid water at the same temperature. For example, at -10°C air that is saturated with respect to liquid water is supersaturated relative to ice by 10% and at -20°C by 21%.

The rate of growth of an ice crystal by deposition from the vapor phase is given by \( \frac{dM}{dt} = 4\pi CGS \) where, \( M \) is the mass of the ice crystal, \( S \) the supersaturation of the environment with respect to ice, \( C \) the electrostatic capacity of the crystal, and \( G \) is a temperature dependent function. The rates of growth of ice crystals by deposition in an environment at water saturation calculated from this equation are shown in Fig. 1 (see end of Lecture #5).

The curves in Fig. 1 show that by growing by deposition alone an ice crystal may reach a mass of a few tens of micrograms at -5°C (or, assuming the crystal to be a dendritic plate, it will grow to a diameter of about 2mm) in about 30 minutes. Such a crystal has a terminal velocity of about 30 cm/sec and would be unable to fall toward the ground in the vigorous updrafts which exist in active cumulus clouds. In less active clouds such a crystal might reach ground level as precipitation. Therefore, growth of ice crystals by condensation alone is insufficient to explain precipitation in active cumulus clouds.

C. Growth by aggregation of ice crystals

Rough calculations show that a plate having an initial diameter of 1mm can grow by collecting other crystals (in concentration of 1 gm/m³) to a diameter of 1 cm in about 20 minutes. Such a crystal has a fall velocity of about 150 cm/sec and can therefore fall out in moderate updrafts. Hence, with updrafts of about 100 cm/sec, cloud depths of about 1500 m, and fairly
high concentrations of ice particles, particles can grow to precipitable size in about 40 minutes by a combination of deposition and aggregation.

The mechanism by which ice particles adhere together is of interest. It is observed that when two ice particles are kept in contact for a few minutes, they stick together and some force is required to separate them. The force that is required to separate two ice spheres at ice saturation is shown as a function of temperature in Fig. 2 (see end of Lecture #5). It is clear from the figure that the force of adhesion decreases with decreasing temperature. The explanation for this adhesion of ice particles and its variation with temperature has been given by Hobbs and Mason (1964).

If two spheres are kept in contact for a few minutes a solid neck of ice forms between them due to sintering. This neck grows with time as shown in Figs. 3 and 4 (see end of Lecture #5) and it welds the particles into contact.

D. Accretion of droplets

In a supercooled cloud we must also consider the growth of ice particles due to the accretion of supercooled droplets. Rough calculations indicate that the collision efficiency of an ice crystal 1mm in diameter for droplets greater than 10μ diameter is reasonably high. With a collection efficiency of about 0.5, it can be shown that an ice crystal can grow to a spherical shape of about 1mm in radius in about 30 minutes. This will have a terminal velocity of about 100 cm/sec, and will be able to fall through moderate updrafts in clouds to reach the ground as precipitation.

E. Ice crystal habits

Observations both in the field and the laboratory have shown that ice crystals may grow in a variety of habits, the basic shapes of which are determined mainly by temperature (Nakaya, 1951). This fact is demonstrated
by growing ice crystals in a diffusion chamber. The crystals are grown on a thin fiber which hangs vertically in a chamber in which the gradients of temperature and supersaturation are accurately controlled. (see Fig.5 at the end of Lecture #5).

The crystal habit varies along the length of the fiber in the following manner (Hallett and Mason, 1958):

- 0 to -3°C Thin hexagonal plates
- -3 to -5°C Needles
- -5 to -8°C Hollow prismatic columns
- -8 to -12°C Hexagonal plates
- -12 to -16°C Dendritic, fern-like crystals
- -16 to -25°C Hexagonal plates
- -25 to -50°C Hollow prisms

Some photographs of different ice crystals in the diffusion cloud chamber are shown in Figs. 6, 7 and 8. (see end of Lecture #5).

The effect of suddenly changing the temperature and supersaturation on the growth habit of a particular crystal may be observed simply by raising or lowering the fiber in the chamber. When a crystal is transferred to a new environment, the continued growth assumes a new habit characteristic of the temperature of the environment. For example, Fig. 7 shows a case where needles were grown at -5°C and were then lowered in the chamber to where the temperature was about -14°C, stars then grew on the ends of the needles.

If a crystal grows under thermodynamic equilibrium the shape of the crystal can be predicted from Wulff's theorem which states that in equilibrium the distance of any crystal face from the center of the crystal is proportional to the surface energy of the face. Exact values of the surface energies for different faces of ice are not known. However, approximate
values can be calculated by assuming that the molecules in ice interact only with their nearest neighbors. Using these values it may be shown from Wulff's theorem that ice crystals should grow in the form of rather thick hexagonal plates in which the ratio \( \ell/r \) of the thickness of the plate to the radius of the circumscribed circle about the hexagonal end is 0.82. Some ice crystals are in the shape of hexagonal prisms but at different temperatures \( \ell/r \) takes on values from less than 0.1 to greater than 10. We conclude, therefore, that ice crystals do not grow under equilibrium conditions but are probably controlled by surface kinetic effects. Considerable insight into the mechanism by which ice crystals grow has been obtained from studies of the epitaxial growth of ice crystals using the apparatus shown in Fig. 9 (see end of Lecture #5).

Hallett (1961) found that when ice crystals grew epitaxially on the basal plane of cupric sulfide (CuS) the crystals thicken in a direction normal to the basal plane by steps of ice (a few 100 Å in height) sweeping across the basal plane. The velocity of these steps varied with temperature in the manner shown in Fig. 10 (see end of Lecture #5). It is believed that the relative growth velocities of such steps on the basal and on the prism faces determine the habit of ice crystals growing from the vapor phase. Thus, if at a particular temperature the velocity on the basal plane were greater than that on the prism face prisms would form, and if the reverse were true plates would form. The velocities of steps on the prism face of ice have not been measured. However, if we postulate that this velocity varies with temperature in the manner indicated by the dotted line in Fig. 9, then the basic changes in ice crystal habit with temperature would be explained. For example, between about -3 and -8°C the velocity of steps on the basal plane would exceed that on the prism face and the ice crystals would grow prism-like
as observed experimentally. However, this theory does not explain the more complicated habits such as dendrites.

Notes taken by:
J. Presley
M. R. Rao

REFERENCES

FIGURE CAPTIONS

Figure 1 - Growth of ice crystals by deposition in a cloud at water saturation (A) plane dendritic crystal at -15°C (B) hexagonal plate at -5°C (from Houghton 1950).

Figure 2 - Force required to separate ice spheres at ice saturation against temperature (from Jensen, 1956).

Figure 3 - Two ice spheres placed in contact at -6°C.

Figure 4 - Same two ice spheres as in Fig. 3 sometime later.

Figure 5 - Diffusion cloud chamber.

Figure 6 - Ice crystals of differing shapes growing on a filament suspended in a diffusion chamber with controlled temperature gradient. The crystals take characteristic forms at different temperatures as indicated along the right edge of the photograph. Reading from the top, the symbols represent: thin hexagonal plates, needles, hollow prismatic columns, hexagonal plates, branched fern-like crystals (or dendrites), and hexagonal plates. At temperatures below -25°C, prisms appear again (from Mason, 1962).

Figure 7 - Hollow prismatic columns (from Mason, 1962).

Figure 8 - Crystal hybrids showing how the form is dictated by temperature. Needles grown at -5°C developed stars on the ends when shifted to a temperature of -14°C (from Mason, 1962).

Figure 9 - Apparatus for studying epitaxial growth of ice crystals (from Shaw and Mason, 1955).

Figure 10 - Velocity of steps on ice as a function of temperature. Basal plane (measured); -- -- -- -- Prism face (postulated).
Growth of ice-crystals by sublimation in a cloud at water saturation. A: plane dendritic crystal at -15 C; B: hexagonal plate at -5 C. Figures entered at intervals along curves give fall distances in kilometres (after Houghton, 1950).

Lecture 5, Figure 1
Force required to separate ice spheres at ice saturation against temperature (after Jensen, 1956)

Lecture 5, Figure 2
Lecture 5, Figure 5  The diffusion cloud chamber
Lecture 5, Figure 6
Lecture 5, Figure 8
Velocity of steps on ice as a function of temperature. ——— Basal plane (measured); ———— Prism face (postulated).
VI. CLOUD ELECTRIFICATION

In this lecture an outline is given of several theoretical models which have been proposed to explain the generation and separation of electrical charges in clouds and thunderstorms and some experimental evidence for and against these models is presented.

A. Requirements of a satisfactory theory of thunderstorm charge generation

Mason (1953) has listed the conditions which a satisfactory theory of thunderstorm electrification must meet. They are:

1. The average duration of precipitation and lightning from a single-cell thunderstorm is about 1/2 hour.

2. The average electric moment destroyed in a lightning flash is about 110 coulomb km. Corresponding charge is 20 to 30 coulombs.

3. The magnitude of the charge being separated immediately after a flash, by virtue of the fall speed \( v \) of the precipitation elements, is of the order of \( \frac{8000}{v} \) coulombs, where \( v \) is in m/sec.

4. In a large extensive thundercloud this charge is generated and separated in a volume bounded by the -5 and -40°C levels and has a radius of about 2 km.

5. The negative charge is centered near the -5°C level while the main positive charge is situated some kilometers higher up, near the -20°C level.

6. Charge generation and separation processes are closely associated with the development of precipitation in the ice phase (probably soft hail). These precipitation particles must be capable of falling through upcurrents of several meters per second.

7. Sufficient charge must be generated and separated to supply the first lightning flash within 12 to 20 mins. after the appearance of precipitation particles of radar-detectable size.
8. Rate of generation of charge is about 1 C km$^{-3}$ min$^{-1}$ (1000 C in 50 km$^3$ in 20 min).

Some workers might question these requirements on two grounds, namely, that lightning sometimes occurs in warm clouds and that convective activity and not precipitation may be the prerequisite for charging.

An adequate theory of thunderstorm electrification based on the hypothesis that precipitation is responsible for generation and separation of the charge must explain how the heavier precipitation elements acquire a negative charge and the smaller cloud particles (which are assumed to be carried along by the updrafts) acquire positive charges.

B. Outline of a simple theory for charge separation by interaction of cloud and precipitation particles.

Suppose that the electrification is produced by collision between cloud particles (ice crystals or water droplets) present in number density $n$ and hailstones of radius $R$ present in number density $n_h$. The number of collisions per cc per second between hailstones and cloud particles is given by

$$\frac{dN}{dt} = E \pi R^2 n_h n V$$

where $V$ is the full speed of the hailstone and the $E$ the collision efficiency. The precipitation rate $p$ due to the hailstones is

$$p = \frac{4}{3} \pi R^3 n_h V$$

when $\rho$ is the density of the hail. If each collision, on the average, results in a charge transfer $q$, the rate of production of charge per unit volume of cloud is

$$\frac{dQ}{dt} = \frac{3}{4} q \frac{E n p}{R \rho}.$$

If we take $dQ/dt = 1$ C km$^{-3}$ min$^{-1}$, $p = 5$ cm/hr, $R = 0.2$ cm, $\rho = 0.5$ gm/cc and $E = 1$. Then for ice crystals $n = n_c = 0.1/\text{cc}$ (diam. $> 80\mu$) yields

$q = q_c = 5 \times 10^{-5}$ esu/collision. For water droplets, $n = n_d = 1/\text{cc}$ (diam. $> 30\mu$) so that $q = q_d = 5 \times 10^{-6}$ esu/collision.
C. Ice crystal - hailstone charge transfer theory of Reynolds, Brook and Gourley (1957).

Laboratory measurements by Reynolds et al. (1957) found in laboratory experiments that the charge separated during the collision of ice crystals with an ice surface was of the order $5 \times 10^{-4}$ esu per collision. If the hailstone was warmer than the crystal, it received a negative charge. In a natural cloud hailstones growing by collecting supercooled water drops might be expected to be warmer than ice crystals which collide with them. Thus the hailstones would acquire negative charges which would be carried to the lower levels of the cloud.

However, rather similar laboratory measurements made by Latham and Mason (1961) yielded a value for $q_c$ of $5 \times 10^{-9}$ esu/collision. This would produce a negligible $dQ/dt$.


Laboratory experiments carried out by Latham and Mason indicated that when supercooled drops collided with an ice surface a charge $q_d = 5 \times 10^{-6}$ esu/collision was separated, the ice surface becoming negatively charged. They explained this charging on the basis of the explosion or fragmentation of supercooled droplets during freezing onto the ice surface. As we have shown above, if this amount of charge is separated when supercooled cloud droplets collide with natural hailstones it would be a powerful enough mechanism to explain the generation of charges in thunderstorms. However, the laboratory observations of Latham and Mason are open to criticism on a number of points:

1. Reynolds et al. observed negligible charging when supercooled droplets alone collided with ice (they observed charging only with mixture of droplets and ice crystals).

2. Field measurements by Hobbs and Burrows (1966) agreed with the laboratory measurements of Reynolds et al.
3. Supercooled drops are unlikely to explode or fragment after colliding with a hailstone (see Lecture #4).

E. Recent observations on charging resulting from ice particle interactions

A detailed study of the charging of ice surfaces due to collisions with natural ice particles has been carried out by a group at the University of Washington during the past few years. A brief summary of the results of these measurements will now be given.

Shown in Fig. 1 (see end of Lecture #6) are four cases of the spectrum of charges transferred to an ice surface due to natural ice crystals colliding with it. The magnitude of the charges received by individual collisions range from \(-10^{-2}\) to \(+10^2\) esu/collision.

Results fall into two types of distributions:

1. **Symmetrical distributions** for positive and negative charges. This is generally observed for graupel and rimed particle. For graupel at warm temperatures, net charge is generally positive or zero. For rimed spatial dendrites the net charge is negative or zero at \(-8^\circ\text{C}\). Figure 2 (see end of Lecture #6) shows examples of symmetrical distribution for rimed spatial dendrites at \(-8^\circ\text{C}\). (In two cases, the median charge is zero and in one case it is \(-5 \times 10^{-4}\) esu/collision).

2. **Asymmetrical distribution** in which charges of one sign dominate. This is observed with most other types of ice crystals. Charges follow a log-normal distribution. Figure 3 (see end of Lecture #6) shows a log-probability plot for the charging events when stellar crystals collided with an ice surface at \(-12^\circ\text{C}\). Net charge may be positive or negative but this appears to be temperature dependent as summarized in Fig. 4 (see end of Lecture #6) which shows the temperature dependence of the net charge acquired by an ice sphere per cm of path length due to collisions with ice particles (values below dotted
line, i.e., between -2 and -8°C, are more than sufficient to give $dQ/dt = 1$ coulomb/km³/min).

The observed charging when ice particles collide is probably due to two basically different mechanisms, (see, for example, Loeb, 1958).

(a) Symmetrical charging which occurs on the basis of pure statistics whenever two surfaces come into contact across a small area in which, by chance, more ions of one sign diffuse across the boundary in one direction than the other. In this case the direction of charge transfer can go both ways.

(b) Asymmetrical charging which leads to the transfer of a series of charges predominately of one sign. This may be due to direct transfer of existing charges on particles, thermoelectric effect, impurities, etc.

F. Effect of a polarizing electric field on charging

In the work described so far, we have neglected the effects of electric fields on the charging of interacting particles. Such fields can polarize the particles and enhance the transfer of charges between colliding particles. It may be shown that the charge transferred by contact and separation between conducting spheres of radii $R$ and $r$, whose line of centers is at an angle $\theta$ to field $F$, is given by (Davis, 1964).

$$q = \left( \gamma_1 F \cos \theta + \gamma_2 \frac{q_R}{R^2} \right) r^2$$

where $q_R$ is the charge carried by the sphere of radius and $\gamma_1$ and $\gamma_2$ are functions of $r/R$. If $R >> r$,

$$q = \left( \frac{\pi^2}{2} F \cos \theta + \frac{\pi^2}{6} \frac{q_R}{R^2} \right) r^2.$$  

Due to this polarizing effect, if the electric field is directed towards the ground, the hailstone will receive a negative charge and the
cloud particle a positive charge. The field builds up as charge separation by this process continues until the downward force acting on the cloud particles due to the electric field gives them a downward velocity comparable to the fall velocity of the hailstones. The probability of collisions between cloud and precipitation particles is then reduced.

Figure 5 (see end of Lecture #6) shows some experimental measurements of the charges transferred to a sphere by the collision of stellar ice crystals as a function of the applied electric field. We see from these results that the charge transfer increases rapidly with increasing fields; even fair weather electric fields of 1 volt/cm give a charge per collision of $10^{-5}$ esu. For fields up to about 50 volt/cm, the results agree well with Davis's theory based on the collisions of conducting spheres in an electric field. For higher fields the experimental results increase more rapidly with the applied field than the theory predicts.

G. Some possible effects of electrical forces on cloud physical processes

1. Collisions and coalescence of droplets.

Recent calculations show that for highly charged droplets interacting in fields approaching the dielectric breakdown of air, the collision efficiencies are drastically affected (generally increased). The effects are most pronounced for smaller drops. Fields below about 1000 V/cm have insignificant effects on collisions. Thus in many clouds, electrical fields will have little effect.

There is evidence that electrical forces can modify coalescence efficiency. Woods (1965) found that the coalescence rate for drops of radius less than 40 μm increases linearly with applied charge above a threshold value of $5 \times 10^{-5}$ esu. If the drops carried charges of the same sign, then coalescence could be totally inhibited.
Moore et al. (1964) observed intense rainfall from highly electrified clouds within a very short time of the detection of the radar echo. The estimated collection efficiencies must have been 4 to 10 times greater than normal values. They have suggested that gushes of rain or hail which sometimes follow lightning are caused by drops capturing ions and then travelling at high velocities in intense local fields, colliding and coalescing with numerous oppositely charged droplets.

2. Collisions and aggregation of ice crystals

Figure 6 (see end of Lecture #6) shows the increase in mass acquired by an ice sphere exposed to a stream of ice particles as function of applied electric field.

3. Nucleation of ice

The role of electrical charges and fields on the action of ice nuclei is unknown. There is some experimental evidence that electric fields can cause nucleation of supercooled droplets. It has been suggested that freezing occurs if electrical forces are of sufficient strength to disrupt the surface of the drop, drawing it out in a liquid filament of thickness around $10^{-6}$ cm. Molecular aggregates of water of these dimensions, possibly orientated by the field, take the form of minute crystallites which could cause the drop to nucleate. Recent experiments by Abbas and Latham (1969) lend support to this idea.

Notes taken by:

M. S. Sher

W. G. Slinn
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Figure Captions

Figure 1: Spectra of charges received by an ice surface exposed to natural ice crystals. (From Scott and Hobbs, 1968).

Figure 2: Example of a symmetrical distribution of charging events for rimed spatial dendrites colliding with ice at -8°C. (From Scott and Hobbs, 1968).

Figure 3: Log-probability plot for charges received by an ice surface exposed to natural stellar crystals at -12°C. (From Scott and Hobbs, 1968).

Figure 4: Temperature dependence of net charge acquired by an ice sphere for cm of path length as it moves through a cloud of natural ice crystals (From Burrows and Hobbs, 1969).

Figure 5: Measurements of the charges transferred to a sphere by the collision of natural ice crystals (From Scott and Levin, 1970).

Figure 6: Increase in the mass of ice acquired by an ice sphere exposed to a stream of ice particles as a function of the applied electric field (From Saunders, 1968).
The charging of an ice sphere exposed to a stream of snowflakes. (Upward deflections correspond to the ice surface receiving a positive charge, and vice versa.)
RUN TG 4
Ice Island T3, Arctic
May 10, 1967
Rimed Spatial Dendrites
Temperature = -8°C

Cumulative Spectrum of Charging Events for Rimed Spatial Dendrites Colliding with a Simulated Hailstone
RUN Y1
Yellowstone Park
January 24, 1967
Stellar Crystals
Temperature = -12°C
700 Charging Events
Average Charge = -1.7 x 10^4 e.s.u.
1.2 events/sec

Cumulative Spectrum of Charging Events for Stellar Crystals Colliding with a Simulated Hailstone.
Lecture 6, Figure 4

Charge acquired by an ice sphere per cm of path length due to collisions with ice particles in the air.

Lecture 6, Figure 4
Lecture 6, Figure 5
% Increase in aggregation at -7°C due to an electric field.

- Volume increase determination
- Mass increase determination
  (a) 20 cm sec$^{-1}$  (b) 800 cm sec$^{-1}$
VII. ARTIFICIAL MODIFICATION OF CLOUDS,_precipitation, hailstorms, and thunderstorms

In this lecture a brief review is given of work which has been carried out in the past twenty years or so on the artificial modification of clouds, precipitation, hailstorms and the electrical properties of clouds.

A. Early history of artificial modification of clouds and precipitation

In 1938 the German cloud physicist Findeisen suggested that the introduction of artificial ice nuclei into supercooled clouds might enhance the formation of rain. In July 1946 Schaefer (1946) accidently discovered that a tiny piece of dry ice when dropped into a cold box filled with a supercooled cloud resulted in the formation of millions of tiny ice crystals. (N.B. Dry ice forms ice crystals by homogeneous nucleation.) On November 13, 1946, Schaefer made the first field tests using 3 pounds of crushed dry ice which was dropped into a layer of supercooled altocumulus cloud. Observers on the ground saw snow fall out of the cloud for about 2000 feet before it evaporated.

In November 1946 Vonnegut (1947) discovered that silver iodide particles acted as ice nuclei at about -5°C. These particles could be produced in large numbers (about $10^{15}$/gm of AgI) by vaporizing an acetone solution of AgI in a hot flame. This suggested that if large quantities of the particles were released from the ground that they might be carried in updraughts to cloud levels. On December 21, 1948, Vonnegut dropped lumps of burning charcoal impregnated with silver iodide from an aircraft into 6 square miles of supercooled stratus about 1000 feet thick. The cloud was converted into ice crystals using less than 1 ounce of AgI!

B. Experimental seeding of cumulus clouds

Kraus and Squires (1947) seeded large supercooled cumulus clouds with dry ice. They observed that in some cases the clouds showed "explosive"
vertical growth following seeding. In one spectacular case a cloud with a base at 11,000 feet and summit at 23,000 feet was seeded with 150 pounds of dry ice. In 13 minutes it grew to an elevation of 29,000 feet and after 21 minutes heavy rain fell while radar echoes within a 100 miles showed no other precipitation activity. The explosive growth can be explained by the liberation of latent heat as the supercooled droplets freeze, but this only occurs under certain environmental conditions. This has led in recent years to fairly detailed studies of the dynamics of cumulus clouds using the latent heat released by seeding to check certain aspects of theoretical models.

C. Artificial seeding to increase rainfall

1. Warm clouds

We have seen in a previous lecture that the presence of fairly large water droplets is necessary in order to initiate the growth of precipitable drops by coalescence, and that these large droplets probably form by condensation onto large cloud condensation nuclei. Hence, the introduction of hygroscopic particles into a cloud might initiate the coalescence mechanism. A few experiments of this type were carried out in the 1950's and appeared to be partially successful.

A recent experiment carried out in India in which clouds were seeded with salt particles has been reported by Biswas, et al. (1967). Ground based salt seeding was carried out in three climatologically similar regions surrounding Delhi, Agra, and Jaipur in northwest India. Control and target areas for each seeding day were defined as the 90° sectors upwind and downwind, respectively, of the central seeding location. Hygroscopic particles were introduced by the spraying of a known concentration of salt solution or by compressed air dusting of a finely powdered salt solution. In spraying, particles having dry masses of $4 \times 10^{-10}$ to $10^{-8}$ grams and diameters of 7 to 25μ were spread with a generation rate of $10^9$/sec. In dusting the particles
were about $10^{-9}$ grams with a radius of 5μ and were dispersed at a rate of $2 \times 10^{10}$/sec.

The experiments were carried out for eight seasons in Delhi, six seasons in Agra, and four in Jaipur. Comparisons were made between target and control areas for seeded and nonseeded days. In 16 out of the total 18 seasons an average increase of rain of 41.9% occurred in the target area.

(2) **Cold clouds**

It takes about a million cloud droplets 10μ in radius to form a raindrop 1000μ in radius. One possible way to achieve this is to introduce into the cloud about 1 ice nucleus per million supercooled droplets. Since there are about 100 cloud droplets per cubic centimeter of air, this means introducing about 1 ice nucleus per 10 liters of air. An average isolated cumulus cloud might contain $10^{12}$ liters of air and therefore about $10^{11}$ ice nuclei are needed to remove all the droplets. To achieve this number only a few grams of AgI are needed as one gram yields about $10^{13}$ nuclei.

In some early Australian experiments about 10 grams of AgI were introduced into isolated supercooled cumulus clouds. These clouds had summit temperatures ranging from -2.5 to -10°C and depths of 4000 to 17,000 feet. The results indicated that 72% of the clouds which were seeded precipitated within 20 to 25 minutes, 21% evaporated, and 7% showed no change.

In large-scale experiments where AgI is dispersed into stratiform or layer cloud systems (warm fronts, occlusions, orographic clouds) by either aircraft or from the ground, statistical evaluations of the results of seeding become mandatory. A number of such well-conducted experiments have been carried out and the results indicate that precipitation may be increased, decreased, or be unaffected by artificial seeding. A summary of the results
obtained in a few projects of this kind is given below.

**Israel Experiment**

This experiment is based on a randomized cross-over design with a north and a center target area separated by a buffer zone. Seeding is done from aircraft just below the cloud base and upwind of the target area at a rate of 800 to 900 grams of AgI per hour resulting in about $10^{12}$ nuclei at $-10^\circ$C. The clouds are assumed to be convective and seeding is started only when the cloud tops have reached the $-5^\circ$C level. Six seasons of data have been evaluated since 1967. The results indicate an average increase of 15.2% over both target areas with individual yearly increments ranging from 1.2 to 65%. These results are significant at the 5% level (i.e. there is a 5% probability that the results are merely chance). The study of seeding effects versus temperature at the 700 mb level (as a substitute for cloud top temperature) indicate that the largest ratios of rain increase occurred for cloud tops in the $-5^\circ$ to $-6^\circ$C range.

**Project Whitetop**

This experiment, which was rather similar in design to the Israel experiment, was carried out in summertime in Missouri over a period of 6 years. Seeding was carried out from aircraft along 10 mile tracks upwind from the test area during a 6 hour period (1100-1700 CST) at or below the cloud base. The three aircraft involved, each with two acetone burners, dispersed about 2700 grams of AgI per hour into afternoon convective clouds, neither single thunderstorm or stratus clouds were included. This resulted in about $10^{14}$ to $10^{16}$ nuclei per gram at $-17^\circ$C.

Many different statistical evaluations of the results of Project Whitetop have been made by different groups. Rainfall amounts inside and outside the target area were considered on seeded and non-seeded days. It was found that the largest average rainfall occurred in the seeded areas on the **nonseeded**
days (Decker and Schickendanz, 1967). Neyman et al. (1969) extended the analyses up to distances of 180 miles from the seeded area and found that the average precipitation on 102 seeded days was less than on the 96 experimental days without seeding. For distances less than 30 miles there was 32% less rain on seeded days. Estimated average rainfall over the whole region of about 100,000 square miles was 21% less on seeded days.

Climax Project

This project is being carried out in orographic cloud systems in one of the Colorado River watersheds. Silver iodide ground generators are placed at distances of 13, 20, 22, 30, 47, and 59 km from the target area. These produced 20 grams of AgI per hour resulting in about $10^{14}$ nuclei per gram of AgI at $-12^\circ$C. The normal concentrations of freezing nuclei in the target area is 1 per liter and this is increased to between 10 and 100 per liter at $-20^\circ$C during seeding. The criterion used in choosing the experimental days is that Leadville (located upwind from the target area) be forecast to have over 0.01 inches of precipitation within the 24 hour period.

A simple theoretical model has been developed which equates the diffusional growth of ice crystals to the rate of formation of condensate in an orographic cloud. Rimming is not considered to be important. Computations have been made for various cloud temperatures and updraft velocities to determine the concentrations of ice crystals needed in order for all the water vapor released in the updraft to be accumulated as snow. This concentration of crystals is called the optimum concentration.

The results so far have shown that nearly 100% more precipitation has fallen on seeding days when the temperature at the 500 mb level was $-11$ to $-20^\circ$C (significant at the 5% level). However, when the 500 mb temperature was less than or equal to $-26^\circ$C there was a 30% decrease in precipitation in the target area (significant at the 1% level). These results are in
agreement with the predictions of the theoretical model. At lower temperatures there are sufficient natural nuclei to effectively release precipitation, and the artificial nuclei cause overseeding and therefore a reduction in precipitation.

**King's River Watershed**

This project was begun in California in 1955 and is still continuing. Ground based AgI-acetone burners (supplemented recently by pyrotechnic AgI flares) consume 12 grams of AgI per hour. The target area is about 3000 km$^2$. The evaluation is based on the streamflow in the King's river and two nearby watersheds are used as controls. The results over 13 years suggest an average annual increase in runoff from the King's river of approximately 6% as a result of the artificial seeding.

**Lake Almanor**

This project was carried out in the watersheds of Lake Almanor in north-east California to determine the effects of seeding on wintertime Pacific storms. An area of about 500 square miles was divided into control and target areas which varied with the wind direction. Six AgI generators were used at or near mountain tops and the outputs were 27 grams of AgI per hour. Fifty-one raingauges were spaced at 2 mile intervals on the watershed. Operations were carried out from January to May of 1963 and included 76 randomized seeding events. The results for these winter orographic clouds indicate 80% more rain on seeded days in a westerly wind and a 10% decrease in precipitation on seeded days when the winds were from the south.

D. **Targeting of Snowfall by Seeding**

Two projects are currently underway, one carried out by the University of Washington in winter storms over the Cascade Mountains of Washington State and the other by ESSA in the Lake Erie area. The main purpose of these projects is to investigate the feasibility of targeting snowfall by seeding
with ice nuclei. For example, in winter orographic storms over the Cascade
mountains the concentrations of natural ice nuclei are quite low (about 1 per
10 liters at -21°C). The ice crystals which form in these clouds increase in
mass mainly by riming and attain fall velocities of the order of 1 m/sec. If
these clouds are completely glaciated by overseeding with artificial ice nuclei
so that all of the supercooled water droplets are removed, the ice crystals
will grow only by condensation from the vapor phase. Such crystals have fall
velocities of about 0.5 m/sec. Therefore, if these crystals fall out in a
westerly airstream they will be carried further east than will the rimed
crystals which form under natural conditions.

E. Hail Suppression

The principle here is that if the number of ice embryos is increased
the size of the hailstones will be correspondingly reduced, since

\[ \frac{4\pi}{3} r_s^3 N_s = \frac{4\pi}{3} r_I^3 N_I \]

or,

\[ r_s = r_I \left( \frac{N_I}{N_s} \right)^{1/3} \]

where the subscripts \( s \) and \( I \) indicate after-seeding and before-seeding
respectively, \( N \) the number of ice embryos and \( R \) the radius of the hail-
stones. Theoretical work indicates that the water content of a hail cloud
becomes effectively depleted by a small number of hailstones (about 10 per m\(^3\)),
so that even modest increases of their concentration of two orders of magnitude
can be expected to decrease their size sufficiently to prevent damage.

Hail modification experiments are currently being carried out in the USSR.
Seeding agents are delivered by guns and shells or by rockets. Guns have
greater range and altitude and can deliver 100 to 200 grams of AgI or PbI.
Rockets can carry larger quantities (3.2 kg of pyrotechnic material). In 1966
more than one million hectares (3900 square miles) were "protected" by artificial
seeding. Hail damage in the protected areas was 3 to 5 times smaller than in the unprotected areas. The cost of protection amounted to about 2 or 3% of the value of the crops saved. However, these experiments were not randomized therefore statistical evaluation is not possible.

F. Modification of Electrical Behavior of Clouds

Lightning is the greatest single cause of forest fires in the Western United States. The U. S. Forest Service has developed a program on lightning research known as Project Skyfire.

The attempts at modifying lightning have centered around experiments in which silver iodide seeding is employed to produce an abundance of ice crystals in that part of a supercooled thundercloud in which most ground strokes are thought to originate (i.e., between the -10 and -15°C isotherms). It is postulated that the increased number of ice crystals produced by the seeding will provide additional corona points; these should increase the leakage current between the charge centers in the form of a corona current and thereby suppress the formation of a stepped leader.

A two-year pilot experiment of the kind outlined above was carried out during the summers of 1960 and 1961 in Western Montana. Seeding was done from aircraft and ground-based generators, and lightning within a selected area was recorded. It was found that there were 30 percent fewer ground discharges on those days when the clouds were seeded than on those days when seeding was not carried out. Intracloud and total lightning discharges showed decreases of 8 and 21 percent, respectively, for seeded clouds during the two-year period. However, statistical analysis of the data showed that the probability of this distribution occurring by chance was as high as 1 in 4.

A new series of seeding experiments were carried out during the summers of 1965 and 1967 using more efficient silver iodide generators, new instruments for identifying the lightning flashes, and an improved physical
and statistical design. Analysis of data on the basis of the life cycle of individual storms showed 66 percent fewer cloud-to-ground discharges, 50 percent fewer intracloud discharges, and 54 percent less total storm lightning occurred during seeded storms than during unseeded storms.

It should be noted here that the physical explanation of the results obtained in Project Skyfire is complicated by the fact that although artificial seeding might cause an increase in the leakage current in a cloud, and therefore tend to decrease lightning activity, it may also affect the electrical behavior of the cloud in other ways. For example, several workers have found that the collision of ice particles can produce significant generation and separation of electric charge and this could be the mechanism for charge generation in thunderstorms (Reynolds, Brook and Gourley, 1957; Hobbs and Burrows, 1966; Burrows and Hobbs, 1969). If this is the case, the artificial glaciation of a cloud might lead to an increase in electrical activity. Moreover, the vigor of a cloud may be significantly increased as a result of the latent heat released by seeding and this might also increase the electrical activity of a cloud.

Chaff Seeding

If leakage currents in a cloud can be increased by the ice crystals produced by artificial seeding, the dispersal of long chaff needles into a thundercloud should have the same effect. Moreover, in the latter case, the experiment is not complicated by an increase in the concentrations of ice in the cloud.

Experiments of this kind were started by the U. S. Army Atmospheric Sciences Laboratory (Kasemir and Weickmann, 1965) and are now being continued in the APCL of ESSA. Laboratory experiments have shown that the onset of corona discharge on a 10 cm long chaff fiber is about 30 KV/m which is about twenty times lower than the field necessary to initiate lightning discharges.
in clouds. If such chaff were dispersed in a large column of cloud about five pounds of it, containing $10^7$ fibers, would produce a 10 ampere corona current in a field of 70 KV/m. This current would be adequate to counterbalance the current output from an average thunderstorm (approx. 3 ampere) and should therefore suppress lightning.

Carefully controlled field experiments to test this idea have yet to be carried out.

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Notes taken by:
B. Wilhelmson
R. E. Forbes
VIII. NUMERICAL MODELING OF CUMULUS CLOUDS

A. Early work and ideas

Early models of cumulus dynamics neglected the interactions between the rising cloud and the environment. Thus, they generally overpredicted cloud height, liquid water content, vertical velocity, and temperature excess. Stommel (1947) showed how entrainment or mixing could be included if the rising air is represented by a conical steady-state jet, and thereby laid the foundation for the present one-dimensional models of cumulus convection.

If a steady-state jet spread angle of the jet is $\alpha$ (Fig. 1, see end of Lecture #8), the rate of entrainment $\mu$ of air into the jet is given by $\mu = 1/M \cdot dM/dz = 2\alpha/R \approx 0.2/R$. The important point here is the inverse dependence of $\mu$ upon the radius $R$.

Scorer and Ludlam (1953) proposed a different model for cumulus convection called "bubble" model in which the convective element is likened to a hot bubble whose top is continuously being eroded away and entrained into the wake. They found that in this case $\mu = 1/M \cdot dM/dz = 9/32 \cdot K/D$ where $D$ is the diameter of the jet in kilometers. The value of $K$ can range from 0.5 to 0.8. (Color slides are shown, illustrating the development and decay of a cumulus tower).

Turner (1962) proposed a "starting plume" model which combines the jet and bubble models. A steady-state jet is topped by a bubble-type cap. In this case $\mu$ is the same as for the steady-state jet, i.e., $\mu = 0.2/R$.

B. The model of Weinstein and Davis

In the one dimensional, steady-state cumulus model by Weinstein and Davis (1967), it is assumed that at cloud base, the rising thermal is just saturated and has the temperature and pressure of its environment,
as well as a prescribed upward velocity and radius. A parcel of air is initially lifted moist adiabatically through one grid interval (usually 200m) to the next level where its temperature is $T_2''$ and its mixing ratio $q_2''$ (Fig. 2, see end of Lecture #8). The environment at this level is cooler and drier than the cloud, and the mixing that is now assumed to take place at this point cools the parcel to a temperature $T_2'$ and lowers its mixing ratio to $q_2'$ given by

$$T_2' = \frac{T_2'' + \mu dz T_e}{1 + \mu dz} \quad \text{and} \quad q_2' = \frac{q_2'' + \mu dz q_e}{1 + \mu dz}$$

where $T_e$ and $q_e$ refer to the environment. These expressions are weighted means of the mass $M$ of the parcel mixed with an amount $dM$ of environmental air. The parcel is now undersaturated, but since it contains some liquid drops these must be allowed to evaporate until the parcel is once more saturated, which will further cool it. This adjustment can be accomplished graphically by lifting the parcel dry adiabatically to saturation and bringing it back moist adiabatically to its original level. The final result, $T_2'$ and $q_2'$, is that which would have resulted from the evaporation of enough water to saturate the air. The resulting lapse rate of the parcel $\gamma_c$ shown in Fig. 2, is greater than $\gamma_m$, the pseudo-adiabatic lapse rate. The process is then repeated for higher levels.

The principal source of energy for the buoyancy of cloudy air is the latent heat of condensation. A second important source is available if freezing occurs, namely, the latent heat of freezing. After freezing occurs
it is assumed that vapor condenses directly from the vapor to the solid phase so that the ice adiabatic lapse rate is then used before mixing is allowed to occur. Glaciation is assumed to take place at one particular level, where all of the latent heat of freezing is released.

The model assumes that water in the cloud consists of two components, cloud droplets of mixing ratio $Q_c$ and hydrometeor water having a mixing ratio $Q_h$. The droplets are formed by condensation and removed by three processes: evaporation (due to mixing with the environment), conversion to hydrometeor water, and collection by hydrometeor water. The hydrometeor water can be created only by conversion and collection and is the only water that goes into precipitation.

Details of the conversion process are not taken into account; rather the process is parameterized in the manner suggested by Kessler (1965). Conversion is not allowed to occur until $Q_c$ reaches a critical value $a$; after this, conversion is assumed to take place at a constant rate given by $\frac{dQ_h}{dt} = K_1(Q_c - a)$ where $a = 0.5 \text{ g/m}^3$.

Collection is treated on the assumption of a Marshall-Palmer drop size distribution for the hydrometeor water, namely, $N = N_0 e^{-\lambda D}$ where $N$ is the concentration of drops of diameter $D$. All drops are given a fall velocity appropriate to the volume mean diameter and coalescence growth is considered to be continuous.

C. Applications of the model

The model has been used in connection with field experiments involving the artificial seeding of isolated cumulus clouds. Inputs that are needed include an environmental vertical sounding of temperature and relative humidity, the height of the cloud base, and certain microphysical parameters
such as K, a and the ice nucleation temperature. Some of the outputs of the model are shown schematically in Fig. 3 (see end of Lecture #8). They include vertical profiles of temperature, vertical velocity, Qc and Qn. The temperature at which ice is nucleated can be varied to model a natural cloud say (-25°C) or a seeded cloud say (-8°C) and the effects of cloud seeding upon the physical properties of the cloud can be assessed. The results are sensitive to environmental stability, ice nucleation temperature, and the radius R.

The field procedure consists of taking an early morning sounding, using this as input for the model, comparing unseeded computations with seeded computations, and looking at the range of initial cloud radii that will yield a maximum difference between growth with and without seeding. Then an aircraft is taken up to look only at clouds in the size range of interest. Randomly selected clouds in this range are seeded and their changes in height are noted. Another sounding is taken by the plane and this is once again run on the computer with two ice nucleation temperatures for verification. Also radar reflectivity observations give a measure of rainfall produced by the cloud.

Figure 4 (see end of Lecture #8) shows a comparison between observed and predicted changes in cloud top height for seeded and unseeded clouds. It can be seen that the model exhibits considerable skill in its predictions for both kinds of clouds. However, the model may be somewhat unrealistic in that its predictions of liquid water do not correspond to those usually observed; this may be a serious defect because latent heat of condensation is the main driving force for the cloud.

Notes taken by:
P. M. Caplan
S. K. Chan
REFERENCES


Figure Captions

Figure 1 - Illustration of the angle of a divergent jet.
Figure 2 - Graphical description of thermodynamical calculations (From Weinstein and Davis, 1967).
Figure 3 - Steady-state outputs of the model (From Weinstein and Davis, 1967).
Figure 4 - Predicted versus observed cloud top heights for 19 clouds in Arizona (From Weinstein and Davis, 1967).
Lecture 8, Figure 1
Graphical Description of Thermodynamic Calculations.

Lecture 8, Figure 2

\[ \gamma_m = \text{MOIST ADIABATIC LAPSE RATE} \]
\[ \gamma_c = \text{CLOUD LAPSE RATE AFTER MIXING} \]
\[ \gamma_d = \text{DRY ADIABATIC LAPSE RATE} \]
\[ \gamma_e = \text{ENVIRONMENTAL LAPSE RATE} \]
\[ \gamma_q = \text{LAPSE RATE OF DEW POINT TEMPERATURE} \]
ADDITIONAL RESULTS: HEIGHT INCREASE FROM SEEDING ($\Delta HT$)

TOTAL PRECIPITATION ($\int_{\text{BASE}}^{\text{TOP}} Q_h$) SEEDED OR NON-SEEDED

DURATION OF PRECIPITATION, SEEDED AND NON-SEEDED

NOTE: CLOUD DYNAMICS RESULTS PARTICULARLY SENSITIVE TO:

1. ENVIRONMENTAL STABILITY (STRENGTH AND ALTITUDE OF INVERSIONS).
2. ICE NUCLEATION TEMPERATURE.
3. UPDRAFT RADIUS, MIXING PARAMETER $\mu$.

$Q_c, Q_h$, TOTAL PRECIPITATION AMOUNT AND DURATION, ARE IN ADDITION SENSITIVE TO $K_1, K_2, \alpha$.

STEADY STATE MODEL OUTPUT

Lecture 8, Figure 3
Predicted Vs Observed Cloud Top Heights for 19 Clouds in Arizona.

Lecture 8, Figure 4
IX. UNPLANNED WEATHER AND CLIMATE MODIFICATION

We have long known that cities can have an effect on certain aspects of the weather. Thus the temperatures in large cities are generally several degrees higher than those in areas just outside of the cities. However, in recent years there has been a growing body of evidence that cities and certain industrial activities may be influencing precipitation.

A. A study of precipitation by day-of-the-week over the eastern United States

Frederick (1970) analyzed fifty years of daily precipitation data from twenty-two Weather Bureau stations in the eastern United States to see if the precipitation on week days (Monday through Friday) differed from that on week-end days. The results are shown in Figs. 1 and 2. (See end of Lecture #9).

It can be seen from these figures that during the cool season (October through March) Sundays, Saturdays and Mondays had, on an average, less than 1/7 (14.3%) of the total weekly precipitation while on Tuesday through Thursday this daily precipitation was greater than 1/7. However, during the warm season (April through September) the precipitation on each day was within 0.2% of 14.3%.

These results show that the precipitation during the cool season in urban areas of the United States is not randomly distributed but is less on weekends and greater on weekdays. Since there is not a natural seven-day cycle, the implication is that this difference in precipitation is due to the "higher level" of man's activities on weekdays than on weekends.

B. The La Porte weather anomaly

La Porte, Indiana, is a small town situated about 30 miles east of the industrial complex of East Chicago and Gary. Changnon (1968)
Dr. Peter V. Hobbs has pointed out that during the period 1920 to 1945 there was an apparent pronounced increase in precipitation at La Porte, but during the same period nearby stations experienced little precipitation change.

The indicated increases in precipitation at La Porte occurred during the warm season of the year. The increases in summer rainfall were accompanied by an increase in the number of thunderstorm days. The suggestion is that these changes were brought about by the increasing industrial activities in the Chicago area. However, since 1945 there has been a general downward trend in the precipitation at La Porte although the industrial activity in Chicago has continued to increase.

C. Cloud condensation nuclei from industrial sources and their apparent effects on precipitation in Washington State

Hobbs, et al. (1970) have reported on survey measurements of cloud condensation nuclei (CCN) in Washington State. These measurements showed that certain industries are prolific sources of CCN. Some of the more important sources identified were:

<table>
<thead>
<tr>
<th>Source</th>
<th>Output per sec of CCN active at 1% supersaturation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large paper mills (Kraft process)</td>
<td>$10^{17} - 10^{19}$</td>
</tr>
<tr>
<td>Small paper mills (Kraft process)</td>
<td>$10^{14} - 10^{15}$</td>
</tr>
<tr>
<td>Small paper mills (Sulfite process)</td>
<td>$10^{14} - 10^{15}$</td>
</tr>
<tr>
<td>Large sawmills with wood-waste burners</td>
<td>$10^{18}$</td>
</tr>
<tr>
<td>Aluminum smelters</td>
<td>$10^{14} - 10^{16}$</td>
</tr>
</tbody>
</table>

All of the above sources produced sufficient numbers of CCN to cause appreciable changes in the concentrations of CCN in the air for distances extending up to 100 km downwind. Many of the major sources of general air pollution are not appreciable sources of CCN. Thus the
exhaust from automobiles, oil refineries and oil-fired power plants produce relatively few CCN. Consequently, the city of Everett, Washington, which has four large paper mills and a sawmill produces more CCN than Seattle, although Seattle has a population ten times greater than Everett. The locations of some of the principal sources of CCN in Washington State are shown in Fig. 3. (See end of Lecture #9.)

The effluents from industrial sources of CCN can have an appreciable influence on the visible structure of clouds and precipitation. Clouds forming downwind of these sources (proposed name: "fumulus") have been observed to produce precipitable particles very efficiently. In addition to emitting CCN these industries emit varying amounts of heat and water vapor into the air which also aid the formation of clouds and precipitation.


An analysis has been made of the precipitation and streamflow records in Washington State for the periods 1929 to 1946 and 1946 to 1966. Figure 4 (see end of Lecture #9) shows values of the Student "t" statistic when a comparison is made between the mean annual precipitations and streamflows for the period 1929 to 1946 with those for the period 1946 to 1966. Positive values of "t" indicate that the mean annual precipitation was greater in the second period than in the first period. "t" values of 2.0, 2.7 and 3.0 are significant at the 0.05, 0.01 and 0.001 levels respectively.
The principal conclusions to be drawn from the results shown in Fig. 3 are:

1) Over most of the State of Washington the mean annual precipitation during the period 1946 - 1966 was higher than in the period 1929 - 1946. The increases in precipitation were generally more pronounced over higher elevations than lower. This conclusion is substantiated by high "t" values for streamflow from those rivers fed by high elevation watersheds.

2) Contrary to the general trend described above, there are two lowland pockets in the Puget Sound Basin which have highly significant positive values of "t". One of these is centered south of Pt. Townsend at Chimicum and the other just N.E. of Victoria. Also, the North Forth of the Stillaguamish River (NFS) which has a low elevation watershed shows the most significant "t" value (3.18) of all the rivers in the Puget Sound. The "t" values of 5.6 at Chimicum and Victoria reflect the fact that the mean annual precipitations at these locations during the period 1946-1966 were 33.7% greater than during the period 1929-1946. The mean annual runoff from the NFS was 20.8% greater during 1946-1966 than 1929-1946.

3) The two regions west of the Cascade Mountains which have the highest "t" values (Chimicum and Victoria) are both in the vicinity of large sources of CCN from paper mills (see Fig. 3). Also, the NFS is directly N.E. and therefore generally downwind of paper mills at Everett, Pt. Townsend and Port Angeles.

4) There is a region in the S.W. corner of the Olympic Peninsula (located at Raymond) where the "t" value is 3. This is also adjacent to a source of CCN associated with the burning of woodwastes in open furnaces (Fig. 3).
Several industrial sources of CCN along the Columbia River may be associated with fairly high "t" values.

5) Several high "t" values are found in the eastern part of the area shown in Fig. 4 (see end of Lecture #9). The largest of these (t = 6.03) is at Trail, Canada, where the largest non-ferrous smelter in the world is located. Near the center of the "t" value of 4.31 at Newport is a large wood-burning match factory. At Kellogg, Idaho, where the "t" value is 4.29, there is a large non-ferrous smelter.

It appears from these results that there is a high correlation between industrial sources of CCN and areas which have experienced anomalously high precipitation in the past twenty years.

E. Climate modification

We turn now to the possible effects of man's activities on worldwide climate.

From 1880 to 1940 the mean temperature over the earth's surface averaged for one year increased by 0.6°C while in the last 25 years it has decreased by 0.3°C. These apparently small changes in temperature caused dramatic changes in the position of frost and ice boundaries over the earth. The question must be asked: Were these changes in temperature caused by man's activities on this planet? We consider below several possible ways in which man's activities might cause a change in world climate.

The radiation balance of the earth is very sensitive to the amount of CO₂ in the air. In the nineteenth century the concentration of CO₂ in the air was about 290 parts per million and in 1970 it was 330 parts per million. This increase in CO₂ in the air is probably due to the burning of fossil fuels by man.
Calculations by Manabe and Wetherald (1967) show that for a period of relative humidity a 5% increase in CO₂ would lead to an increase in the mean temperature of the earth of 0.1°C. Hence, the warming trend up to 1940 may have been due in part to the increase in CO₂ in the atmosphere. However, since 1940 the CO₂ content has continued to increase while the temperatures have decreased.

The thermal balance of the earth is also affected by particulates. Bryson (1968) estimates that a decrease in atmospheric transparency, due to increased particulates, of 3% would reduce the surface temperature by 0.4°C. As we have seen above, particulates can also affect cloud cover. At present, about 31% of the earth's surface is covered by low cloud. If this increased to 36% the average temperature over the earth's surface would fall by 4°C! The transparency of the atmosphere is certainly decreasing and this is in part due to man's activities.

Manabe and Wetherald (1967) estimate that a unit increase in the average albedo over the earth's surface would result in a decrease in average surface temperature of 1°C. The albedo of the earth is being changed by cultivation, de-forestation and urbanization. However, the net magnitude (and even the direction) of the change in albedo is unknown.

It is clear from these brief remarks that the changes in the environment caused by man's activities might be producing modifications in world climate. However, at the present time we are unable to predict the direction of these changes let alone their magnitude.

Notes taken by:

J. R. Travis

Y. M. Chang
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Figure Captions

Figure 1 - Per cent of total precipitation by day-of-the-week for 22 stations in the Eastern United States for the period 1912 - 1961 (From Frederick, 1970)

Figure 2 - Per cent of total cool-season precipitation by day-of-the-week for 22 stations in the Eastern United States for the periods 1942 - 1961 and 1912 - 1931 (From Frederick, 1970)

Figure 3 - Locations of some industrial sources of CCN in Washington State

Figure 4 - Values of Student "t" statistic for comparison of mean annual precipitations and streamflows for period 1929 - 1946 with 1946 - 1966.
Lecture 9, Figure 1
Lecture 9, Figure 2