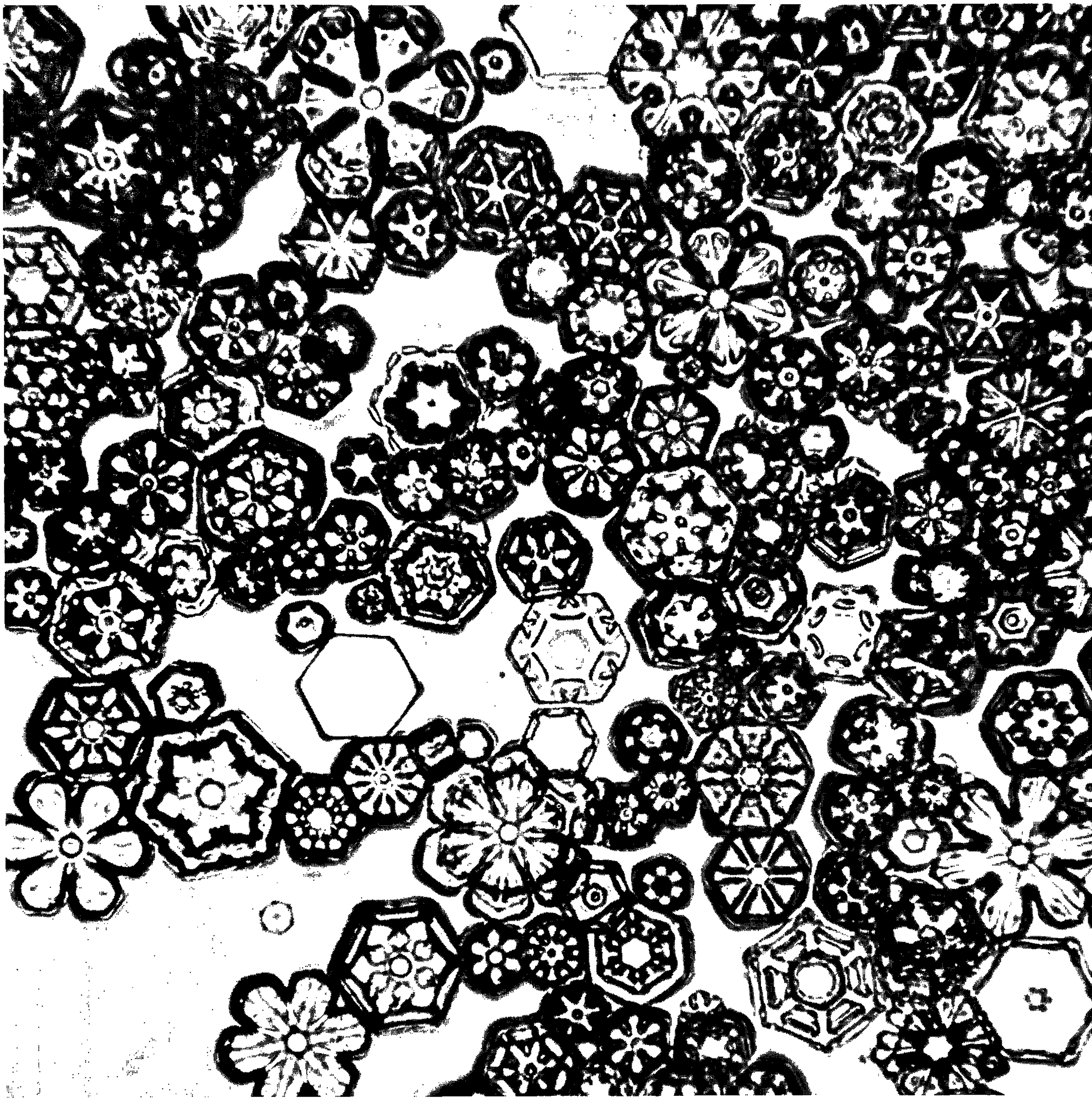
WMA



The Journal of Weather Modification

Volume 3 Number 1 April 1971

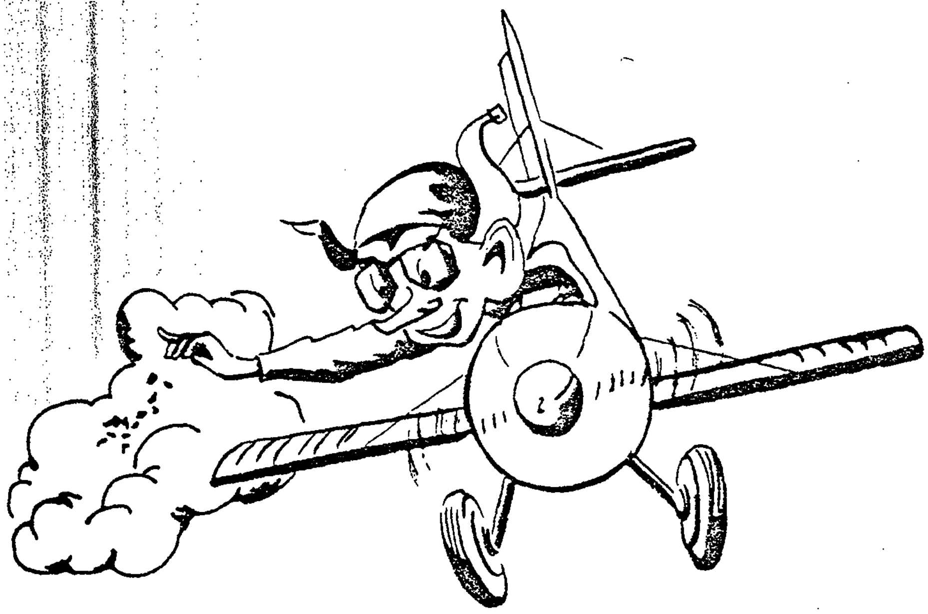
SECOND PRINTING

THE JOURNAL OF WEATHER MODIFICATION

Cover Photograph

Ice crystals produced from pyrotechnic generated silver iodide smoke into a supercooled (-2C.) cloud in a cold chamber. Crystals replicated by the Schaefer method using the Formvar 15-95 in ethylene dichloride. The photograph was made with a Leitz orthomat automatic microscope camera through a Leitz ortholux polarizing microscope with Xenon intensity light source. Optical magnification: 100x.

Photograph by Roger Cheng, Research Associate, Atmospheric Sciences Research Center, State University of New York at Albany.



WEATHER MODIFICATION ASSOCIATION
c/o Mr. Jay Bibby, Exec. Sec.
P.O. Box 490
Coronado, California
92118

Additional copies available at $6.00 each

"NON-REVIEWED"

- THE JOURNAL OF WEATHER MODIFICATION -

WEATHER MODIFICATION ASSOCIATION

Volume III Number 1 April 1971

Table of Contents

Table of Contents

Table of Contents | *Page*

- - - - -

PYROTECHNIC PRODUCTION OF NUCLEANTS FOR CLOUD MODIFICATION

Part VII. Nucleation Processes

Pierre St.-Amand
William G. Finnegan
F. Kirk Odencrantz

Naval Weapons Center
China Lake, California

ABSTRACT

Glacogenic nuclei function by (1) condensation of water upon the nucleant followed by freezing, (2) sublimation, (3) contact nucleation.

The temperature of activation of pyrotechnically generated, relatively pure AgI is close to -2.0°C.

The maximum usable number of nuclei per gram, for contact nucleation is determined from Fletcher's theory.

Some experimental results are discussed.

INTRODUCTION

Augmentation or stimulation of rainfall by seeding of clouds with nuclei intended to induce formation of the ice phase is the most common form of intentional weather modification. This process may be considered to be a series of steps in a chain of events. The chain must be considered step by step and at each step the details are of consequence. One must ask a series of questions: Did the material enter the critical part of the cloud? Was the material a viable catalytic agent upon its entry? Was the material what it was thought to be? Was the cloud type such that nucleation could occur in a time short compared to the lifetime of the cloud? What is the reaction of the cloud once the ice phase is formed? What effect will the wind regime have upon the cloud?

In order to assess some of these, let us examine these questions in some detail.

NUCLEATION PROCESSES:

The ice phase may be initiated in a cloud by conversion of either water vapor or liquid water to solid water. The conversion process depends upon the exact nature, chemical and physical, of the nucleant and upon the history of the nucleant, as well as upon the characteristics of the cloud.

The physical theory of catalysis is well developed and reasonably satisfactory, except in microscopic detail and is used every day by chemists and others interested in catalysis in industrial processes. Reviews of nucleation processes as applied to the atmosphere were written by Horace Byers (1965), Fukuta (1967), Long (1969), and St.-Amand (1967).

Three processes by which the ice phase can be nuclated are: (1) Contact between the catalyst and supercooled water either internally or superficially; (2) Condensation of liquid water upon the catalyst, followed by freezing; (3) Sublimation of water vapor directly to the ice phase.

1. Contact Nucleation: Nucleants actually touch, or become imbedded in cloud droplets, causing them to freeze. If ice is the catalyst, freezing can take place at $0^{\circ}C$ or colder; if silver iodide is the nucleating agent, the process can be expected to cause freezing if the nucleation material comes in contact with droplets colder than $-2^{\circ}C$. If the droplets are colder than the air in which they are imbedded, the freezing process can take place in air somewhat warmer than this. Such temperature differentials between droplets and air can arise if the droplets are evaporating.

In general, this process is usually thought to be less important than the diffusional processes in real clouds. If however, the number of nuclei per unit volume; the number of droplets per unit volume are high; and the droplets large, the process overrides all the others in importance. If the nuclei are electrically charged so that they are attracted to water droplets, the probability of this reaction taking place becomes higher. This subject will be discussed in detail in a later section of this series.

Because the frozen droplets are large compared to ice embryos, once contact nucleation occurs, the release of heat from frozen droplets by initial freezing and subsequently by diffusional growth is more rapid than it is when growth occurs from a tiny ice embryo by the diffusional processes alone and considerably greater effects can be observed upon the dynamics of cloud systems. Although the same amounts of energy may be released in the end, the rate of release is much higher owing to the larger sizes of the ice particles produced initially.

2. Nucleation by Condensation: At temperatures above and below the freezing point, both AgI and PbI_2 nuclei adsorb water vapor and produce condensation. In the case of silver iodide, condensation is followed by freezing if the temperature is below $-2.0^{\circ}C$. The water is probably deposited, first as structured liquid water which then gradually freezes through loss of heat and continued ordering of the structure. If only pure AgI, incapable of adsorption, is considered, it is unlikely that condensation will occur at all in the regimes of saturation pressure found in nature. Rigorously pure AgI, is seldom, if ever, used as a nucleating

material. Experimental and theoretical work indicates that nuclei greater than one micron are required to produce condensation. Fletcher (1962), page 217, discusses the effects of adsorption of the vapor phase onto the surface of a nuclei at saturation ratios close to and less than unity.

The first experimental clue came from the work of Schaefer (1954) in which he was experimenting with a diffusion cloud chamber. The temperature was set at +10^{o}C at the top and -65^{o}C at the bottom. He waited until all the droplets had disappeared, presumably by fallout, and then introduced into the cloud chamber AgI and PbI_2 made by an electric spark using either silver or lead electrodes in an atmosphere containing iodine vapor. He found that when either smoke was introduced at the -5^{o}C level that ice crystals were produced. These he attributed to sublimation. When the smoke was introduced at the +5^{o}C level, the particles of both AgI and PbI_2 formed droplets which fell and subsequently froze, the AgI at -5^{o}C, the PbI_2 at -20^{o}C. He attributed the difference to solution of the PbI_2 by the water.

Isono and Ishizaka (1968) noted that at -3.5^{o}C water droplets form on an AgI film at the dewpoint and remained liquid for hours. Between -3.5^{o}C and -8^{o}C water drops appeared and subsequently froze. Below -8^{o}C and at saturations below that of liquid water, but presumably above that of ice, ice crystals formed on prism faces (a and b axes) of AgI. Below -8^{o}C and above saturation with respect to liquid water, drops formed on both basal and prism faces.

Birstein (1952, 1955, 1956) showed a remarkable adsorption of water onto dry silver iodide. Using a McBain balance, he placed AgI in the pan, outgassed it and then admitted water vapor from distilled water. The weight increase showed, as did earlier work by Coulter (1952), that for all values of partial pressure, adsorption took place, giving a BET type III curve. At +20^{o}C and a saturation ratio of 0.97 about 183 molecular layers of water were adsorbed. At -20^{o}C, 160 molecular layers were adsorbed although some of this may well have been accretion by sublimation. The binding energy at -20^{o}C approached 18 kilogram calories per mole for the first layer.

Corrin, et al. (1963, 1964) did not find the adsorption of water on AgI that Birstein reports, nor do they think that the type II BET isotherm applied unless the AgI contains some hygroscopic material. They attribute the adsorption to the presence of ammonium nitrate. The method of preparation of the AgI that they used differs from that used by Birstein. Karaz, Champion and Halsey (1956) found the amount of adsorption to be undetectable. Sano and Fukuta (1956) found the adsorption to be very slow. Tompkins, Muus and Pearson (1963) found that the AgI-KI system shows an adsorption isotherm of the type BET III, because this system is deliquescent. Corrin, et al., (1963, 1964) may well be correct in that sorption is taking place on hygroscopic impounds. Zettlemoyer and colleagues (1961, 1963) contend that AgI is hydrophobic and that adsorption takes place on hygroscopic centers. Corrin and Barchel (1970) note that adsorption begins to take place at partial pressure of 0.93.

Quite possibly absorption, rather than adsorption initiates the process and once begun, adsorption is activated. At temperatures adequately low so that liquid water were not deposited first, the beneficial effects of sorption would not take place. Whatever the mechanism it is clear that water is deposited on AgI over and above what might be expected from simple condensation on an insoluble particle. Evans (1970) has begun to investigate this aspect of nucleation.

From such a start, it would be possible to grow viable condensation embryos on the particles far too small to serve as sublimation nuclei and much smaller than the theory, to be discussed later, will indicate to be possible for a pure AgI particle that does not show effects of adsorption or sorption by hygroscopic additives.

If supersaturation of the order of 3 percent is attainable, particles as small as 0.01 microns radius do become effective as condensation nuclei. The process of condensation can be made more effective by treating AgI with another compound which will be effective in reducing the vapor pressure of water over the surface of the material by even a very small amount. If either adsorption or absorption of water vapor on the nucleating agent is possible, then much smaller particles can serve as condensation nuclei. The 0.93 partial pressure noted by Corrin and Barchel (1970) is capable of initiating condensation.

With slightly modified AgI, the condensation process will work at temperatures as cold at $-12.5^{o}C$. If the material used is very hygroscopic, the condensation process will work at much colder temperatures, except that if complexing of the AgI with the hygroscopic material (NaI for example) occurs, partial or complete solubilization of the nuclei may occur before freezing is produced. The subsequent accretion of water upon a viable ice embryo, once formed by freezing of condensed water, will be by continued condensation at temperatures between $0^{o}C$ and $-12.5^{o}C$. At temperatures lower than $-12.5^{o}C$, and at saturation ratios of unity, or less, further accretion is by sublimation. Theoretical work by Krastanov is quite clear in this respect, except that he places the lower limit for the process at $-10^{o}C$.

3. Nucleation by Sublimation: At temperatures colder than $-12.5^{o}C$, nucleation on AgI from water vapor to ice appears to be by direct sublimation. Mason and Van Den Heuvel (1959, p. 754-755) propose:

> ".....At temperatures below $-12^{o}C$, silver iodide can initiate ice crystal formation in an atmosphere subsaturated relative to liquid water, provided that the supersaturation with respect to ice exceeds a critical value of 12%. At temperatures between $-5^{o}C$ and $-12^{o}C$ however, the air has to become water saturated before ice crystals appear; this may be a consequence of the fact that, at temperatures above $-12^{o}C$, the critical supersaturations of 12% cannot be reached without passing water saturation as indicated by the dashed line in the figure." (Figure 1)

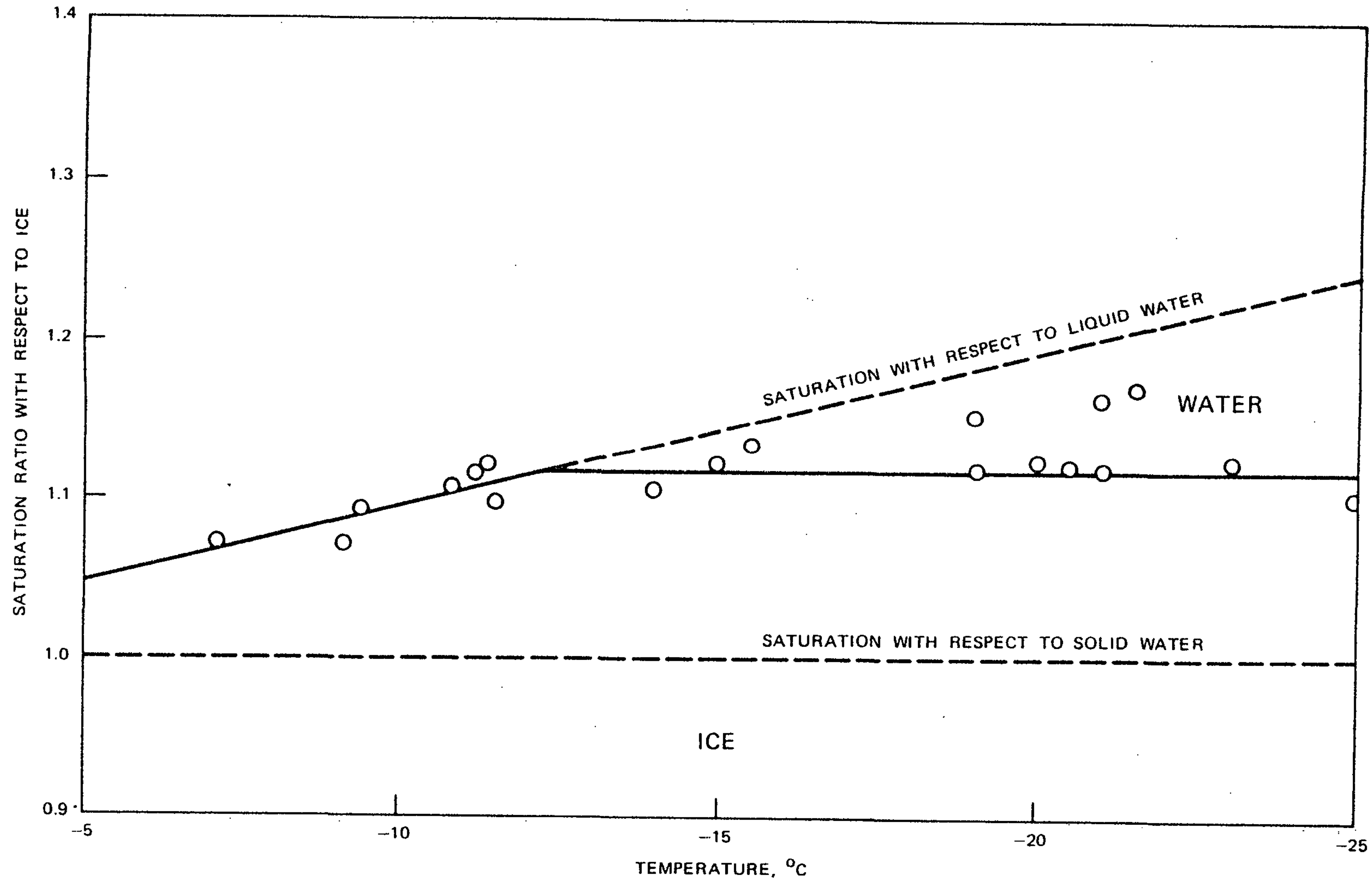


Fig. 1. Taken from Mason and Hallet, 1965.

Temperature of Activation: Temperature of activation is the highest temperature at which a relatively large piece of a substance will serve as a nucleant for ice. Values of -4.5°C or -5°C are commonly cited for AgI. These values appear to have been established by custom rather than experiment. The actual activation temperature will depend markedly upon the state of cleanliness of the material, contaminants, and previous history of exposure. It depends as well upon the state of elastic strain in the material.

If epitaxy is important to nucleation, then the interatomic spacing of the substrate is important and a little elastic strain is all that may be required to bring a portion of the surface into a very close fit to water.

Table 1 reproduced from Mason and Hallet (1956, p. 683) show the closeness of fit between silver iodide and ice.

TABLE 1. Dimensions of unit cells.

Substance	Crystal Form	Unit Dim. Å	
		a	c
AgI	Hexagonal	4.58	7.49
	Cubic	6.47	
	"High Temp. Cubic"	5.03	
PbI_2	Hexagonal	4.54	6.86
Ice	Hexagonal	4.52	7.37

That epitaxy may be important is shown by Turnbull and Vonnegut (1952,p. 1296) where they discuss the relationship between "disregistration" and threshold temperature. Using 0.0145 as "S" the disregistry and -2.5°C as the activation temperature determined by Schaefer (1949) they show an excellent fit between theory and observation, Figure 2.

Davis and Blair (1969) indicate that strain stored in a nucleant may aid in bringing about nucleation.

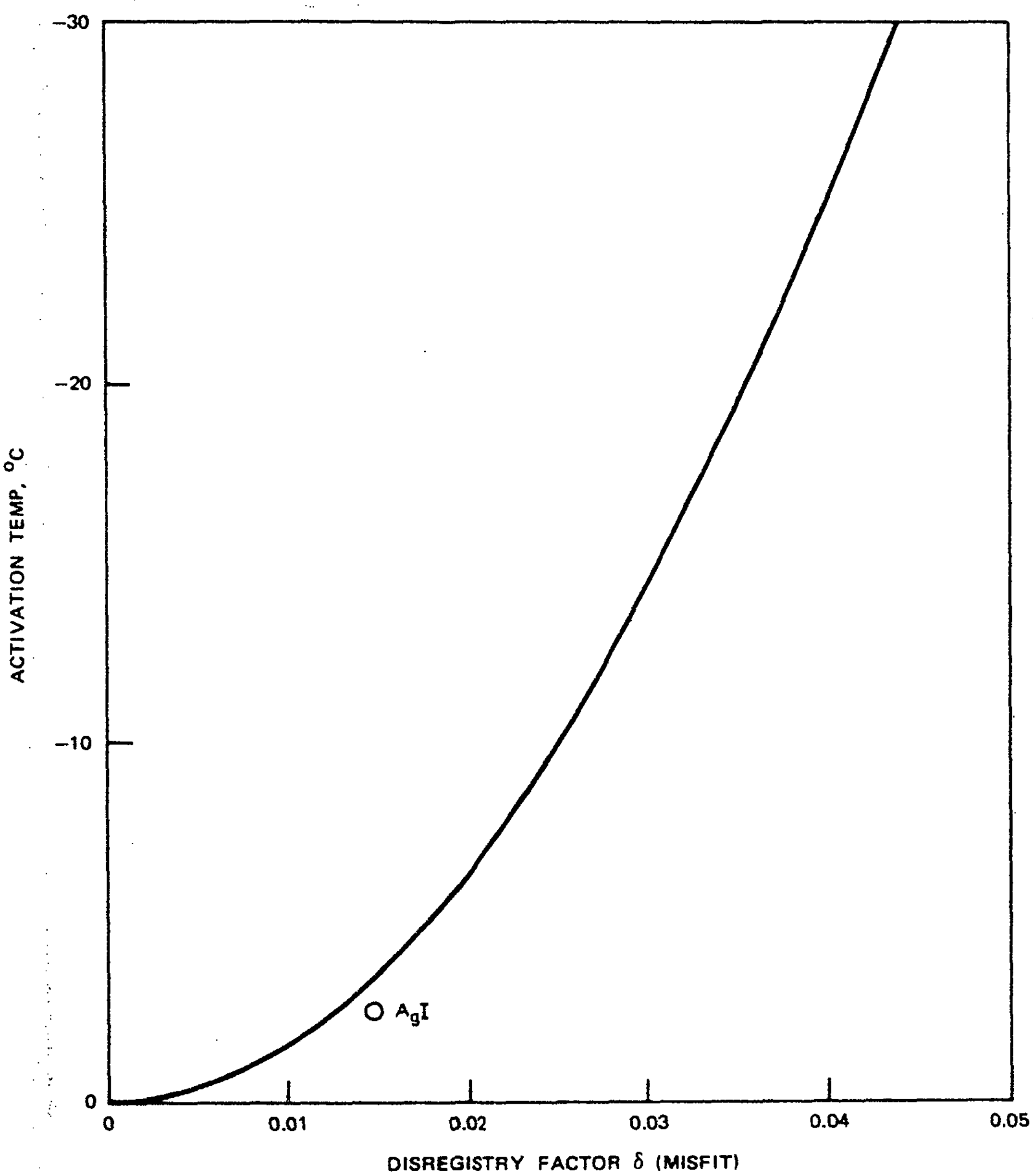


Fig. 2. Activation temperature as function of disregistry, redrawn from Turnbull and Vonnegut (1952, p. 1296)

In Table 2 we summarize the activation temperature from several sources:

TABLE 2. Activation Temperatures, AgI

Author	Activation Temperature, C°
Isono and Ishikawa, 1968	-3.5
Mason and Hallett, 1956	-5
Vonnegut, 1949	> -3.5
Schaefer, 1949	-2.5
Fukuta, 1958, p. 21	-2.6 ± 0.2
Cuilong, 1947	-2.9 ± 0.1
Hosler, 1951	-3
Aufmkampe , 1951	-3
Odencrantz, 1969*	-2

* Pyrotechnically generated

The actual value of threshold depends on the material to a great extent.

Odencrantz (1969) placed drops of multiple distilled water on a teflon substrate and held them at temperature for a protracted interval without freezing. He then exposed them to pyrotechnically generated AgI smoke and noted that some froze at -1°C and most of them at -2°C. He has measured nucleation effectiveness as high as 10^8 ice crystals per gram at -2.1°C and 10^9 at -3.0°C. Because he was using a very finely divided smoke, his temperatures of activation are lower than what could be obtained with similar material in larger pieces.

It seems reasonable therefore to assume that -2.0°C is the activation temperature for pyrotechnically generated AgI using NWC formulations.

The temperature of activation for AgI may also depend on supersaturation, and it may well be different for sublimation than it is for freezing.

Fletcher's Theory of Nucleation: A theory of nucleation by particles, and based on classical thermodynamics taken from the general theory of catalysis has been applied to water by Fletcher (1958a, 1958b, 1959a, 1959b, and 1959c). Basically, he considers the energy barriers necessary for the formation of critical embryos and shows that the nucleation rate is strongly dependent on particle size, saturation ratio and temperature. The Fletcher theory is clearly written and is not reviewed in detail here, but the results are used and discussed. Because the theory is based on thermodynamics and the energy exchanges necessary, it describes the processes on a macroscopic scale without detailed knowledge of the microphysics and sets limits to which the processes must conform regardless of the microscopic details. It gives the values that will produce the most results that can be expected from an ideal spherical nuclei and sets the limits for the number of nuclei that can be produced as a function of temperature. These values are correct as far as they go, and he has even extended the theory to particles of other regular geometric shapes. Fletcher (1968) does not include effects of adsorption but has recently discussed the effects of inoculating the nucleating material with hygroscopic substances. Should the actual nucleation take place only on certain portions of the surface, then the theory will have to be modified, and, indeed, Fletcher (1970) has begun to do this.

Zettlemoyer, et al. (1963, p. 498) sum it up nicely:

> "Most remarkable is the fact that a useful description of nucleation phenomenon has been derived from the classical theory (referring to Fletcher's work) while knowledge of these molecular effects were lacking. What is needed now is a theory to incorporate these concepts to explain the probable lowering of the energy barriers to nucleation by the proper type of adsorption site."

Following classical theory, Fletcher shows that the free energy of formation, ΔG^*, of an embryo of critical size r^* is:

$$\Delta G^* = 16\pi\sigma^3/3(\Delta G_v)^2 \qquad (1)$$

where σ is the surface free energy of the embryo and ΔG_v is the volume free energy difference σ between the old and the new phases.

Thus, from homogeneous nucleation, the energy barrier is related almost exclusively to the surface free energy of the embryo and to the difference in the volume free energy. For passage from the vapor phase to either the liquid or solid phases, these energy differences are greater than from the liquid to the solid phase.

Heterogeneous Nucleation: Fletcher then passes to nucleation on a plane surface of an exotic material. If a foreign particle, phase 3, be introduced in phase 1, (the old phase) and if it has properties identical to the new phase, phase 2, and complete contact can be attained, it will function as a critical embryo of the new phase and nucleation will proceed, see Figure 3. If the exotic particle be of a different material, a lesser nucleating effect is to be expected for a given size particle and the contact angle θ, involving the surface tension is important in determining the effects of the exotic particle. An expression, m, is defined as:

$$m = \cos\theta = (\sigma_{13} - \sigma_{23}) / \sigma_{12} \qquad (2)$$

where σ_{13} is the free surface energy of Phase 1 on the nucleating particle, phase 3; σ_{23} is the free surface energy of phase 2 on phase 3, and σ_{12} is the surface free energy of phase 1 on phase 2, and is the σ used in equation 1. If phase 3 is the same as phase 2, then

$$m = \cos\theta = 1 \qquad (3)$$

It is at this point in the theory that the nature of the material becomes important and is introduced into the argument. In the discussion to follow Fletcher (1958) considers the case of spherical nuclei. Fletcher (1963) considers the case where the nuclei are not spherical but are regular solids; this leads in general to a requirement for larger particles.

Fletcher's development considers next nucleation upon a plane surface, extends it to spherical nuclei and develops the relation between particle size and saturation ratio.

He uses the classical expressions for nucleation rate as based on the energy necessary to form a critical embryo. In this expression, one constant, B, is subject to some question and the actual results will depend to some extent on the value, Fletcher (1970) emphasizes this point.

From the basis then of considering the energy necessary to form embryos by condensing or sublimating the water vapor or by changing liquid to solid, he develops a series of curves showing the relationship between the size of the particle necessary to form one embryo in one second, some of which we reproduce herein.

Figure 4 shows the saturation ratio necessary at 0^{o}C for condensation to occur on a nonsoluble particle of surface parameter, m, radius, r, microns. Even for ice itself, assuming that ice has a contact angle of unity for water, which it probably has only at the triple point, particles of at least 1 micron radius will be necessary for nucleation of the ice phase by condensation. In other words, a particle of ice must be larger than one micron in radius before condensation will occur thereon. The size necessary to induce condensation is not a strong function of temperature because it is reciprocally proportional to the absolute temperature T. Hence, Figure 4 may be used over a reasonable temperature range.

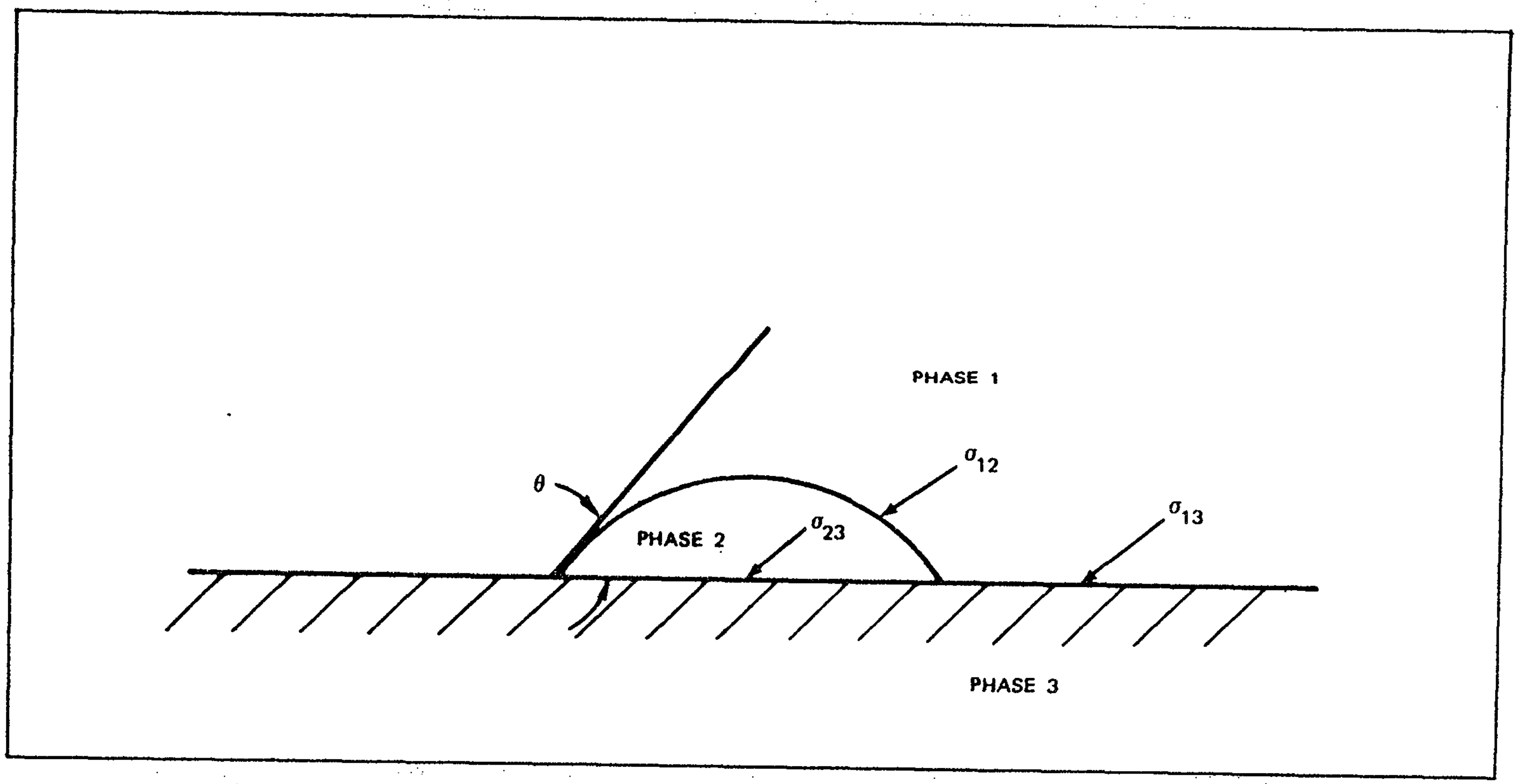


Fig. 3. Relation between contact angle θ and components of a three-phase system.

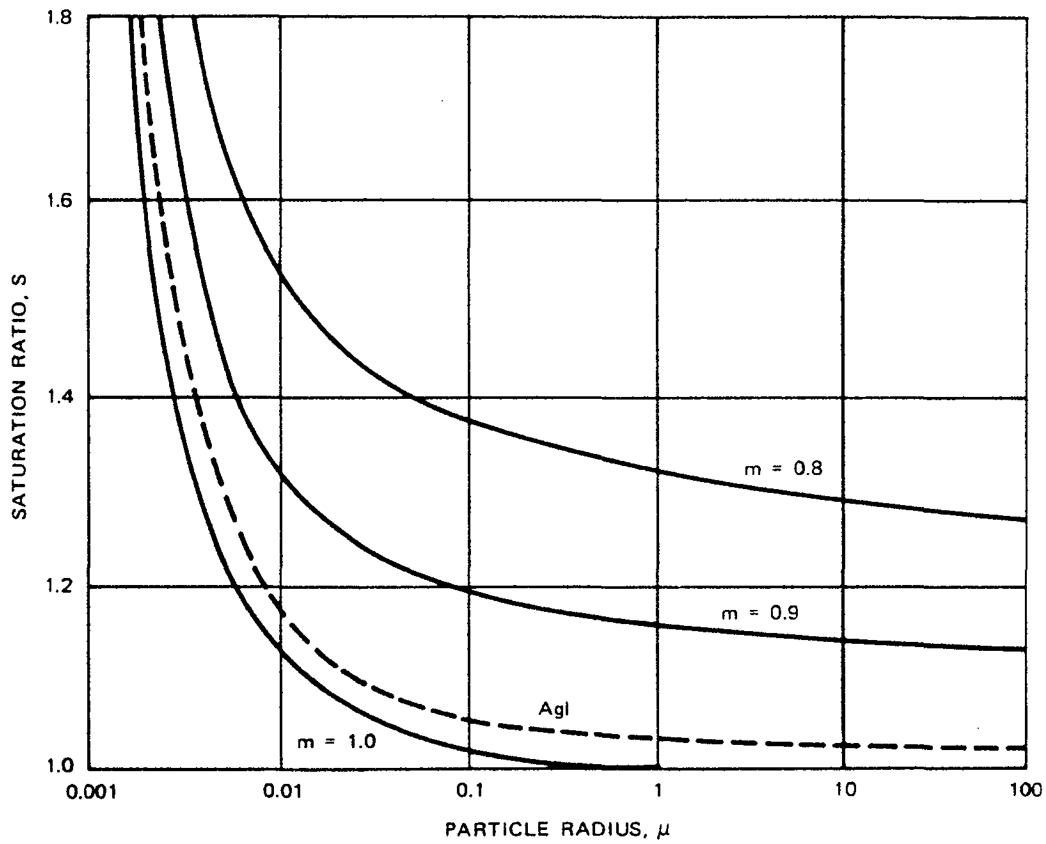


Fig. 4. Supersaturation necessary, with respect to a flat water surface, to induce condensation on a spherical particle of radius, r, in microns for various values of m.

It is clear from Figure 4 that pure, nonadsorbing, silver iodide will not serve as a condensation nuclei under the conditions ordinarily met within the atmosphere. This fact led Fletcher to assume that condensational processes probably were not important and that nucleation in the atmosphere probably took place by sublimation. This is the only serious error in his earlier discussions and was challenged by Edwards and Evans (1960). The statement is correct for pure AgI, except that sublimation would not be possible at temperatures above $-12.5^{\circ}C$ because of lack of adequate vapor pressure, and hence pure AgI having the properties of bulk laboratory reagents and showing no adsorption could only be expected to work by diffusional processes at temperatures colder than $-12.5^{\circ}C$. The difficulty arose, it is suspected, from a misunderstanding of the nature of nuclei produced by acetone burners. This point was clarified in part by Fletcher (1968), but as will be discussed later, the clarification is not quite correct.

Effects of Adsorption and Absorption: A simple manipulation is here permissible for purposes of exploring what effects small changes in the nature of the AgI would involve. (If the AgI were capable of adsorption, or if it were adulterated with an agent that showed hygroscopic properties, then condensation could take place easily because the vapor pressure lowering over the surface would behave very like an increase in vapor pressure over the pure AgI, that is to say, an apparent supersaturation would be produced.) Crude as this argument is, it can be seen that even a slight vapor pressure lowering will result in condensation taking place on AgI and making that material effective as a condensational nucleating agent. For a 20% lowering in vapor pressure, over the surface of the nucleant, a decrease in nuclei radius of 2 and 1/2 orders of magnitude becomes permissible.

Figure 5 taken from Fletcher (1958a), shows the temperature in degrees centigrade necessary to produce ice embryos by nuclei of radius R at ice saturation. The values of m are for the contact angle of ice on the nucleus with no elastic strain. It is interesting to note that for all values of m 1 that the curve does not approach $T = 0^{\circ}C$ for large values of R but instead appraoches some lower temperature determined by the degree of misfit and the elastic constants of ice. Fletcher assumed that the warmest temperature at which unstrained monocrystalline AgI can function as a sublimation or freezing nuclei was $-4.5^{\circ}C$.

The data of Figure 5 are for the vapor pressure over ice, the curves are not directly usable in the real atmosphere. At $0^{\circ}C$, the vapor pressure over ice is the same as over water. At all colder temperatures, the vapor pressure over water is greater than that over ice. At $-12.5^{\circ}C$ the vapor pressure difference is just about that necessary to produce the excess over ice saturation required for sublimation on AgI. Hence, at warmer temperatures condensation will take place first. Fletcher's sublimation curve for AgI, as shown in Figure 5 here, is valid for atmospheric application only at temperatures below $-12.5^{\circ}C$. The supersaturation needed for sublimation does not usually exist in nature at warmer temperatures because it is greater than that for liquid water saturation.

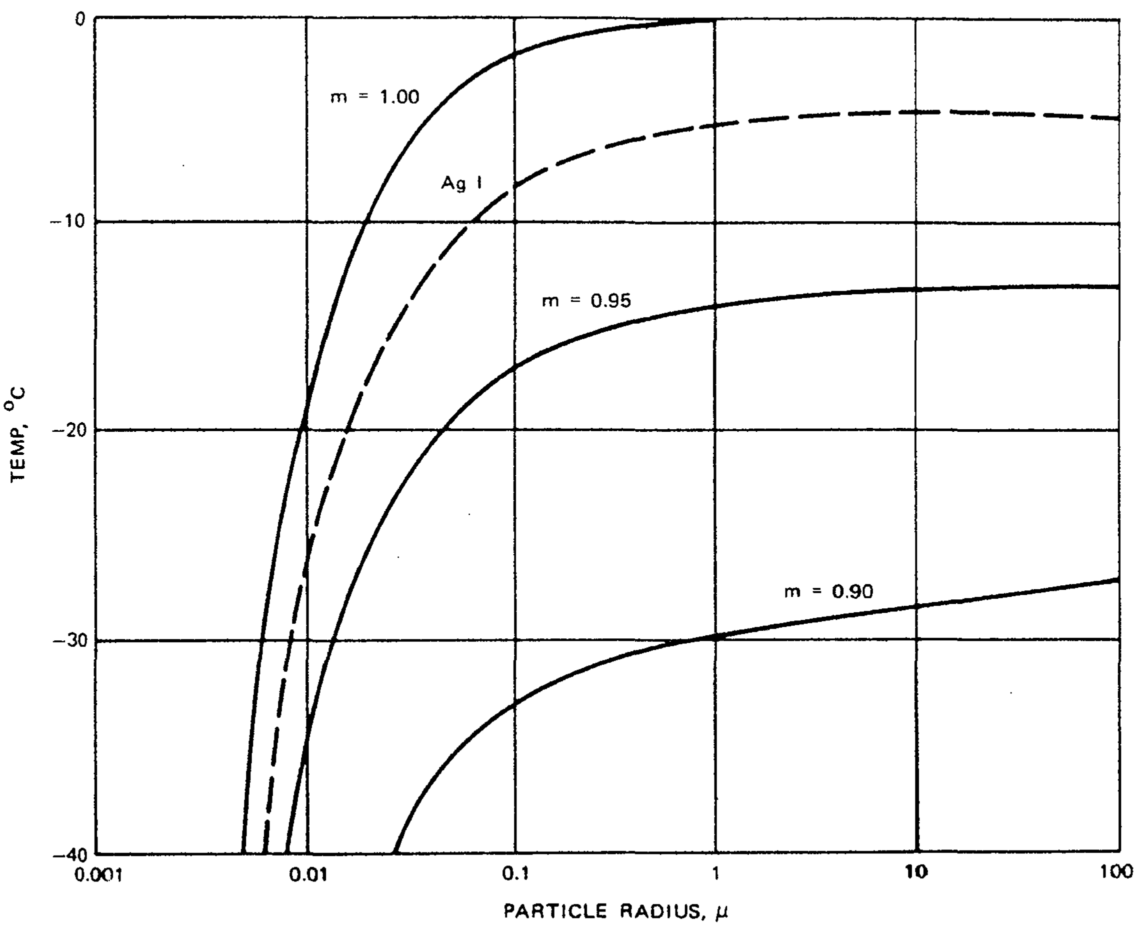


Fig. 5. Temperature at which nucleation by sublimation will occur on spherical particles of radius, r, and surface parameter, m, within one second at saturation with respect to ice. It is unlikely that in the atmosphere sublimation will occur above -12.5°C. From Fletcher, 1959, p. 195.

Freezing of Liquid Water: Figure 6 shows the particle sizes necessary for freezing, once condensation has taken place on the nucleating agent, or if the nucleating agent comes in contact with a water droplet. Much smaller particles can be effective in inducing freezing than are necessary for induction of sublimation. If condensation can be induced on smaller particles by means of adsorption or some process for lowering vapor pressure, that does not also greatly depress the freezing point, then the results that can be expected by means of contact nucleation or condensation followed by freezing will be much more interesting to the weather modifier than the effects of sublimation. Figure 7 is important to our argument because it shows the relationship between particle sizes necessary for the two processes, sublimation and freezing or simply contact nucleation. It shows the curves for AgI plotted on the same scale. The outstanding difference is that particles far too small to initiate sublimation are effective in initiating freezing if the material can be placed in contact with liquid water either by condensation or by coming into contact with a droplet. There will be a difference, it is true, in the numerical effectiveness, depending on whether the particles become imbedded in droplets and are totally wetted, or whether they merely touch the surface.

Determination of the Maximum Number of Nuclei from One Gram of AgI: Fletcher (1959a) goes on to show how the maximum number of nuclei producible from one gram of a nucleating agent may be determined. His argument is applicable to any kind of nucleant that functions as does his hypothetical silver iodide. It determines the maximum number of nuclei produced by one gram of AgI. Any nucleant produced by a real AgI generator will always have to produce less than this amount if it is functioning in accordance with the theory upon which the determination is based.

From Figure 5, the graph showing the size of particle necessary to induce sublimation and from the density of silver iodide, it is an easy matter to calculate the maximum number expected to be active. Figure 8 shows Fletcher's famous curve. We have here dashed the portion between $-5^{\circ}C$ and $-12.5^{\circ}C$ because for the reasons stated earlier, we feel that sublimation should not occur in this temperature regime.

This curve is one of the most misunderstood items in weather modification. People who have read Fletcher's papers superficially or who have looked only at the figures have concluded that this represents the performance curve for a properly functioning generator material. Not so, it represents only the maximum number of nuclei of a given diameter that can be produced and that the limiting upper temperature at which they are effective. For example, if a monodispersed agent is of a given size, say 1,200 A, these particles will not work by sublimation in less than a second at temperatures warmer than $-10^{\circ}C$. They will produce about 1×10^{14} nuclei per gram and no more at any colder temperature. Each time that the observed curve for a nucleant touches Fletcher's curve, it means that one gram of material is represented (acting as sublimation nuclei). It is possible to analytically determine the amount of material

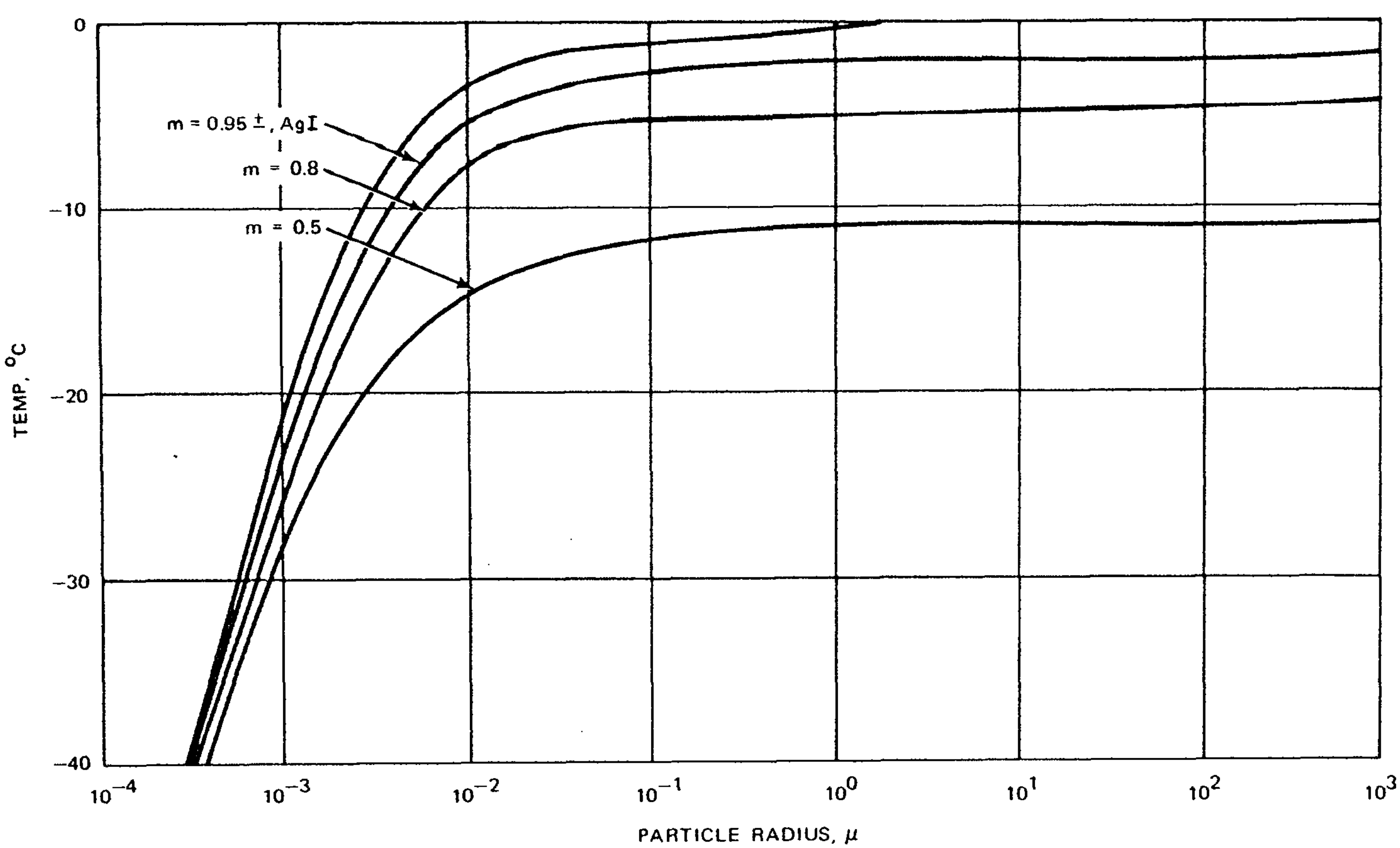


Fig. 6. Temperature at which a particle of radius, r, will cause liquid water to freeze as a function of surface parameter.

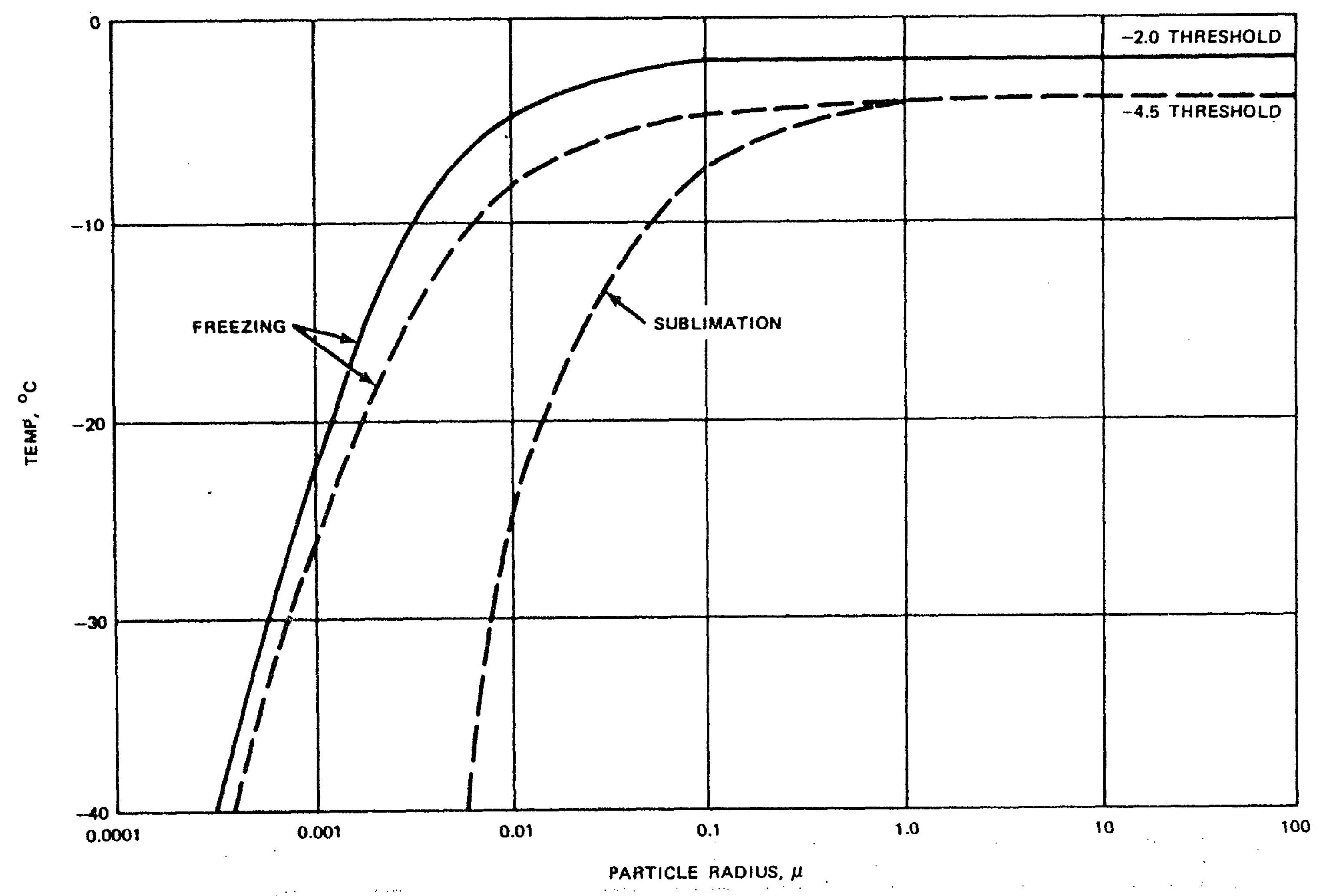


Fig. 7. Showing comparison of particle sizes necessary for freezing by sublimation and by freezing.

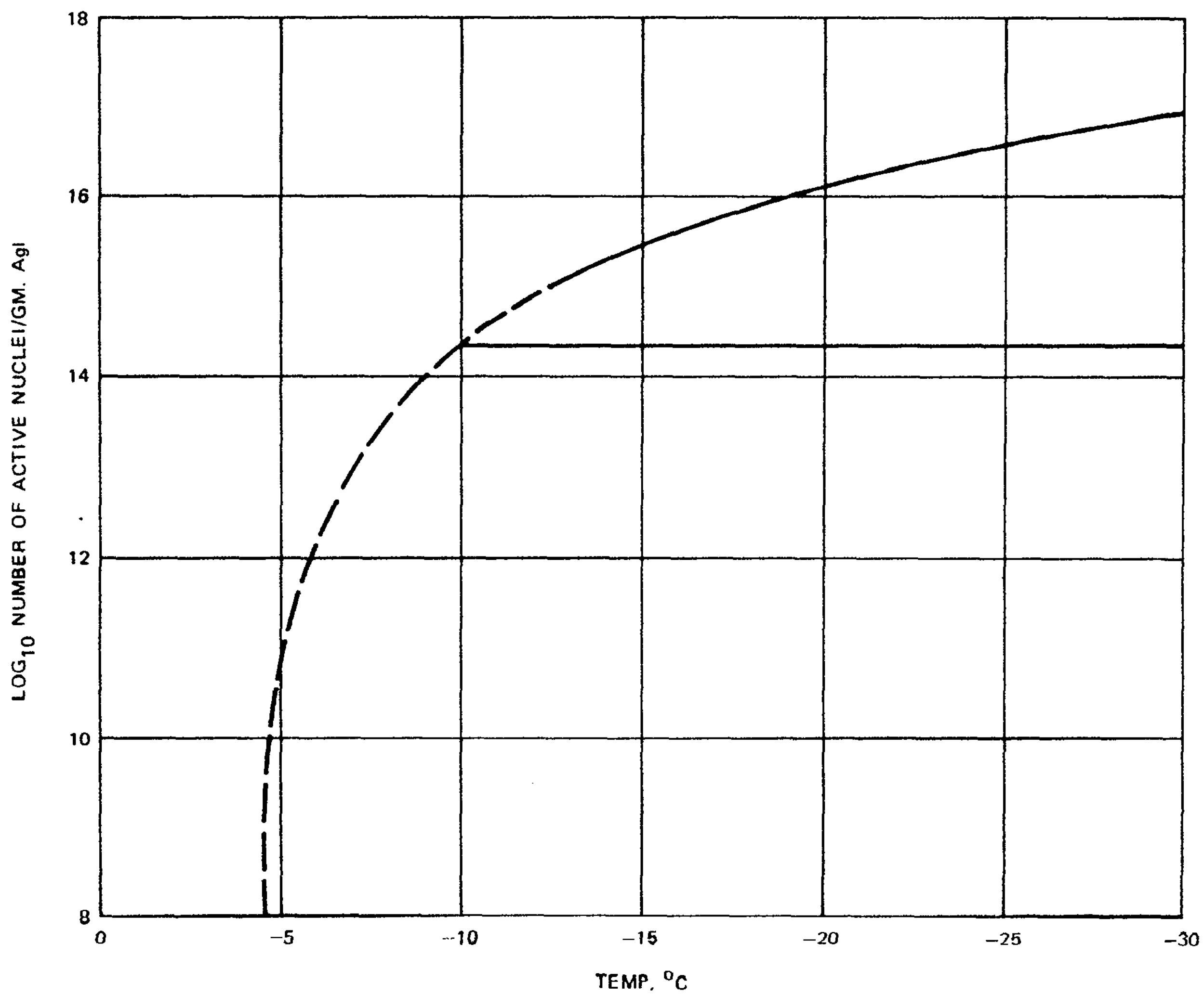


Fig. 8. Fletcher's curve for estimating the maximum number of AgI particles capable of producing sublimation, counting one nucleation event per sec. per particle. The solid line at $-10^{0}C$ gives the activity curve for a monodispersed smoke capable of inducing sublimation at $-10^{0}C$. Thus for such an agent 10^{14} nuclei will be produced at $-10^{0}C$ and only that number at all colder temperatures.

necessary to produce a curve of any given shape. By studying published ice nuclei activity curves, it soon becomes apparent that amounts far in excess of one gram are represented by the area under most of the curves. This means that some mechanism other than sublimation is working, that the bulk density chosen for the nucleation material is wrong, or that nucleation events requiring more than one second are being counted.

If sublimation is the important process, one cannot have a material with a high numerical effectiveness per gram and still expect it to work at the warmer temperatures. For purposes of cloud seeding for precipitation, we have found that nucleants active in the -5°C region are more effective than the numerically superior smokes that have a high yield in the -20°C regime. For many years, it was felt that measurements at -20°C were the most important indicators and that every attempt should be made to maximize this number. This can only be done at the expense of effectiveness at the warmer temperatures. Thus by trying to overmaximize the system, the overall effectiveness may be degraded.

Figure 9 shows our version of the Fletcher type curve. The usual curve for sublimation is shown, dashed in the regions where it will not be applicable. The novelty herein lies in the addition of curves for freezing. This curve is similar to that shown by Fletcher (1968, 1970) except that this argument, in those references, is applied to the AgI-NaI complex produced by acetone burners and is probably valid for pure AgI but not for the complex. Our curve is dashed at temperatures colder than -12.5°C and applies only to agents contamined with very small amounts of a hygroscopic material.

The solution offered by the Fletcher theory for any of the nucleation processes is strongly dependent upon the various surface free energies. For condensation and for sublimation it is reasonably clear that the value of m used, about 0.95, is probably correct. The actual value will depend to a great extent on the material. For freezing, once liquid water is present on the substrate, the matter is not so clear.

In the first case, differences between the surface free energy of the nucleating particle and the liquid, the nucleating particle and the vapor, and the liquid and vapor can be estimated from the contact angle. In the second case, the difference in surface free energy between the liquid, the solid and the substrate are more difficult to estimate. The differences in surface free energy between the solid and the vapor are about 5 times that between the solid and the liquid. Hence, it is much easier for liquid water to freeze than to sublimate.

In actual fact, Fletcher in all his work, simply uses -4.5°C as the maximum temperature at which AgI will produce freezing. Use of this value assumes a value of m for the processes. The same value of m cannot be applicable for condensation, sublimation or freezing, because in each case the free energy relations are different.

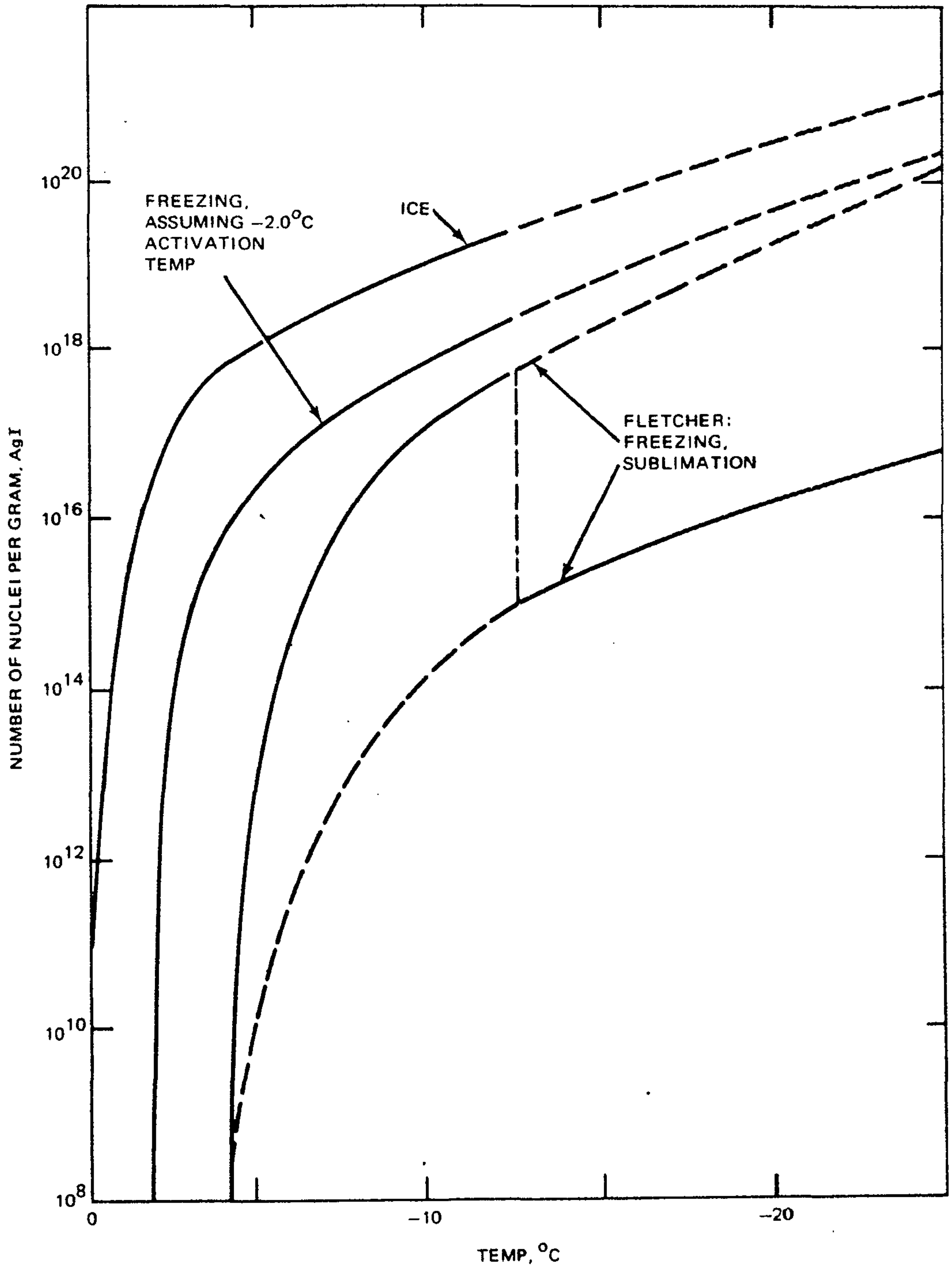


Fig. 9. Curves similar to Fig. 8 for AgI assuming -4.5°C activation temperature for freezing and for sublimation and assuming -2.0°C activation temperature for freezing.

At this point the epitactical theory and Fletcher's effort come together. The asymptote, determined by a particular value of m on the left hand side of Figures 5, 6, and 7 is actually the activation temperature, and so m is determined by this value without detailed knowledge of the surface free energies.

In view of the earlier discussion of the activation temperature for AgI, we feel that the use of -2.0°C is justified, this is equivalent to an m of 0.98 and is applicable to the freezing processes. On Figure 9 we show a curve for freezing based on this value. This indicates that particles of 0.05 microns radius are active at -5°C and particles of 0.01 microns radius at -10°C.

On basis of Fig. 7 we recalculate the curves of Figure 9 and add a third curve to show the expectable yield of AgI when completely surrounded by water. In actual practice, as contact nucleant, the spherical particle of AgI will not completely touch the water surface and only a part of its surface will be available as a nucleant. Hence in practice, the new curve in Figure 9 will be a maximum and all real nucleants will function somewhat less effectively. The exact difference will depend upon the size of the particle and the size of the drop as well as upon the temperature and upon orientation in the gravitational field. While we will not try to calculate the effects at this time, it seems as if, for a given particle size that large drops will nucleate more easily than small drops and that the new curve in Figure 9 for contact nucleation will become perhaps an order of magnitude lower and less dependent upon temperature.

The matter has been summed up by Fletcher in the three-axis diagram shown in Figure 10. He here assumes a material similar to silver iodide but not necessarily AgI. The condensation surface is placed at a slight supersaturation so that in the regimes of saturation found in nature the condensational or the freezing phase is not likely to be activated. The space in which the sublimation regime will function intersects the condensation regime at -10°C. According to this diagram, now rendered obsolete by his most recent paper (Fletcher, 1969), the only regime in which pure, nonadsorbent AgI will work is the wedge-shaped sublimation space and the maximum temperature at which it will work is -12.5°C, because saturation ratios in nature rarely exceed 1.0.

Figure 11 shows a more up-to-date version, superseding ours of 1966 (St.-Amand, 1967). The chief difference is that the space in which condensation occurs is larger, the almost vertical condensation surface is at the saturation ratio of 1.0 instead of 1.05 and the size particle capable of producing freezing is now shown to be smaller. We assume that the AgI is capable of adsorption, or that it has been treated with a nonreacting vapor pressure suppressant. Initially the embryo growth is possible by sorption and will occur at low relative humidities; the surface for condensation being actually moved forward on the diagram until an embryo of water or ice is formed, the surface gradually being

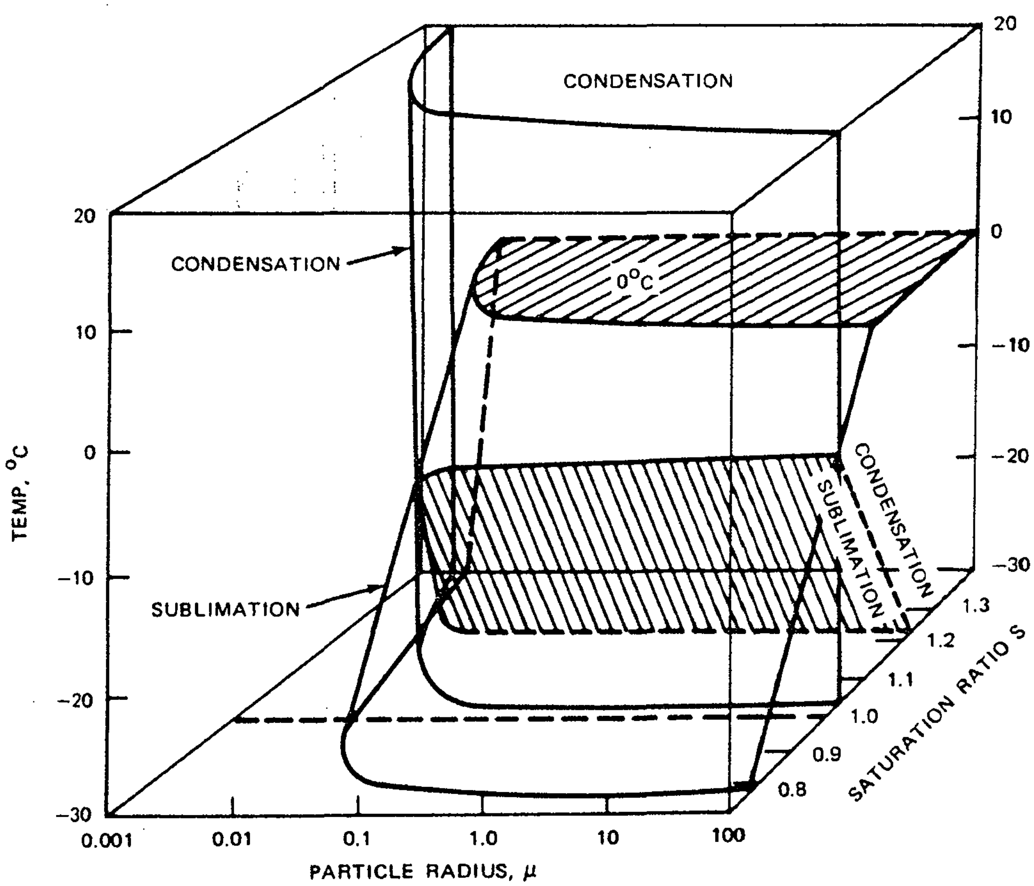


Fig. 10. Fletcher's three-dimensional diagram showing the relation between condensation, freezing, and sublimation as a function of saturation ratio, temperature, and particle size.

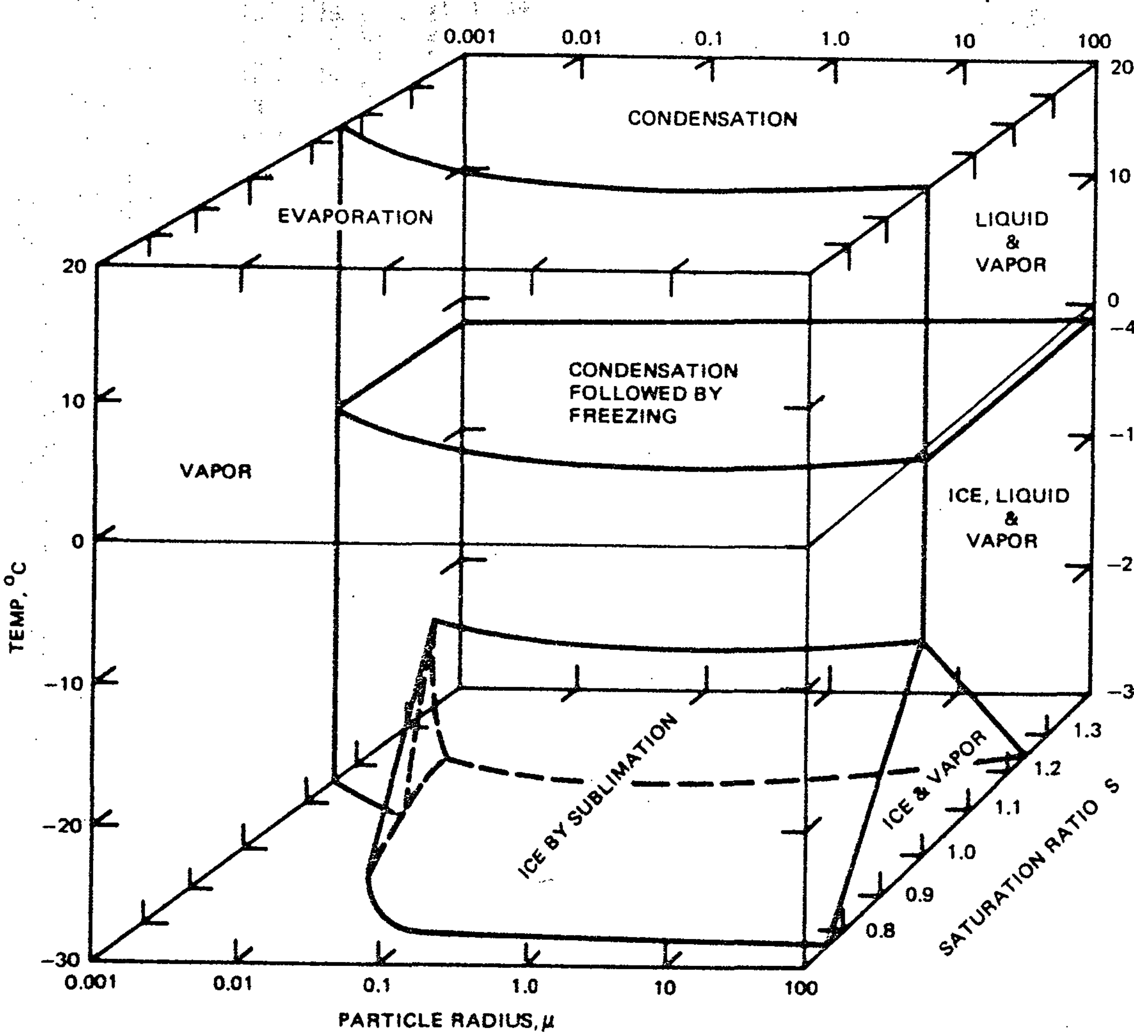


Fig. 11. Variation of Fletcher's three-dimensional diagram showing the relation between condensation, freezing, and sublimation as a function of saturation ratio, temperature, and particle size. This differs from Fletcher's primarily in that the condensation surface begins at a saturation ratio of unity rather than 1.05.

moved back until it is in the position shown. The intersection of the sublimation-condensation regime is now at -12.5°C. We will show experimental curves later that seem to apply to this matter.

NUMERICAL EFFECTIVENESS DATA FOR SOME PYROTECHNICS:

Figure 12 shows the effectiveness curve for two types of pyrotechnic generators of AgI. The data were determined by Colorado State University and are for the Alecto units used in Project Stormfury and elsewhere. There are two types of materials represented, CY 21 and Cy 35. These are described in Vetter, et al. (1970). The curves are interesting, because both show that there is a very linear portion of the curve between -5°C and -12.5°C. At the latter temperature, the numerical efficiency for CY 21 drops markedly by about two orders of magnitude, and the curve then begins to rise again, reaching about 10^{15} nuclei per gram at -20°C. This is a result not to be expected from simple application of Fletcher's theory. We feel that the difference in behavior is caused by a change from condensation-freezing reactions in the warmer portion to a purely sublimation mechanism in the colder portion. It may be an artifact to the nuclei counting process, but neither the people at CSU nor ourselves have been able to figure out what it might be. These materials were tested by burning full-scale units in a wind tunnel that very effectively removed all the electrically charged particles or else discharged them. The aerosol was quite dilute and the fog tenuous, rendering contact nucleation less rapid than the diffusion processes. No measurements were made of the saturation ratios. Thus, the curve represents perhaps, nucleation mainly by the diffusional processes. A similar effect has been found by Davis (1970), using an NaI-AgI complex.

Figure 13 shows some results of testing LW83, an agent that produces smoke similar to that of Electo, CY 21.

The curve by Donnan (1970) is for a liquid water content of 3 grams/m^3 and was made in his multibox facility. This arrangement consists of dilution by a wind tunnel followed by aspiration into 5 cold boxes, each of which is run at a different temperature. He uses a droplet fog made by a nebulizer and maintains careful control of LWC.

The curve by Odencrantz is from burning small amounts of material in his walk-in refrigerator. He uses a water cloud made from condensation of steam. No control of LWC is attempted.

The curve by Grant was made at CSU in their isothermal chamber, before it was moved to its new location. Material was diluted by a wind tunnel, sampled by a syringe and deposited into the cold chamber, LWC is not known.

The curve by Steele was produced in the same way after the device was moved. LWC for this test is not known.

The curve by Grant shows the same inflection at -12.5 as do his

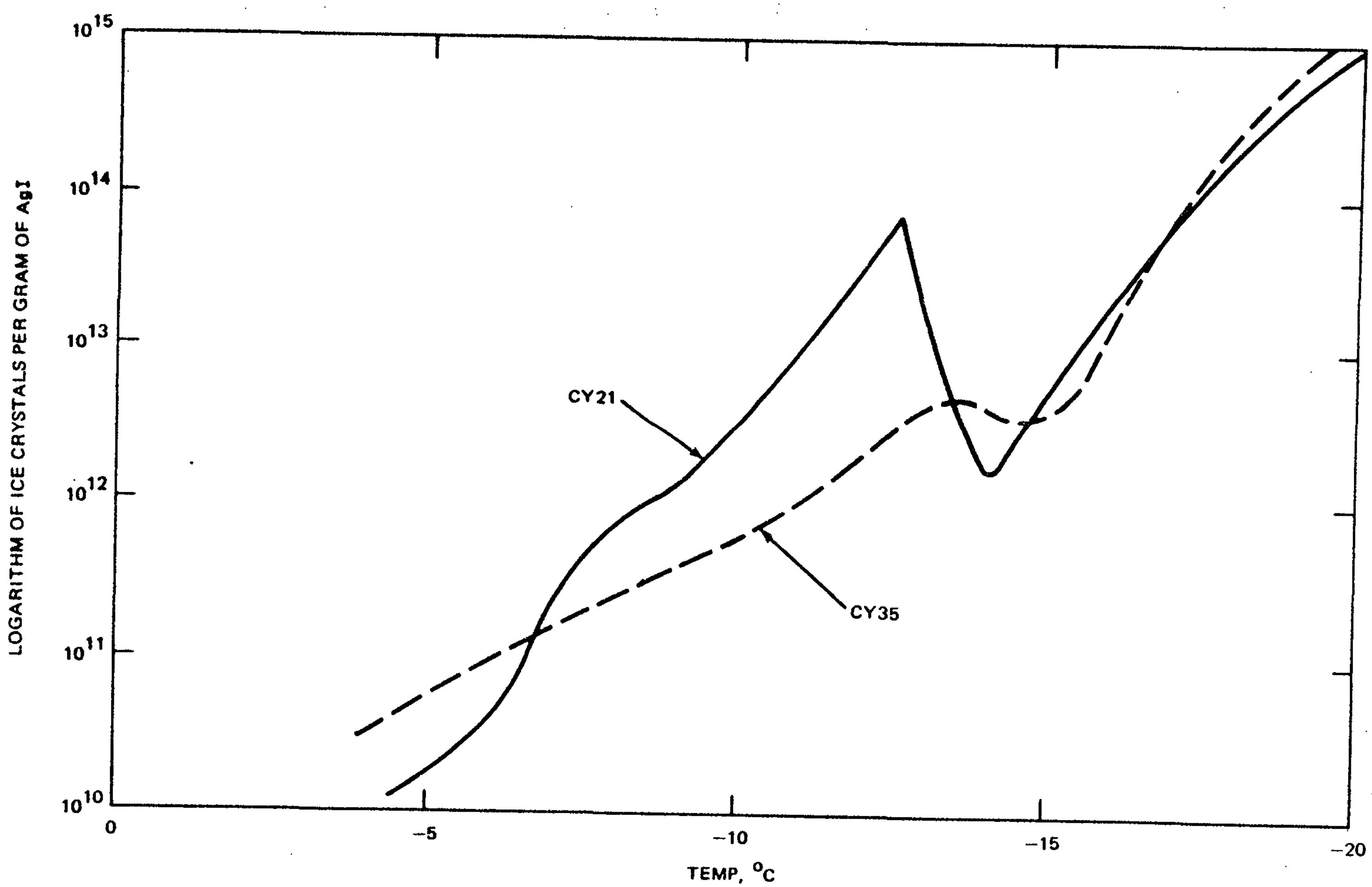


Fig. 12. Nucleation effectiveness curves for two propellant type seeding agents.

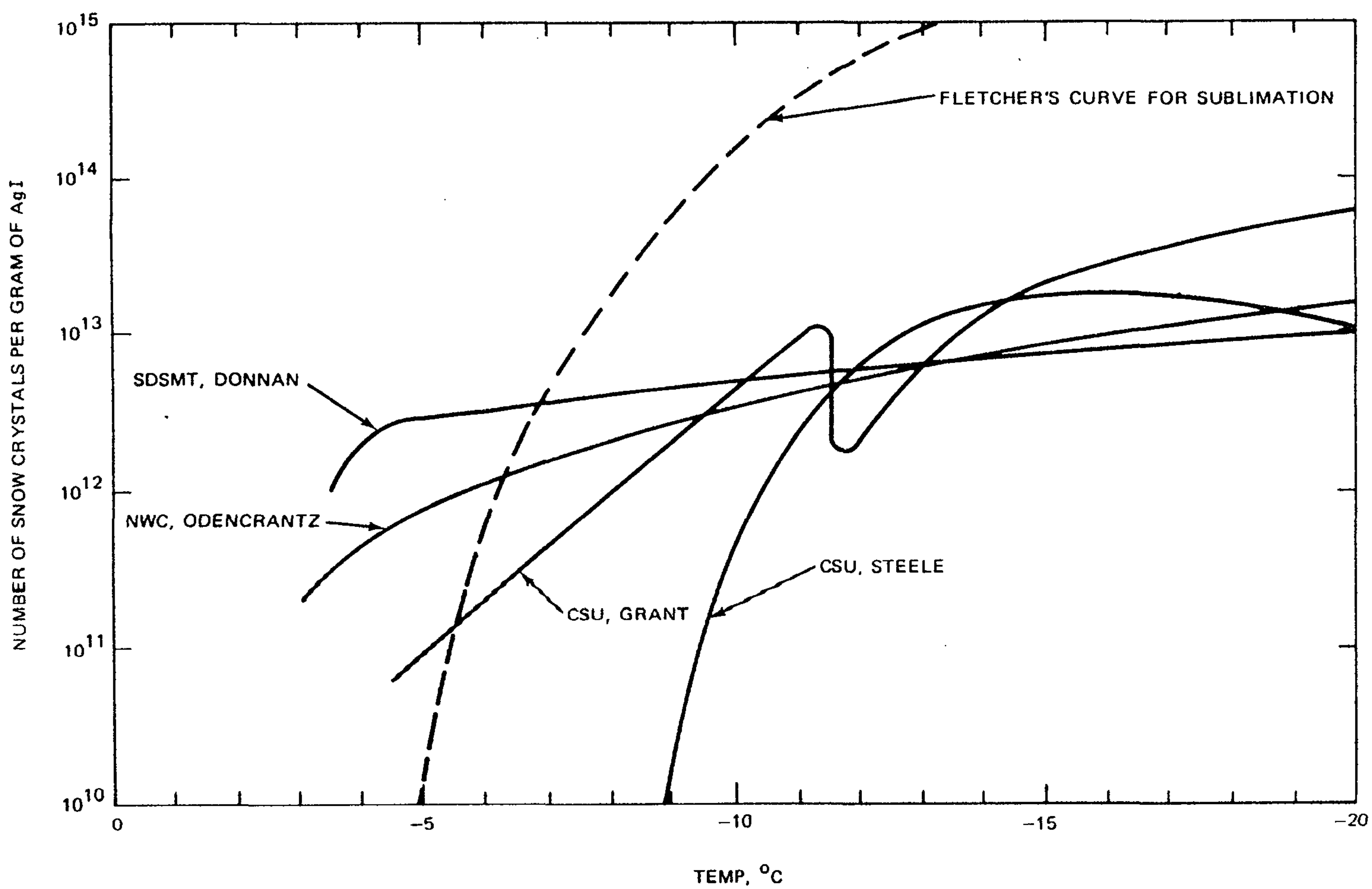


Fig. 13. Activity curves for LW 83, a pyrotechnic producing a relatively pure AgI.

curves for CY 21 and CY 35. This type of curve has been obtained for AgI in nitrasol and in gun cotton. In all cases a variable amount of KI has been present, usually less than 3% of the weight of the AgI.

Within the limits of measurement, the results from -15^{o}C to -20^{o}C are remarkably similar, but at the warmer temperatures it is clear that the data presented by Steele are totally different from the others. Because the cause of this difference is not now known it is difficult to decide who is correct, if that is the proper expression.

The numerical effectiveness derived by Donnan, Odencrantz and Grant is clearly much greater than we should be getting from sublimation and is in line with what should be deducible from contact nucleation of droplets or from condensation-freezing of vapor. Some light might be thrown upon the situation by an examination of the differnt techniques in cold box determination of nuclei counts.

The theory of nucleation is at present adequate for decision making in planning experiments. There remains a gap of several orders of magnitude between theory and cold box measurements. This will be the subject of the next paper in this series.

Effects of Dry Ice and Compressed Air Seeding: It seems worthwhile to discuss to some extent the effects of dry ice and compressed air seeding in the induction of precipitation. It has been recognized for decades that dry ice can be used to induce the ice phase formation in clouds. It does this by cooling the surrounding air by expansion of the carbon dioxide gas from the evaporating dry ice. The air is cooled so much that homogenous nucleation, or nucleation upon materials not capable of producing freezing at any reasonable temperature occurs. The temperature of the air immediately adjacent to the dry ice is reduced to the neighborhood of about -70^{o}C. The water vapor in a given volume of such air collects into ice embryos which continue to grow in the very cold region but which perhaps find survival difficult in the less hospitable warmer air remaining once the dry ice has been removed. Thus, it has been shown that the number of nuclei produced by dry ice is high until temperatures around -10^{o}C are encountered, Eadie and Mee (1963). Thereupon at warmer temperatures, the effectiveness drops off rapidly. Ice possesses a coefficient of expansion and ice produced by extremely cold temperatures will not have the same lattice structure as the ice at the tmperature of the rest of the air. This means that, within the nomenclature of Fletcher's theory, the embryo will have a value of m less than unity and condensation cannot take place upon it until such time as it reaches the temperature of the ambient air. By the time this occurs, a large percentage of these embryos will certainly have evaporated because they are too small to survive in the ambient air. Once again, only a very few droplets will be frozen in comparison to the large numbers of smaller ice embryos and so growth will have to occur on the embryos by diffusional processes and will be slower. Thus, dry ice will not work as well as or as rapidly in inducing precipitation as will the other agents such as silver iodide. There too, the best effects of the dry ice will occur only at the coldest temperatures, taking the major part of the water in those regions out of cirulation and removing it from any possible reaction involving precipitation.

Dry ice may well work to induce precipitation if it is used as an optimal situation. This would seem to be one where it was dropped in far greater quantities than is ordinarily used in the portion of the cloud below about the -10°C level.

REFERENCES

Aufmkampe, H. J. and H. K. Weikmann, "The effectiveness of natural and artificial aerosols as freezing nuclei," J METEOROL, Vol. 12, (1951), pp. 68-73.

Birstein, S. J., "The Effect of Relative Humidity on the Nucleating Properties of Photolyzed Silver Iodide", BULL METEOROL SOC., Vol. 33 No. 10 (1952), pp. 431-434.

________. "The Role of Adsorption in Heteorogeneous Nucleation, I: Adsorption of Water Vapor on Silver Iodide and Lead Iodide," J METEOROL, Vol. 12 (1955), pp. 324-331.

________. "The Role of Adsorption in Heteorogeneous Nucleation, II: The ADSORPTION OF Water Vapor on Photolyzed Silver Iodide," J METEOROL. Vol. 13 (1956), pp. 395-398.

Byers, Horace R., "Nucleation in the Atmosphere," Third Nucleation Symposium, IND ENG CHEM, Vol. 57, No. 11 (1965), pp. 32-40.

Corrin, M. L., Harry W. Edwards and John A. Nelson, "The Surface Chemistry of Condesnation Nuclei: II. The Preparation of Silver Iodide Free of Hygroscopic Impurities and Its Interaction with Water Vapor," J ATMOS SCI. Vol. 21 (Sept 1964), pp. 565-567.

Corrin, M. L. and N. S. Storm, "The surface chemistry of condensation nuclei: I. The Sintering of Silver Iodide," J PHYS CHEM, Vol. 67 (1963), pp. 1509-1511.

Coulter, L. V., "A Preliminary report on the adsorption of water vapor on silver iodide," Presented at a meeting of the Bunsen Gesellschaft, Berlin, 1952.

Cuilong, B. M., "Sublimation in a Wilson Chamber," ROY SOC LOND, PROC, SER A, Vol. 190 (1947), pp. 137-143.

Davis, B. L., and D. N. Blair, "Role of Substrate Strain in Ice Nucleation," J GEOPHYS RES, Vol. 74 (1969), pp. 4571-4580.

Davis, M. H., J. D. Klett, and M. Neiburger, "Collision Efficiencies of Cloud Droplets at Small Reynolds Numbers," Conference on Cloud Physics, PREPRINTS, Ft. Collins, Colo. (Aug 24-27, 1970), pp. 115-116

Donnan, J. A., Donald N. Blair, William G. Finnegan, and Pierre St.-Amand, "Nucleation Efficiencies of AgI-NH_4I and AgI-NaI Acetone Solutions and Pyrotechnic Generators as a Function of LWC and Generator Flame Temperature, A Preliminary Report," J WEA MOD, Vol.II, No. 1 (May 1970), pp. 155-164.

Eadie, W. J. and T. R. Mee, "The effect of dry ice pellet velocity on the generation of ice crystals," J APPL. METEOROL, Vol. 2, (1963) pp. 260-265.

Edwards, G. R., and L. E. Evans, "Ice Nucleation by Silver Iodide: I. Freezing vs Sublimation," J METEOROL, Vol. 17 (1960), pp. 627-634.

Fletcher, N.H., "Size Effect in Heterogeneous Nucleation," J CHEM PHYS, Vol. 29, No. 3 (1958a), pp. 572-576.

________. "Erratum: Size Effect in Heterogeneous Nucleation (J CHEM PHYS, Vol. 29 (1958), p. 572) J CHEM PHYS, Vol. 31 (1958b) pp. 1136-1137.

________. "On Ice-Crystal Production by Aerosol Particles," J METEOROL, Vol. 16 (1959a), pp. 173-180.

________. "A Descriptive Theory of Photo De-activation of Silver Iodide as an Ice-Crystal Nucleus," J METEOROL, Vol. 16 (1959b), pp. 249-255.

________. "The Optimum Performance of Silver-Iodide Smoke Generators," J METEOROL. Vol. 16 (1959c), pp. 385-387.

________. "The Physics of Rainclouds", Cambridge University Press, 1962, 386 pp.

________. "Nucleation by Crystalline Particles," J CHEM PHYS, Vol. 38, No. 1 (Jan. 1963), pp. 237-240.

________. "Ice Nucleation Behavior of Silver Iodide Smokes Containing a Soluble Component," J ATMOS SCI, Vol.25 (1968), pp. 1058-1060.

________. "Physical Basis of Ice Nucleation: Developments Since 1960," Second National Conference on Weather Modification, PREPRINTS, Santa Barbara, Cal., (April 6-9, 1970), pp. 320-324.

Fukuta, Norihoko, "Experimental investigations on the ice forming ability of various chemical substances," J METEOROL.. Vol. 15 (Feb. 1958), pp. 17-26.

________. "Review of Physics of Ice Nucleation and Its Application to Weather Modification," Prepared for Bureau of Reclamation, Office of Atmospheric Water Resources, Denver, Colo., by MRI, Altadena, Calif. P.O. 7-D-3053 and P.O. 7-D-4133 (May 8, 1967) pp. 4-55.

Hosler, C. L., "On the Crystallization of Supercooled Clouds," J METEOROL Vol. 8 (1951), pp. 326-331.

Isono, K. and Y. Ishizaka, "On nucleation properties of different faces of silver iodide crystals," Jour. de Recherches Atmospherique, Vol. III, 2^{o} annee, No. 1 and 2 (Jan-June 1968), pp. 139-140.

Karaz, F. E., W. M. Champion, and G. D. Halsey, "The growth of ice layers on the surfaces of anastase and silver iodide," J PHYS CHEM, Vol. 60, (1956) pp. 376-378.

Long, Alexis B., "A Thermodyanmic Study of Homogeneous Nucleation," REV GEOPHYS, Vol. 7, No. 3 (Aug 1969), pp. 595-621.

Mason, B. J. and J. Hallett, "Artificial Ice-forming Nuclei," NATURE, Vol. 177, No. 4511 (1956), pp 681-683.

Mason, B. J. and A. P. van Den Heuvel, "The Properties and Behavior of Some Artificial Ice Nuclei," PHYS SOC PROC, Vol. 74 (July to Dec 1959), pp 744-755.

Odencrantz, F. Kirk, "Freezing of Water Droplets: Nucleation Efficiency at Temperatures above $-5^{o}C$," J APPL METEOROL, Vol. 8, No. 3 (1969) pp 322-325.

St. Amand, Pierre, "Nucleation by Silver Iodide and Similar Materials," Skywater Conference I, PROC, Bureau of Reclamation, Denver, Colo. (1967), pp. 305-346.

Schaefer, V. J., "Ther formation of ice crystals in the laboratory and the atmosphere," CHEM REV, Vol. 44 (1949), pp. 291-320.

________, "Silver and Lead Iodides as Ice-Crystal Nuclei," J METEOROL, Vol. 11 (1954), pp. 417-419.

Thompkins, L. M., D. A. Muus, and T. Pearson, "Water Adsorption in the system AgI-KI-H_2O," J GEOPHYS RES, Vol. 68 (1963), pp. 3537-3539.

Turnbull, David, and Bernard Vonnegut, "Nucleation Catalysis," IND ENG CHEM, Vol. 44, no. 6 (1952), pp. 1292-1298.

Vetter, Ronald F., William G. Finnegan, Lohr A. Burkardt, Pierre St.-Amand, H. Sampson, and Martin Kaufman, "Pyrotechnic Production of Nucleants for Cloud Seeding, Part III. Propellant Compositions for Generation of Silver Iodide," J WEA MOD, Vol. 2, No. 1 (May 1970), pp. 53-64.

Zettlemoyer, Albert C., Noubar Tcheurekdean, and Charles L. Hosler, "Ice Nucleation by Hydrophobic Substrates," Z ANGEW MATH PHYS, Vol. 14 (1963), pp. 496-502.

UNDERSTANDING OF THE USE OF SIMPLE AND COMPLEX ICE NUCLEI GENERATED FROM PYROTECHNICS AND ACETONE BURNERS

P. St.-Amand
W. G. Finnegan
L. Burkhardt

Naval Weapons Center
China Lake, California

ABSTRACT

Silver iodide is solubilized in acetone by formation of complexes with iodides of the alkali metals and ammonia. The solution is burned yielding AgI and/or complexes of AgI and the alkali metals. Use of NH_4I as solubilizing agent results in production of pure AgI. Use of the alkali iodides results in a series of complexes of AgI and the alkali iodides.

Formulations for the NH_4I-2AgI complex in acetone are given.

Pyrotechnic formulations using the complexes are discussed and methods of cloud seeding utilizing the complexes are outlined based on the behavior of the materials following exposure to moist air.

Many cloud seeding experiments using alkali iodides and silver iodide lead to marginal or negative results because the clouds were not seeded at temperatures high enough to affect the clouds to a greater extent than the naturally occurring nuclei.

INTRODUCTION

Acetone burners are valuable devices for the production of nuclei and have an important place in cloud seeding. The exhaust products may or may not be silver iodide, however, and an understanding of the chemistry of the nucleant is important in deciding how to use the devices.

In his original attempts to produce silver iodide, Vonnegut (1949, 1950) made use of the fact that silver iodide could be made to dissolve in a mixture of acetone and alkali iodides or ammonium iodide. He originally used ammonium iodide as a solubilizing agent, Vonnegut (1949), and later, for reasons that are not clear, chose to use sodium iodide instead (Vonnegut, 1950). This change, however simple, is responsible for a good deal of the confusion regarding the effectiveness of cloud seeding.

A wide variety of devices were subsequently invented that use the same principle, these include strings, wicks, tissue paper, charcoal and similar materials impregnated, dried and burned. The arguments that follow are applicable to any or all of these.

Silver iodide is readily soluble only in solvents such as ammonia or the aliphatic amines. When mixed with iodides of ammonia or the alkali metals, it forms complexes soluble in acetone and to some extent in water. These are not solutions in the sense that material exists as ions of silver and iodine, but rather as solvated complexes. The chemistry of silver iodide-potassium iodide complexes was discussed in 1883 by Berthelot.

COMPLEXES:

While the following discussion is not applicable to acetone solutions in its entirety, owing to the formation of acetonates and complexes with acetone, it is included to shed some light on the formation of complexes. *Sensu-strictu* we should be speaking of coordinate-bonded substances in this connection instead of complexes, but for purposes of clarification we will retain the already established nomenclature.

A simple polar compound dissolved in water ionizes to some extent. The concentration of the ions is expressed in moles per liter. If the product of the concentration of any two ions exceeds a value known as the solubility product constant, precipitation of those ions, as a compound of the two materials occurs, and material is precipitated until the product of the concentrations of those two ions is equal to the solubility product constant. As an example, if a solution of NaCl is at or below saturation, NaCl may be precipitated by adding HCl to increase the concentration of the chloride ion. If NaCl and KCl are in solution together and the solution be evaporated until the solubility product constant for Na+ and Cl- is reached, NaCl precipitates, leaving KCl in solution. This is known as the common ion effect, a corollary of LeChatliers principle. It is widely used in industrial processes.

The solubility of AgI in water is low. The solubility product constant is 1×10^{-16} moles ^{2}/liter^2. Any time the product of the concentrations of Ag^+ and I^- exceeds this value, AgI is precipitated.

Silver, in common with a number of other elements, forms complexes. A complex molecule is one which contains more than two different atoms. Sulphuric acid, H_2SO_4, is made up of H_2 and SO_4. It ionizes to H+ and $SO_4^=$. The $SO_4^=$ is a complex ion. It dissociates but little, if at all, into its component elements. Thus while the sulphur and oxygen are united in $SO_4^=$, no appreciable amount of sulphur or oxygen ions are found in the solution.

Silver forms complexes readily with any alkali metal, iodides and with ammonia. If a solution of AgI is inoculated with KI one might expect the common ion reaction to cause AgI to precipitate. On the other hand, more dissolves. This apparent contradiction to the common ion effect is brought about because a new material is formed taking the silver ions out of the solution. The new material may be in a number of combinations of Ag, I and K.

Burkardt, et al., 1970, gives a long list of such complexes.

For example, if we let RI be any compound with which AgI can complex, a series of complexes can form.

$$nRI + mAgI \rightarrow Ag_mR_nI_{(m+n)}$$

Ionically

$$nR^+ + nI^- + mAg^+ + mI^- \rightarrow$$

$$Ag_mR_n^{++} + (n+m)I^-$$

The ionizations may also result in

$$Ag_mR_nI_o^{+} + I^-$$

The only effect that need concern us here is that the concentration of the iodine ion can increase, with a concomitant loss of the Ag^+ ion because it is now coordinate bonded with R and/or I. The new species is more stable in solution than the AgI, and therefore, the otherwise insoluble AgI dissolves with reaction to produce the new species. AgI is thus not precipitated but is retained in the liquid, bound up in the new form.

The stability of the new form is dependent upon the concentration of the iodide ion. If the iodide ions be removed from a solution of one of these complexes or if the concentration be reduced by dilution, the complex breaks down in part or in whole, and AgI is again precipitated.

METHOD OF FUNCTIONING OF ACETONE BURNERS

In operation, the acetone solution is atomized by a nozzle and then burned, either in air alone or in a flame such as propane. The droplets evaporate, leaving behind a sintered residue of the unburnable material dissolved in the droplets (St.-Amand, 1967). The evaporation of the acetone cools the particles to some extent, and until the acetone is consumed, the particles remain cooler than the flame. If the flame temperature is too high some of the solid constituents may vaporize and later recondense producing a much finer aerosol. This may be disadvantageous in that the very small particles produced in this way will not function as freezing nuclei at the warmer temperatures.

If the flame is too hot, and/or underoxidized, some of the iodine compounds may be reduced with formation of free iodine, silver, hydrogen iodide and similar products. The elemental halogens and elemental silver recombine but incompletely and for this reason maintaining proper stoichiometry is very important in that potentially useful material is wasted.

TABLE 1, PHYSICAL PROPERTIES OF AgI, NaI and NH_4I

Material	Mol. Weight	Density	Melt Pt.	BP
AgI Hex	234.8	5.68	558	1506
AgI Cu	234.8	6.01		
NH_4I	144.9	2.51	551 sublimates	
Na I	149.9	3.67	651	1304
K I	166.0	3.13	683	1330

An evaluation of the importance of flame temperature can be determined by simple calculation. TABLE 1 gives some of the physical properties of the various compounds to be considered in this discussion. The density of an acetone solution of NaI·2AgI with no water is given in TABLE 2. To two significant figures, these values can be used for all of the complexes discussed here.

Assume that a nozzle in an acetone burner makes droplets of solution of radius, r_s. The mass of solution in each droplet, M_s, is

$$M = \frac{4}{3} \pi \rho_s r_s^3 \qquad (1)$$

where ρ_s is the density of the solution. The mass of AgI in the droplet is

$$M_A = \frac{4}{3} \pi \rho_s r_s^3 C \qquad (2)$$

where C is the fractional concentration by weight of the AgI.

The volume of AgI, V_A in each drop is

$$V_A = \frac{4\pi}{3} r3\rho_s C/\rho_A \qquad (3)$$

where ρ_A is the density of AgI.

The radius, r_A, of this volume of AgI, if the acetone were evaporated, and the AgI melted without loss of mass, would be

$$r_A = r_s \left(C \frac{\rho_s}{\rho_A} \right)^{\frac{1}{3}} \quad (4)$$

TABLE 2 gives values of ρ_s and $\left(C\frac{\rho_s}{\rho_A}\right)^{\frac{1}{3}}$ for several concentrations of AgI in acetone.

TABLE 2

Solution density ρ_s and ratio of AgI particle radius to original drop radius as function of concentration of solution

C	ρ_s	$\left[\frac{\rho_s}{\rho_A} C\right]^{\frac{1}{3}}$
0.0 x 10	7.8×10^{-1}	1.12×10^{-1}
1.0×10^{-2}	8.0×10^{-1}	1.12×10^{-1}
5.0×10^{-2}	8.5×10^{-1}	1.96×10^{-1}
1.0×10^{-1}	9.2×10^{-1}	2.52×10^{-1}
1.5×10^{-1}	1.0×10^{0}	2.98×10^{-1}
2.0×10^{-1}	1.1×10^{0}	3.40×10^{-1}

TABLE 3

Radius of AgI particles as function of Concentration and solution droplet radius

C %	Radius of solution droplet, microns		
	1	10	100
0.1	5.17×10^{-2}	5.17×10^{-1}	5.17×10^{0}
1	1.12×10^{-1}	1.12×10^{0}	1.12×10^{1}
5	1.96×10^{-1}	1.96×10^{0}	1.96×10^{1}
10	2.52×10^{-1}	2.52×10^{0}	2.52×10
15	2.98×1	2.98×10^{0}	2.98×10
20	3.40×10^{-1}	3.40×10^{0}	3.40×10

TABLE 4

Mass of AgI particles from Table 3 , grams

C %	Radius, r , of solution drops		
	1	10	100
0.1	5.80×10^{-10}		
1	7.95×10^{-15}	7.95×10^{-12}	7.95×10^{-6}
5	4.28×10^{-14}	4.28×10^{-11}	4.28×10^{-5}
10	9.09×10^{-14}	9.09×10^{-11}	9.09×10^{-5}
15	1.50×10^{-13}	1.50×10^{-10}	1.50×10^{-4}
20	2.23×10^{-13}	2.23×10^{-10}	2.23×10^{-4}

TABLE 5

Particles of AgI/gm

C %	Radius, r, of solution drops 1	10	100
0.1	1.73×10^{15}	1.73×10^{12}	1.73×10^{6}
1	1.26×10^{14}	1.26×10^{11}	$1.26 \times 10^{+5}$
5	2.34×10^{13}	2.34×10^{10}	2.34×10^{4}
10	1.10×10^{13}	1.10×10^{10}	1.10×10^{4}
15	6.66×10^{12}	6.66×10^{9}	6.66×10^{3}
20	4.50×10^{-12}	4.50×10^{9}	4.50×10^{3}

Tables 3, 4, and 5 give the size mass and number of the AgI particles that would be produced by uniform droplets if they were in no way broken up in the flame. Most nozzles so used produce droplets in the range of 10 to 100 micron. Therefore, it seems certain that some droplet break-up occurs, that some AgI is evaporated and recondensed, or that the droplets in the 1 to 10 micron range produce most of the nuclei.

Because the NH_4I-AgI-acetone system yields pure AgI as an output and is simplest, we will begin with that one.

Use of Ammonium Iodide as Solvation Agent for Silver Iodide: Relatively pure AgI may be produced with acetone burners by reverting to the formulation originally given by Vonnegut (1950). This uses ammonium iodide as a solvation agent.

During combustion at any reasonable flame temperature, the NH_4I is first sublimed (at about 554°C) and then destroyed, yielding nitrogen, water and free iodine, together with HI and a few other species involving combinations of O, C, H, N and I. The exact nature of the products depends on the flame temperature and degree of oxidation. In properly oxidized flame, the presence of the iodine vapor resulting from the destruction of NH_4I invokes the common ion effect and protects the AgI from disassociation. The result is that a nucleating product, identified by Burkardt

(1970) as relatively pure AgI, is produced.

Studies are going forward to optimize the concentration of solution, flame temperature and degree of oxidation. At present, it seems as if a flame temperature in the vicinity of 1200°F and 15% solution will give excellent results at temperatures near -5°C. The addition of KI in the amount of 1 to 3 percent of the weight of the AgI may aid in increasing the response at temperatures between -2°C and -5°C, with little change at lower temperatures.

The use of ammonium iodide as a solubilizing agent in acetone burners yields a glacogene as good or better than can be obtained with pyrotechnics. The exhaust product, being uncomplexed AgI will not be deteriorated by atmospheric water vapor, nor should immersion of the particles in cloud droplets impede their effectiveness, although some of the smaller will dissolve given enough time. This means that the activity spectrum will not markedly deteriorate at the higher temperatures following introduction below the freezing level.

The NH_4I-2AgI complex can be made up as a 25% stock solution and then be diluted with acetone to any desired strength (Donnan, et al 1970).

A ten percent solution can also be made according to TABLE 6.

TABLE 6

Material	Percent by weight	Weight		Volume
		pounds	grams	
AgI	10.0	3.86	1,750	
NH_4I	3.1	1.11	540	
H_2O	3.0	1.10	500	500 cm^3
Acetone	84.5	33.10	15,000	5 gals

This gives 5.1 gallons of solution.

Mixing is simple. Five hundred cm^3 of water, heated to 150-180°C, is used to dissolve the NH_4I. This produces a clear, light brown solution of 700 cm^3 volume. The AgI is then placed in suspension in the acetone by vigorous stirring at room temperature. When a uniform milky, yellowish suspension is formed, add the NH_4I solution rapidly and stir until a clear, transparent light brown solution is formed with no residual AgI. This usually takes 3 to 5 minutes. The material may be indefinitely in an air tight glass or plastic gasoline jug. It is highly flammable and should be stored away from open flames, bright light and sunlight.

The material is corrosive to iron, steel and galvanized surfaces. It may be corrosive to brass, copper and cadmium on long exposure. It does not corrode aluminum or stainless steel. Care should be used to keep iron salts and rust out of the solution. The solution will soften many plastics and rubber, but does not seem to affect polyethylene.

Solutions up to 25 percent can be burned in any good acetone burner without modification to the burner.

Silver Iodide-Sodium Iodide Complexes: Upon combustion of the acetone in air, the acetone is removed from the droplets of solution leaving behind motes of the sodium iodide-silver iodide complex, the remainder is a sintered mass of microcrystalline AgI and NaI, or a fused complex of the two. Some free silver, iodine, sodium carbonate or sodium oxide may also be present. If the flame temperature be adequately high some AgI may vaporize and later condense as very small particles of relatively pure AgI. At temperatures usually used, this reaction is not important.

It has long been apparent that the product of acetone burners using AgI and alkali iodides is not pure AgI. Apart from the simple consideration of conservation of mass, which should suffice, several demonstrations have revealed that some other material was present. This aspect was discussed by Vonnegut (1957) and by St.-Amand (1967).

Mason and Hallet (1956, p. 683) show an electron diffraction pattern, taken by Lisgarten, that demonstrates the presence of a mixed crystal of AgI and KI in which no lines identifiable with AgI can be found. Similar work with NaI and AgI indicates an admixture of cubic crystals of both substances. Because electron diffraction patterns are usually made in a vacuum and most water would be removed, the mixed crystals could exist as discreet entities. In the presence of water in even small amounts, they would combine to form a new substance.

Koenig (1964, p. 308) showed that a soluble component was present in the smoke from an acetone burner and clearly indicated that the effects of liquid water would modify the agent.

R. G. de Pena (1964) identified by electron diffraction of smoke from an acetone burner, hexagonal AgI, cubic AgI and NaI and pointed out that a double salt of AgI and NaI could be present but would be indistinguishable from the cubic AgI and NaI. She indicates (p. 129) that the double salt is likely because certain lines from AgI are absent. The exact molar ratio of the two salts that went into the test, the conditions of combustion and similar considerations would greatly alter the situation and permit a variety of interpretations, as will be seen later.

Figure 1 shows detailed work by Dr. Burkardt (Burkardt, et al., 1970) on the phase diagram for the AgI-NaI-H_2O system. Usually people intend to use the NaI-2AgI mixture shown by the dotted line (a) on the graph. Following line (a) upward on the graph permits an analysis of the reaction as the smoke sojourns in the atmosphere. At low relative humidities, the particle begins as a microcrystalline mixture of 2AgI + NaI. Upon pickup of the slightest amount of water the material in Space II is formed, AgI and NaI coexist with sodium iodide dihydrate. Upon acquiring 5% water by weight, AgI coexists with the complex sodium iodide-silver iodide-trihydrate. At about 8% water by weight the complex goes into solution leaving motes of AgI in a saturated droplet. These will freeze at a temperature determined by the size of the largest particle of AgI in the droplet and the freezing point depression brought about by the solute.

In practice, however, it is difficult to get a satisfying amount of AgI in solution rapidly and more NaI is sometimes added. Exposure of the resulting combustion products to water vapor initiates an even more complex reaction, line b, Fig. 1. At 2Na + AgI all the material temporarily dissolves at about 25% by weight of water, and then AgI again precipitates as the material picks up more water. The 2NaI + AgI mixture passes rapidly into a state in which at about 15% water pickup the whole mass goes into solution and stays there until extreme dilutions are reached.

Thus, almost any acetone burner product will function as a contact nucleant if the freezing process can proceed at a rate greater than solution, hydration and other reactions occur. It serves well if introduced into a cloud chamber, or a real cloud colder than, say, -5°C. The reagent, however, will not work well--if at all--when it passes a considerable time in moist air or in clouds too warm to freeze. Preliminary laboratory experiments by Davis (1970) and Blair and Davis (1969) tend to support this view, while those of St.-Louis and Steele (1968) do not.

Fletcher (1968) implicitly assumes that the whole of the AgI remains in a little ball within the drop formed around the particle. This is almost certainly not the case because the AgI must be disseminated in colloidal particles throughout the drop. The temperature of freezing is then determined by the largest particle and will occur at a much lower temperature than for the same amount of AgI if it were in one lump. Fletcher's (1968) treatment, while excellent for a particle of AgI surrounded by a relatively dilute solution or by water, is not applicable to the product of an acetone burner.

Comparative Activities for AgI and AgI Complexes: Figure 2 shows activity curves, as developed by John A. Donnan (Donnan et al., 1970) at the South Dakota School of Mines and Technology.

The smoke produced from the NH_4I-AgI solution is markedly more effective at the higher temperatures , as is the smoke from the pyrotechnics, than is that from the NaI-AgI solution. These differences

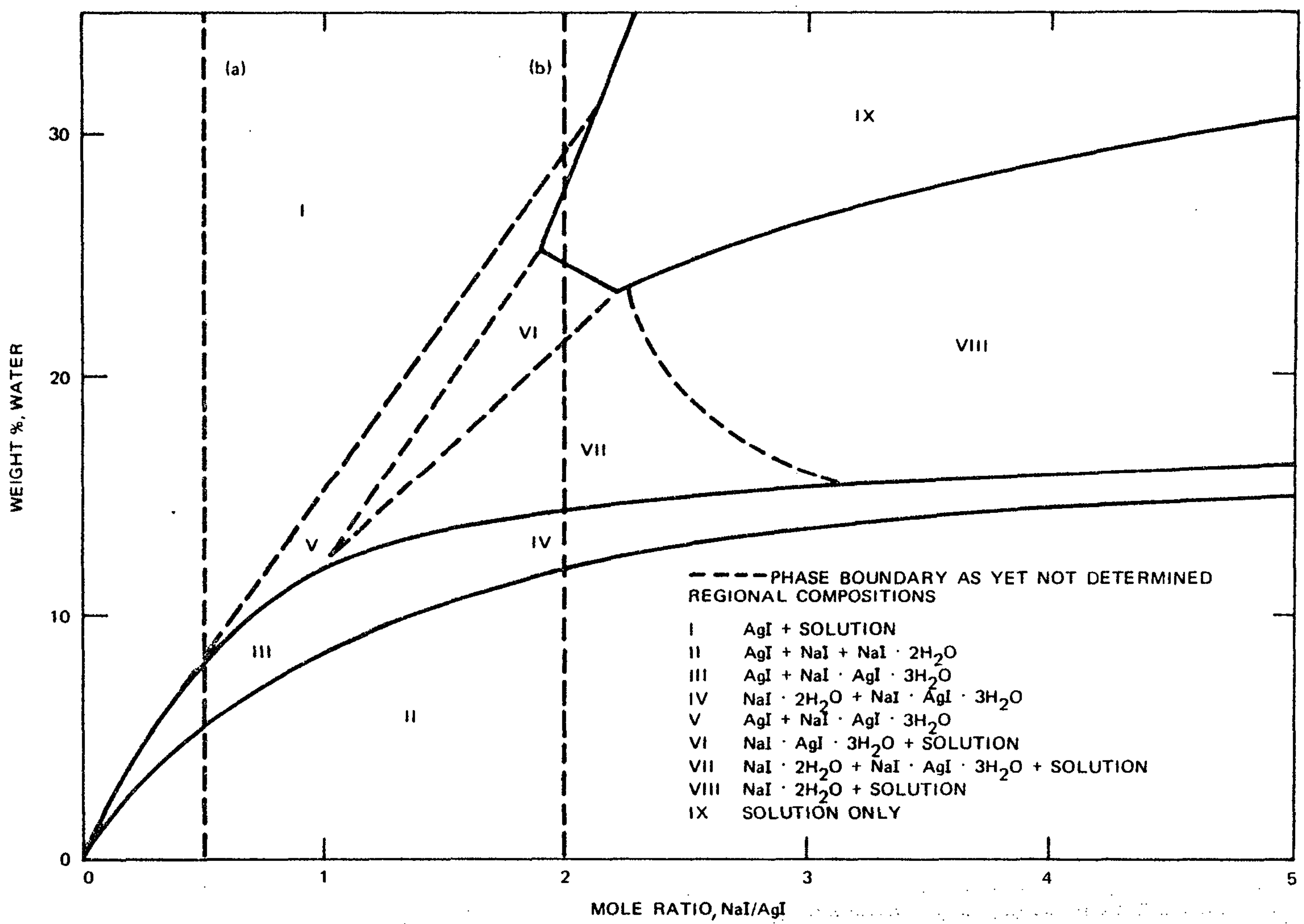


Fig. 1. Partial Phase Diagram AgI + NaI + H_2O

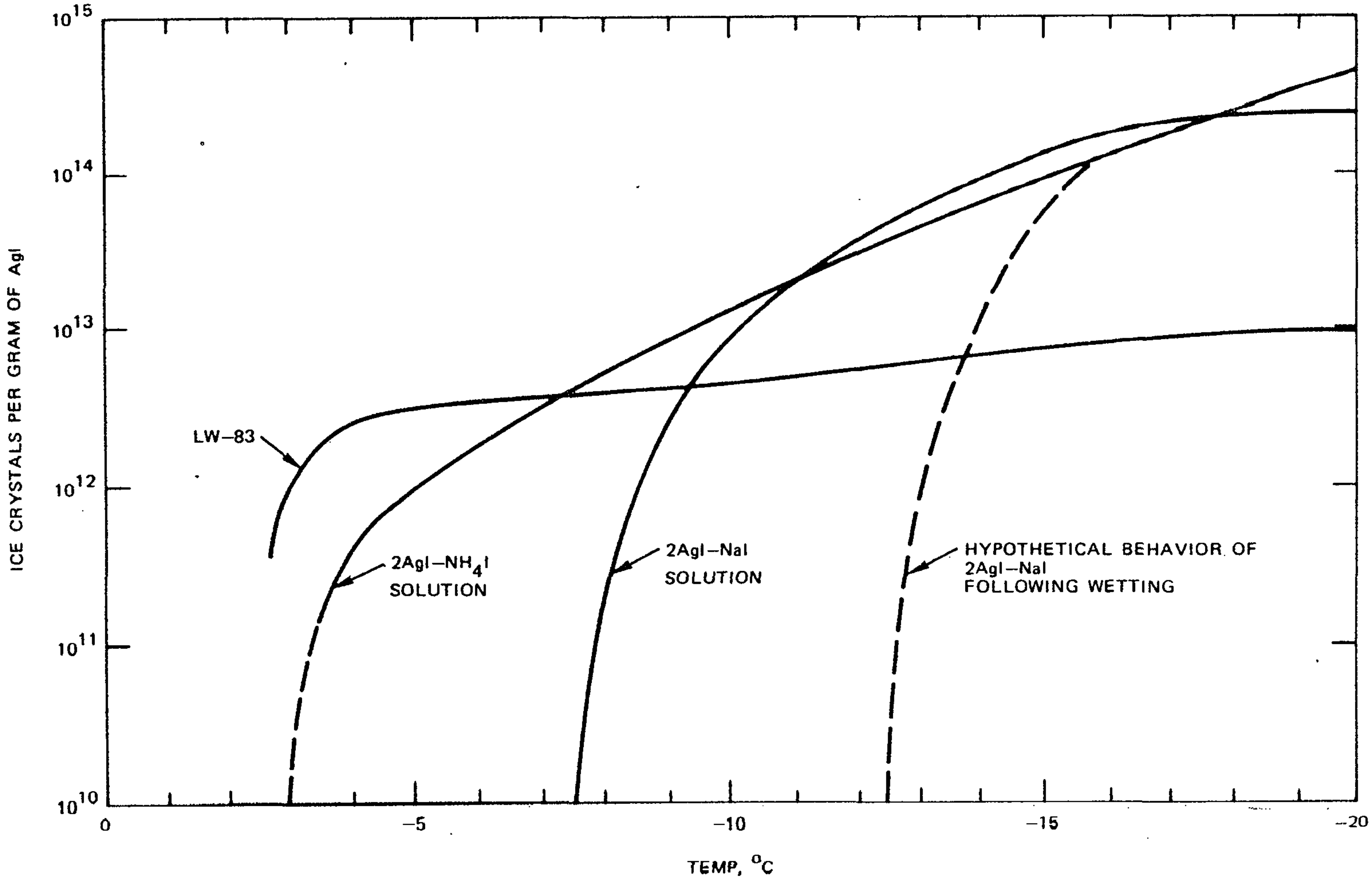


Fig. 2. Activity curves developed by J. Donnan at South Dakota School of Mines and Technology, comparing LW 83, acetone burner using $2AgI\text{-}NH_4I$ and producing AgI smoke and acetone burner using 2AgI-NaI and producing the complex 2AgI-NaI. The dashed curve is for behavior of the complex following wetting or solution.

will be accentuated when the material is used in a real cloud in the atmosphere.

Complexing with Potassium Iodide: The complexing properties of AgI and KI are equally interesting in that a series of complexes are possible. Burkardt, et al. (1970) lists a number of complexes recognized to date for the alkali metals.

A unimolar mixture of AgI and KI will remain as a simple mixture indefinitely, but once exposed to water vapor or liquid water will react rapidly to produce the complex AgI-KI. Tompkins, Muus and Pearson (1963) studied the reaction in detail. They showed that it occurred rapidly at the values shown in Table 6.

TABLE 6

P/P_∞	Temperature	P/P_∞ for saturated
0.578 ± 0.004	0°	0.74
0.480 ± 0.015	25°	0.69
0.43 ± 0.03	35°	0.67

The reaction product is a colorless crystalline material which became visibly wet at $P/P_\infty = 0.55$. Isolated KI crystals become visibly wet at $P/P_\infty = 0.67$ or higher. Similar reactions were noted at lower partial pressures with NaI.

Upon the addition of a little more water, AgI is precipitated. If the AgI is filtered off and the material recrystallized, at least three complexes may be formed, each of which progressively has more KI and less AgI.

Pyrotechnically Generated Complexes: Experiments using the mixtures of KI and AgI in varying relative amounts in pyrotechnic mixtures yielded an interesting series of nucleating agents, Burkardt, et al., (1970). Figure 3 shows the results of adding progressively more KI to the mixture. Used in a mixing type cold chamber, the yields per gram of AgI were higher for higher ratios of KI to AgI up to a value of 12. Above this value up to a ratio of 80, the activity at higher temperatures began to fall off but increased markedly at lower temperatures. Between the ratios of 3 and 12, the behavior was not markedly different, and the activity curve was almost independent of temperature. The 3KI-AgI mix appears to induce freezing at temperatures as high as -1°C, provided

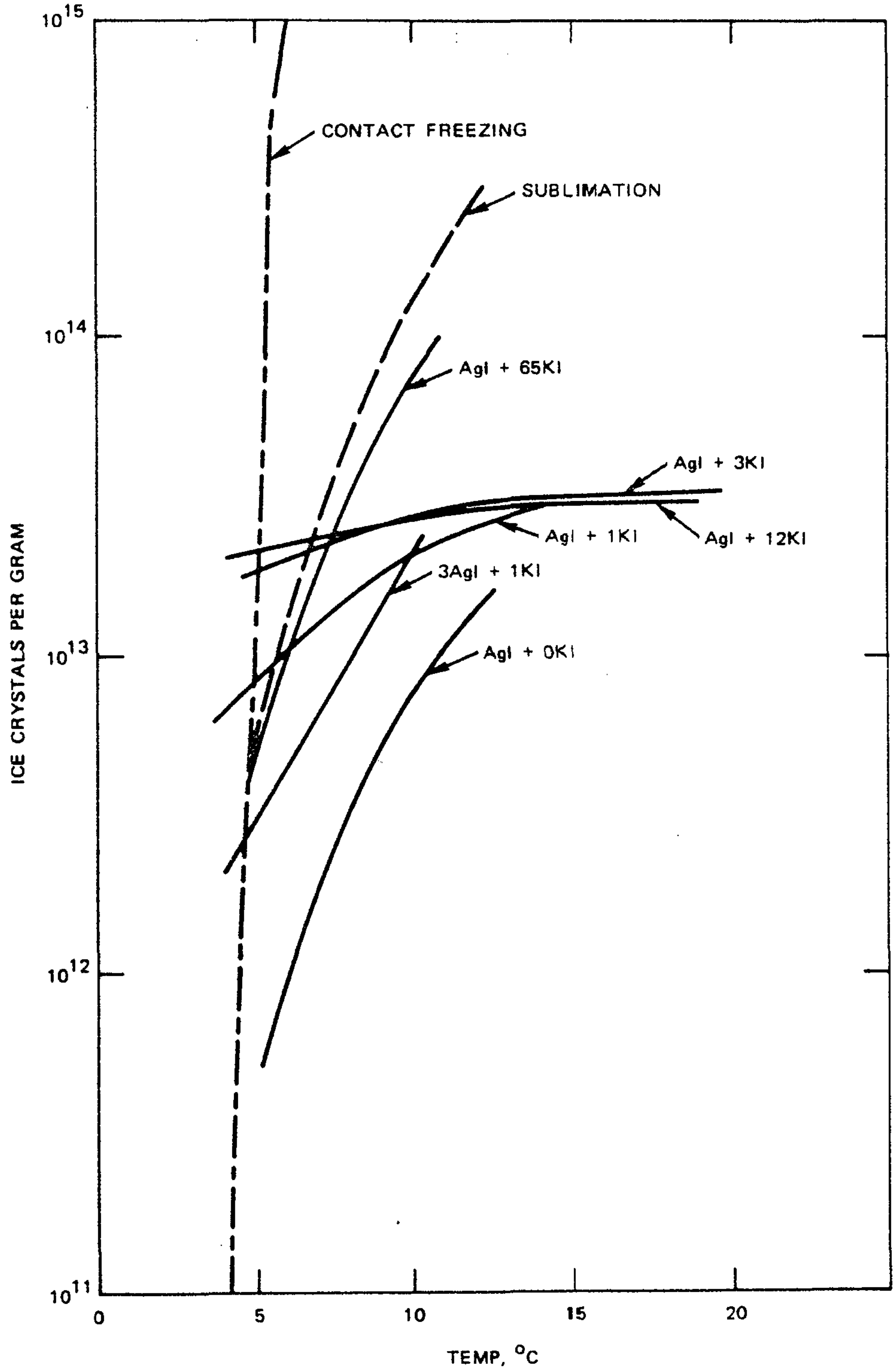


Fig. 3. Effects of admixing various amounts of KI with AgI.

it is directly introduced into the cold cloud chamber.

The nAgI-mKI series may well prove to be very useful as a pyrotechnic cloud seeding agent because by that means it can be directly introduced into the cloud and quite importantly will be rendered inactive by solution once it has been exposed to warm moist air or becomes involved in droplets. Thus, it may be possible to use the agent without having it contaminate the clouds in immediate or surrounding areas.

The material probably works by contact nucleation at the higher temperatures because it is so soluble that freezing would likely have to occur before protracted wetting took place. Indeed, the behavior at extremely high temperatures must be due to contact nucleation because of the long time required for diffusional growth of ice crystals near the triple point. Quite possibly the freezing properties depend more on the iodine-iodine spacing than upon the iodine-silver spacing in the crystalline lattice.

<u>Behavior of Complex Nucleants in Clouds</u>: If a microcrystalline mixture or fused complex of AgI and an alkali iodide is used as a nucleant, its behavior will depend markedly upon the history of the material before it is introduced into a cloud. Thus, one may measure the numerical effectiveness in a cloud chamber, and, if nothing is wrong with the techniques, demonstrate that it will serve as a freezing nucleant at quite warm temperatures. However, if it is subjected to protracted exposure to warm moist air, the whole particle will dissolve and serve no further useful purpose as a freezing nucleant until such time as it forms a droplet so dilute that AgI is again precipitated. Once this occurs freezing will take place at a temperature determined by the size of the largest particle of precipitated AgI in the droplet, modified of course by the freezing point lowering properties of the solute. Hence, the behavior in a real cloud will become markedly different from that in a cold chamber. For all practical purposes therefore, seeding by acetone burners, using alkali iodide-silver iodide complexes, on the ground or at cloud base in an air temperature regime above freezing, will almost certainly yield poorer results than might be obtained when the same device is employed directly in the clouds. Not only will the effectiveness of the nucleating material be reduced, the behavior will be changed so that seeding only occurs in the very uppermost parts of the cloud system. It has been our observation that seeding the coldest parts of the clouds rarely results in any precipitation at all. Hence, we feel that many otherwise well conducted experiments yielded no results or inconclusive results, as indeed they should have, because the experimenters were either not seeding at all or were doing so in a deleterious manner. In re-evaluation, such experiments intended to show that precipitation can be increased by seeding, the experiments should be carefully reviewed to see if seeding did indeed really occur at high enough temperatures to affect the dynamics of the clouds or to make a contribution to the ice content more important than that produced by naturally occuring nuclei.

That is should be so difficult and take so long for general acceptance of the idea that a complex of AgI with an alkali-iodide is not

AgI, nor behaves like AgI is puzzling in view of the published comments by Vonnegut (1957), Tompkins, Muus and Pearson (1963), Koenig (1964), dePena and Caimi (1967) and St.-Amand (1967). It is perhaps proper to quote dePena and Caimi, page 386: "Whatever it is, we are certain that it is a hygroscopic compound, at least at the saturation point, and that the drops formed contain suspended silver iodide. In planning cloud seeding experiments the freezing temperatures of these droplets must be taken into account."

And to repeat Tompkins, Muus and Pearson's succinct remarks, p. 3539: "A thorough and quantitative study of the two systems (AgI-KI and AgI-KI-H_2O) and a new choice of seeding system are alternatives deserving of serious consideration in any attempt to describe the mechanism of cloud seeding."

None of the foregoing is intended to say that under the proper conditions that acetone burner seeding with alkali iodide-silver iodide complexes may not prove valuable. Bowen (1967, p. 70) has used such a technique very effectively in Australia, where he has sent his seeding aircraft directly into the clouds at the proper temperature to insure that seeding does, indeed, work. Also of interest are his comments, p. 70, "There appears to be a distinct difference in the behavior of seeded clouds when the cloud top temperature is warmer or colder than -10°C. When the cloud top temperature is warmer than -10°C, rainfall is stimulated, but the amount is not significantly different in seeded as compared to unseeded clouds. However, at temperatures colder than -10°C, the amount of rain which falls is two or three times greater in seeded than in unseeded clouds."

A similar phenomenon has been noted by Professor Grant in his work in Colorado. One wonders how much more effective it would be to have used real AgI instead of the sodium iodide complex, so that rain could have been obtained from clouds warmer than -10° or -14°C.

<u>Preferred Techniques Based on the Chemistry of Nucleants</u>: It is necessary to tailor the use of the seeding agent nucleant to the chemical nature of the nucleant. For seeding from beneath a cloud or from the ground, we prefer to use a relatively pure silver iodide, or one doped with 3% or less of potassium iodide. We have had good results and commonly use AgI doped with up to 3 times its own weight of potassium oxide, St.-Amand, et al. (1970). This material works better when introduced directly into the cold part of the cloud, but can be used effectively from beneath. The advantage of the doping with K_2O is that it forms no complexes, known to us, and results very rapidly in the formation of large frozen droplets, probably by contact nucleation. The K_2O is rapidly changed to KOH and eventually to K_2CO_3.

We have, at present, under development and in the first stages of experimental use (Vetter, et al., 1970, page 63), a composition that produced AgI with about 3% KI and a small amount of K_2SO_4. This material produces about 3×10^{13} nuclei per gram at -5°C with no appreciable increase at colder temperatures. It consists of a mixture of nitroglycerin and nitrocellulose with 10% $AgIO_3$. AgI would work just as well. The material is not suitable for use from aircraft because of the difficulty of keeping it lighted at altitude and so will be exploited as a ground seeding agent. It would probably be fallacious to try for a larger numerical count for ground seeding because of the likelihood of having the AgI particles entrapped in droplets and dissolved if they were any smaller.

REFERENCES

Berthelot, M., "Sels haloids doubles de l'argent et du potassium," ANN CHIM PHYS, No. 29 (1883), pp. 271-288.

Blair, Donald N., and Briant L. Davis, "Aging of Silver Iodide-Sodium Iodide Generator Effluent in Moist and Dry Air," J APP MET, Vol. 8, No. 4. August 1969. pp. 551-555.

Bowen, E. G., "Cloud Seeding," SCIENCE JOURNAL (August 1967), pp. 69-73.

Burkardt, Lohr A., and William G. Finnegan, "Complex Ice Nuclei: The Silver Iodide-Sodium Iodide System," 2nd National Conference on Weather Modification PREPRINTS, Santa Barbara, Calif. (April 6-9, 1970). pp. 325-328.

Burkardt, Lohr A., William G. Finnegan, F. Kirk Odencrantz, and Pierre St.-Amand, "Pyrotechnic Production of Nucleants for Cloud Modification, Part IV: Compositional Effects on Ice Nuclei Activity," J WEA MOD, Vol. 2, No. 1 (May 1970), pp. 65-76.

Davis, Bryant L., "Ice Nucleation Effeciencies Obtained by X-ray Diffration," Conference on Cloud Physics PREPRINTS, Ft. Collins, Colo. (Aug. 24-27, 1970), pp. 103-104.

de Pena, R. G., "Étude des Noyaux Glacogenes Artificiels au Microscópe Electronique par la Methode des Répliques," Jour. De Recherches Atmosphérique, Vol. 1, 2e annee, No. 8 (1964), pp. 121-131.

de Pena, R. G. and E. A. Caimi, "Hygroscopicity and Chemical Composition of Silver Iodide Smoke used in Cloud Seeding Experiments, "J ATMOS SCI, Vol. 24 (July 1967), pp. 383-386.

Donnan, J. A., Donald N. Blair, William G. Finnegan, and Pierre St.-Amand, "Nucleation Efficiencies of AgI-NH_4I and AgI-NaI Acetone Solutions and Pyrotechnic Generators as a Function of LWC and Generator Flame Temperature, A Preliminary Report," J WEA MOD, Vol. 1, No. 2 (May 1970), pp. 155-164.

Fletcher, N.H., "Ice Nucleation Behavior of Silver Iodide Smokes Containing a Soluble Component," J ATMOS SCI, Vol. 25 (1968) pp. 1058-1060.

Koenig, L. R. "Some Chemical and Physical Properties of Silver Iodide Smokes," J APPL METEOROL. Vol. 3 (June 1964), pp. 307-310.

Mason, B. J. and J. Hallett, "Artificial Ice-forming Nuclei," NATURE, Vol. 177, No. 4511 (1956), pp. 681-683.

St.-Amand, Pierre, "Nucleation by Silver Iodide and Similar Materials," Skywater Conference I, PROC, Bureau of Reclamation, Denver, Colo. (1967), pp. 305-346.

St.-Amand, Pierre, Lohr A. Burkardt, William G. Finnegan, L. Lee Wilson, Shelden D. Elliott, Jr., and Paul T. Jorgensen, "Pyrotechnic Production of Nucleants for Cloud Modification, Part II: Pyrotechnic Compounds and Delivery Systems for Freezing Nucleants, "J WEA MOD, Vol. 2, No. 1 (May 1970), pp. 33-52.

St. Louis, P., and R. L. Steele, "Certain Environmental Effects on Silver Iodide Ice Nuclei," 48th Annual Meeting of the American Meteorological Society, 29 January - 1 February 1968, San Francisco, Calif. abstracted in AMER METEOROL SOC, BULL, Vol. 48, No. 11 (1967), p. 844.

Steele, Roger I., and Charles I. Davis, "Variation of Ice Nuclei Effectiveness with Liquid Water," J ATMOS SCI, Vol. 26, No. 2 (March 1969), pp. 329-330.

Thompkins, L.M., D.A. Muus, and T. Pearson, "Water Adsorption in the system AgI-KI-H_2O." J GEOPHS RES, Vol. 68 (1963), pp. 3537-3539.

Vetter, Ronald F., William G. Finnegan, Lohr A. Burkardt, Pierre St.-Amand, H. Sampson, and Martin Kaufman, "Pyrotechnic Production of Nucleants for Cloud Seeding, Part III: Propellant Compositions for Generation of Silver Iodide," J WEA MOD, Vol. 2, No. 1 (May 1970), pp. 53-64.

Vonnegut, B., "Nucleation of Supercooled Water Clouds by Silver Iodide Smokes," CHEM REV, Vol. 44 (1949), pp. 272-289.

__________., "Techniques for Generating Silver Iodide Smoke," J COLLOID SCI, Vol. 5, No. 1 (Feb. 1950), pp. 37-48.

__________., "Early Work on Silver Iodide Smokes for Cloud Seeding," Final Report of the Advisory Committee on Weather Modification, Vol. 2 (1957), pp. 283-355; p. 421.

EFFECTS OF CONTACT NUCLEATION ON CLOUD SEEDING METHODS

Pierre St.-Amand
William G. Finnegan
Larry A. Mathews

Naval Weapons Center
China Lake, California

ABSTRACT

Contact nucleation is defined as any means by which a glacogenic aerosol touches a water drop or becomes enclosed in one. It can take place by Brownian Diffusion, Smoluchowski Coagulation, turbulent coagulation, diffusiophoresis, electrophoresis and direct condensation.

Brownian diffusion is important for small aerosol particles and large droplets, rapidly decreasing with increases in aerosol radii. Smoluchowski, or impact collection is important for all sizes of aerosol and drops, becoming more important for the larger members of both classes. It is especially valuable once ice embryos have begun to form on nuclei. The values of the capture coefficient for Smoluchowski coagulation is estimated for water drops with water drops and for water drops with AgI particles. It is no where zero, but finite values exist for all sizes of aerosol and droplet sizes.

Diffusiophoresis capture is proporitional to total liquid water condensed. For seeding at or beneath cloud base, it is adequate to assure nucleation of all drops by the time freezing level is reached, provided an aerosol density of 10^5 particles per cm^3 is established and a liquid water content of 1 gram per cubic meter is reached.

Tables are given for the combined effects of Brownian and Smoluchowski capture and for diffusiophoresis.

Contact through condensation on the AgI particles is assumed for seeding below cloud base, but in this mode the smaller AgI particles are dissolved and only the larger particles survive to produce freezing near the triple point.

Electrophoresis aids capture by Brownian and Smoluchowski coagulation and by diffusiophoresis.

Cloud seeding strategy based on contact nucleation is discussed, and is shown to be most useful in updrafts in convective clouds and is by far the most important nucleation process in this regard.

CONTACT NUCLEATION:

Water drops may be contacted by an aerosol by one or more of several processes. All these processes work independently and sometimes synergistically. A great deal of work has been done in connection with the scavenging of debris from atomic explosions and a few examples of which are Goldsmith, Delafield and Cox, 1962; Greenfield, 1957; and Vittori and Prodi, 1967. This work was done with an eye to calculating removal of material and because the collection rates are not impressive it has been occasionally indicated that contact nucleation is not important. For our purposes, it is enough that only one glacogenic nucleus touch a water droplet in order to initiate freezing. We will investigate several processes and estimate the nuclei density necessary to cause glaciation in a cloud.

Some of the processes by which droplets can capture aerosol particles are:

1. Brownian Diffusion of aerosol particles to cloud droplets caused by random motion of the particles.
2. Turbulent coagulation
3. Impact coagulation
4. Diffusiophoresis
5. Electrophoresis
6. Direct condensation

BROWNIAN MOVEMENT:

An excellent discussion of Brownian motion and coagulation is given by Green and Lane, 1964, pp. 73-75 and pp. 138-178. Small particles in suspension in gas or liquid undergo continual movement because of bombardment by molecules of the materials in which they are suspended. It is generally considered, that if, in the course of their peregrinations, two particles strike each other they will coagulate and the two particles become one. It is by no means clear that they should do so but experimental results indicate that this frequently happens.

The measure of movements of particles is the Diffusivity,

$$D = \overline{\Delta X}^2 / 2t \qquad (1)$$

where $\overline{\Delta X}^2$ is the movement of a particle along an axis in interval t. An approximate formula for D, derived by Einstein is

$$D = \frac{RT}{N 6 \pi \eta r} \qquad (2)$$

where R is the gas constant, T is the absolute temperature, N is Avagadros number, η is the viscosity of the gas and r is the particle radius, assuming a spherical particle.

Where r is cimparable to, or smaller than, the mean free path, λ, of the gas molecules a semi-empirical formula may be used:

$$D = \frac{KT}{6 \pi \eta r} \left(1 + \frac{A \lambda}{r} + Q \frac{\lambda}{r} e^{-br/\lambda}\right) \quad (3)$$

Where K is Boltzmann's constant, A and Q are numerical factors that depend on the manner by which gas molecules are reflected from the surface of a particle. The numerical factor, b, depends on the complexity of the flow about the particle due to molecule - molecule and molecule - particle collisions. Fuchs (1964) uses A = 1.246, Q = 0.42 and b = 0.87 as obtained by Millikan (1923). These coefficients are based on the well-known relationship for the viscosity of a gas, $\eta = 0.499 \rho_g c_m \lambda$, where $\eta = 1.830 \times 10^{-4}$ poise at 23°C. Also ρ_g is the atmospheric air density and $\bar{c}_m$ is the mean velocity of the gas molecules at 23°C.

Langmuir, in his unpublished notes, developed an approximate formula used by Vonnegut, 1949, giving

$$D = \frac{2.04 \times 10^{-16}}{r^2} + \frac{1.18 \times 10^{-11}}{r} \quad (4)$$

and this was used for the larger drops in our calculations.

For smaller particles values for D, from Green and Lane and from Fuchs are given in Fig. 1.

BROWNIAN COAGULATION OF AEROSOLS:

Every aerosol changes in size and number distribution with the passage of time because the particles contact each other by Brownian motion and other processes and mutually adhere. A monodispersed aerosol cannot long exist because the radii of the particles increase and the numbers decrease. The smallest particles are the first to go, the size spectrum gradually shifting to fewer and larger particles. For a monodispersed aerosol

$$- \frac{dn}{dt} = \frac{2}{3} \frac{RTS}{n N}\left(1 + \frac{A \lambda}{r}\right) n^2 \quad (5)$$

where S is the ratio of the sphere of influence of a particle to the radius, and n is the number of particles per cm^3.

For a polydispersed aerosol

$$- \frac{d n}{dt} = \frac{RTS}{6nN} \left(\frac{r_1 + r_2}{r_1 r_2}\right) n_1 n_2 \left(1 + \frac{A \lambda}{r}\right) \quad (6)$$

This is properly solved by taking into account all possible combinations of aerosol radii and summing the collisions so calculated.

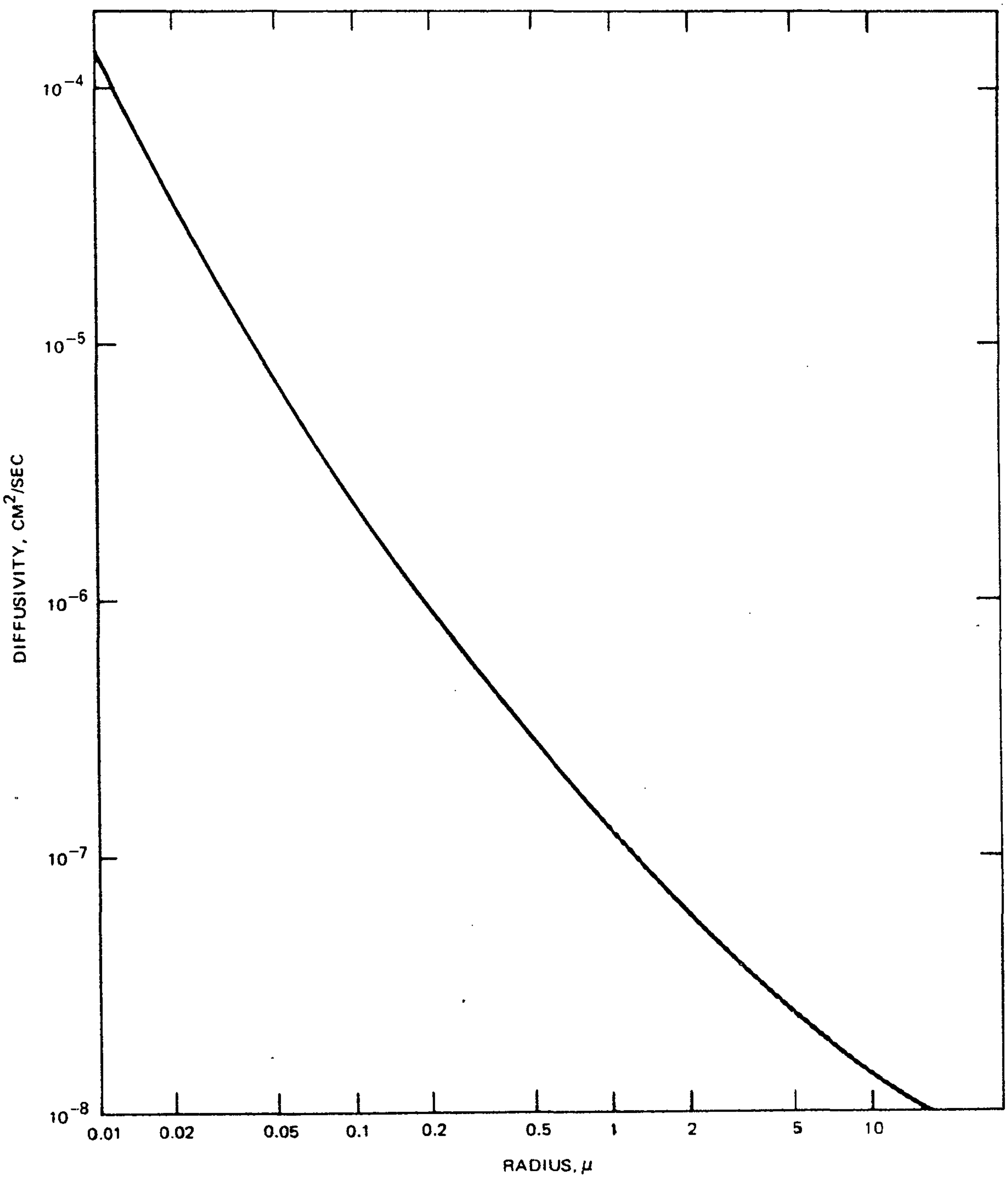


Fig. 1. Diffusivity as a function of particle radius. From Fuchs (1964) and Green and Lane (1964).

Coagulation has a very important bearing on cloud seeding in that the design of a nucleant generator should be such as to try to keep the particle density below, say, 10^6 particles/cm^3 in order to avoid excess coagulation.

BROWNIAN COAGULATION OF AEROSOL PARTICLES WITH DROPLETS:

If n_d spheres of radius r_d, be in a space in which n_a aerosol particles are dispersed per cm^3, and the space is large compared to the sphere and the particles stick to the sphere upon impact, the number, N, removed in unit time by the sphere is

$$N = 4\pi \; (D_a + D_d) \; (r_a + r_d) \; (1 + \frac{A\lambda}{r_a}) \; n_a \; n_d \qquad (7)$$

assuming that D_d is small compared to D_a and that r_a is small compared to r_d and that the term in parenthesis is near unity

$$N = 4\pi \; D_a \; r_d \; n_d \; n_a \qquad (8)$$

In practice, one must calculate the collisions between all particles of a given D_a and all droplets of radius r_d, Fig. 2.

$$N = \sum_{r_d}^{\infty} \sum_{D_a}^{\infty} 4\pi \; D_a r_d \; (n_d)_{r_d} \; (n_a)_{D_a} \; \Delta r_a \; \Delta D_a \qquad (9)$$

If

$$\omega = \sum_{o}^{\infty} D_a \; N_a \; \Delta D_a \qquad (10)$$

and

$$\varepsilon = \sum_{o}^{\infty} r_d \; n_d \; \Delta r_d \qquad (11)$$

Then

$$N = 4\pi \; \omega\varepsilon \qquad (12)$$

ω and ε can be found from Figure 2 if $n_a(D_a)$ and $n_d(r_d)$ can be evaluated. Such evaluation must await detailed measurements of the particle and drop size spectra. A grossly simplified procedure for calculation follows:

The liquid water content LWC of a cloud, usually given in grams per cubic meter is,

$$LWC = \frac{4}{3} \pi\rho \sum_{r_d}^{\infty} n_d \; r_d^3 \Delta r_d \times 10^6 \text{ gms/m}^3 \qquad (13)$$

where ρ is the density of H_2O, and Δr_d is the class interval of r_d.

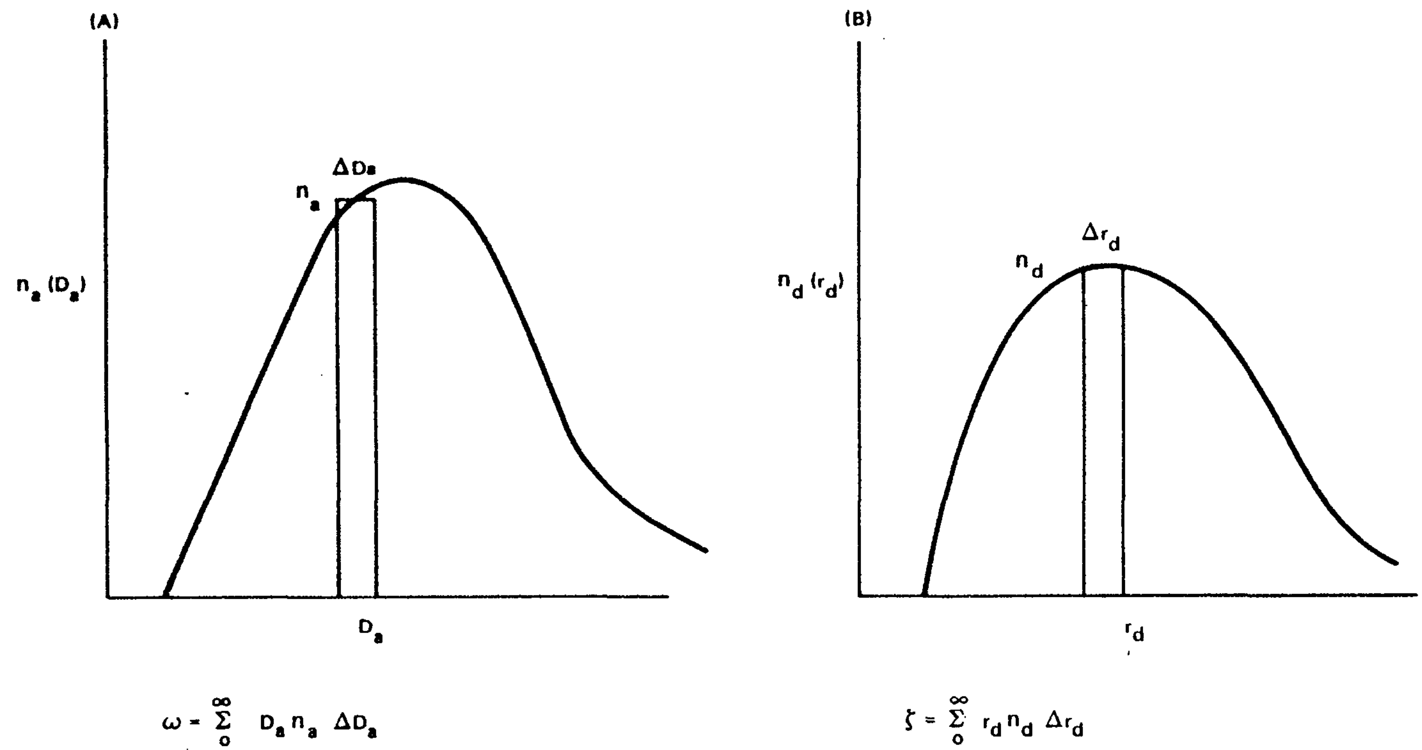


Fig. 2. Distributions for (A) number of particles as a function of diffusivity, (B) number of drops as a function of radius.

To simplify matters we could assume a water cloud of uniform droplet size, $\overline{r}_d$, and number n_d, then

$$LWC = \frac{4}{3} \pi \rho \, \overline{r}_d^{\,3} \, n_d \times 10^6 = \frac{4}{3} \pi \rho \, \overline{r}_d^{\,2} \, (\overline{r}_d n_d) \times 10^6 = \frac{4}{3} \pi \rho \, \overline{r}_d^{\,2} \, \varepsilon \times 10^6 \qquad (14)$$

hence

$$\varepsilon = \frac{LWC}{\frac{4}{3} \pi \rho - r_d^2 \times 10^6} \qquad (15)$$

and, approximately

$$N = \frac{4\pi \, D_a \, n_a \, LWC}{\frac{4}{3} \pi \rho \overline{r}^2_d \times 10^6} \qquad (16)$$

or

$$N = \frac{3D_a \, n_a \, LWC \times 10^{-6}}{\overline{r}\,\frac{2}{d}} \qquad (17)$$

In Table 1 and Figure 3 we have calculated from Equation (7) the values of $N/n_a n_d$ for monodispersed clouds, making no assumptions except that the Cunningham slip term is unity and using the values of Diffusivity for both droplets and particles. This table shows the number of collision by Brownian motion per drop per particle per second. For larger particles, capable of producing freezing at temperatures of the order of -5°C, a very high concentration of particles is needed. Much less is needed at colder temperatures where the smaller particles can function.

Table 2 shows the number of particles per cubic centimeter necessary to nucleate all the drops of a given size in one second. It is, within reasonable bounds, independent of LWC.

As particles remain in the air, they grow nuclei by diffusional capture of water vapor and the Brownian capture by drops decreases in importance as the particles aggregate mass.

If the comments considered earlier (St.-Amand, et al., 1970) that only 10^3 ice crystals can exist per cubic centimeters, then one can consider the time required for nucleation of only 10^3 droplets per cm^3 and the aerosol density necessary for complete nucleation by Brownian contact is only of the order of 10^4 - 10^5 particles/cm^3 for all droplet dispersions of radius less than 20 μ.

Brownian coagulation increases slightly in rate with altitude because, λ , the mean free path of molecules, is longer. Decreases due to temperature decrease will be less important.

TABLE 1

BROWNIAN COLLISIONS PER SECOND PER DROP PER AEROSOL PARTICLE.

THE VALUES ARE TAKEN FROM: $4\pi(D_a + D_d)\ (r_a + r_d) = \frac{N}{n_a n_d}$

r_a	r_d						
μ	10	20	50	100	200	500	1000
0.01	1.70×10^{-6}	3.39×10^{-6}	8.48×10^{-6}	1.70×10^{-5}	3.39×10^{-5}	8.48×10^{-5}	1.70×10^{-4}
0.02	4.51×10^{-7}	9.03×10^{-7}	2.26×10^{-6}	4.51×10^{-6}	9.03×10^{-6}	2.25×10^{-5}	4.51×10^{-5}
0.05	8.57×10^{-8}	1.71×10^{-7}	4.29×10^{-7}	8.57×10^{-7}	1.71×10^{-6}	4.28×10^{-6}	8.57×10^{-6}
0.1	2.79×10^{-8}	5.56×10^{-8}	1.39×10^{-7}	2.78×10^{-7}	5.56×10^{-7}	1.39×10^{-6}	2.78×10^{-6}
0.2	1.06×10^{-8}	2.11×10^{-8}	5.26×10^{-8}	1.05×10^{-7}	2.09×10^{-7}	5.23×10^{-7}	1.04×10^{-6}
0.05	3.62×10^{-9}	7.11×10^{-9}	1.75×10^{-8}	3.45×10^{-7}	6.91×10^{-8}	1.72×10^{-7}	3.44×10^{-7}
1	1.77×10^{-9}	3.42×10^{-9}	8.40×10^{-9}	1.61×10^{-8}	3.22×10^{-8}	7.98×10^{-8}	1.60×10^{-7}
2	9.40×10^{-10}	1.76×10^{-9}	4.15×10^{-9}	7.82×10^{-9}	1.55×10^{-8}	4.52×10^{-8}	7.68×10^{-7}
5	4.72×10^{-10}	8.25×10^{-10}	1.81×10^{-9}	3.14×10^{-9}	6.13×10^{-9}	1.77×10^{-8}	3.00×10^{-8}
10	3.47×10^{-10}	5.73×10^{-10}	1.18×10^{-9}	1.88×10^{-9}	3.62×10^{-9}	1.03×10^{-8}	1.72×10^{-8}

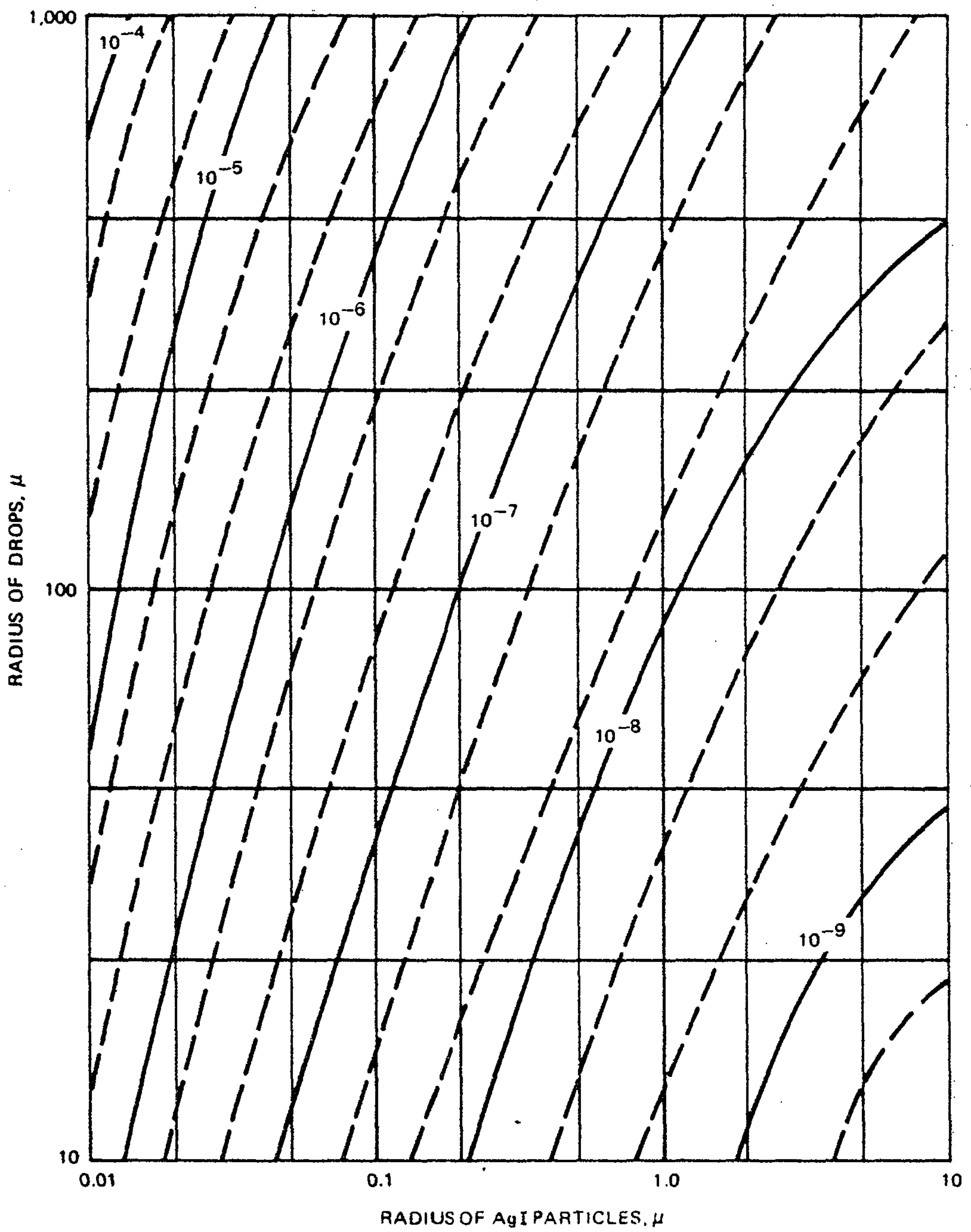


Fig. 3. Values of $N/n_a n_d$ per second for Brownian capture.

TABLE 2

NUMBER OF NUCLEI REQUIRED TO FREEZE ALL DROPS OF RADIUS r_a FOR MONODISPERSED CLOUD.

r_a	r_d							D
u	10	20	50	100	200	500	1000	cm^2/sec
0.01	5.9×10^5	2.9×10^5	1.2×10^5	5.9×10^4	2.9×10^4	1.2×10^4	5.9×10^3	1.35×10^{-4}
0.02	2.2×10^6	1.1×10^6	4.4×10^5	2.2×10^5	1.1×10^5	4.5×10^4	2.2×10^4	3.59×10^{-5}
0.05	1.2×10^7	5.8×10^6	2.3×10^6	1.2×10^6	5.8×10^5	2.3×10^5	1.2×10^5	6.82×10^{-6}
0.1	3.6×10^7	1.8×10^7	7.2×10^6	3.6×10^6	1.8×10^6	7.1×10^5	3.6×10^5	2.21×10^{-6}
0.2	9.4×10^7	4.7×10^7	1.9×10^7	9.5×10^6	4.8×10^6	1.9×10^6	9.6×10^5	8.32×10^{-7}
0.5	2.8×10^8	1.4×10^8	5.7×10^7	2.9×10^6	1.4×10^7	5.8×10^6	2.9×10^6	2.74×10^{-7}
1	5.6×10^8	2.9×10^8	1.2×10^8	6.2×10^7	3.1×10^7	1.3×10^7	6.3×10^6	1.27×10^{-7}
2	1.1×10^9	5.7×10^8	2.4×10^8	1.3×10^8	6.4×10^7	2.2×10^7	1.3×10^6	6.10×10^{-8}
5	2.1×10^9	1.2×10^9	5.5×10^8	3.2×10^8	1.6×10^8	5.7×10^7	3.3×10^7	2.38×10^{-8}
10	2.9×10^9	1.7×10^9	8.5×10^8	5.3×10^8	2.8×10^8	9.7×10^7	5.8×10^7	1.38×10^{-8}
D	1.38×10^{-8}	5.90×10^{-9}	2.36×10^{-9}	1.18×10^{-9}	5.90×10^{-10}	2.36×10^{-10}	1.18×10^{-10}	

Numerical density of nuclei of the order of 100 per liter, or 10^{-1} per cm^3, have been proposed as ideal for cloud seeding. It is clear from the foregoing that no appreciable nucleation by Brownian contact can be expected under these conditions.

Figure 3 shows the Brownian Diffusion contact rate in terms of $N/n_a n_d$. The function plotted is $4\pi (r_a + r_d)(D_a + D_d)$. To find the number of collision between drops and aerosol particles, one multiplies the values on the graph for a given drop size and particle size by the number of drops of that size and the number of aerosol particles of that size and sums these for all size ranges.

ORTHOKINETIC CAPTURE OR SWEEPOUT

Particles moving through a medium or through space with different velocities overtake, and sometimes collide with each other. The differential velocity may be due to any cause at all.

The theory starts off simply enough using arguments based on concepts from molecular physics. A spherical particle moving in nonturbulent air sweeps out a volume V, dependent on its velocity v and radius, in time t:

$$V = \pi r^2 vt \tag{18}$$

If other particles occupy the space at a density of n per unit volume, the number of collisions, N, is:

$$N = \pi r^2 vtn \tag{19}$$

provided that the size of the smaller particles are small compared to the larger and aerodynamic forces are neglected for the moment.

Smoluchowski has developed a general expression for this type of capture as applied to a distribution of particles, see Figure 4.

$$N = \pi \int_{o\,r_i}^{\infty} \int_{o\,r_j}^{\infty} (|V_i - V_j|)\ (r_i + r_j)^2\ E\ n_i\ (r) dr_i\ dr_j \tag{20}$$

where N is the number of collisions per unit time, and E is the so-called collision efficiency.

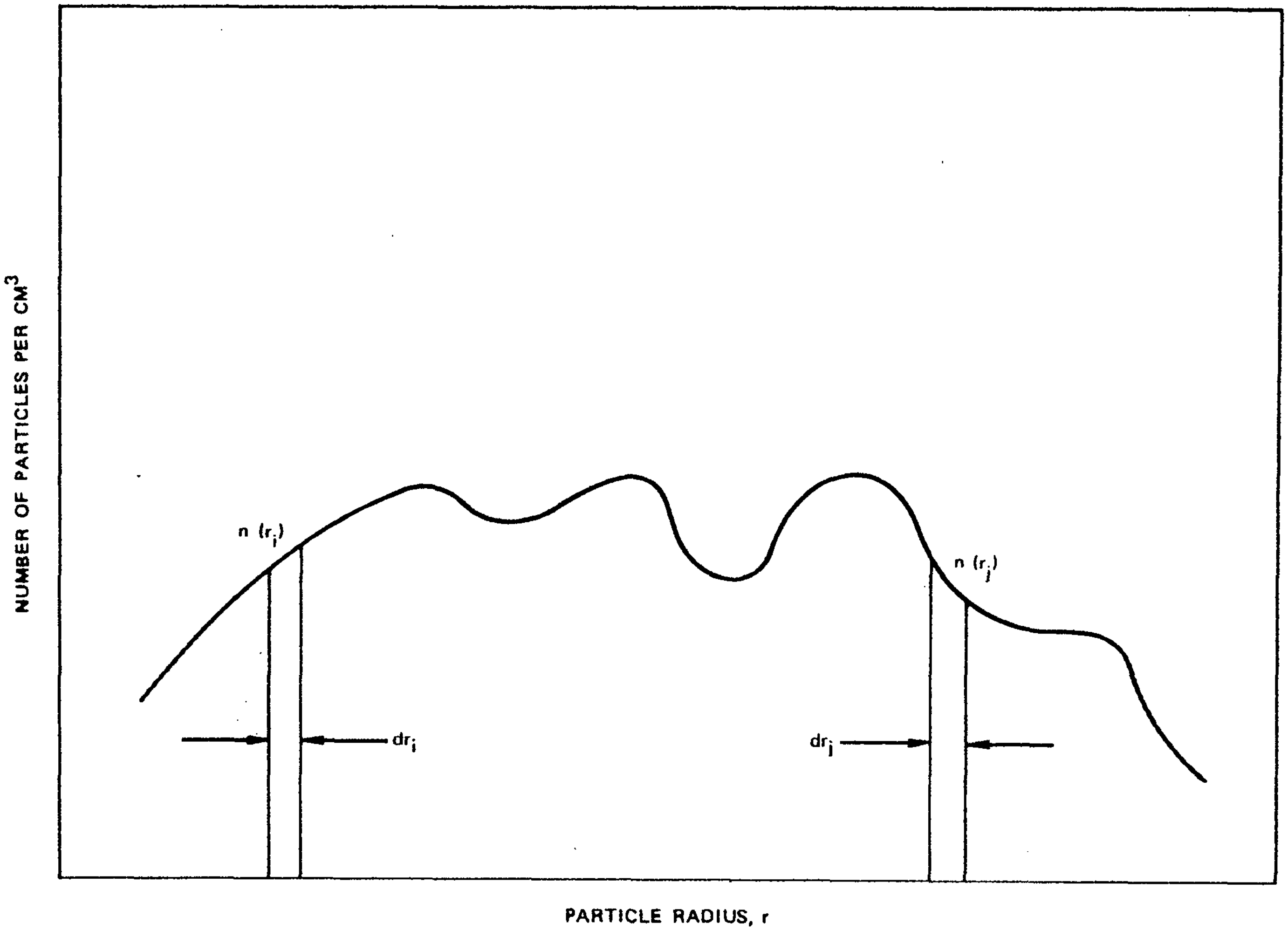


Fig. 4. Nomenclature of Smoluchowski's equation.

Because no constraint has been placed on velocity and particles can be considered to be moving in all directions, this equation could be also used to describe Brownian coagulation.

We will now consider only fall velocity in the earth's gravitational field and thus make the paths parallel. It then suffices to use the terminal velocities as determined by the density and viscosity of the air.

The difficulty now arises of considering the value of E. E. is a complicated function of particle size, velocity, density and the properties of the air.

$$E = f\ (V_1, V_2. r_1, r_2, \rho_p, \rho_g, T, P, \text{Etc.}) \qquad (21)$$

Langmuir (1948) attempted to evaluate this term and has given a set of values for it. These have, for various reasons, been criticized and other sets produced by Pearcey and Hill (1957), Hocking (1959) and others. The state-of-the-art has been reviewed by Fletcher (1962), p. 138-147 and neatly summarized by Fuchs (1964), p. 319-324.

E is now still under some dispute even for the particle size range in which it should be best known. Briefly, as a faster, and hence larger particle approaches a smaller, it displaces the air and the air sweeps the smaller particles apart. The complexity of the situation is enormous because capture of particles by effects of wake turbulence can result in values of E greater than unity as predicted and observed by Pearcey and Hill (1957). The increase in E from the deformation of the surface of a liquid drop by viscous forces is difficult to estimate as is the relative importance of viscous and potential flow around a drop for intermediate Reynolds numbers. Even more complex is the perturbation of the air by a large number of falling drops whose wakes react with each other. Effects of particles of irregular shape, Brownian and aerodynamic rotation and similar factors are almost impossible to estimate.

One of the difficulties is that for particles below a certain size, capture does not seem possible because E is taken as zero. Hocking (1959) indicates that when a collecting drop has a radius of 18 μ or less that E is zero for all smaller particles. Similar limits of about 6 μ and 15 μ were found by Pearcey and Hill (1957) and Langmuir (1948, p. 182), respectively.

For our purpose there is great difference between zero and some small but finite value of E. It may be true that for ordinary cloud physics in which coalescence of cloud droplets into rain drops is a matter of concern, that capture of small particles is infrequent and of little consequence. We, on the other hand, need to estimate the number of droplets of whatever size that become infested with just one glacogenic nuclei, and we may locally make the numerical density of the nuclei quite high compared to the number of droplets, so that even small values of E are important.

The restrictions on particle size considered by Hocking (1959) and Langmuir (1948) have no physical significance and are apparently in error. For example, Langmuir assumed for simplification that the particles were point-sized, and thus failed to account for the interception effect (i.e., edge of particle making contact) of the finite-sized particles. Fuchs (1964) states that E is not zero for particles of any size because of the interception effect alone. He gives limiting values of collision efficiency E for both viscous and potential flow, which are plotted in Figure 5. For interception only, finite values of E exist for all values of radius ratio, $r_1/r_2 > 0$ and any value of radius, r_1 or r_2. An experimental value of collection coefficient found by Adam and Semonin (1970) is also shown in Figure 5 for very small r_1/r_2.

Although Hocking considered the droplets to be of finite size in his analysis, Davis (1966) has found errors in several of Hocking's drag expressions when the droplets are close together. Davis and Sartor (1967) who used more accurate drag expressions for Stokes flow, found none of the restrictions associated with particle size which were characteristic of Hocking's work. They found no "cut off" in droplet collision existed even for radii as small as 5 μ. In fact, their calculations showed finite collision efficiencies for all values of $r_1/r_2 > 0$. As mentioned above, this finding is reasonable because of the interception effect. Also, the effect of Brownian motion, diffusiophoresis and electrophoresis would cause E to be larger at very small particle sizes.

At the recent Conference on Cloud Physics, Davis, Klett, and Neiburger (1970) presented collision efficiencies, which are calculated by using drag forces based on a new hydrodynamic model. The new model, which was developed by Klett (1968) provides drag forces based on time independent Oseen-type flow. The model allows taking into account fore-aft asymmetry, which definitely exists when Reynolds number approaches one. None of their calculations exhibit a "cut-off" or zero collision efficiency for any droplet size or any size ratio. It was noted that collision efficiencies, which involve the Oseen-type forces, showed a tendency to increase at radius ratios near unity. The values of linear collision efficiencies, Y_c, presented in Figure 6, appear to be more physically real than the previous calculations of Davis in that there is no confluence of curves at $r_1/r_2 \to 1$. The work of Davis, et al. (1970) for small r_1/r_2 is supported by Woods and Mason's (1964) experimental studies of small droplet collection, which show finite collection for $r_1/r_2 \leq 0.1$. Davis et al.(1970) believe that their results are qualitatively sound.

The curves in Figure 6 are extrapolated to zero collision efficiency at $r_1/r_2 = 0$ (Davis, et al., 1970) indicate that finite collision efficiencies exist for any r_1/r_2, but they terminate their preliminary graph curves at $r_1/r_2 = 0.1$). Linear collision efficiency, Y_c, is plotted versus the small radius, r_1, for $r_2 = 10$ to 50 μ in Figure 7. The curves are extrapolated down to $r_1 = 0.01$ μ. A border is shown to distinguish collision efficiencies which are extrapolated (on left side). It is noted that the extrapolated collision efficiencies tend to converge to the interception-only values at small r_1/r_2. Table 3 shows a listing of the linear collision efficiencies. Table 4 shows the sources of collision

- 63 -

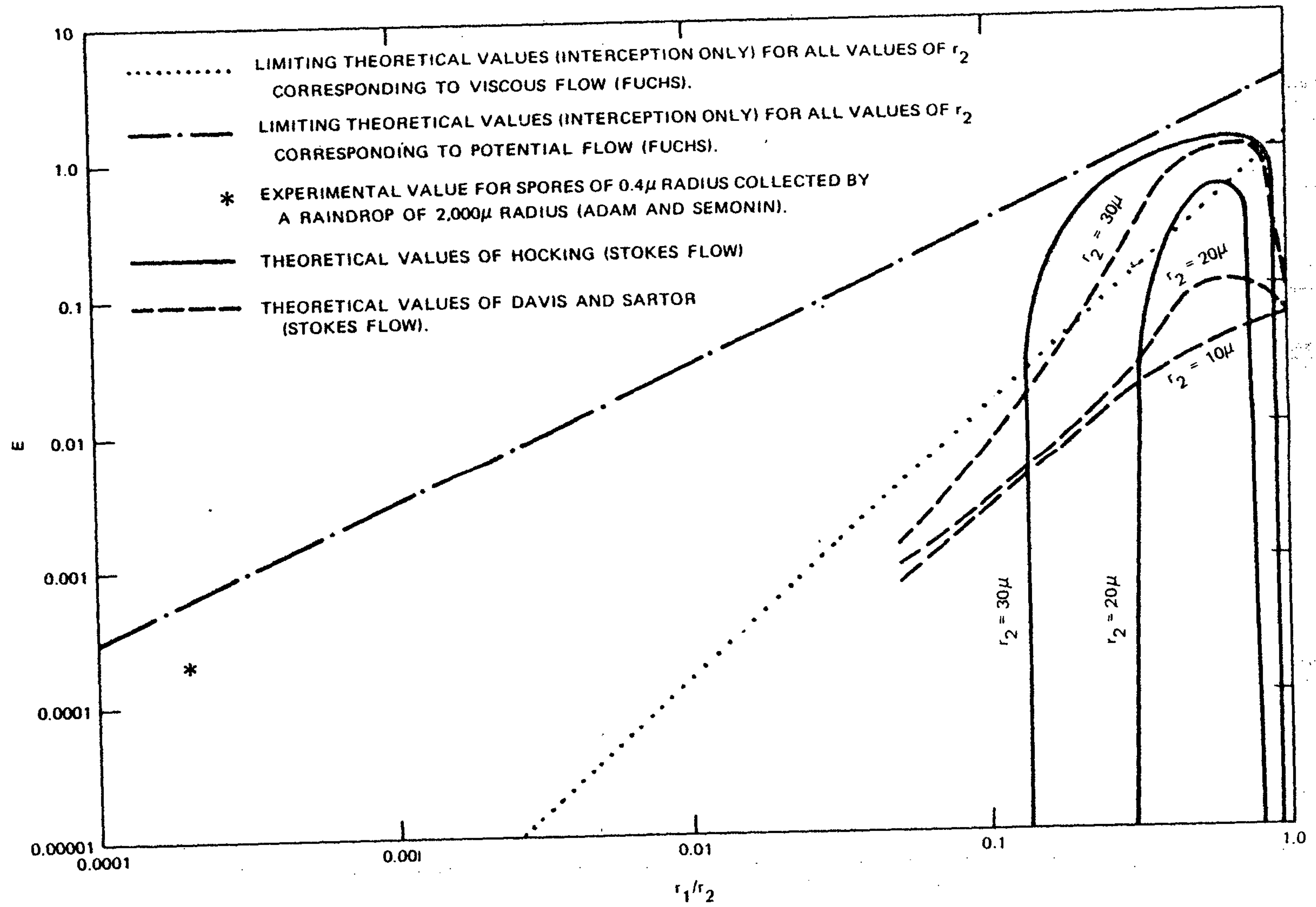


Fig. 5. Values of E as a function of radius ratio for several theories.

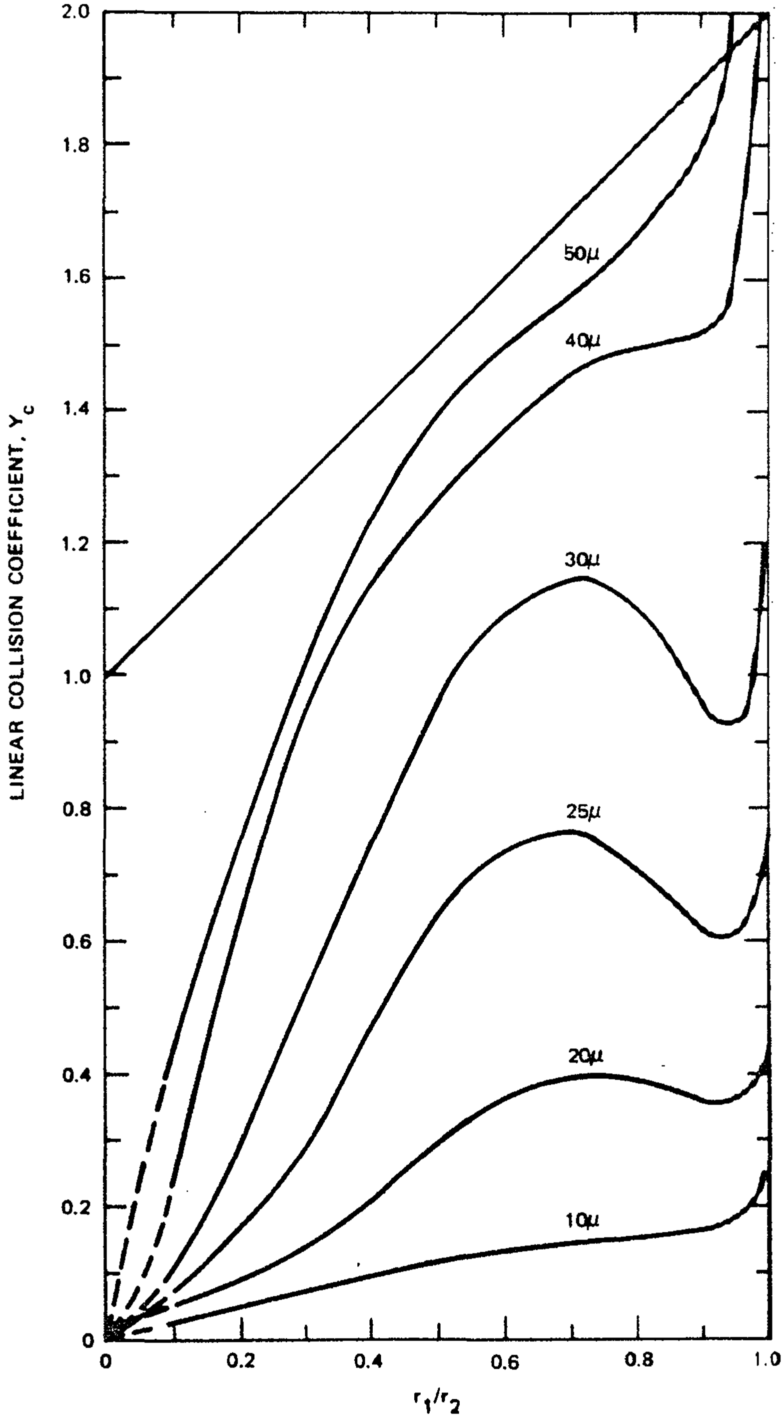


Fig. 6. Preliminary linear collision efficiency versus radius ratio data of Davis.

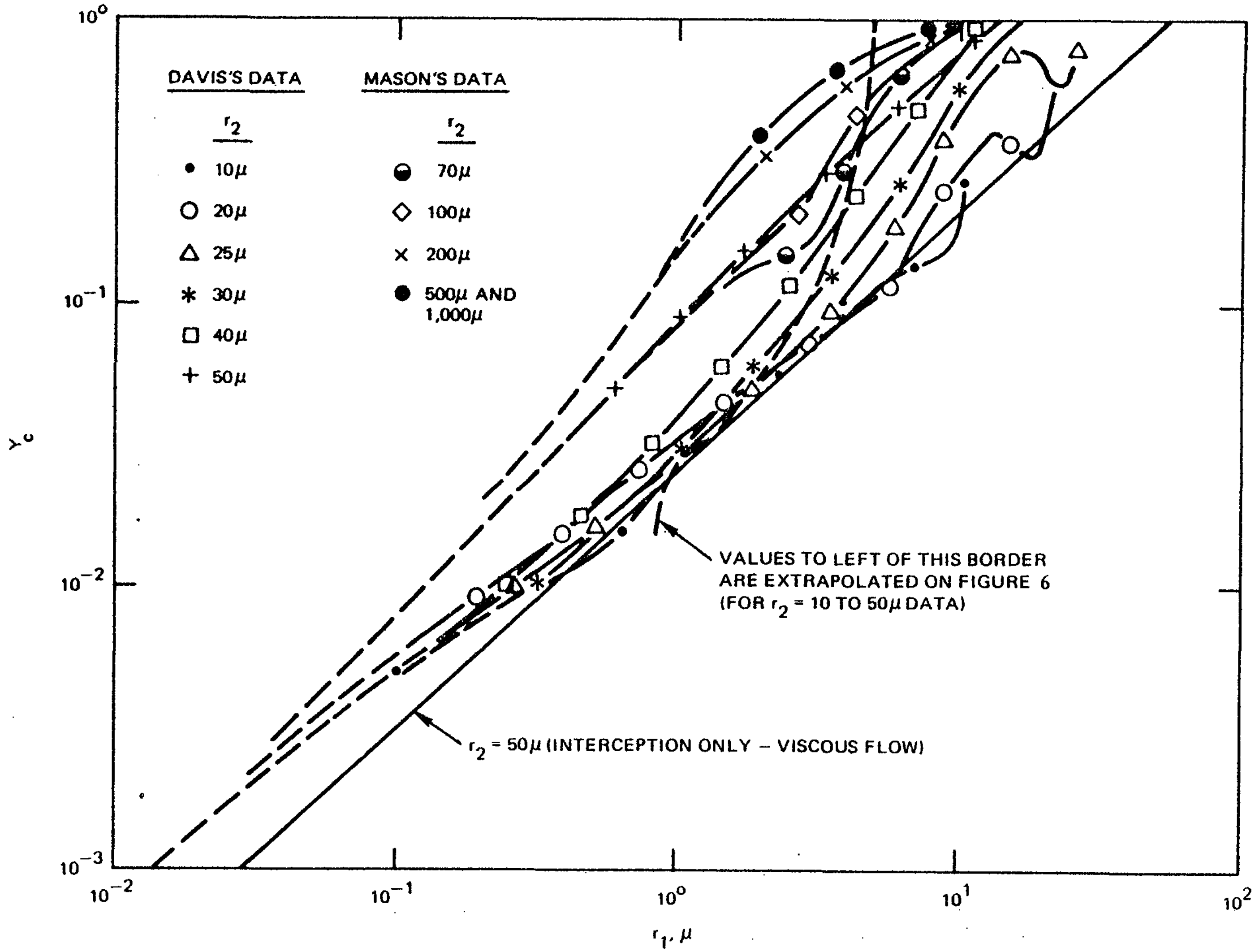


Fig. 7. Calculations by Davis and Mason and extrapolations therfrom for Y_c.

TABLE 3

LISTING OF LINEAR COLLISION EFFICIENCIES

Y_c FOR WATER DROP AND DROPLET SYSTEM

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	8×10^{-4}	1×10^{-3}	1×10^{-3}	1.7×10^{-2}	1.2×10^{-2}	7.7×10^{-3}	5.5×10^{-3}
0.02	1.4×10^{-3}	1.5×10^{-3}	1.5×10^{-3}	2.5×10^{-2}	1.7×10^{-2}	1.1×10^{-2}	7.7×10^{-3}
0.05	3×10^{-3}	3.2×10^{-3}	4×10^{-3}	3.9×10^{-2}	2.7×10^{-2}	1.7×10^{-2}	1.2×10^{-2}
0.1	5×10^{-3}	5.3×10^{-3}	8×10^{-3}	5.5×10^{-2}	3.9×10^{-2}	2.5×10^{-2}	1.7×10^{-2}
0.2	8×10^{-3}	9.4×10^{-3}	1.6×10^{-2}	1.6×10^{-2}	2.0×10^{-2}	2.0×10^{-2}	2.0×10^{-2}
0.5	1.3×10^{-2}	1.9×10^{-2}	4×10^{-2}	4.0×10^{-2}	6×10^{-2}	6×10^{-2}	6×10^{-2}
1	2.6×10^{-2}	3.3×10^{-2}	8.8×10^{-2}	8.5×10^{-2}	1.5×10^{-1}	1.8×10^{-1}	1.8×10^{-1}
2	5×10^{-2}	5.6×10^{-2}	1.8×10^{-1}	1.7×10^{-1}	3.2×10^{-1}	4.0×10^{-1}	4.0×10^{-1}
5	1.1×10^{-1}	1.1×10^{-1}	4.2×10^{-1}	5.7×10^{-1}	6.9×10^{-1}	7.9×10^{-1}	7.9×10^{-1}
10	2.4×10^{-1}	3×10^{-1}	8×10^{-1}	9.3×10^{-1}	9.3×10^{-1}	9.5×10^{-1}	9.5×10^{-1}

TABLE 4

COLLISION EFFICIENCY SOURCES

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	Extrapolation of			Interception effect only —			
0.02	data of Davis, et al.			Potential Flow			
0.05							
0.1							
0.2				Extrapolation of data			
0.5				of Mason			
1							
2				Actual data of Mason			
5	Actual data of						
10	Davis, et al.						

efficiencies for the various size ranges. By considering the inertia parameter (Stokes number) of the smaller droplets, the results of Davis, et al. (1970) (water droplet-drop system) were approximately converted to the AgI particle and water drop system (i.e., assuming the AgI particles are spherical and have a density of 5.7 g/cc). This approximation was only used at conditions where the smaller droplet or particle perturbs the flow field to a small degree and where the relative velocity, U, is effected only by a small amount (i.e., radius ratios of, say, less than 0.25). The values of Davis, et al. were adjusted (i.e., a shift of 2.4 r_1 of the abscissa in Figure 7) to those values shown in Table 5. It can be seen that collision efficiencies sufficient for contact nucleation are obtained for drops of 10 to 50 μ radii interacting with small particles (e.g., a collision efficiency of 3.6 X 10^{-6} is obtained for a 0.01 μ radius particle and a 10 μ radius drop).

The theoretical collision efficiencies of Mason (1957) for the water droplet and drop system were used for our puroses in the case of larger drops (r_2 = 100 to 1000 μ). Mason considered in his calculations the interception effect, the inertia of the small droplets, and Langmuir's interpolation formula between viscous and potential flow. Again, by extrapolation, in Figure 7, and by considering Stokes number, collision efficiencies sufficiently large enough for contact nucleation were obtained from Mason's data for AgI particles down to a radius of 0.2 μ (see Table 5).

Collision efficiencies for smaller particles (0.01 to 0.1 μ) interacting with drops of 100 to 1000 μ were calculated by considering the interception effect only. For example, a collision efficiency of 3 x 10^{-5} was found for 0.01 μradius particle and a 1000 μ radius drop (Note that values shown in Tables 3 and 5 are linear collision efficiencies). The interception-only values in Table 3 or 5 are for the case of potential flow around the water drop. The Reynolds numbers for the 100 and 200 μ radius drops are 7 and 35, respectively. At these Reynolds numbers the flow around the drop is still somewhat viscous. Table 6 shows linear collision coefficients for interception only in the case of viscous flow, and the values can be seen to be lower than those in Table 3 or 5 which are based on potential flow. Thus, collision efficiencies for interception-only in Table 3 or 5 should be a little lower for the 100 and 200 μ radius drops (especially for the 100 μ radius drops). For this intermediate hydrodynamic region, one might interpolate between potential and viscous flow as Langmuir did.

Walton and Woolcock (1960) found experimental collision efficiencies for 1.25 and 2.5 μ radius spherical particles interacting with 250 to 1000 μ radius water drops having flow Reynolds numbers ranging from 70 to 870. Their values tend to be lower than the theoretical collision efficiencies derived by Langmuir (1948) for potential flow. Conversely, Ranz and Wong (1952) found higher experimental collision efficiencies than those given by Langmuir for potential flow. They used a sphere of radius 450 μ (Reynolds number of the flow ranged from 650 to 8000) as the collector of 0.18 to 0.65 μ radius droplets. Thus, the experimental results of Walton and Woolcock may be lower because of viscous effects at the lower Reynolds numbers. This observation supports the above discussion in regards to the collision efficiencies based on

TABLE 5

LISTING OF LINEAR COLLISION EFFICIENCIES

Y_c for AgI Particle and Water Drop System

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	1.9×10^{-3}	2.4×10^{-3}	2.4×10^{-3}	1.7×10^{-2}	1.2×10^{-2}	7.7×10^{-3}	5.5×10^{-3}
0.02	3.3×10^{-3}	3.6×10^{-3}	3.6×10^{-3}	2.5×10^{-2}	1.7×10^{-2}	1.1×10^{-2}	7.7×10^{-3}
0.05	7.1×10^{-3}	7.6×10^{-3}	9.5×10^{-3}	3.9×10^{-2}	2.7×10^{-2}	1.7×10^{-2}	1.2×10^{-2}
0.1	1.2×10^{-2}	1.3×10^{-2}	1.9×10^{-2}	5.5×10^{-2}	3.9×10^{-2}	2.5×10^{-2}	1.7×10^{-2}
0.2	1.9×10^{-2}	2.2×10^{-2}	3.8×10^{-2}	4.0×10^{-2}	5.4×10^{-2}	5.4×10^{-2}	5.4×10^{-2}
0.5	3.1×10^{-2}	4.5×10^{-2}	9.5×10^{-2}	1.0×10^{-1}	1.9×10^{-1}	2.3×10^{-1}	2.3×10^{-1}
1	6.2×10^{-2}	7.9×10^{-2}	2.1×10^{-1}	2.0×10^{-1}	3.7×10^{-1}	4.8×10^{-1}	4.8×10^{-1}
2	1.2×10^{-1}	1.3×10^{-1}	4.3×10^{-1}	5.6×10^{-1}	6.7×10^{-1}	7.7×10^{-1}	7.7×10^{-1}
5	1.1×10^{-1}	2.6×10^{-1}	9.5×10^{-1}	9.6×10^{-1}	9.7×10^{-1}	9.8×10^{-1}	9.8×10^{-1}
10	2.4×10^{-1}	3×10^{-1}	1.4	1.3	1.3	1.06	1.02

Stokes number considered in Mason's and Davis's data.

<u>TABLE 6</u>

LINEAR COLLISION EFFICIENCIES
FOR INTERCEPTION ONLY WITH VISCOUS FLOW

r_1	r_2						
u	10	20	50	100	200	500	1000
0.01				1.6×10^{-4}	6×10^{-5}	2.4×10^{-5}	1.2×10^{-5}
0.02				3×10^{-4}	1.6×10^{-4}	4.9×10^{-5}	2.4×10^{-5}
0.05				6.5×10^{-4}	2.5×10^{-4}	1.2×10^{-4}	6×10^{-5}
0.1				1.4×10^{-3}	7×10^{-4}	2.5×10^{-4}	1.2×10^{-4}
0.2							
0.5							
1							
2							
5							
10							

interception-only for 100 to 200 μ radius drops.

It must be emphasized here that the collison efficiencies for the larger drops, especially those having radii greater than 200 μ, may be minimum values since all calculations have been based on collection on the front side of the drop. Engelmann (1965), Asset and Hutchins (1967) and others have reported collection on the back side of the drop in greater amounts than that on the front side. Asset and Hutchins experimentally have found particles having radii of 2.5 μ to collected 131 times as much on the back side as on the front side. Their results were found with a glass rod having a 2.1-cm diameter which was mounted in a wind tunnel with a wind velocity of 8 m sec^{-1}., i.e., the flow around the cylinder is considered to be potential. Also this ratio of collection on the back side to the front side increased with decreasing particle size.

The above results may be reasonable since it is hypothesized here that drops having Reynolds numbers large enough to have a viscous boundary layer will have collection on the back side. Incidentally, Langmuir failed for simplification purposes to consider the boundary layer in his collision efficiency calculations for potential flow. Particles too small to collide on the front side will penetrate into the viscous boundary layer to a degree depending on their size. Particles which penetrate by the correct degree would then be swept around into the wake. The lower resistance in the wake would allow Brownian diffusion and electrostatic attraction, which had a smaller effect on the front side, to deposit particles on the back side.

Davis (1970) does not calculate collision efficiencies for drops smaller than 5 or 10 μ in radius. Because the drag forces rapidly decrease with decrease in radius, the calculation for smaller drops becomes overwhelmingly sensitive. Since we have need of order of magnitude collision efficiencies for water drops of 1, 2 and 5 μ, the curves of Figure 7 have been cross-plotted and the resulting curves of linear collision efficiency versus the drop diameter, r_2, appear in Figure 8 for particle radii ranging from 0.05 to 2 μ. The curves have been roughly extrapolated out to $r_2 = 1$ μ. The linear collision efficiencies are tabulated in Table 7 for 1, 2, and 5 μ. radius drops. It must be remembered that these are order of magnitude values especially for the particles with $r_1 < 0.5$ μ. These low values of collision efficiency for drops of radii under 10 μ were not used in the following work because of uncertainties in the values. Also they were not necessary because of the small drops and low velocities, and thus, Brownian diffusion would predominate over impact capture to a large degree.

Table 8 gives values of E, rather than Y_c. These are illustrated graphically in Figure 9.

Values of $N/n_a n_d$ are shown in Table 9 and Figure 10. This figure is used in a manner similar to that for Brownian motion.

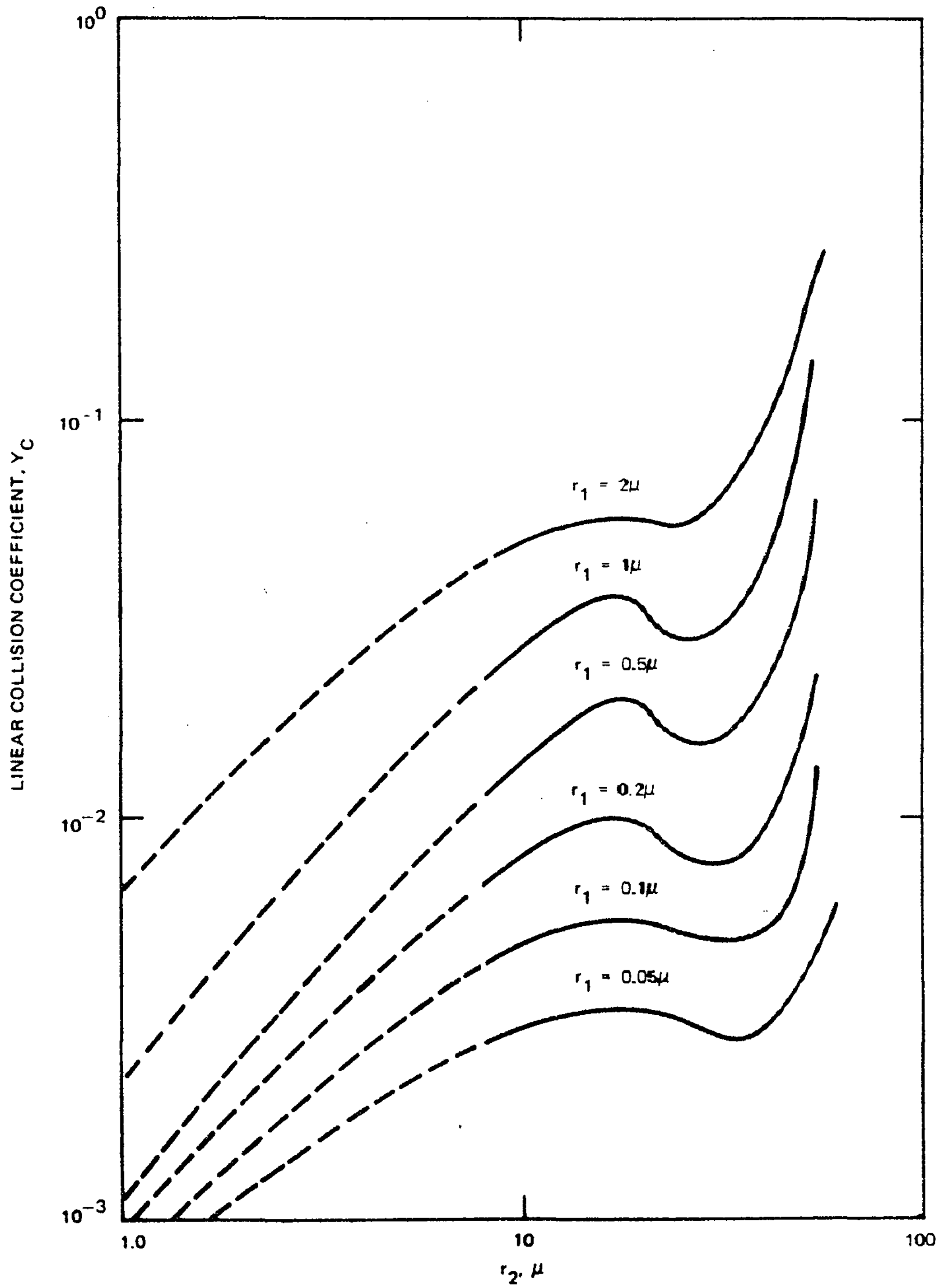


Fig. 8. Y_c as a function of r_2 found by cross plotting Fig. 7.

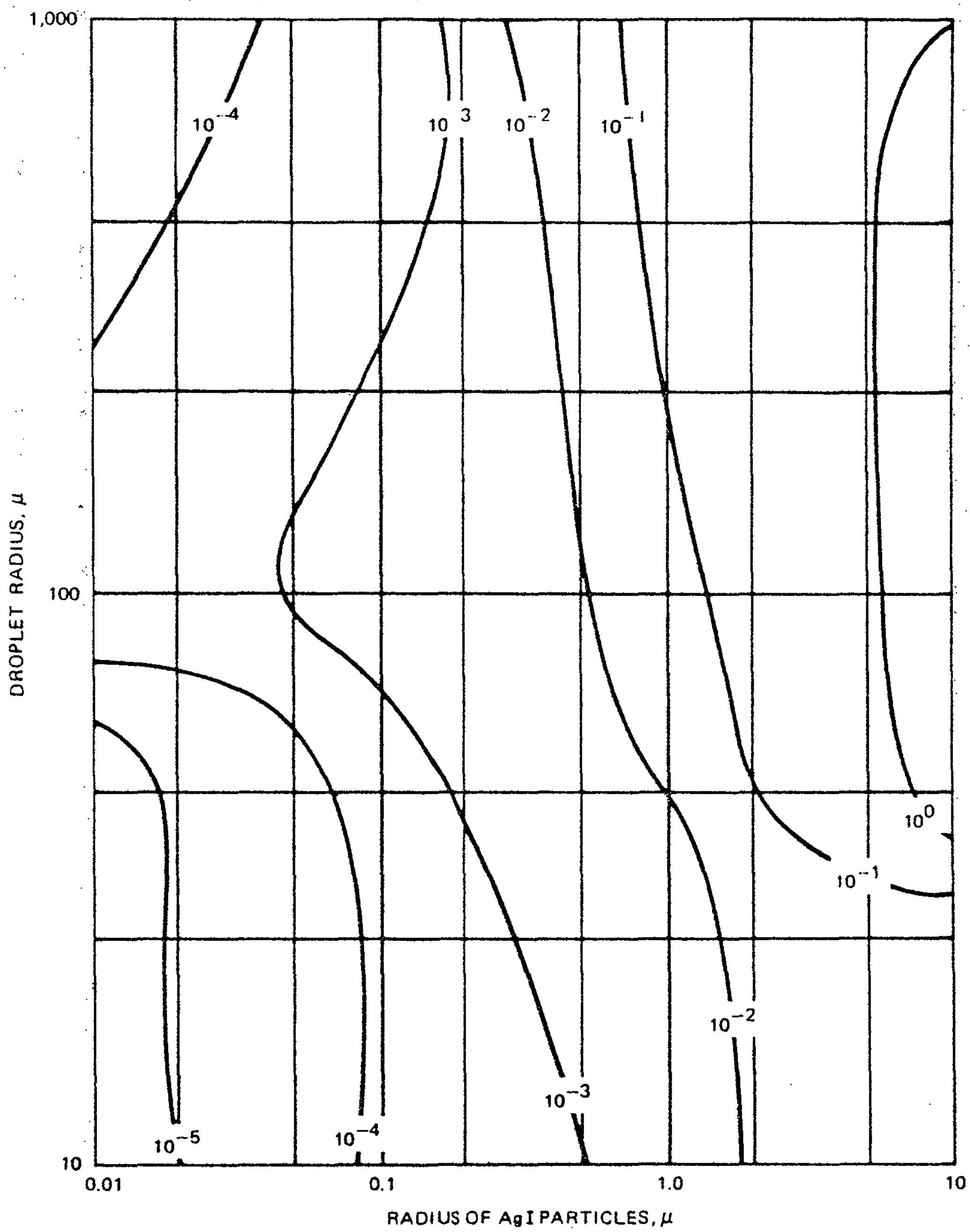


Fig. 9. Values of E as function of r_a and r_d.

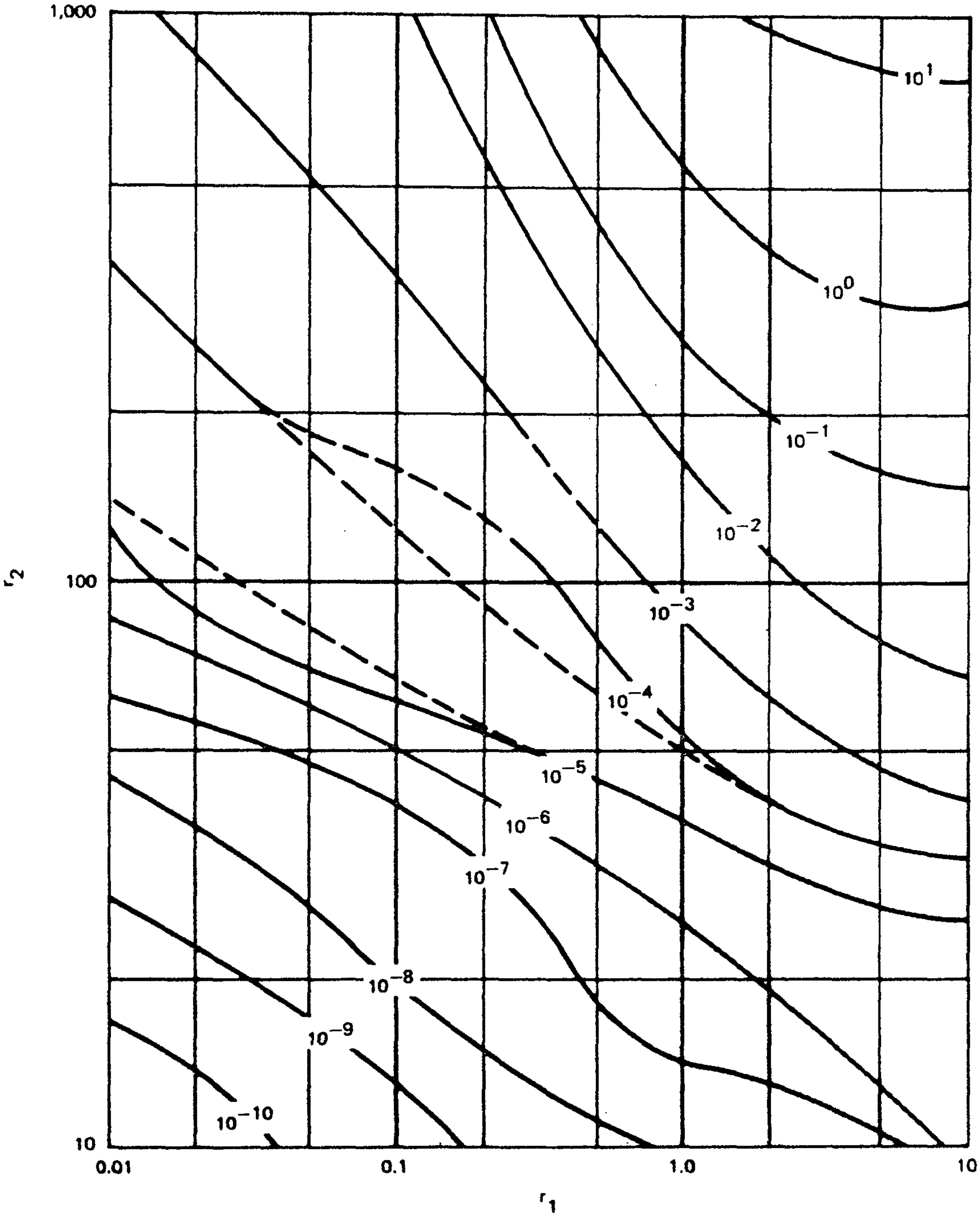


Fig. 10. Values of $\frac{N}{n_a\, n_d}$ as function of r_1 and r_2.

TABLE 7

LINEAR COLLISION EFFICIENCIES OF
FIGURE 8 FOR 1, 2, and 5 μ RADIUS DROPS

r_1	r_2		
μ	1	2	~~3~~ 5
0.05	7×10^{-4}	1.2×10^{-3}	2.2×10^{-3}
0.1	8×10^{-4}	1.5×10^{-3}	3.1×10^{-3}
0.2	1×10^{-3}	1.9×10^{-3}	4.6×10^{-3}
0.5	1.1×10^{-3}	2.5×10^{-3}	7.0×10^{-3}
1.0	2.2×10^{-3}	5.0×10^{-3}	1.4×10^{-2}
2.0	6.5×10^{-3}	1.4×10^{-2}	3.1×10^{-2}

TABLE 8

COLLISION EFFICIENCY E OR Y_c^2 FOR THE AgI PARTICLE AND WATER DROP SYSTEM

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	3.6×10^{-6}	5.6×10^{-6}	5.6×10^{-6}	2.9×10^{-4}	1.4×10^{-4}	5.9×10^{-5}	3.0×10^{-5}
0.02	1.1×10^{-5}	1.3×10^{-5}	1.3×10^{-5}	6.2×10^{-4}	2.9×10^{-4}	1.2×10^{-4}	5.9×10^{-5}
0.05	5.0×10^{-5}	5.7×10^{-5}	9.0×10^{-5}	1.5×10^{-3}	7.2×10^{-4}	2.9×10^{-4}	1.4×10^{-4}
0.1	1.4×10^{-4}	1.7×10^{-4}	3.6×10^{-4}	3.0×10^{-3}	1.5×10^{-3}	6.2×10^{-4}	2.9×10^{-4}
0.2	3.6×10^{-4}	4.8×10^{-4}	1.4×10^{-3}	1.6×10^{-3}	2.9×10^{-3}	2.9×10^{-3}	2.9×10^{-3}
0.5	9.5×10^{-4}	2.0×10^{-3}	9.0×10^{-3}	1.0×10^{-2}	3.6×10^{-2}	5.3×10^{-2}	5.3×10^{-2}
1	3.8×10^{-3}	6.2×10^{-3}	4.4×10^{-2}	4.0×10^{-2}	1.4×10^{-1}	2.3×10^{-1}	2.3×10^{-1}
2	1.4×10^{-2}	1.7×10^{-2}	1.8×10^{-1}	3.1×10^{-1}	4.5×10^{-1}	5.9×10^{-1}	5.9×10^{-1}
5	1.2×10^{-2}	6.7×10^{-2}	9.0×10^{-1}	9.1×10^{-1}	9.3×10^{-1}	9.5×10^{-1}	9.5×10^{-1}
10	5.7×10^{-2}	9.0×10^{-2}	2.0×10^{0}	1.7×10^{0}	1.4×10^{0}	1.1×10^{0}	1.0×10^{0}

TABLE 9

NUMBER, N, OF COLLISION BETWEEN AgI PARTICLES OF RADIUS, r_1, AND WATER DROPLETS OF RADIUS, r_2, PER DROPLET, PER AEROSOL PARTICLE PER SECOND ACCORDING TO $\frac{N}{n_a n_d} = E (V_a - V_a) (r_1 + r_2)^2 \pi$

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	1.3×10^{-11}	3.7×10^{-10}	1.1×10^{-8}	6.6×10^{-6}	2.8×10^{-5}	1.9×10^{-4}	6.2×10^{-4}
0.02	4.1×10^{-11}	8.5×10^{-10}	2.5×10^{-8}	1.4×10^{-5}	5.8×10^{-5}	2.8×10^{-4}	1.2×10^{-3}
0.05	1.9×10^{-10}	3.7×10^{-9}	1.8×10^{-7}	3.4×10^{-5}	1.4×10^{-4}	9.6×10^{-4}	2.9×10^{-3}
0.1	5.3×10^{-10}	1.1×10^{-8}	7.1×10^{-7}	6.8×10^{-5}	3.0×10^{-4}	2.0×10^{-3}	6.0×10^{-3}
0.2	1.4×10^{-9}	3.2×10^{-8}	2.8×10^{-6}	3.6×10^{-5}	5.8×10^{-4}	9.6×10^{-3}	6.0×10^{-2}
0.5	4.3×10^{-9}	1.4×10^{-7}	1.8×10^{-5}	2.3×10^{-4}	7.2×10^{-3}	1.7×10^{-1}	1.1×10^{0}
1	1.6×10^{-8}	4.4×10^{-7}	8.9×10^{-5}	9.2×10^{-4}	2.8×10^{-2}	7.6×10^{-1}	7.1×10^{0}
2	5.8×10^{-8}	1.3×10^{-6}	3.8×10^{-4}	6.9×10^{-3}	9.2×10^{-2}	1.9×10^{0}	$1.2 \times 10^{+1}$
5	*4.3×10^{-8}	*4.6×10^{-6}	2.0×10^{-3}	2.2×10^{-2}	1.9×10^{-1}	3.2×10^{0}	$2.0 \times 10^{+1}$
10	*4.0×10^{-6}	*4.0×10^{-6}	4.1×10^{-3}	4.1×10^{-2}	3.0×10^{-1}	3.7×10^{0}	$2.1 \times 10^{+1}$

* AgI overtakes the drops.

Table 10 and Figure 11 include the effects of Brownian diffusive capture and orthokinetic impact capture. This does not include any synergism that may exist between the two, but only the combined effects. If the aerosol are of less than 0.1 μ, the effect of drop size is only three orders of magnitude or so. If the particles have a chance to grow embryos by diffusional processes, to radii greater than one micron, the capture probability improves markedly.

Table 11 shows the number of AgI particles per cm^3 necessary to cause a number of contacts in one second equal to the number of drops present, so long as the number of drops per cm^3 is small compared to the number of particles per cm^3. Within limits, this is independent of the liquid water content, so long as the water cloud is monodispersed. By inspection, the number of nuclei of 0.05 μ radius, large enough to cause freezing at -5^0C is 5 orders of magnitude larger for a cloud with 10 micron radius droplets than for one containing 1-mm radius droplets. Values for necessary nuclei concentration are not available for smaller droplets because of uncertainties in the values of E but these are much larger than for 10 μ drops.

ELECTROPHORESIS:

Electrophoresis is the tendency of particles to move under the influence of electrically produced forces. If a water drop is in the vicinity of a charged aerosol particle or vice versa, the two will be attracted if the charges are unlike, or at a distance repelled if the charges are like, on the other hand, the development of an induced charge in the larger particles may at very short distances cause like charged particles to attract each other. Work by Cochet (1952) and Levin (1954) address the problem. It appears that unlike charges can, for large charges, cause the collection efficiency, E, used in Smoluchowski's equation to rise rapidly. In the case of 5 μ drop, mutually attracted to a 20 μ drop and having a charge of 4×10^{-4} esu C.G.S.; E approaches 100. This is a very large charge, however.

No values of charge are available for AgI nuclei, although those produced by pyrotechnics appear to be positively charged by pyrolytic stripping of electrons. No attempt will be made here to evaluate the effects of electrophoresis due to charges except to point out that the collection coefficients used in the preceeding section will all be increased by any charge on the smoke particles.

DIFFUSIOPHORESIS:

As a droplet grown, water vapor is deposited thereon in liquid form. The flow of water vapor towards the growing drop pushes particles of micron and sub-micron size towards the drop. Conversely, an evaporating drop has an efflux of water vapor that tends to push such particles away. Goldsmith, Delafield and Cox (1962) experimentally estimated the effect of water vapor pressure gradients upon the velocity of such particles and then extended the results theoretically to calculate the washout of radioactive particles from the air by diffusiophoresis.

TABLE 10

BROWNIAN AND IMPACT, COLLISIONS PER SECOND PER DROP PER PARTICLE

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	1.70×10^{-6}	3.39×10^{-6}	8.49×10^{-6}	2.36×10^{-5}	5.16×10^{-5}	2.75×10^{-4}	7.90×10^{-4}
0.02	4.51×10^{-7}	9.04×10^{-7}	2.28×10^{-6}	1.85×10^{-5}	6.70×10^{-5}	3.02×10^{-4}	1.25×10^{-3}
0.05	8.59×10^{-8}	1.75×10^{-7}	6.09×10^{-7}	3.49×10^{-5}	1.42×10^{-4}	9.64×10^{-4}	2.90×10^{-3}
0.1	2.84×10^{-8}	6.66×10^{-8}	8.49×10^{-7}	6.83×10^{-5}	3.00×10^{-4}	2.00×10^{-3}	6.00×10^{-3}
0.2	1.20×10^{-8}	5.31×10^{-8}	2.85×10^{-6}	3.61×10^{-5}	5.80×10^{-4}	9.60×10^{-3}	6.00×10^{-2}
0.5	7.92×10^{-9}	1.47×10^{-7}	1.81×10^{-5}	2.30×10^{-4}	7.20×10^{-3}	1.70×10^{-1}	1.10×10^{0}
1	1.78×10^{-8}	4.43×10^{-7}	8.90×10^{-5}	9.20×10^{-4}	2.80×10^{-2}	7.60×10^{-1}	7.10×10^{0}
2	5.89×10^{-8}	1.30×10^{-6}	3.80×10^{-4}	6.9×10^{-3}	9.20×10^{-2}	1.90×10^{0}	$1.20 \times 10^{+1}$
5	4.35×10^{-8}	4.6×10^{-6}	2.00×10^{-3}	2.2×10^{-2}	1.90×10^{-1}	3.20×10^{0}	$2.00 \times 10^{+1}$
10	4.10×10^{-6}	4.0×10^{-6}	4.10×10^{-3}	4.1×10^{-2}	3.00×10^{-1}	3.7×10^{0}	$2.1 \times 10^{+1}$

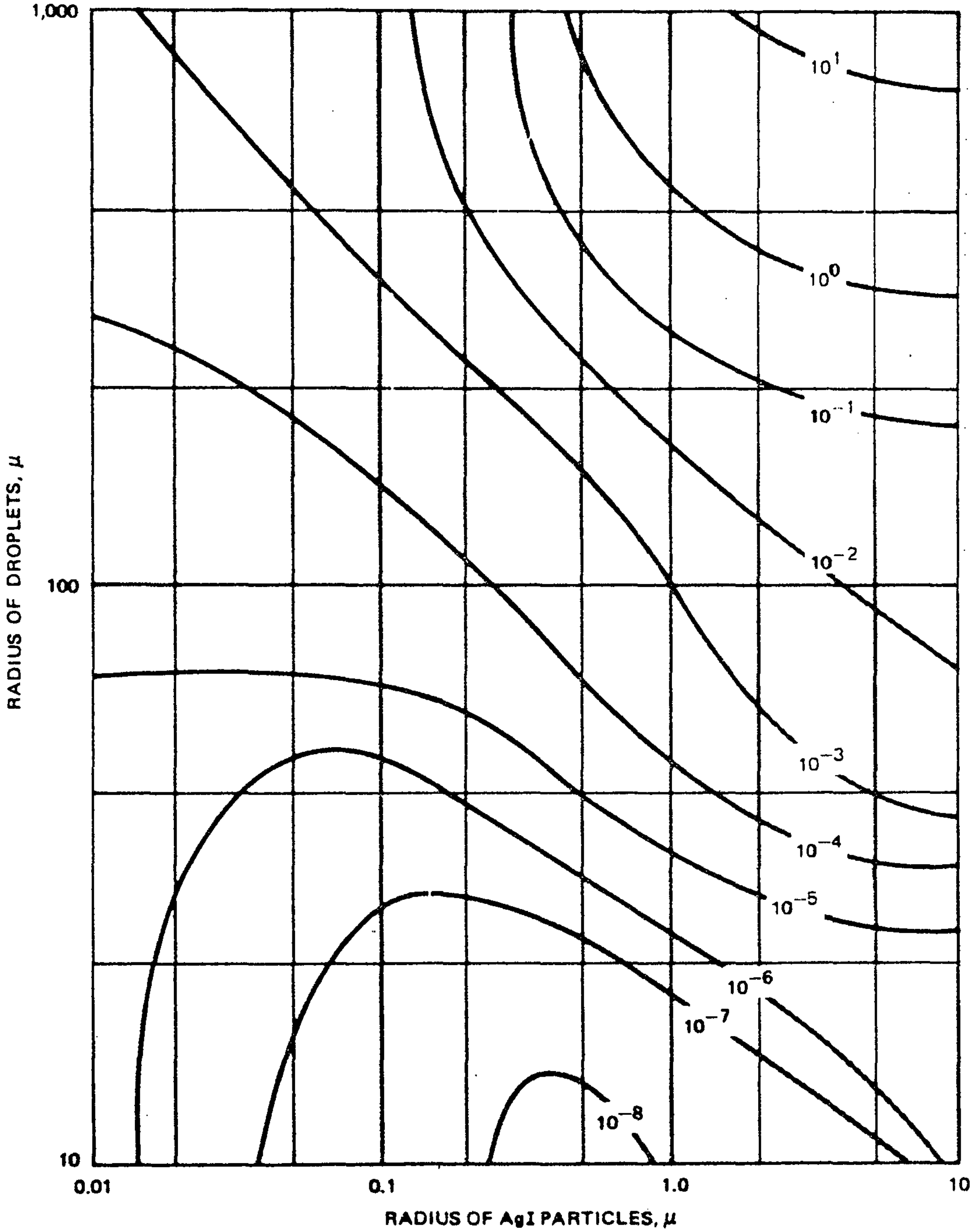


Fig. 11. Values of $N/n_a n_d$ for combined Brownian and Smoluchowskian collisions.

TABLE 11

NUMBER OF NUCLEI REQUIRED TO FREEZE ALL DROPS FOR MONODISPERSED CLOUD BY BROWNIAN AND IMPACT COLLISION

r_1	r_2						
μ	10	20	50	100	200	500	1000
0.01	5.9×10^5	3.0×10^5	1.2×10^5	4.2×10^4	1.9×10^4	3.6×10^3	$1.3 \times 10^{+3}$
0.02	2.2×10^6	1.1×10^6	4.4×10^5	5.4×10^4	1.5×10^4	3.3×10^3	8.0×10^2
0.05	1.2×10^7	5.7×10^6	1.6×10^6	2.8×10^4	7.0×10^3	1.0×10^3	3.4×10^2
0.1	3.5×10^7	1.5×10^7	1.2×10^6	1.5×10^4	3.3×10^3	5.0×10^2	1.7×10^2
0.2	8.3×10^7	1.9×10^7	3.5×10^5	2.8×10^4	1.7×10^3	1.0×10^2	1.7×10^1
0.5	1.3×10^8	6.8×10^6	5.5×10^4	4.2×10^4	1.4×10^2	5.9×10^0	9.1×10^{-1}
1	5.6×10^7	2.2×10^6	1.1×10^4	1.1×10^3	3.6×10^1	1.3×10^0	1.4×10^{-1}
2	1.7×10^7	7.7×10^5	2.6×10^3	1.4×10^3	1.1×10^1	5.3×10^{-1}	8.3×10^{-2}
5	2.3×10^7	2.2×10^5	5.0×10^2	4.5×10^2	5.3×10^0	3.1×10^{-1}	5.0×10^{-2}
10	2.4×10^5	2.5×10^5	2.4×10^2	2.4×10^2	3.3×10^0	2.7×10^{-1}	4.8×10^{-2}

They determined, on p. 57 of this paper, that

$$N = \frac{K^1}{D} \; (n_a n_d m) \qquad (34)$$

where N is the total number of aerosol particles removed from air containing n_a aerosol particles per cm^3, and n_d droplets per cm^3, having grown to mass m. K^1 is a constant equal to -2.55×10^2, and D is the diffusivity of water vapor, 2.4×10^{-1}.

Hence

$$N = 1.06 \times 10^3 \; n_a \; n_d \; m \qquad (35)$$

The liquid water content in grams per cm^3 is $n_d m$, hence

$$N = 1.06 \times 10^{-3} \; n_a \; LWC \qquad (36)$$

where LWC is the liquid water content in grams per cubic meters.

Thus, a cloud having a liquid water content of 1 gram per m^3 and an aerosol density of 10^5 particles per cubic centimeter will have removed some 10^3 particles per cm^3 or about one particle per drop by diffusiophoresis alone. One such particle is adequate to cause a drop to freeze if the temperature is correct.

Table 12, taken from their paper, shows the "Efficiency," E, given as the portion of the aerosol scavenged by droplets, assuming 100 droplets per cm^3.

TABLE 12

SCAVENGING RATIO FOR DIFFUSIOPHORESIS

r, radius of droplets, micron	E, "Efficiency"	LWC g/m^3
1	4.5×10^{-7}	4.2×10^{-4}
5	5.6×10^{-5}	5.2×10^{-2}
10	4.5×10^{-4}	4.2×10^{-1}
20	3.5×10^{-3}	3.3×10^{0}

Thus in growing to 20 μ, the 100 drops, corresponding to a liquid water content of 3.3 grams per cm^3 will remove 350 particles from an aerosol of 10^5 particles per cm^3, or about 3 particles per droplet. These values are typical of what could be expected in a tropical cumulus cloud of medium size.

While diffusiophoresis is not the most effective means of assuring contact nucleation, it is of adequate magnitude to play an important role when clouds are seeded from beneath in updrafts feeding the clouds of that the material is present during droplet growth.

The time rate of contact nucleation by this process is difficult to estimate because the rate of mass accumulation by droplets is extremely variable and depends upon cloud type and history, notably on updraft velocity and duration. It would be important for cumulus clouds but relatively unimportant for stable stratus. It is not affected by droplet number alone, but only on the increment in the liquid water content.

The diffusiophoretic forces will to some extent aid capture by Brownian diffusion and will increase the impact collection efficiency during periods in which droplets are growing. Hence contact nucleation will be augmented in updrafts and slightly decreased in downdrafts, by the effects of diffusiophoresis.

Vittori and Prodi (1967, p. 538) points out that diffusiophoresis of particles to growing ice crystals is about 16 times that to condensing droplets. Hence, diffusiophoresis is an important factor in removal of glacogenic nuclei from a cloud once freezing has begun.

DIRECT CONDENSATION:

Fletcher (1958a) shows that for condensation to occur at all on AgI that a small supersaturation must exist. Such a supersaturation does exist in most updrafts and in an atmosphere with a paucity of condensation nuclei.

Mordy (1959) and others have shown that particles as small as 0.1 μ in radius or even 0.05 may serve as condensation nuclei provided that there are few nuclei and that a sufficiently strong updraft to assure supersaturation is present. We, St.-Amand, 1967, have examined the effects of adsorption on AgI and of hygroscopic contamination of AgI particles in which it is clear that condensation can take place on even small particles of AgI.

Thus, for material injected below cloud base in relatively clear air most of the particles in an aerosol distribution such as those from a pyrotechnic will produce water droplets that will freeze upon reaching an altitude at which the temperature falls below the activation temperature for the glacogenic agent. Some of the smaller particles will dissolve but the larger ones will survive. Freezing of the already treated droplets will be an important source of heat release because the droplets have had time to grow to a reasonable size.

Some of th AgI particles will dissolve totally in raindrops. The solubility of AgI is not well known but some values are available in the literature. Using these and extrapolating one gets 3×10^{-9} grams of AgI per gram of water at $+30^{o}C$, 2×10^{-9} at $+20^{o}C$, 1.2×10^{-9} at $10^{o}C$ and 1×10^{-10} at $0^{o}C$.

Using these values the curves of Fig. 12 were calculated as follows:

$$C = \frac{MAgI}{MH_2O} = \frac{\frac{4}{3}\ r_a^{\ 3}\ P_a}{\frac{4}{3}\ r_d^{\ 3}\ P_d} = \frac{r_a^{\ 3}\ P_a}{r_d^3}$$

and

$$r_a = \left[\frac{C}{P_a}\right]^{\frac{1}{3}} r_d$$

where C is the solubility of AgI in water

r_a is the radius of a completely soluble spherical AgI particle

r_d is the radius of a drop of pure water

P_a is the density of AgI, 5.67 grams/ cm^3

P_d is the density of H_2O, taken as 1.00.

All of the particles will lose some mass if imbedded in drops too warm to freeze. Depending on the drop and particle size some will dissolve completely. These are not toally lost because as the drops cool during ascent, some of the AgI will be precipitated because the solubility decreases with decreasing temperature. The precipitated AgI will be in a colloidal form and freezing temperature will be determined by the largest particle precipitated.

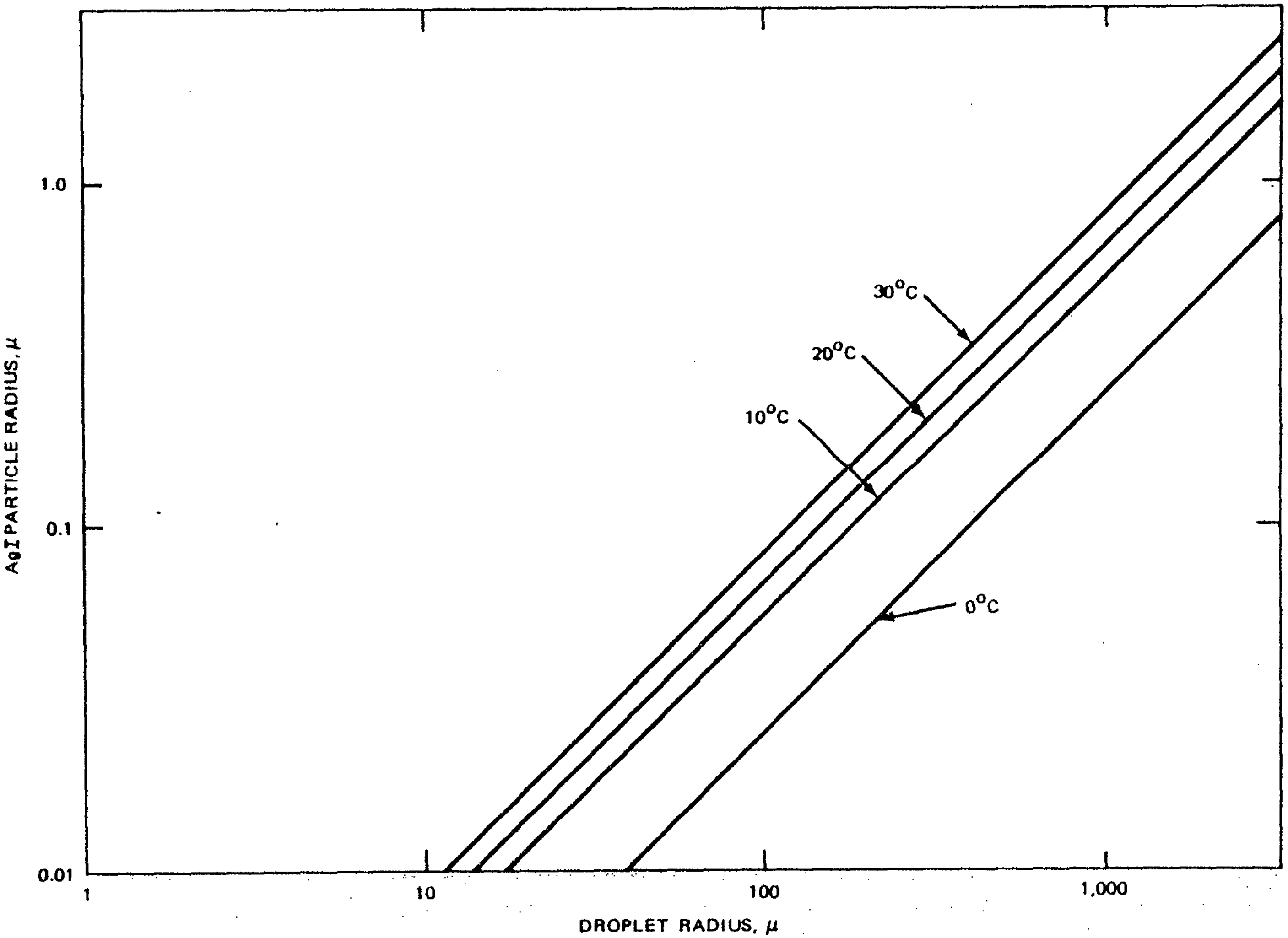


Fig. 12. Radius of AgI particle that will dissolve in droplets of pure water of given radius.

For seeding from the ground or from beneath cloud base, it is clear that larger particles are needed than when it is injected at temperatures below the activation temperature. Indeed, very high yield generators giving 10^{16} per gram may well be totally ineffective for seeding from the ground because of the solubility of the very fine AgI. In any case, the numerical effectiveness in the cold part of the cloud will be different than it would be in a cold box test.

If the material is injected at below freezing temperatures, condensation followed by freezing will occur at temperatures above $-12.5^{\circ}C$, but this process will not proceed fast enough to compete with the vapor demand of already frozen droplets and we are back in the diffusional growth regime discussed earlier in this paper. Capture of such particles by impact will be rapid if they have gained enough water to reach radii of a few microns. If the nuclei and droplet concentration is so low as to preclude much contact nucleation, this process, in this temperature regime, will result in slow growth of ice crystals.

Recent work by Roger Cheng (1970) at State University of New York at Albany, shows clearly that upon freezing of large drops, a number of small drops are formed upon a microscope slide in the vicinity of the large drop undergoing freezing on the slide. Cheng attributes this to ejection of the smaller droplets. Whether or not he is correct in his interpretation, the freezing of the large drop produces a temperature change in the drop, raising its temperature to near the triple point. This produces a momentary supersaturation in the vicinity of the freezing drop well above that of the surroundings. This supersaturation can be as high as 15 to 20 percent and will cause activation of many nuclei too small to function at any reasonable temperature or saturation, thus leading to condenation and freezing on many nuclei that otherwise could not function. The mass of the large drop causes it to have a terminal velocity high compared to the smaller particles so that they would be able to grow once the big frozen drop had fallen away.

APPLICATIONS TO CLOUD SEEDING

We have reviewed the nucleation behavior, chemistry and contact mechanisms of some ice producing agents. It remains to be seen how this technology should be applied to actual clouds. The techniques of seeding will be included in another paper in this series and only a few comments made here.

Stratus Clouds:

Stratus clouds, such as cold fogs, are often seeded, either to produce clearings or to induce precipitation. While some stratus have relatively large amounts of liquid water, the usual cold fog has only about 0.1 grams of liquid water per cubic meter and the drop sizes peak at 7 to 10 microns or perhaps less. If one is not in a hurry this type of cloud could be seeded with just enough glocogenic nuclei to cause all the water, liquid and vapor down to the level of saturation over ice to be deposited as diffusionally grown ice crystals. This process requires a considerable time, and the fall of ice from the top of the cloud would probably precede the complete growth of the ice crystals to the point of using up all the water.

If on the other hand, 10^7 nuclei were introduced per cubic centimeter, almost all the drops would be frozen in one second. These drops would more or less rapidly adsorb by diffusion enough water to grow large enough to begin to fall effectively in a few minutes. The size to which they grew would depend upon the temperature because the vapor pressure differential depends upon the temperature, and the only water available for growth would be that in the vapor phase between supersaturation over water and that over ice.

If freezing occurred at 0^{o}C, there would be no crystal growth at all because the vapor pressures are the same. At -5^{o}C the excess vapor density over ice at sea level is about 0.4 grams/m^3. Thus freezing of all droplets would, on an average, result in quintupling of the mass of the frozen droplets, resulting in a radius of 1.7 times the original. Hence in a cloud of 7 to 10 micron drops one could expect the fall rate to increase from about 1 cm/sec balanced by rising air, to about 5 cm per second airspeed or a net velocity increase to 4 cm/sec. Thus, a 300 meter thick stratus so seeded would require some 16,000 seconds or 100 minutes to fall away, barring aggregation of droplets due to electrical forces. Coagulation due to gravity would not be important because of the small differential in particle size. This is close to the 45 minutes usually observed.

New moisture introduced from below would bring in some 4×10^{-3} gms per second per square meter available water vapor so that growth from the bottom would be a little faster than in a totally static situation.

The tactic in this case would be to make a mini-max decision using fewer nuclei than that necessary to nucleate all the drops in one second. If only 10^5 nuclei were introduced only one percent of all the drops would be nucleated in one second and these would grow more rapidly than the others. resulting in destruction of some of the unfrozen drops by evaporation. The excess nuclei would not grow embryos of any great size by diffusion so that the presence of a large number of pure AgI nuclei in this temperature regime would not make any difference in visibility.

The exact solution of this problem would depend upon computer analysis and we will not discuss it further here except to point out that the balance between liquid water available and vapor water will make an enormous difference in response of the cloud, depending on mainly how the liquid water is distributed in droplets.

Cumulus Clouds:

Seeding of cumulus clouds to enhance rainfall is, because of wind shear, a complicated problem that will be discussed in a separate publication. In general, the tactic is to seed in updrafts with relatively small amounts of material. Updrafts, at the freezing level for tropical clouds with bases in the vicinity of 5,000 feet contain large droplets, high liquid water contents and between the drops some degree of supersaturation.

Droplets of the order of 200 microns radius will remain suspended in updrafts of 300 feet per second, and even larger drops, up to 1-mm in radius, are frequently observed striking aircraft windshields in higher updrafts. No really good measurements are available from the updraft cores, but the drop sizes are definitely larger there. Liquid water commonly ranges between 1.5 and 3.5 grams/m^3, occasionally rising as high as 5 grams/m^3, in tropical clouds of the sort encountered in the Philippines.

Under this regime only a few hundred to a few thousand nuclei per cm^3 are necessary to cause nucleation of almost all the drops within a second. This results in release of heat of the order of 100 to 400 calories per cubic meter in a few seconds, followed by release of a great deal more in a matter of minutes by condensation as moist air is relatively rapidly carried past the frozen particles.

In the downdrafts and outlying portions of the cloud, droplets are generally of the order of 10 to 20 microns hence seeding in these places requires some 10^7 particles/cm^3 to release much less heat in the same time period.

Hence in seeding a cumulus, one does just as well from a standpoint of heat release to seed only the updrafts because the change in buoyancy will come only from the parts of the cloud containing the largest drops. In addition, release of heat in downdrafts may well serve to disturb the delicate balances in the circulation within the cloud.

In regions of high air pollution where many more condensation nuclei are available than in clean air, drops do not grow as large in a cumulus updraft and so nucleation should be much slower. It is often observed that in continental type clouds much higher seeding doses are required than are needed in maritime clouds or in tropical clouds.

Clearly seeding dosages of the commonly used 200 particles per liter will be of no great use in nucleating cumulus clouds, except in updrafts where some contact nucleation can occur. Elsewhere, as in downdrafts or in virtually stationary air so few contacts take place that ice growth occurs only by diffusion onto the nucleating agent and is hence much slower than it is on frozen droplets. Heat released by slow growth is soon lost by radiation and by ventilation so that, even though the same amount of heat be released in the end, the temperature of the air may not rise markedly, and the cloud might well collapse before any notable change could take place.

If seeding from beneath clouds, almost all the nuclei serve to produce water droplets by condensation, and upon arrival at the -2 to -4°C level, the droplets freeze. Those not going into updrafts reaching the freezing level are lost for the time being. Such seeding is effective, but requires skill and judgement in selection of updrafts and estimating the cloud duration so that material will arrive in the freezing regime before the cloud begins to collapse.

The seeding technique we prefer is to drop pyrotechnic flares from the -4°C level in updraft cores. The flares are so constructed as to fall for several thousands of feet so that seeding is done continuously for 5 to 10 minutes as the material is carried upward by the rising air. In this case, much of the AgI arrives already encapsulated in liquid water.

Some compounds such as the KI-AgI complexes work at temperatures of only -1 to -2°C. These can be used on marginally cold clouds but will not work well, if at all, if the agent is permitted to reside for any length of time in the part of the cloud warmer than 0°C, or if long residence in moist air below cloud base is permitted. For this sort of application, that is to say, seeding marginally cold clouds, a compound grain could be used, emitting a small amount of soluble complexing agent in the cold part, and then a relatively pure AgI below that, and perhaps even a third agent capable of assuming condensation on the AgI so that already nucleated drops could be carried up later.

Seeding of cumulus whose bases are warmer than -5°C with any of the complexes now used is not very effective unless the material is implaced at the -5°C level or colder because they go into solution when used below cloud base and only become effective at much colder temperatures. The use of an acetone burner from below cloud base is responsible for most of the uncertainty that has plagued weather modification for the last twenty years. A number of well carried out experiments having no results or even negative results have resulted from this practice because the clouds were not really seeded in excess of what would have occurred from dust and nuclei already present in the atmosphere. Such experiments are valid tests of the particular technique used but should not be interpreted to be a test of weather modification in general.

Seeding with the NH_4I-AgI system in acetone burners will probably result in more striking effects, especially in seeding of potential hail storms from below cloud base. The rain yield from such operations will probably increase markedly. The NH_4I-AgI acetone system will prove very useful in winterstorm seeding for rain production, and will be well suited for clearance of supercooled fog.

In summary, the theory of nucleation is now well enough developed to be used intelligently for decision making even though the details can be elavorated with profit over the years ahead. The chemical nature of the nucleant is extremely important, as is the history and environment of the nucleant from its moment of production until it is used in clouds. The materials should be selected carefully for the type of work and the use should be confined to those situations in which the nucleant can be expected to perform as intended.

REFERENCES:

Adam, J. R. and R. G. Semonin: "An Experimental Determination of the Collection Efficiencies of Raindrops for Submicron Particles," Presented at 1970 Precipitation Scavenging Meeting, Richland, Wash. (June 2-4, 1970)

Asset, G. and T. G. Hutchins, "Leeward Deposition of Particles on Cylinders from Moving Aerosols," AMER IND HYGINE ASSOC. J. (1967), pp. 348-353.

Cheng, R. J., "Generation of Micro-Droplets by Freezing a Supercooled Water Drop," Conference on Cloud Physics, PREPRINTS, Ft. Collins, Colo. (Aug. 24-27, 1970), pp. 53-54.

Cochet, R., "L'evolution d'une goutelette d'eau chargee dans un nauge a temperature positive," ANN GEOPHYS, Vol. 8 (1952), pp. 33-54.

Davis, M. H., "Collisions of Very Small Cloud Drops," J GEOPHYS RES, Vol. 71, (June 1966), pp. 3101-3104.

Davis, M.H., J. D. Klett, and M. Neiburger, "Collision Efficiencies of Cloud Droplets at Small Reynolds Numbers," Conference on Cloud Physics, PREPRINTS, Ft. Collins, Colo. (Aug 24-27, 1970), pp. 115-116.

Davis, M. H., and J. D. Sartor, "Theoretical Collision Efficiencies for Small Cloud Droplets in Stokes Flow," NATURE, Vol. 215 (Sept. 1967) pp. 1371-1372.

Engleman, R. J., "Rain Scavenging of Zn5 Particles," J ATMOS SCI, Vol. 22 (1955), pp. 719-727.

Fletcher, N. H., "Size Effect in Heterogeneous Nucleation," J CHEM PHYS, Vol. 29, No. 3 (1958a), pp. 572-576.

________. "Erratum: Size Effect in Heterogeneous Nucleation," (J CHEM PHYS, Vol. 29, (1958), p. 572), J CHEM PHYS, Vol. 31 (1958b), pp. 1136-1137.

Fletcher, N. H. The Physics of Rainclouds, Cambridge University Press, 1962, 386 pp.

Fuchs, N.A., The Mechanics of Aerosols, MacMillan Co., New York, 1964, 408 pp.

Goldsmith, P., H. J. Delafield, and L. C. Cox, "The Role of diffusiophoresis in the scavenging of radio-active particles from the atmosphere," ROY METEOROL SOC, QUART J, Vol. 89 (1962), pp. 43-61.

Green, H. L. and W. R. Lane, Particulate Clouds: Dusts, Smokes and Mists, 2nd ed., D. Van Nostrad, Inc., Princetone, N. J., 1964, 471 pp. 8 plates.

Greenfield, S. M., "Rain Scavenging of Radioactive Particulate Matter from the Atmosphere," J METEOROL, Vol. 14, No. (April 1957), pp. 115-125.

Hocking, L. M., "The Collision Efficiency of Small Drops," ROY METEOROL SOC, QUART J. Vol. 85 (1959) pp. 44-50.

Klett, J. D., "The Interaction and Motion of Rigid Spheres Falling in a Viscous Fluid at Low Reynolds Numbers," Unpublished PhD. Thesis, UCLA (1968).

Langmuir, I., "The Production of Rain by a Chance Reaction in Cumulus Clouds at Temperatures Above Freezing," J METEOROL. Vol. 5 (1948) pp. 175-192.

Levin, L. M., "The Coagulation of Charged Cloud Droplets," Dak. Akad, Nauk, SSR, Vol. 94 (1954), pp. 467. Trans. T263R of D.R.B. Canada.

Mason, B. J., The Physics of Clouds, Oxford University Press, Inc., New York, 1957, 424 pp.

Millikan, R. A., "The General Law of Fall of a Small Spherical Body Through a Gas, and Its Bearing Upon the Nature of Molecular Reflection from Surfaces," PHYS REV, Vol. 22 (1923), pp. 1-23

Mordy, W. A., "Computations of the Growth by Condensation of a Population of Cloud Droplets, TELLUS, Vol. 11 (1959), pp. 16-42.

Pearcey, T., and G. W. Hill, "A Theoretical Estimate of the Collection Efficiencies of Small Droplets," ROY METEOROL SOC, QUART J. Vol. 83, No. 355 (1957), pp. 77-92; and Vol. 83, No. 358 (1957), pp. 555-556.

Ranz, W. E. and J. B. Wong, "Impaction of Dust and Smoke Particles on Surface and Body Collectors," IND ENG CHEM, Vol. 44 (1952), pp. 1371-1381.

St.-Amand, Pierre, "Nucleation by Silver Iodide and Similar Materials," Skywater Conference, I, PROC, Bureau of REclamation, Denver, Colo. (1967), pp. 305-346.

St.-Amand, Pierre and William G. Finnegan, "The relevance of cloud chamber tests to ice nuclei activity," 2nd National Conference on Weather Modification, PREPRINTS, Santa Barbara, Ca., (April 6-9, 1970), pp. 361-365.

Vittori, Ottavio A., and Vittorio Prodi, "Scavenging of Atmospheric Particles by Ice Crystals," J ATMOS SCI, Vol. 24, (Sept. 1967) pp. 533-538.

Vonnegut, B., "Nucleation of Supercooled Water Clouds by Silver Iodide Smokes," CHEM REV, Vol. 44 (1949), pp. 272-289.

Walton, W., and A. Woolcock, Aerodynamic Capture of Particles, Pergamon Press, London, 1960, p. 129.

Woods, J. D. and B.J. Mason, "Experimental Determination of Collection Efficiencies for Small Water Droplets in Air," ROY METEOROL SOC, QUART J., Vol. 90 (1964), pp. 373-381.

COUNTING OF GLACOGENIC NUCLEI

Pierre St.-Amand
William G. Finnegan
Larry A. Mathews

Naval Weapons Center
China Lake, California

ABSTRACT

Considerable divergence in determination of the nuclei yields of various generators leads to questions as to which methods are the most reliable and whether such determinations are actually possible.

Even before the nuclei enter a cold box, they are usually diluted, collected, stored and transported. By the time a smoke is ready to be introduced into the measuring device, coagulation has caused the nuclei count to be less than the original and caused a larger particle size. Also particle inertia, diffusion, and gravitational settling will cause differences in concentration and size distribution from the original smoke during sampling. Thus, dilution and handling techniques should be standardized for intercomparison purposes.

It appears as if only about 10^3 ice crystals per cm^3 can exist in a cold box at temperatures above -20°C at any one time. The Ostwald Ripening process causes some nucleated drops or the larger embryo to grow at the expense of their neighbors so that a determination of the actual number of nucleation events may not be possible by present-day methods.

It is quite likely that mixing type cold boxes yield results too low by perhaps 3 to 5 orders of magnitude at temperatures near the triple point and perhaps 3 orders too low at -20°C. This may be the reason that more ice crystals are counted in clouds than there should be for the number of nuclei implaced.

INTRODUCTION

One of the most difficult things to do in theoretical work involving freezing nuclei in general is to develop enough confidence in the measurement of ice nuclei by the several cold box techniques to use the results in deciding between alternative hypothesis. For years, it has been recognized that no two cold boxes gave identical results and that the reproducibility in any one box from one test to the other was not impressive. This difficulty may stem in part from imperfections in the measuring system but is most probably caused by the nature of ice crystal growth and these same difficulties may well be involved in measurement of condensation nuclei as well. The nuclei workshop conducted at Fort Collins in August 1970 brought these problems clearly to the attention of the scientific world. A discussion of one class of problem has been presented by St.-Amand and Finnegan (1970).

Firstly, the nuclei are usually collected, diluted and stored before introduction into the measuring device. The results of the measurement may depend more upon the technique involved than upon the nature of the nucleant. Smokes and aerosols are usually electrically charged to some extent and these properties are altered by dilution and storage so that they are not the same following processing as they would be in a cloud. Even more important, is the coagulation of the aerosol.

Coagulation: For an uncharged aerosol, Whytlaw-Gray and Patterson (1932) show that

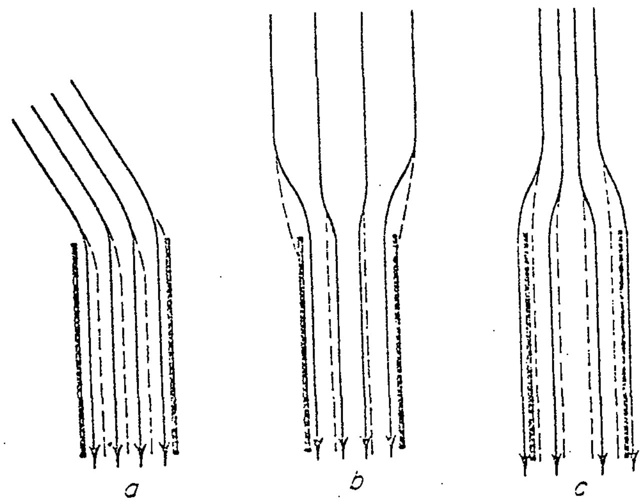
$$-\frac{dn}{dt} = Kn^2$$

where n is the number of aerosol particles per cubic centimeter, t is time in seconds and K is a constant in the range of 5 to 9 x 10^{-10} cm^3/sec. In clouds of high concentration, coagulation is very rapid so that if the initial concentration is greater than 10^{10} particles per cm^3 the smoke soon coalesces to a number density that is independent of the original number density in a short time. By the time a smoke is ready to be examined, the number of particles available is dependent more upon the dilution and storage technique than upon the original aerosol. This results in a count that is always less than the original and holds no clear hope for improved nucleation because of larger particle size. The particles are loose aggregates with no systematic improvement in overall surface large enough to sustain an embryo any bigger than that which the largest component particles would have sustained alone.

Effects of Sampling Technique upon Capture of Aerosol Particles:

If an ideal sampling technique is desired, the difference in concentration and particle-size distribution must be as small as possible between the sample and the original aerosol. Differences do occur because of entrance effects at the inlet of the sampling tube or instrument and of deposition in the tube or instrument itself.

a. Entrance Effect. If particles are of sufficient size, they will tend to travel in a more or less straight trajectory relative to the flow streamlines. Fuchs (1964, p. 142) neatly describes entrance effects on aerosol concentration as follows: "If the sampling tube is at an angle to the flow direction (Fig. 38a) some particles, because of their inertia, will either be deposited on the inside wall of the tube, or else fail to enter it, so that the concentration of the aerosol in the sample will be less than the true concentration. If the sampling tube is parallel to the flow but the velocity in it is greater (Fig. 38b) than in the main flow, particles from the streamlines directed just outside the tube will not enter the tube; if the velocity in the tube is less than in the main flow (Fig. 38c) particles from the streamlines directed just inside the tube but passing outside it, will enter the tube. The aerosol concentration in the sample will be too low in the first case and too high in the second. In correct or isokinetic sampling, the flow velocities in the tube and in the main flow must be equal. In addition the wall at the mouth of the tube must be so thin that deposition of particles on its butt end can be neglected."



<u>Fig. 1:</u> Taken from Fuchs (1964), his Fig. 38 entitled, "Various Cases of Aerosol Sampling."

Even when the isokinetic condition was maintained, Sehmel and Schwendiman (1968) have found an effect on sample concentration due to sampling probe diameter. By using sampling tubes (0.135 to 0.402 in. I.D.) having sharp edged inlets, the measured concentration ranged from 0.8 to 1.2 times the average concentration for single size uranine particles ranging from 28 down to 1 μ diameters.

Ideal isokinetic sampling can be observed only if a long narrow sampling tube with very thin walls is used.

Watson (1954) experimentally determined sampling efficiency versus the ratio of windspeed to inlet air speed, $\frac{U_o}{U}$, for sampling tubes aligned parallel with the aerosol windstream. For example, at $\frac{U_o}{U} = 2$, the sampling efficiency was found to be 1.05, 1.2, and 1.5 for particles having radii of 2, 9, and 19 μ. His data indicate that higher concentrations (or higher sampling efficiencies) would be obtained at higher ratios of $\frac{U_o}{U}$.

Donnan and Wright (1969) collect aerosol through sampling tubes which protrude about 1.5 inches forward of the leading edge of an air foil which is mounted in their wind tunnel. The intake ends of their tubes have been sharpened to a knife edge so that deposition of particles on blunt surfaces can be neglected. However, their sampling flow rates are far from isokinetic with $\frac{U_0}{U}$ somewhat greater than 10. Thus, higher concentrations of larger particles than the true concentration enter their sampling tubes as shown by the data of Watson.

As indicated by Dahl of Colorado State University (1970), they are presently using a 100-liter syringe to extract aerosol samples from their dilution tunnel. The syringe has a 2-inch diameter intake orifice whose axis is directed parallel to the dilution tunnel stream. By considering the time of intake (5 sec) and the windspeed in the dilution tunnel (60 to 130 knots), the ratio $\frac{U_0}{U}$ varies approximately from 2.5 to 6. Again, higher concentrations of larger particles than true may be collected for the 100 liter syringe. However, more or less of the larger particles may be deposited in the intake tube of the syringe due to inertia effects if a 90^o elbow is present.

CSU (1970) also uses syringes of 1, 2 and 10 liter capacity which withdraw a sample in about two seconds. However, the 1-inch diameter orifices are at a 90^o angle to the flow direction of the tunnel. Watson (1954) has shown experimentally that the sampling efficiency for particles having a radii greater than one micron is decreased by an increase of angle between the axis of the sampling tube and the flow direction of the windstream. For example, in the case of the angle being 90^o and the flow inside and outside the tube being equal, the sampling efficiencies for particles having radii of 2, 6 and 19 μ are 70, 45, an 12 percent according to the data of Watson. This effect may be even greater in the case of CSU's sampling since the flow rate through the orifice is somewhat smaller than that of the stream.

However, CSU seems to be taking a desired approach in varying their sampling technique. Yet more of an attempt should be made to obtain isokinetic sampling conditions since, as indicated above, an effect on particles in the 1 to 10 μ radius size may be substantial.

b. Deposition. Deposition of small particles may occur through gravitational or diffusional means during the laminar or turbulent flow of aerosol through sampling tubes or instruments.

As an example, let us consider the apparatus of Donnan and Wright (1968). The flow through their 0.25-inch O.D. sampling tubes is laminar, i.e., the Reynolds number is about 50 for their average flow rate of 200 cc per minute. Also the horizontal length of their sampling tubes is of the order of 15 feet.

Fuchs (1964, p. 112) gives the critical length for particle gravitational deposition in the case of laminar flow as

$$L_{cr} = \frac{8R\overline{U}}{3V_s}$$

where R is tube radius, $\overline{U}$ is the mean velocity of flow, and V_s is the terminal velocity of settling of a particular particle size. As calculated from this equation, particles having radii of 1, 2 and 5 μ will precipitate out at lengths of 30, 7.5 and 1.2 feet in the sampling tubes of Donnan and Wright. These particles would not be re-entrained since flow near the wall is near zero for laminar flow. Although Donnan and Wright collect higher concentrations of larger particles than true at the inlet, as noted previously, it is concluded all particles of radii 2 μ or greater never reach their cloud chamber due to gravitational deposition.

Diffusion can also cause a significant amount of deposition of submicron-size particles in small diameter tubes. From the theoretical expressions of Gormley and Kennedy (1949), Fuchs (1964, p. 205) presents a table which shows the fall in concentration of an aerosol due to diffusion to the walls for laminar flow through a circular tube. For the sampling tubes of Donnan and Wright, the table shows that for particles having radii of 0.1 and 0.01 μ that only 2.5 and as much as 35 percent of the particles are lost due to diffusional deposition, respectively.

Deposition will also occur due to the inertia of the particles, for example in curved sampling tubes. This process, which is important for particles with radii greater than one micron, has been experimentally studied by Sehmel and Swendiman (1968). They found deposition was significant for all flow rates and must be considered for any sample passed through a curved tube in either laminar or turbulent flow. In conclusion, attempts should be made to avoid unnecessary lengths and bends in sampling tubes in order to obtain representative aerosol concentrations.

Thus, intercomparison of devices for counting, or intercomparison of different measurements of the same generator should be made only with the same dilution and handling technique. Indeed the output of a generator, as measured, will depend markedly upon the delivery rate used during testing.

Following dilution, collection, storage, and transport, the aerosol is usually injected into an expansion chamber or into a mixing chamber; each have their own class of problems.

Expansion Chambers: An expansion chamber relies upon production of a momentary supersaturation, usually unknown, to cause deposition of water vapor upon the nuclei. If the supersaturation is high enough, and long enough sustained, particles too small to ever react in nature are caused to be counted. Because these devices do not duplicate actual cloud conditions, they are useful in measuring embryo formation and growth by diffusional processes under less than ideal conditions.

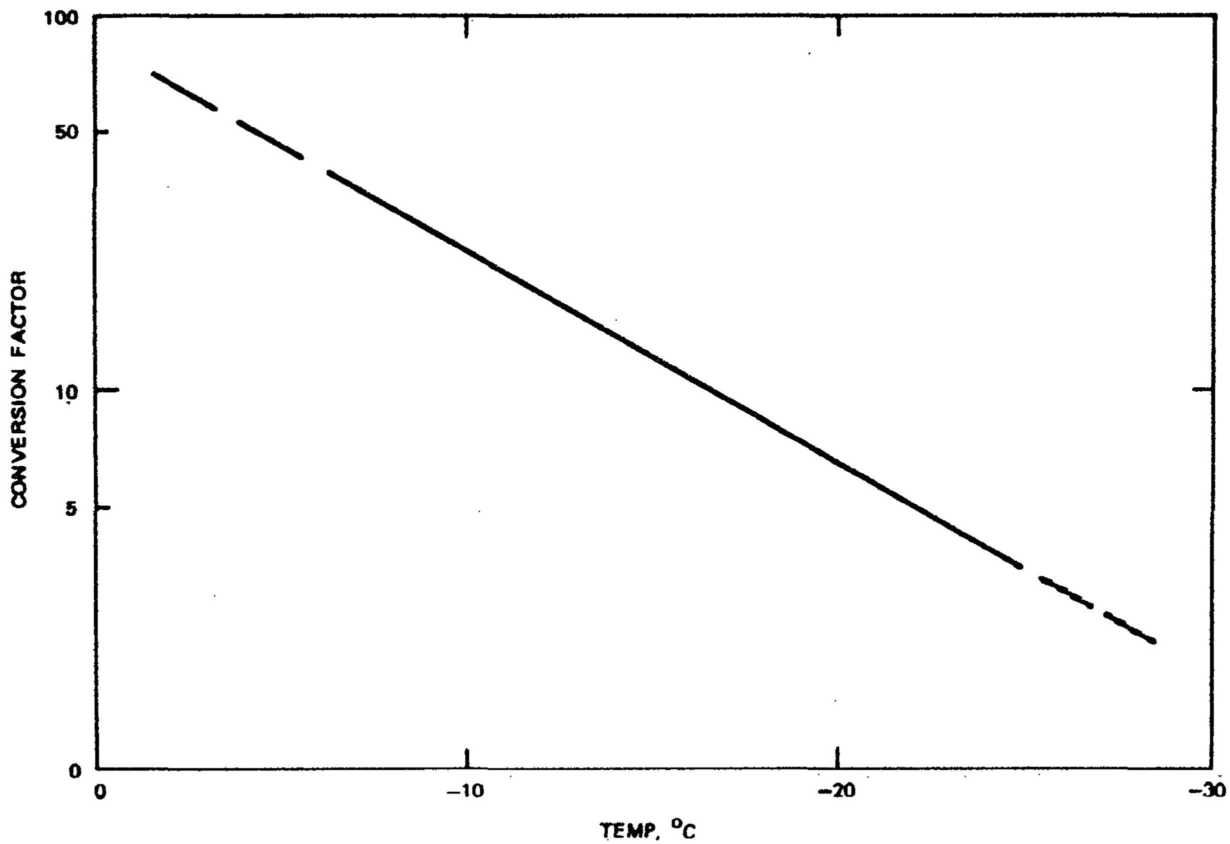


Fig. 2. Ratio of ice crystals observed optically to those counted in a cold box. From Smith and Hefferman (1954).

Mixing Chambers: Mixing type chambers use a water droplet fog. This fog is usually produced by condensation of steam, by dynamic expansion or by a spray from a very fine nozzle. Occasionally, hygroscopic substances are added to the air to induce droplet growth. Usually no account is taken of liquid water content, drop size distribution, or state of evaporation or condensation. Recently, measurement of liquid water content has been incorporated in some experiments. The apparent nuclei content is strongly dependent upon liquid water content. Steele, et al., (1969) and Donnan, et al., (1970) have found similar results. The processes whereby droplets, nuclei, dust particles and ice particles interact are complicated, as discussed by St.-Amand and Finnegan (1970).

Most mixing type cold boxes are so designed that a large numerical density of ice crystals is needed in order to have enough fall onto the microscope slide to count. There seems to be a limit to the number of ice crystals that can be grown in unit volume. Eadie and Mee (1963, p. 264) noted that Langmuir (1948) predicted some 10^{17} ice crystals could be produced from a gram of dry ice but that only 10^{10} crystals were noted per gram. They comment that in their 2-liter cold box, that 4×10^7 crystals were produced, being 2×10^4 per cm^3. They then speculate, "It is possible that this may represent the upper limit to the ice crystal concentration that can exist for any length of time in the 2-liter cold chamber. Even though the ice cloud was continuously supplied with moisture, competition for moisture by the ice crystals may have resulted in such a limit."

Balabanova (1959) noted that in a 14 m^3 chamber that increasing the AgI particle count from 1500 to 3000 particles per cm^3 resulted in no increase in ice crystal product at -6°C. At -10°C a similar change resulted in a 60% increase. Shiskin (1964) and Balabanova both conclude that not all AgI crystals manifest their ice-forming capabilities in the fog of the chamber.

Smith and Heffernan (1954, p. 185) in discussing the ice cloud developed in their testing device: "The upper limit to the concentration of ice crystals which can be reliably estimated with this apparatus is about 10^5 crystals per liter. Water then diffuses rapidly to the ice crystals and it is difficult to maintain any surplus fog. For this reason the estimated concentration of crystals never exceeds 10^6/liter." A liter contains 10^3 cm^3 hence they too find a limit of about 10^3 ice crystals/cm^3.

By making an optical count of the ice crystals in their cold box and comparing that to the total number of crystals estimated by collection, they were able to obtain the graph reproduced in Fig. 2. Extrapolation to -5°C indicates that they saw at least 50 times as many as were counted and that even at -30°C there were at least twice as many.

From this observation, it seems clear that nuclei in excess of 10^3 per cm^3 will not likely be counted unless a new cloud forms following fallout of the ice crystals. This is a consequence of an important phenomena known as the Ostwald Ripening Process. Once a nucleation event occurs, the ice embryo begins to grow, reducing the vapor around it to such a level that nearby particles are deprived of adequate vapor pressure to permit nucleation. Even if nucleation takes place, the larger drop or crystal will grow at the expense of near neighbors. In a cloud chamber, the first droplets to be nucleated dominate the scene. If the number of aerosol particles per unit volume is large, contact nucleation of drops will completely dominate the scene. The frozen droplets are already larger than an ice embryo and so even if an embryo forms by condensation or by sublimation on a particle, it will not be able to grow. Hence, it will not fall down and be counted in a time short compared to the experimenters patience span.

Kampmann and Kahlweit (1967) discuss this in detail for nucleation and growth in general. Briefly stated: If the growth of embryos is rapid once they are formed, the number of growing particles decreases rapidly so that the number of particles found in experiment is, in general, no measure for the nucleation rate of the beginning of precipitation. This process is illustrated by the scanning electron microscope photograph shown in Fig. 3, where a field of 50-micron ice crystals is surrounded by a number of smaller particles consisting of nuclei with one-micron ice crystals affixed thereto. The smaller particles have not grown because the larger crystals, which are formed presumably from frozen droplets, have competitively reduced the water vapor pressure. The nuclei count which is taken by counting only the larger crystals is bound to be several orders of magnitude less than the actual number present and available for nucleation were there space, time and water enough.

On the other hand, if the numerical density of the aerosol particles is kept below $100/cm^3$, the probability of nucleation by collision of aerosol particles with drops is reduced, and the predominate processes will be that of diffusional growth of ice crystals from embryos. Diffusional growth is quite slow near the triple point and counts made at temperatures warmer than -10^oC will be unrealistically low. At colder temperatures these processes are far more rapid and better counts will be obtained.

It seems clear that cold boxes will always count less than the number of nuclei present. Independent evidence that not all particles are counted comes from Langer, Lieberman and Rosinski (1967) who actually counted the AgI aerosol particles entering the NCAR counter at -20^oC and counted the ice crystals produced. They found (p. 963) that between 1000 and 1600 particles entered for each ice crystal counted. The great majority of these particles were big enough to have served as nuclei at the temperature of the test. It would be interesting to have determined the ratio of particles of other temperatures warmer than -20^oC.

Our own experience with the type of aerosol counter used by Langer, et al, indicates that it is relatively inefficient at counting particles smaller than about 0.2 microns radius, and because particles of the order of 0.05 micron radius will work well at -20^{o}C, they may have easily been undercounting the effective particles by several orders of magnitude, and thus the ratio of nuclei to ice crystals may be made larger than their measurements indicate.

It is basically wrong to consider the number of crystals counted by a cold chamber technique as nucleation events. The number of nucleation events per particle of catalyst may well be in excess of unity, in fact, at temperatures below that at which the whole area of the nucleant is needed for one ice embryo, more than one nucleation event can take place. Scavenging of smaller particles by diffusiophoresis and by collision onto the growing ice crystals also removes many nuclei, some multiple nucleated from the air, hence, further reducing the apparent number of nucleation events. This is a problem common to all testing techniques now in use.

Extension of cold box techniques to field experiments lead to an interesting anomaly. Many observers have noted that the number of ice crystals formed in a cloud is often in excess of the number of freezing nuclei available. Recent examples are Mossop, Ruskin and Heffernan (1968), Smith, Chien and MacCready (1968), and MacCready and Baughman (1968), Mason (1969), Koenig (1964, 1966), Braham (1964), and others. This has led to the proliferation of numerous ad hoc hypotheses to explain ice crystal multiplication, some of which may be true.

Smith, Chien and MacCready (1968) and more recently Mossop (1968) are inclined to question the applicability of cold box testing to measurements in real clouds. Mossop (1968, p. 124) says, after pointing out that six out of seven clouds studied at temperatures near the melting point of ice, had several orders of magnitude more ice crystals than nuclei, and after eliminating most possibilities of ice crystal multiplications, "One can therefore only say that in certain conditions (not yet understood) ice crystals appear in supercooled clouds in numbers higher by several orders of magnitude than conventional cloud chamber measurements would indicate."

The phenomena appear to be limited to clouds warmer than -20^{o}C, indicating that the results of cold box measurements may be more useful at -20^{o}C and colder than at higher temperatures.

It is difficult to see what this means in a real cloud except that, perhaps, the nuclei remaining to be counted have not yet worked in the temperature regime in which they are found. It is also likely that they were undercounted to begin with. A common practice appears to be to use an NCAR counter operating at -20^{o}C to determine the nuclei present and to compare this with ice crystal counts at the ambient temperature, Hobbs, et al., (1969-1970, p. 36). Certainly, if the nuclei had been used up making snow flakes, they would not be available for counting. The fact that they are available, probably means that it is not cold enough for them or that there is not enough water available, but the number not yet used has nothing to do with the number of ice crystals existing in the same air.

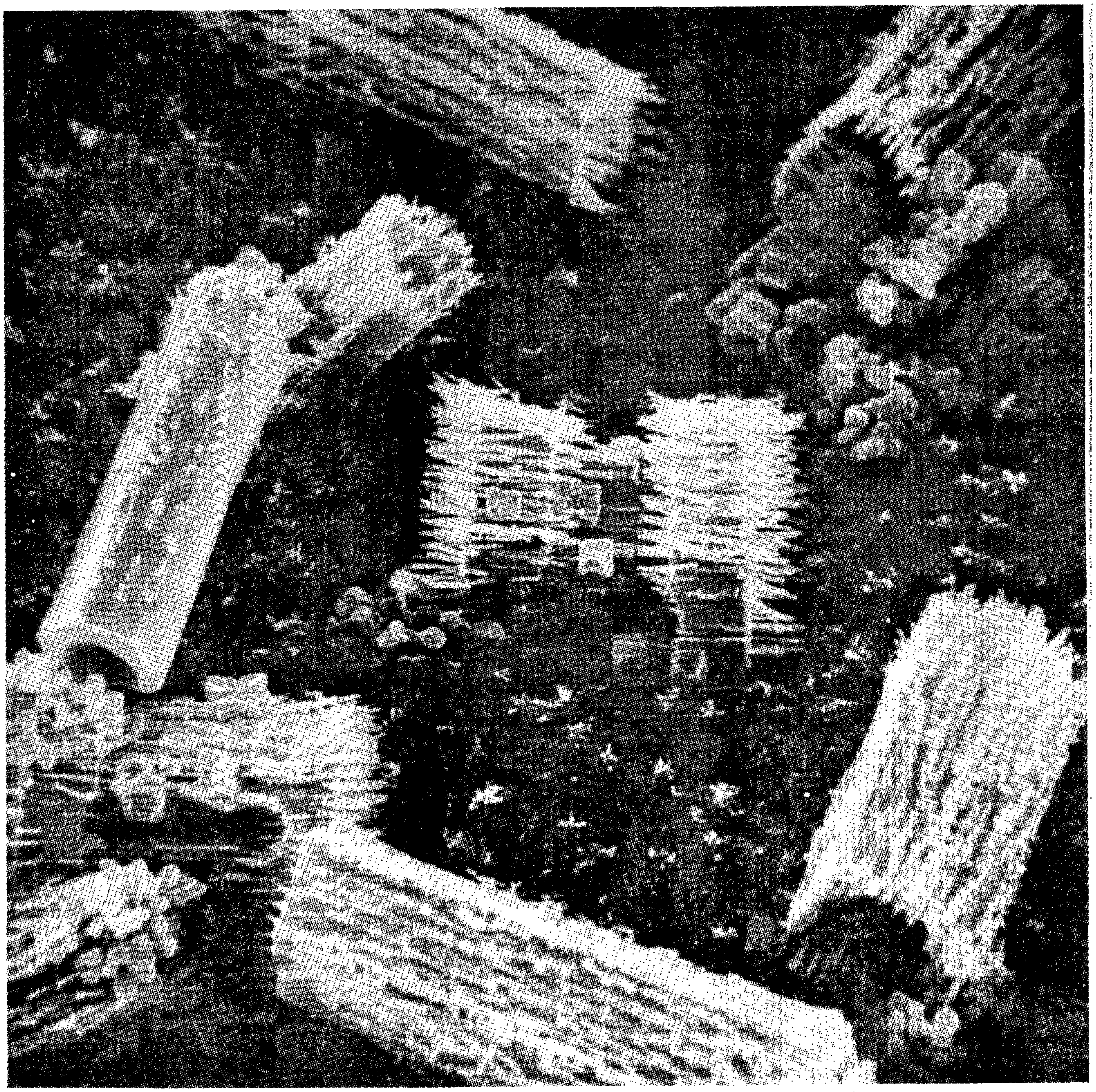


Fig. 3. Scanning electronmicroscope photograph of ice crystals seeded with AgI showing a background of much smaller particles, presumably ice growing diffusionally on nuclei.

REFERENCES

Balabanova, V. N., "On the Effect of the Temperature of Supercooling of Fogs on their Crystallization with Silver Iodide Aerosol," ACAD SCI USSR, BULL, GEOPHYS SER (English ed.) (1959), pp. 658-662.

Braham, R. R., "What is the role of ice in rainshowers?" J ATMOS SCI, Vol. 21 (1964) pp. 640-645.

Dahl, D., Private Communication, Colorado State University, (December 18, 1970).

Donnan, J. A., Donald N. Blair, William G. Finnegan and Pierre St.-Amand, "Nucleation Efficiencies of AgI-NH_4I and AgI-NaI Acetone Solutions and Pyrotechnic Generators as a Function of LWC and Generator Flame Temperature, A Preliminary Report," J WEA MOD, Vol. 1, No. 2 (May 1970), pp. 155-164.

Donnan, J. A., and D. A. Wright, "A Wind Tunnel/Cloud Chamber Facility for Research on Cloud Modification Materials," Inst. of Atmos. Sciences, South Dakota School of Mines and Technology, Rapid City, South Dakota, Report 69-8, Prepared for U.S. Dept. of the Interior, Bureau of Reclamation, Contract No. 14-06-D-5979, 1969.

Eadie, W. J. and T. R. Mee, "The Effect of Dry Ice Pellet Velocity on the Generation of Ice Crystals," J APPL METEOROL. Vol. 2 (1963) pp. 260-265.

Fuchs, N.A., The Mechanics of Aerosols, MacMillan Co., New York, 1964, 408 pp.

Gormley, P. and M. Kennedy, "Diffusion from a Stream Flowing through a Cylindrical Tube," ROY IRISH ACAD, PROC, Vol. 52A (1949), pp 163-169.

Hobbs, Peter V., L. F. Radke, J. Locatelli, C. Robertson, F. Farber, and D. A. Burrows, "Research Report V. Studies of Artifically Seeded and Unseeded Winter Storms over the Cascade Mountains (1969-1970)," Contributions from the Cloud Physics Group, U. of Washington, Seattle, Wash.

Kampmann, L., and M. Kahlweit, Max-Planck Inst., Goetingen. "Zur Theorie von Fallungen." Ber Bunsenges Physik Chem. (formerly Z. Electrochem.) Vol. 71, No. 1, pp. 78-87 (1967).

Koenig, L. R. "Some Chemical and Physical Properties of Silver Iodide Smokes," J APPL METEOROL. Vol. 3 (June 1964), pp. 307-310.

__________, "Numerical test of the validity of the drop-freezing/splintering hypothesis of cloud glaciation, J. ATMOS SCI, Vol. 23 (1966) pp. 726-740.

Langer, G., A. Liberman, and J. Rosinski, "Ice Nucleation Efficiency of Silver Iodide at -20C on a Particle Count Basis," J APPL METEOROL. Vol. 6 (Oct. 1967) pp 963-964.

Langmuir, I., "The Production of Rain by a Chain Reaction in Cumulus Clouds at Temperatures Above Freezing," J METEOROL. Vol. 5 (1948), pp. 175-192.

MacCready, P.B. Jr., and R. Baughman, "The Glaciation of an AgI-seeded Cumulus Cloud," J APPL METEOROL. Vol. 7 No. 1 (1968), pp. 132-135.

Mason, B. J., "Some Outstanding Problems in Cloud Physics -- the interaction of microphysical and dynamical processes," ROY METEOROL SOC, QUART J. Vol. 95, No. 405 (July 1969) pp. 449-485.

Mossop, S. C., "Comparisons Between Concentration of Ice Crystals in Cloud and the Concentration of Ice Nuclei,"Jour. de Recherches Atmospherique, Vol. III, 2e annee, No. 1 and 2 (Jan-June 1968), pp 117-124.

Mossop, S.C., R.E. Ruskin and K.J. Heffernan, "Glaciation of a cumulus at approximately -4C" J ATMOS SCI, Vol. 25, No. 5 (Sept. 1968) pp. 889-899.

St.-Amand, Pierre and William G. Finnegan. "The relevance of cloud chamber tests to ice nuclei activity," 2nd National Conference on Weather Modification PREPRINTS, Santa Barbara, Ca., (April 6-9, 1970), pp. 361-365.

Sehmel, G. A. and L. C. Schwendiman, "Particle Deposition Within a Curved Sampling Probe," Battelle-Northwest Annual Report for 1967 to the USAEC Division of Biology and Medicine, Vol. 2: Physical Sciences Part 3: Atmospheric Sciences, BNWL-715-3, pp. 88-92, October 1968.

__________. "The Effect of Sampling Probe Diameter on Sampling Accuracy," Battelle-Northwest Annual Report for 1967 to the USAEC Division of Biology and Medicine, Vol. 2: Physical Sciences. Part 3: Atmospheric Sciences, BNWL-715-3, pp. 92-95, October 1968.

Shiskin, N. S. "Cloud Modification," Oblaka Osadi i Grozovage Electrichestvo (Clouds, Precipitation and Thunderstorm Electricity), Chapt. 10, (1964), pp. 286-317.

Smith, E. J. and K. J. Heffernan, "Airborne Measurements of the Concentration of Natural and Artificial Freezing Nuclei," COMMONWEALTH SCI IND RES ORGAN, Vol. 80, No. 344 (April 1954), pp. 182-197.

Smith, T. B., C. W. Chien, and P.. B. MacCready, Jr., "Study of the Engineering of Cloud Seeding," Prepared for U.S. Navy Weather Research Facility, Norfolk, Va., by MRI, Altadena, Calif. Final Report Contract N00189-68-C-0100, NWRF 43-0688-139 (Aug. 1968), pp. 8-9.

Steele, Roger L., and Charles I. Davis, "Variation of Ice Nuclei Effectiveness with Liquid Water," J ATMOS SCI, Vol. 26, No. 2 (March 1969), pp. 329-330.

Watson, H., "Errors due to Anisokinetics Sampling of Aerosols," AMER IND HYG ASSOC QUART, Vol. 15 (1954), pp. 21-25

WhytlawGray, R., and H. S. Patterson, Smoke, Edward Arnold, London, 1932, 192 pp.

EFFECTS OF SOLUBILITY OF AgI NUCLEATION EFFECTIVENESS

P. St.-Amand, L. Mathews, D. Reed, L. Burkardt,
and W. Finnegan

Naval Weapons Center
China Lake, California

In considering seeding with silver iodide from beneath cloud base, scant attention has been paid to the history of a nucleant subjected to the warm moist environment in which it resides for a period of some minutes or tens of minutes.

Nuclei consisting of complexes of AgI and alkali metals are specially vulnerable because of the high solubility of these compounds. This aspect has been well dealt with in a number of publications and presentations: St.-Amand, 1967; St.-Amand, Finnegan, Burkardt, and Odencrantz, 1968; Burkardt, Finnegan, Odencrantz, and St.-Amand, 1970; St.-Amand, Finnegan, and Burkardt, 1971, etc. It is not well realized that AgI itself has similar problems.

AgI is slightly soluble in water. Only a few values are available for the solubility and these are not too reliable because of the propensity of AgI to form complexes with sodium and potassium ions dissolved from the container and because impurities present in the AgI sometimes aid solution.

For purposes of calculation we will assume that the solubility, S, of the AgI in pure water is 1×10^{-9} grams of AgI per gram of water at $+10^{\circ}C$. This value is quite reasonable. For $0^{\circ}C$ there is no published value and we make a linear extrapolation to 1×10^{-10} grams of AgI/gram of water. There is no sound basis for this and this value is almost certainly too low by a factor of 6 to 8. Furthermore the values cited are for bulk AgI and may not apply to a very small single particle.

Let r_s be the spherical radius of a particle of AgI that will just dissolve in a spherical drop of water of radius r_D. Then:

$$\frac{4}{3} \pi r_s^3 \rho_A = S \frac{4}{3} \pi r_D^3 \rho_D$$

where

$$\rho_A = 5.67\text{, density of AgI}$$

$$\rho_D = 1.00\text{, density of water}$$

$$r_s = \left(\frac{S}{\rho_A}\right)^{\frac{1}{3}} r_D$$

This function is shown for +10°C and 0°C on Figure 1.

If the AgI is not all dissolved, the particles are decreased in size. The radius of the particle remaining, r_R is

$$r_R = \left(r_A^3 - \frac{S}{\rho_A} r_D^3\right)^{\frac{1}{3}}$$

values of this function are shown for +10°C and 0°C in Figure 1.

The upshot of this is that seeding material imbedded in droplets, or even in contact with and partially wetted by liquid water, begin to dissolve. If the particles are smaller than, r_s, the size that will just dissolve in a given droplet, the particles will completely dissolve. If the particles are larger they will be reduced to the size r_R at which the AgI particle is at equilibrium in the droplet.

Should the temperature increase, r_R will become smaller. If the temperature decreases, AgI will be reprecipitated onto the remaining particle. If there is no particle remaining, the AgI solution may become supersaturated or may be reprecipitated as a colloidal suspension that will grow very slowly by an Ostwald ripening process into one or more crystals large enough to cause freezing, but only at temperatures in the vicinity of -15°C or so. Thus AgI particles that are too small for the size droplets in which they are injected will produce the same inconclusive seeding effects as may be expected from an alkali iodide-silver iodide complex, if they are used below warm cloud base.

Fletcher's theory gives the radius of a particle of AgI such that, for a given temperature one nucleation event can take place on that surface in one second. It could have been as easily calculated for longer or shorter time intervals. For one nucleation event to occur in a shorter time than one second, a larger particle would be required.

Now consider an AgI particle of exactly critical radius for the formation of one viable ice embryo in one second at a given temperature, imbedded in a water droplet at that temperature. If any mass is lost by solution in less than one second, nucleation can not occur at the end of the first second of immersion at that temperature. Hence, a particle always larger than that required by Fletcher's theory will be needed for contact nucleation to occur.

The rate of solution will depend upon temperature, radius of the AgI particle, radius of the water drop and upon the contaminants present in the water and in the AgI. Almost any compound involving the ammonium ion, iodide ions, or alkali ions will increase the solubility of the AgI and the rate of solution.

The matter is further complicated by the possibility that before becoming imbedded in a droplet, the AgI particle may have grown an ice embryo by one of the vapor diffusion processes. If this occurs, the nucleation of the water droplet will be determined by the ice already growing on the nucleus. In this case, the size of the nucleus will have to be adequate to ensure nucleation by condensation followed by freezing or by sublimation.

The size of an AgI particle that will permit condensation following freezing, is probably the same as that needed for contact nucleation provided the particle is completely wet. Some, but not a very considerable, loss of mass will occur before freezing of the condensed water begins, and the AgI particle is protected by the ice. The mass loss will be greatest at the highest temperature at which freezing can occur.

At tempratures below $-12.5^{\circ}C$ or so sublimation will occur first and the AgI will be protected by ice upon entrance to a drop. The particle size required for sublimation is however much larger than that necessary to produce contact freezing and so the advantage gained at the colder temperatures is not so great. Indeed, solution of the smaller particles in a time short compared to nucleation may well still occur, and in the temperature regime $T < -12^{\circ}C$ the basic contribution will still be by the larger particles in cloud containing larger droplets ($r > 10\mu s$).

Calculations of the time required for solution of an AgI particle of radius, r_A, in a droplet of radius, r_d, are underway. At present, the only available information seems to indicate that 0.001 μ and possibly 0.01 μ particles will dissolve in less than one second in bulk water. These times will not be markedly different for the larger droplets ($r > 100\mu$).

The effect of solution on nucleation effectiveness can be estimated only after the rate of solution of AgI in water is known. It does appear now that it will be adequately fast to set a limit on the number of nuclei per gram that can be measured in a mixing type cold box. It may be adequately larger to explain the difference between the observed nucleation effectiveness and that which is estimated as being theoretically possible. Clearly, the effects on nucleants used in sub-cloud seeding, especially when the clouds have high liquid water contents, is very appreciable. Nucleating agents used for this purpose should be designed to give about 5×10^{13} nuclei per gram and no more. Indeed, a monodispersed smoke of 0.1 μ particles would be about optimum.

It is quite probable that most nuclei larger than 0.005 μ serve as condensation nuclei in updrafts and may very well all be encased in water by the time they reach the freezing level. In any case, an appreciable number will be captured by diffusiophoresis, Brownian or Smoluchowskian collision. Most of these smaller than 0.01 or 0.05 μ will be subsequently dissolved and a number of the larger particles reduced in radius.

REFERENCES

St.-Amand, Pierre, "Nucleation by Silver Iodide and Similar Materials," Skywater Conference I, PROC, Bureau of Reclamation, Denver, Colo., 1967, pp. 305-346.

St.-Amand, Pierre, William Finnegan, Lohr Burkardt, and F. K. Odencrantz, "Effects of the Type of Nucleant on Modification of Clouds for Stimulation of Rainfall," presented at the First National Conference on Weather Modification of the American Meteorological Society, Albany, N.Y. 28 April - 1 May 1968.

Burkardt, Lohr A., William G. Finnegan, F. Kirk Odencrantz, and Pierre St.-Amand, "Pyrotechnic Production of Nucleants for Cloud Modification, Part IV. Compositional Effects on Ice Nuclei Activity," J Weather Modification Association, Vol. 2, No. 1, May, 1970, pp. 65-97.

St.-Amand, Pierre, William G. Finnegan, and Lohr Burkardt, "Understanding of the Use of Simple and Complex Ice Nuclei Generated from Pyrotechnics and Acetone Burners," To be published in J. Weather Modification Association, Vol. 3.

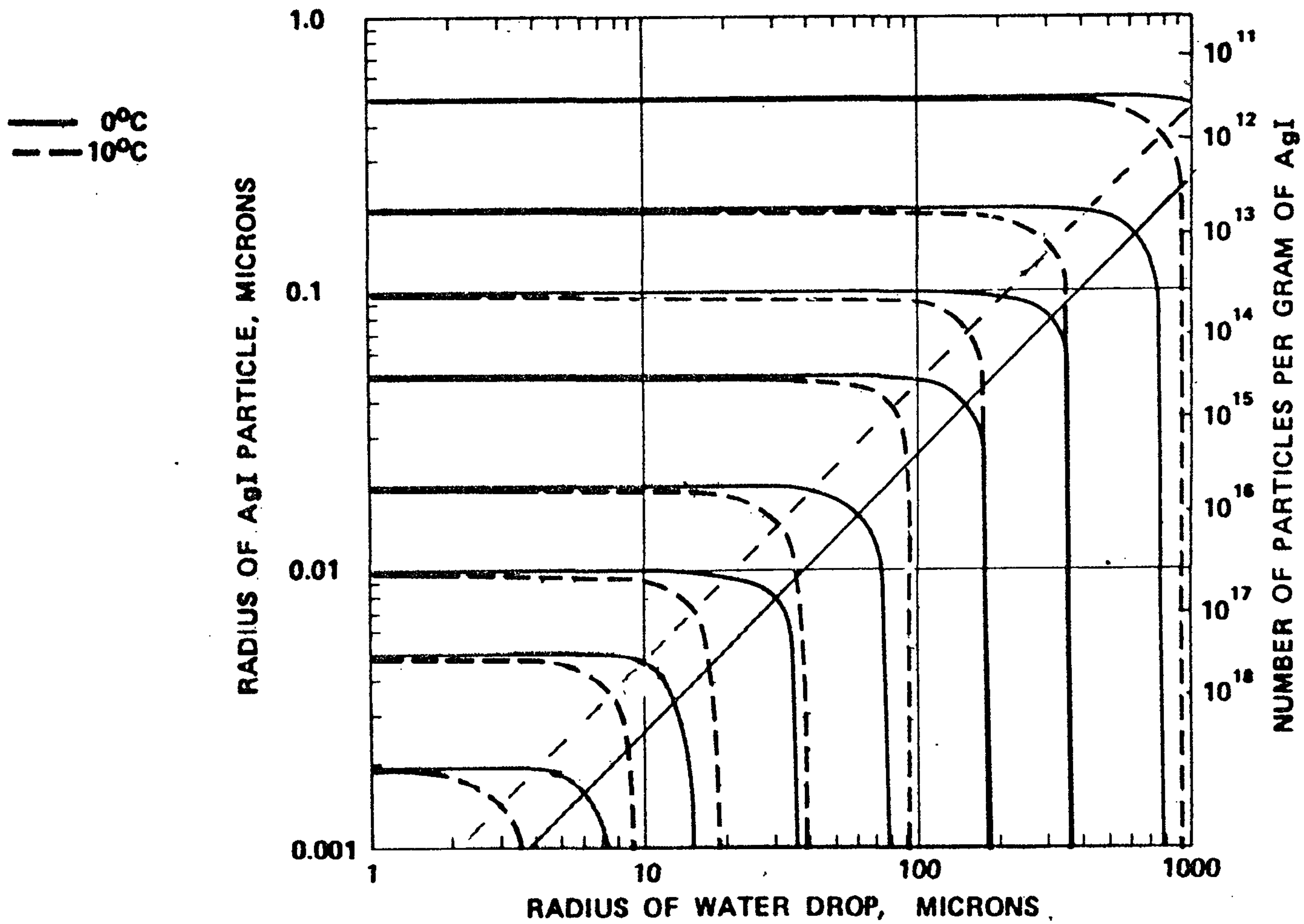


Fig. 1. Two diaganol lines show the maximum AgI particle size for complete dissolution in a given water drop size at 0 and 10^{0} C. The other curve shows the equilibrium particle size as a function of drop size.

ON THE ACTIVATION TEMPERATURE OF AgI-PARTICLES IN CLOUD

A. J. Alkezweeny

Meteorology Research, Inc.
Altadena, California 91001

I INTRODUCTION

Silver iodide particles are widely used in supercooled cumulus cloud modification for the purpose of initiating the ice phase. When the particles are dispersed beneath the cloud base in the region of an updraft, the material is carried upward, mixes with cloud droplets and diffuses horizontally by turbulence. Its concentration is diluted. Other techniques involve introducing the AgI material any place in the cloud. Here again the particles suffer mixing and dilution. In any case, the particles find themselves in an environment of temperature changing from 15C down to -20C or colder, and a maximum of one percent supersaturation with respect to water. Two questions which need to be answered are: (1) When do AgI particles form ice crystals by sublimation, contact or freezing? (2) If the particle makes contact with a cloud droplet at some temperature, or forms a droplet by condensation, what is the nucleation temperature of the droplet? Answers to these questions may be helpful in particle generator design, method of seeding and its optimization. In the following sections, the results of some recent experiments on the nucleation of AgI particles under real cloud conditions are discussed in an effort to answer the above questions.

II ICE CRYSTAL FORMATION BY SUBLIMATION

The temperature at which an ice nucleus becomes active as a sublimation nucleus is dependent upon particle size, properties of the particle surface, and the environmental supersaturation. According to Fletcher's nucleation theory (Fletcher, 1959), if the particle environment is water saturated, in order for it to act as a sublimation nucleus it must have a diameter greater than about 300 Å and 500 Å for activation temperature -20C and -15C, respectively. Based on laboratory experiments, Edward and Evans (1960) have concluded that particles of sizes up to 200 Å in diameter are ineffective as sublimation nuclei in clouds, and they are only useful if they collide with the droplets. Gerber et al. (1970) reported upon a recent experiment in which AgI particles were drawn into Goetz' Aerosol Spectrometer and the particles deposited on chrome-plated foil according to their sizes. After the foil is cooled to a fixed temperature and exposed to a water-saturated environment, the particles which are active grow to visible ice crystals. The authors reported that particles smaller than 400 Å and 320 Å in diameter failed to form ice crystals at temperatures of -15C and -20C, respectively, in agreement with the Fletcher Theory.

Weickmann et al. (1970) introduced AgI smoke from various generators in a large cold chamber and examined the ice crystals formed in the chamber at different temperatures. If the crystal contained a circle at its center, it was assumed to have grown from a frozen droplet. They concluded that, down to a temperature of -17C, the particles formed ice crystals through droplets freezing rather than sublimation.

Since most of the particles produced by any AgI generator are of sizes below 1000 Å, we can expect these particles to be ineffective as sublimation nuclei especially at warm temperatures.

III WATER DROPLET FREEZING

If AgI particles are produced by burning AgI and KI (or NaI) in acetone solution, the particles acquired hygroscopic properties (de Pena and Caimi, 1967). Therefore, if the particles are exposed to saturated environment, water droplets may form by condensation. In an experiment similar to that of Gerber et al. (1970), Alkezweeny (1969) produced AgI particles by burning rope which had been dipped in AgI- and KI-Acetone solution in the ratio 1:1 by weight of AgI and KI. When the foil carrying the particles was introduced into a saturated atmosphere, water droplets were formed. The droplets did not freeze until the foil temperature fell below -10C. Mossop and Jayaweera (1969) have shown experimentally that burning a solution of AgI and NaI in acetone produced particles which act as condensation nuclei first and then as freezing nuclei. The nucleation temperature which was observed, between -7.5C and -11.5C, agrees with Alkezweeny's observation.

Sulakvelidze (1969) and Gokhale (1970) found that the freezing temperature of distilled water droplets contacted by AgI particles was found experimentally to be about -5C. However, this warm freezing temperature was observed only when the droplet was supercooled to this temperature before the contact. If the particle made contact and the droplet temperature was warmer than -5C, the nucleaation temperature was lowered by about 8 to 10 degrees. The same freezing temperature reduction was also observed if the particle was embedded in a distilled water droplet (Gokhale and Goold, 1968).

Sulakvelidze also reported that if the droplets were made up of rain water, they froze somewhere between -5C and -10C. The colder the contact temperature, the warmer the freezing temperature.

Since cloud droplets are not made up of distilled water, the shift in the freezing temperature, due to warm temperature of the droplets at the instant of contact, cannot be applied to the cloud. However, if the rain water used by Sulakvelidze is accepted as representative of that near cloud base, and in the case of cloud base seeding, we can conclude that droplets making contact with AgI-particles at a temperature level warmer than -5C do not freeze until they reach the level of -8C to -10C.

In fact, this nucleation temperature was used by Alkezweeny (1970) in the construction of a mathematical model for clouds seeded with AgI particles below the base for the Flagstaff cloud seeding operation. The results of the ice concentration computed compare well with the field observations.

Rain water is different from water near the cloud base because it has gone through a cycle before it reaches the ground. Therefore, a new experiment is needed in which water is collected near the cloud base, and a study similar to that of Sulakvelidze may be carried out to determine the freezing temperature of the droplets as a function of contact temperature.

IV CONCLUSIONS

From the limited experimental observation on the nucleation of silver iodide particles, it appears that:

1) AgI-particles of small size such as those produced by the available generators are not good sublimation nuclei.
2) AgI-particles act as contact nuclei if they contact the droplets at the temperature level of -5C or colder.
3) If the contact is made at temperatures warmer than -5C the freezing temperature is not well determined but is probably around -10C.

ACKNOWLEDGEMENT

This work is supported by the U. S. Department of the Interior, Bureau of Reclamation, Division of Atmospheric Water Resources Management, under Contract No. 14-06-D-6581.

REFERENCES

Alkezweeny, A. J., 1969: Preliminary results of the nucleation of AgI. J. Appl. Meteor., 8, 993-994.

__________, 1970: A contact nucleation model for seeded clouds. Research Report No. IV, Arizona Weather Modification Research Program, Annual Report FY 1970, Meteorology Research, Inc., Cont. No. 14-06-D-6581.

de Pena, R. G., and E. A. Caimi, 1967: Hygroscopicity and chemical composition of silver iodide smoke used in cloud seeding experiments. J. Atmos. Sci., 24, 383-386

Edwards, G. R., and L. F. Evans, 1960: Ice nucleation by silver iodide: I. Freezing vs. sublimation. J. Meteor., 17, 627-634.

Fletcher, N. H., 1959: On ice-crystal production by aerosol particles. J. Meteor., 16, 173-180.

Gerber, H. E., P. A. Alee, U. Katz, C. I. Davis, and L. O. Grant, 1970: Some size distribution measurements of AgI nuclei with an aerosol spectrometer. J. Atmos. Sci., 27, 1060-1067.

Gokhale, N. R., and J. Goold, Jr., 1968: Droplet freezing by surface nucleation. J. Appl. Meteor., 7, 870-874.

_________, 1970: Ice formation by contact nucleation. Final Report June 1970 to NSF, Dept. of Atmospheric Science, State Univ. of New York at Albany.

Mossop, S. C., and K. O. L. F. Jayaweera, 1969: AgI-NaI aerosol as ice nuclei. J. Appl. Meteor., 8, 241-248.

Sulakvelidze, G. K., 1969: Rainstorms and Hail. Translated from Russian by Israel Program for Scientific Translations. 310 pp.

Weickmann, H. K., U. Katz, and R. Steele, 1970: AgI - sublimation or contact nucleus? Second Conf. on Weather Modification, Santa Barbara, Calif., April 6-9, 332-336.

THE MODIFICATION OF RAIN PARAMETERS BY PYROTECHNIC CLOUD BASE SEEDING

J. L. Sutherland, L. W. Cooper and D. R. Booker

Weather Science, Inc.
Norman, Oklahoma

ABSTRACT

A pyrotechnic, cloud base seeding experiment in Oklahoma has demonstrated increases of more than 100% in the total volume of rain as a result of seeding. The raindrop spectra of seeded rain show significantly more drops having diameters less than 1 mm for all rainrates and generally fewer drops larger than 2.50 mm diameter for rainrates below 50 mm hr^{-1}.

1. INTRODUCTION

During the summer of 1970, a cold cumulus cloud seeding experiment was conducted in Oklahoma. Sponsored by the Naval Weapons Center, the experiment was designed to monitor seeding-induced changes of rain parameters using an airborne mass-sensing raindrop spectrometer. This paper discusses the results of the experiment.

2. TECHNIQUE

A Piper Apache aircraft was used for both seeding and data collection. Seeding was accomplished using Navy-furnished LW-83 flares (300 gm AgI) at cloud base. A mass-sensing raindrop spectrometer was used to collect rain data.

The raindrop spectrometer operates on piezoelectric principles. When a raindrop strikes the raindrop spectrometer's sensor head, a pressure is exerted on a quartz pressure transducer. Reacting to the pressure, the transducer emits a voltage pulse, the magnitude of which is proportional to the pressure. An electronic calibration relates the magnitude of the voltage pulse to raindrop size. The raindrop spectrometer is sensitive to drops larger than 0.5 mm diameter. A more detailed explanation of the raindrop spectrometer can be found in Sutherland and Booker (1970).

Data from the raindrop spectrometer is computer-processed to yield observations of raindrop spectra, rainwater content, and rainfall rate for every five seconds of a rain-measuring pass. Rainwater contents and rainfall rates are computed from the raindrop spectra.

3. TEST CASE SELECTION

The selection of a cloud as a test case was based on one criterion: an updraft at cloud base having a diameter of at least 1.25 n mi. This criterion is a result of previous experiments involving natural cold cumulus clouds where it was found that those clouds having cloud base updraft diameters less than 1.25 n mi seldom produced significant rain. The minimum updraft of 1.25 n mi was further verified during the research discussed here in that all test cases produced rain at cloud base.

After a cloud was selected as a test case, an envelope containing a randomized seeding decision was opened. If the decision dictated seeding, a minimum of two and a maximum of five LW-83 flares were ignited in the updraft at cloud base. The size and intensity of the updraft subjectively determined the number of flares to be used.

After seeding, orthogonal rain-measuring passes were made under the cloud base throughout the life of the shower. Generally, three to four minutes elapsed between passes. Rain-measuring passes halted after the test cloud had stopped raining or when hail or turbulence forced abandonment of the test case.

4. RESULTS

Twenty-five test cases were studied during the experiment. Five of those were declared in an early stage of the experiment that involved seeding at -7C with vertical fall pyrotechnics. For that reason, those five test cases are not included in the following analysis. Of the remaining 20 test cases, 11 were seeded at the cloud base and 9 were not seeded.

A. The Rainwater Volume from Natural and Seeded Test Cases.

Rainwater volume is defined in this paper to be an estimate of the total volume of water precipitated by a shower and is the product of the average rainrate throughout the shower, the average rainshaft size, and the duration of the shower. The average rainrate and rainshaft size are calculated from the individual passes under a shower. The rainshaft is assumed to be a circle with a diameter equal to the product of true airspeed and the time interval during which raindrops are sensed on each pass. The duration of the shower is simply the time from the first rain-measuring pass to the last rain-measuring pass.

Rainwater volumes computed under the assumptions discussed above tend to be more subjective than objective. However, the computational method is the same for natural and seeded showers, so that comparisons between the two can be made regardless of the absolute accuracy of the calculation. New definitive techniques to calculate rainwater volume are presently being developed.

Rainwater volumes have been computed for seven of the seeded test

test cases and seven of the natural test cases. Of the other six test cases, one (intentionally overseeded) did not rain at all and five were abandoned because of hail or excessive turbulence.

The seven seeded test cases (all seeded at cloud base) had an average rainwater volume of 489.2 acre ft., and the seven natural test cases rained an average of 80.4 acre ft. (Table 1). A modified Student's t test (Steel and Torrie, 1960) of the difference between the average seeded and natural test cases showed no significance because of the very large standard deviation of seeded rainwater volume. The ratio of seeded rainwater volume to natural rainwater volume was 6.08.

TABLE 1

Average Test Case Parameters

Test Case	Seeding Amt (gm)	Avg. Rainrate ($mm\ hr^{-1}$)	Avg. Rain-shaft (Km)	Duration (min)	Rainwater Volume (acre-ft)
12	900	3.5	11.9	70	372.2
13	300	27.8	8.3	72	1447.4
14	900	3.4	6.4	36	53.7
15	600	11.7	6.4	38	196.5
16	600	14.2	5.8	49	252.3
18	1200	13.3	10.2	75	1101.8
21	600	0.3	2.3	21	0.3
7	0	2.8	5.9	65	67.2
9	0	2.1	6.6	35	34.0
10	0	13.3	5.8	40	227.9
11	0	0.4	4.1	44	4.2
17	0	12.6	3.3	40	57.2
19	0	7.4	6.4	43	135.8
25	0	6.9	4.1	30	36.6
Average Seeded:		10.6	7.3	51.6	489.2
(without Cases 13 and 18)		6.6	6.6	42.8	175.0
Average Natural:		6.5	5.2	42.4	80.4

When Test Cases 13 and 18 are excluded from the sample of seeded test cases (because of their unusually large rainwater volumes), the average rainwater volume of the remaining five seeded test cases is 175.0 acre ft. The modified Student's t test of the difference between this seeded average rainwater volume and the natural average is significant at 7.5%. The ratio of seeded to natural rainwater volumes here is 2.18.

Increases in rain of more than 100% as a result of seeding have been reported recently by North American Weather Consultants (NAWC) in their analysis of raingage data from a project involving orographic seeding near Santa Barbara, California (Elliott and Thompson, 1970). The same LW-83 type pyrotechnic devices used by NAWC for orographic ground-based seeding were used by WSI in aircraft seeding. Both experiments seeded at or near the bases of convective showers. The cloud types and seeding techniques of the NAWC project and the WSI project were somewhat different, but it is interesting that data from both projects indicated rainfall increases of more than 100% as a result of seeding.

In addition to the rainwater volumes given in Table 1, the average rainrate, average rainshaft size, and duration of each test case are given. The average rainrate of the seven seeded test cases is 10.6 mm hr^{-1}, compared to 6.5 mm hr^{-1} for the seven natural test cases. The average seeded rainshaft is larger than the average natural rainshaft by 2.1 Km (39%), and the average duration of the seeded test cases is longer by 9.2 min (22%).

Excluding Cases 13 and 18 from the seeded sample results in approximately equal durations and rainrates for the five remaining seeded test cases compared with the seven natural test cases. However, the seeded rainshafts averaged 1.4 Km larger (27%) than the average natural rainshafts.

B. Comparison of Seeded and Natural Raindrop Spectra.

The computer-processed data from the RR40 raindrop spectrometer consists of observations of raindrop spectra for every five seconds of a rain-measuring pass. Data in this form have been grouped according to certain rainrate classes, such as 1 (±0.3) mm hr^{-1}, 4 (±0.5) mm hr^{-1}, etc. From these groupings of raindrop spectra at selected rainrates, average spectra have been computed for seeded and natural rain. The average spectra are given in Table 2. Also shown in this table is the number of observations of raindrop spectra used in computing the average. The larger number of observations of natural rain seen in most rain classes of Table 2 is the result of the inclusion of rain data from natural showers that were not declared test cases. All data used to compute the spectra shown in Table 2 were taken from cold convective showers.

The seeded and natural raindrop spectra shown in Table 2 indicate that seeding increases the number of drops less than 1 mm diameter. The significance of the change in the raindrop spectra attributable to seeding was tested by performing an analysis of variance on each raindrop class column in Table 2. Before the analysis of variance was performed, the data were normalized such that all observations of natural raindrop concentrations had the numerical value of 1.0. The seeded concentration associated with each natural concentration was expressed as a fraction of the natural concentration. Thus, the seeded and natural concentrations at 0.75 mm diameter and 1 mm hr^{-1} were changed from 390.6 $m^{-3}mm^{-1}$ and 194.5 $m^{-3}mm^{-1}$, respectively, to 2.01 and 1.0, respectively. Similar operations were performed on the other 79 pairs.

TABLE 2

Comparison of Seeded and Natural Rain Spectra

(S = Seeded; N = Natural)

Rain Type	Drop Concentration ($m^{-3}mm^{-1}$) .75	1.25	1.75	2.25	2.75	3.25	3.75	4.25	Rainrate ($mm\ hr^{-1}$)	No. of Observations
S	390.6	37.2	6.0	1.2	0.08	0.24	0	0	1.0 (±.3)	44
N	194.5	47.9	8.3	1.9	0.69	0.25	0	0	1.0	82
S	661.3	69.0	14.0	3.6	1.1	0.28	0.07	0	2.0 (±.3)	49
N	303.0	83.4	20.4	2.9	1.4	0.42	0.16	0.05	2.0	64
S	771.3	121.9	32.6	6.7	2.9	1.4	0.49	0.34	4.0 (±.5)	40
N	354.8	116.1	32.8	10.5	3.1	1.5	0.77	0.40	4.0	64
S	1024.6	189.6	59.2	17.4	7.0	4.5	1.8	0.95	8.0 (±1.0)	54
N	605.5	209.5	63.3	20.2	7.9	3.3	1.3	1.2	8.0	61
S	1131.5	325.8	114.1	41.9	16.7	8.6	4.2	2.8	16 (±4.)	58
N	717.7	266.6	90.8	35.1	20.2	11.0	4.6	3.7	16	89
S	1394.0	380.5	125.6	53.0	30.5	16.5	8.1	6.2	25 (±5.)	48
N	1023.8	418.4	122.4	54.1	34.8	17.6	8.2	6.2	25	86
S	1486.8	452.2	143.9	67.3	39.4	25.1	12.9	10.9	35 (±5.)	59
N	1272.6	486.2	139.4	67.4	39.4	25.3	11.9	11.9	35	74
S	1782.1	581.6	158.0	86.2	55.8	38.7	20.3	21.4	50 (±10)	33
N	1548.4	732.7	163.2	92.3	59.0	36.1	20.8	18.6	50	39
S	2184.9	846.4	175.2	125.1	87.8	60.7	35.8	35.7	75 (±10)	30
N	1888.9	1044.7	178.9	134.5	94.6	51.1	34.8	26.7	75	18
S	2101.4	1265.8	180.6	148.8	118.4	75.0	43.2	45.9	100 (±10)	25
N	1882.2	1334.3	187.5	175.2	117.6	71.6	44.0	50.6	100	10

The analysis of variance of the normalized data of Table 2 shows the increase of 0.75 mm raindrops caused by seeding to be significant at more than 0.5%. The number of 0.75 mm drops in seeded rain (averaged overall 10 rainrate classes) is over 50% more than the natural average. The average seeded concentration of 1.25 mm drops was 9% lower than the natural concentration, and this difference was significant at more than 10%. More than 10% significance was also found at the 2.75 mm drop class. The average seeded concentration of 2.75 mm drops was lower by 17% than the natural concentration. The remaining five raindrop classes showed no significant differences in natural and seeded raindrop concentration.

The increase in drops less than 1 mm caused by seeding is shown also in Fig. 1, which is based on Table 2 and which shows that seeded rain has at least 5% more drops of diameter 0.75 mm for all rainrate classes. The ratio of seeded to natural raindrop concentration at 0.75 mm decreases from approximately 2 in the lighter rainrate classes to about 1.1 at 100 mm hr^{-1}. There is an indication that seeding produces more drops of 3.25 mm and larger at rainrates above 50 mm hr^{-1}. Underseeding of showers capable of producing these heavier rainrates might explain this phenomenon.

The raindrop spectra at 1 and 25 mm hr^{-1} for natural rain is compared with spectra measured by Best (1950) and Caton (1966) in Fig. 2. The natural rain from the Hotshot showers shows more large drops.

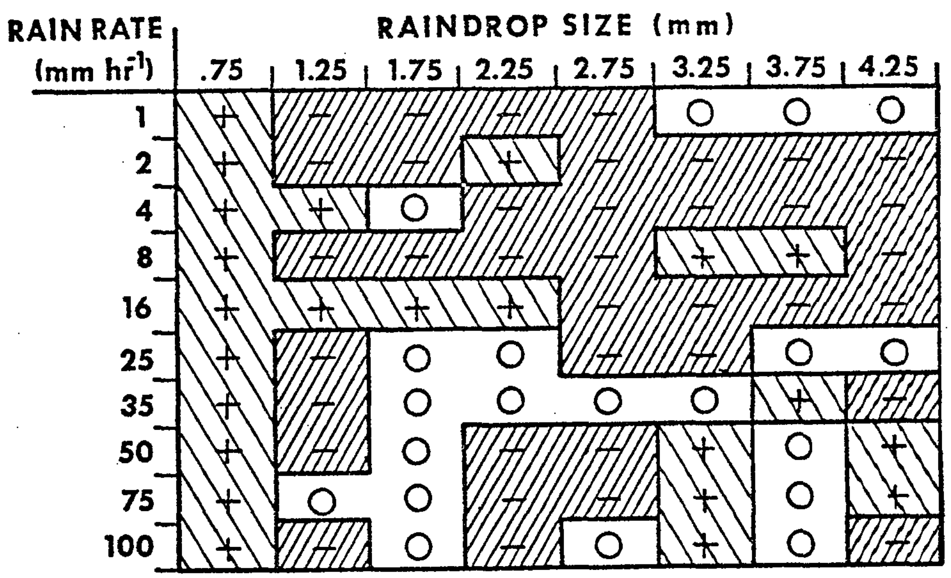


Fig. 1. Increase in drops less than 1 mm caused by seeding.

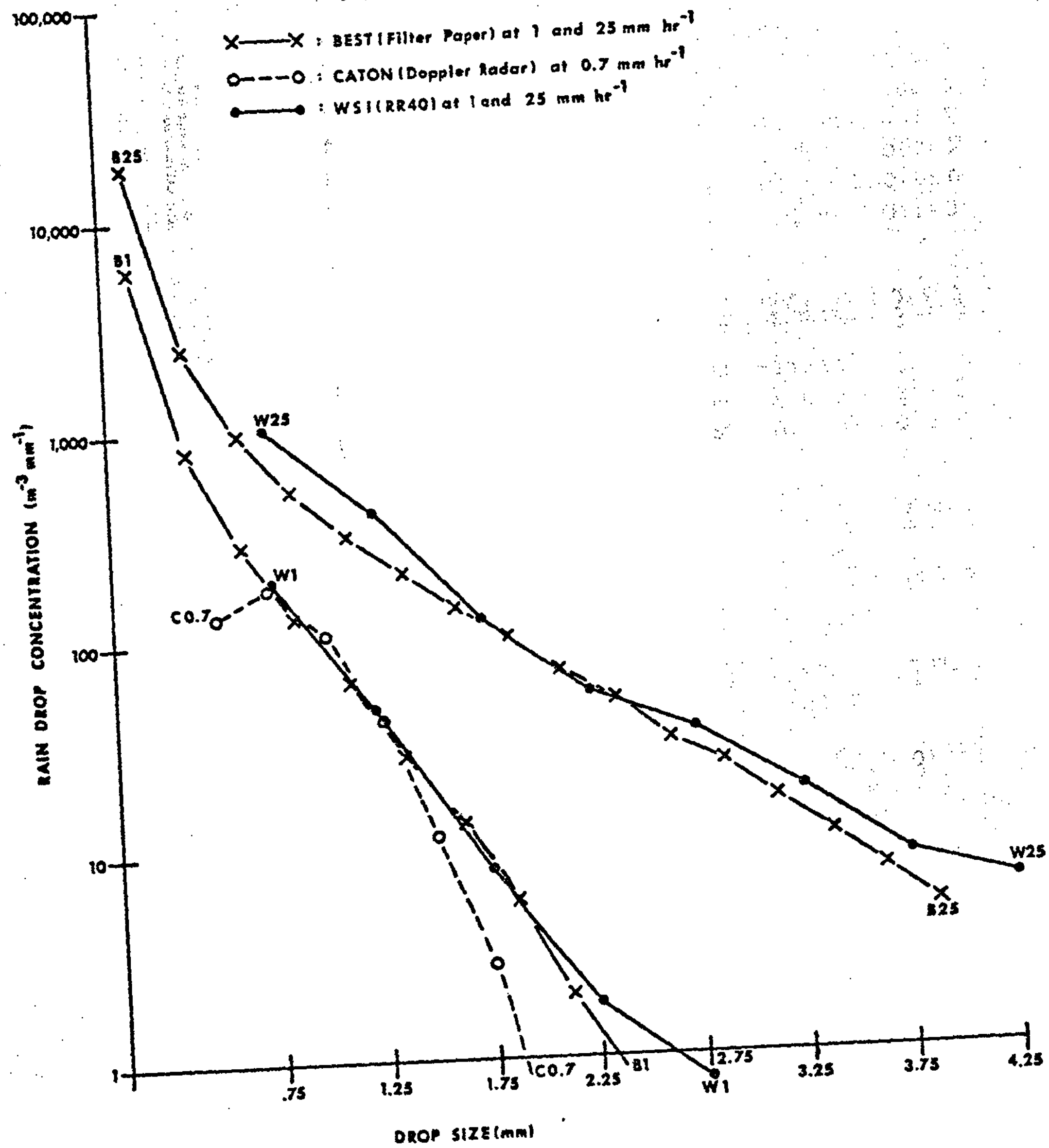


Figure 2. Natural raindrop spectra compared with the spectra of Best and of Caton.

5. CONCLUSIONS

Cloud base seeding of cold convective clouds in Oklahoma has resulted in increases of rainwater volume of at least 100%. The inclusion of two seeded test cases which produced extremely large amounts of rain into the seeded sample results in an increase volume of better than 500% over the average rainwater volume of natural test cases.

The comparison of seeded and natural raindrop spectra has shown that seeding produces more drops of 0.75 mm diameter at all rainrate classes and generally fewer drops of diameters larger than 2.75 mm at rainrates below 50 mm hr^{-1}. There is a trend toward more seeding-produced drops larger than 3.25 mm at rainrates above 50 mm hr^{-1}, perhaps indicating that showers capable of producing rainrates such as this were underseeded.

ACKNOWLEDGEMENTS

This research was sponsored by the Earth and Planetary Sciences Division, U.S. Naval Weapons Center, China Lake, California, under provisions of Contracts N00123-69C2216 and N66001-71-C-0148.

REFERENCES

Best, A.D., 1950: The size distribution of raindrops. Quart. J.R. Meteor. Soc., 76, 16-36.

Caton, P.G.F., 1966: A study of raindrop-size distributions in the free atmosphere. Quart. J.R. Meteor. Soc., 92, 15-30.

Elliott, R.D. and J.R. Thompson, 1970: Santa Barbara Pyrotechnic Seeding Device Test Program 1969-70 Season and 1967-70 Summary. Final Report, Code 602, U.S. Naval Weapons Center, China Lake, California.

Steel, R.G.D. and J.H. Torrie, 1960: Principles and Procedures of Statistics with Special Reference to the Biological Sciences. McGraw-Hill Book Company, Inc., New York, p 81 and 173.

Sutherland, J.L. and D.R. Booker, 1970: An Airborne Momentum-Sensing Raindrop Spectrometer. Proceedings of the Conference on Cloud Physics, Fort Collins, Colorado, 101-102.

A WIND TUNNEL/CLOUD CHAMBER FACILITY FOR CLOUD MODIFICATION RESEARCH

John A. Donnan, D. N. Blair, and Dean A. Wright

Institute of Atmospheric Sciences
South Dakota School of Mines and Technology
Rapid City, South Dakota 57701

ABSTRACT

A wind tunnel/cloud chamber facility for research on cloud seeding materials has been in operation for several months at the Institute of Atmospheric Sciences at the South Dakota School of Mines and Technology at Rapid City, South Dakota.

Among the unique features incorporated into the system are:

1) utilization of five independently temperature-controlled cold boxes;
2) a fog system which maintains any desired fog density in each cold box;
3) a highspeed, precisely metered, sampling system which provides exact sample volumes of nucleants to each cold box, and
4) a data recording system which photographs accumulated ice crystals without removing them from their respective cold boxes.

Full utilization of the above features permits simultaneous sampling into all five cold boxes, each at any desired temperature and each with fog of known density and liquid water content. The resultant crystals may then be simultaneously photographed ane evaluated, making it possible to determine the efficiency of any type of cloud-seeding generator or seeding material at five different temperatures at the same time.

I INTRODUCTION

The use of cloud chambers to simulate cloud physics processes and to test the effectiveness of ice nucleants dates back to the very inception of the modern cloud-seeding technology, beginning with the early work of Vonnegut and Schaefer in the mid-1940's,

Recognition that processes in small chambers may not duplicate those in natural clouds, because of wall effects and other complications, has led to the use of some very large cloud chambers. Among these may be mentioned the Swiss hail tunnel (List, 1961) and several chambers >15 meters high reportedly in existence in the Soviet Union (Battan, 1965).

This report describes a facility built by the Institute of Atmospheric Sciences at Rapid City, South Dakota, beginning in 1966, principally for the purpose of studying ice nucleating materials and processes (Donnan and Wright, 1969).

2. DESCRIPTION OF FACILITY

The facility is made up of two primary components; namely, (a) the wind tunnel and (b) the cloud chamber (or cold box) complex. Fig. 1 is a schematic diagram showing how the various components of the facility are tied together.

Fig. 2 is a photograph of the wind tunnel from the intake end, which better illustrates its size and its proximity to the 12 ft. x 50 ft. trailer which houses the cloud chamber complex. Fig. 3 is an interior view of the cloud chamber complex. Shown on the right are the five cold boxes, each identically configured, with the microscope/photographic systems mounted horizontally in front of each box. The only other readily identifiable component is the fog manifold, standing vertically at the right front corner of each box. The large solenoid in the fog manifold is activated by the light source/photo detector system which senses the fog density. The film drive motor is shown directly under the camera mechanism. Shown on the left is the side of the master control panel. On the shelf above the panel are the two 12-point recorders which monitor the temperatures at four levels in each cold box.

a. Wind Tunnel

The primary purpose of the wind tunnel is to dilute the effluent from the generator to a known and acceptable concentration. The component parts of the wind tunnel, from the intake end to the exhaust end, are as follows:

Intake Filter: An 85% efficient bag type filter with fiberglass prefilter. This filter is intended to remove only the larger ambient dust particles from the air passing into the system.

Fan: A 21-inch diameter vane axial fan capable of producing 6800 CFM against 3 inches of static pressure.

Burn Chamber: A cylindrical chamber 6 feet in diameter and 6 feet long. The chamber is fitted with specially designed amounts which suspend the various generators along the axis of the tunnel, directly in the air stream, to provide maximum quenching and disperson of the generator effluent.

Sampling System: Downwind from the burn chamber there is a 2-foot diameter tube 10 feet long which contains an air foil section mounted vertically along the diameter of the tube. The air foil section houses the sampling tubes through which the nuclei samples are pumped to each cold box. The intake end of the sample tubes protrudes from the leading edge of the air foil in the wind tunnel test section. From the air foil, the tube goes to a pump, then to a 3-way solenoid valve located 2 inches from the cold box, at which point the sample may be directed into the cold box or into a bypass tube back to the wind tunnel. The sampling systems utilize 0.25 inch OD tubing throughout and the solenoid valves are standard 3-way solenoids. The pump is a 3100 rpm rotating-reciprocating piston type pump with variable output from 0 to 2000 cc per minute. The flow rate of each pump is continuously monitored with a flow meter.

<u>Exhaust Stack</u>: A 3-foot diameter 40-foot high stack to carry effluent away from the facility.

b. <u>Cloud-Chamber -- Cold Box Complex</u>

There are 5 cold boxes for operation at 5 different temperatures and liquid water content* during a run. The cold boxes are basically standard, commercially available freezers which have been modified for this use. Each cold box has its own temperature control and is capable of maintaining temperatures as low as -40C. The effective working volume of each is one cubic foot. The interior of the chamber is completely lined with black velvet, except for the several tubes which extend through the walls. The velvet liner is about 0.5 inch from the wall of the cold chamber, which provides a dead air space between the chamber walls and the interior of the test chamber. Black velvet is used to provide a permeable barrier to the supercooled fog, which must be maintained in the test section of the chamber so that the rate of dissipation of the fog to the cold metal walls of the cold box is held within acceptable limits.

The test chamber temperature distribution is continuously monitored on potentiometric recorders connected to four thermocouples per cold box located at comparable positions in each box.

Fog is produced by an ultrasonic medical type humidifier, with fog droplets in the 1 to 8 micron range. The fog passes into a 1.5-inch plastic pipe manifold which carries the aerosol past each of the 5 cold boxes. At each cold box a 1.5 inch solenoid valve permits fog to be automatically injected into each of the 5 boxes through a "T" in the manifold. The fog detection system is composed of a photo-electric system to automatically maintain a fog of a preset fog density (and thus a fog of constant LWC) in each box. The LWC of fog can be accurately maintained at any predetermined value in the range of 1 to 10 gm m^{-3} over the temperature range of -3C to -20C with a 3 to 5-sec injection of fog every few seconds depending on temperature and LWC conditions in the boxes.

The LWC is determined by evaporating the fog and measuring the

*Hereinafter referred to as LWC

resultant dew point. The moist air samples which are drawn from the cold boxes are passed over a heated nichrome wire which is coiled inside a Pyrex glass tube to vaporize all liquid water before it enters the dew point hygrometer. The saturation mixing ratio is read for this dew point temperature and also for the temperature inside the cold box. The difference in these two readings gives the liquid water in grams per kilogram of dry air. Using the factor 1.17 (density of air at 900 mb, the normal surface pressure) the LWC may be calculated in grams per cubic meter. Curves showing the relation between cold box temperature, dew point, and LWC of the air inside the boxes is shown in Fig. 4.

c. Sample Collection and Recording System

This system consists of a 250-foot spool of 16-mm clear film, a film drive assembly, a film track, a settling area, a light source, and an optical-photogaphic system in each box.

The film track lies on the floor of the cold box along the front to back centerline of the box. The spool of clear 16-mm film is mounted in a tight container at the back end of the track. The film is passed through the film track and exists from the cold box through the film slit at the front of the box. It then passes over the drive sprocket and on to a take-up reel. Near the center of the cold box there is an 0.5 x 1 inch opening in the film track, which is the area whre the ice crystals settle and are collected on the 16-mm film. Near the front of the cold box on the film track is a brass cylinder housing the microscope objective and focusing mount. This is positioned directly above a window for the light source in the bottom of the box. As the ice crystals form and settle evenly to the floor of the cold box, some fall on the film. When the film is advanced 3.5 inches, the collected crystals are brought into position under the microscope objective for viewing and/or photographing. An Amici erecting prism located immediately above the objective directs the image through the wall of the cold box and into the microscope-camera system. Four by five-inch black and white pictures are obtained at magnifications of about 200 X using direct development film.

The light source which provides illumination of the crystals for viewing or photographing consists of a 150-watt illuminator with a 3-foot fiber optics light guide. The fiber bundle is inserted to within about two or three inches of the film plane. Fiber optics are used because of the convenience of a small diameter, high intensity light source and because of the low heat radiation. Even though very little heat is produced, it has been determined that after about 5 sec of illumination sufficient heat is produced to cause melting of some of the ice crystals on the film. Therefore, it is necessary to photograph the crystals very quickly after the light source is turned on.

3. DATA COLLECTING PROCEDURES

For a routine efficiency determination, the generator unit is mounted in the wind tunnel burn chamber. For acetone generators, a small auxiliary blower is mounted in the intake end of the generator and adjusted to simulate the desired ram air speeds. Most airborne acetone generators use ram air to pressurize the acetone solution, which controls the solution flow rate, and to produce a high velocity flow past the nozzle which atomizes the solution stream. The air flow from the wind tunnel fan flows past the generator, approximating flow around the generator during flight conditions.

When the cold boxes are stabilized at predetermined temperature and LWC conditions, ignition of the generator is accomplished and the AgI nuclei are pumped into the cold boxes at a known rate and for a known time.

The ice crystals form and settle to the bottom of the cold boxes and are photographed about 3 min after the sample is first injected into the cold boxes, and again about 5 min after the first picture, and as necessary, at 3 to 6-min intervals until no further crystals are observed. The series of photographs thus obtained from each cold box are collected and all crystals appearing on each photograph are counted. The total crystal count from a given series of photographs constitutes another of the parameters which enter into the computational scheme.

At the termination of each test run the following data are summarized for each cold box:

Q total flow through the wind tunnel

A_1 horizontal cross-sectional area of each cold box

A_2 area of film actually viewed

B mass rate of flow of AgI

V volume of sample injected into each cold box

C total crystal count

L sampling system loss factor

From the above data the "efficiency" of a given generator is determined for each cold box.

Finally, then, upon completion of a full "test run", the efficiency of a given generator may be determined at 5 different temperatures and LWC at the same time with burn characteristics exactly alike at all 5 temperatures of the cold boxes, each at known water content.

The question of coagulation in the sampling tubes or deposition on the sampling tube walls has arisen often. By actually taking a sample at the intake end of the sampling tube and another near the cold box, and using a technique for determining efficiencies as developed by Blair and David (1969), a loss factor has been determined. Data show no significant difference in the loss factor when flow rates varied from 200 cm^3 min^{-1} to 2000 cm^3 min^{-1} nor was there a significant difference with variation in the tube length. It has been concluded that there is a loss factor of about 3 which must be applied to all efficiency results, and which remains essentially constant as sample tube length and sampling rates vary, within the limits stated above.

It should be noted that the operation of the system is done from a central master control panel. The human factor is completely eliminated. The variables in a given test are determined and set before a test is started and during a run all the parameters must remain fixed, including the area of the film that is photographed.

4. TEST DESCRIPTION

In the past six months, about 300 separate nucleation efficiency tests have been run giving nearly 1500 data points at different temperatures and LWC in the boxes. The tests have included the evaluation of numerous pyrotechnic seeding generators supplied by several different manufacturers as well as extensive study of the acetone solutions of AgI-NaI and AgI-NH_4I.

Tests are classified as standard tests or LWC tests. A standard test is defined as a test in which each of the five cold boxes is at a distinctly different temperature, and may cover the range from -3 to -20C. At the same time LWC is controlled so that the LWC decreases with colder temperatures in an attempt to simulate the decreasing water content in a natural cloud with increasing altitude. Sample size may be regulated to simulate a pyrotechnic device falling vertically through a cloud, in which case the same size sample is injected in all 5 cold boxes. Or if it is desired to simulate cloud base seeding, a large sample is injected in the warmer boxes and much smaller samples in the colder boxes.

In an LWC test, all five cold boxes are held at the same temperature as closely as possible, and the LWC is varied from about 1 gm m^{-3} in the driest box to about 8 gm m^{-3} in the wettest box. For these tests, the same size (volume) sample is injected into each cold box and the results are plotted as nuclei count vs LWC.

Special tests which have been run include the selection of a pair of cold boxes in which the temperature and LWC gradients are as closely matched as possible, and then injecting sample into the boxes at known, but different levels. Such tests are providing interesting results with regard to the effect on nucleation efficiencies to short exposures to different environmental conditions.

ACKNOWLEDGEMENTS

We wish to acknowledge the contributions of Mr. Charles Tollinger whose patience, advice, and interest has contributed greatly to making this facility a working reality. The following agencies supported the research: The U.S. Department of the Interior, Bureau of Reclamation under Contract No. 14-06-D-6796; the Department of Naval Research under Themis Contract No. N00014-68-A-0160; and the Naval Weapons Center, China Lake, California under Contracts No. N60530-67-C-1289 and N60530-68-C-0006.

REFERENCES

Battan, L. J., 1965: A view of cloud physics and weather modification in the Soviet Union. Bull. Amer. Meteor. Soc., 46, 309-316.

Blair, D. N., and B. L. Davis, 1969: Aging of silver iodide-sodium iodide generator effluent in moist and dry air. J. Appl. Meteor. 8, 551-555.

Donnan, J. A., and D. W. Wright, 1969: A wind tunnel/cloud chamber facility for research on cloud modification materials. Report 69-8, Institute of Atmospheric Sciences, South Dakota School of Mines and Technology, Rapid City, South Dakota. 31 pp.

List, R., 1961: Influence of silver iodide on the atmospheric ice forming process. Zeitschrift Für Auguwandte Mathematik and Physik, Basel, 12, 474-476.

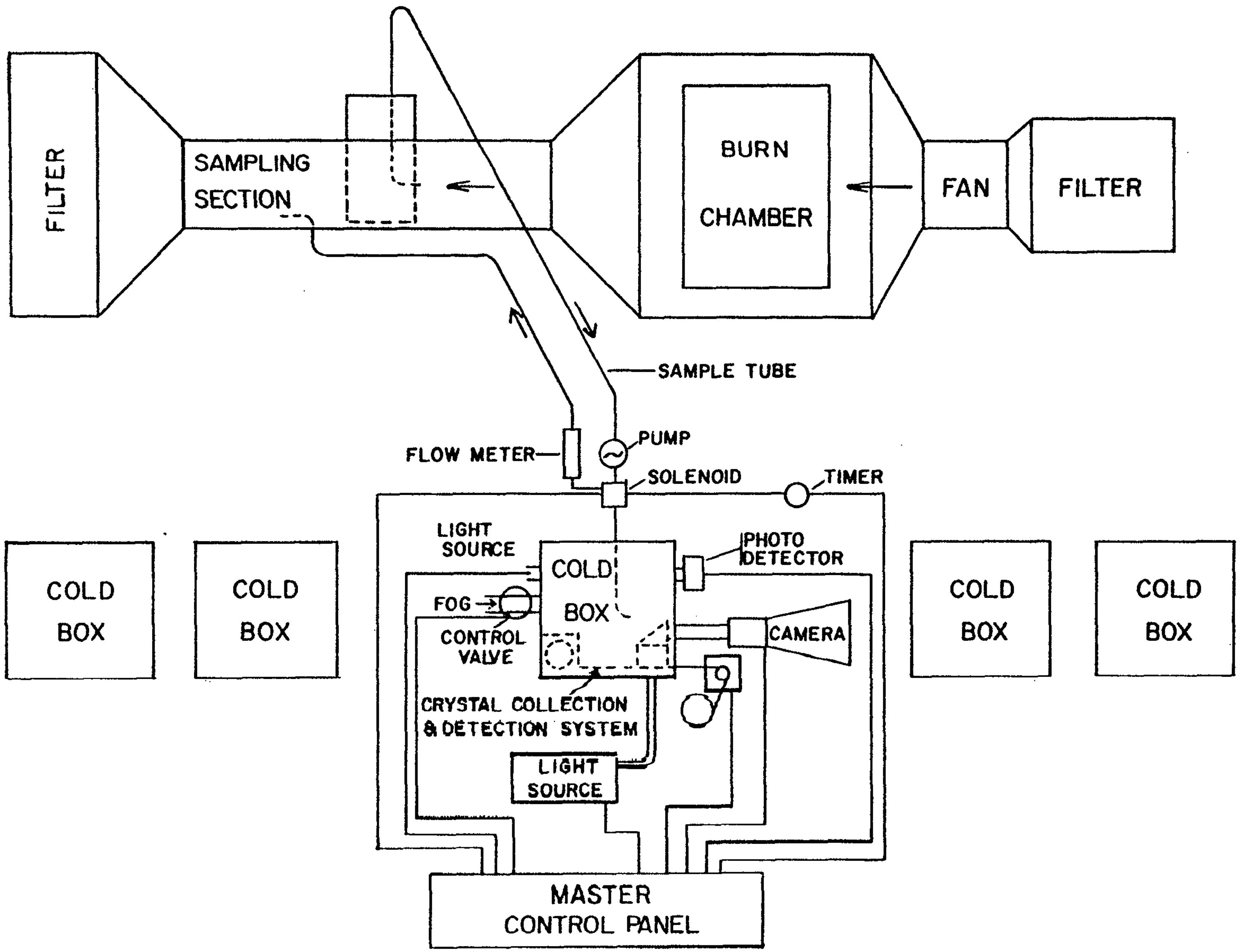


Fig. 1. A schematic diagram of the wind tunnel/cloud chamber facility.

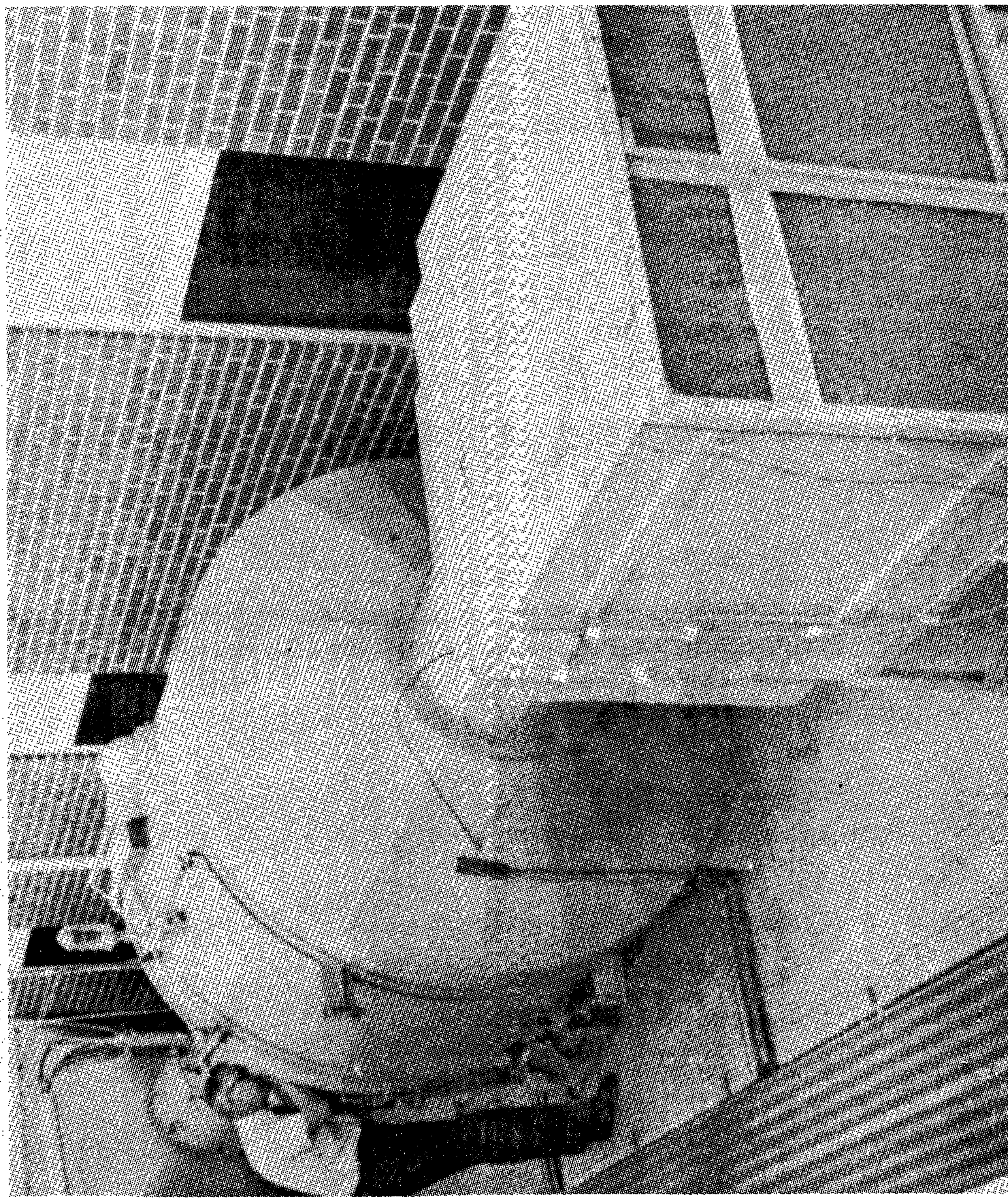


Fig. 2. Photograph of the wind tunnel from the intake end.

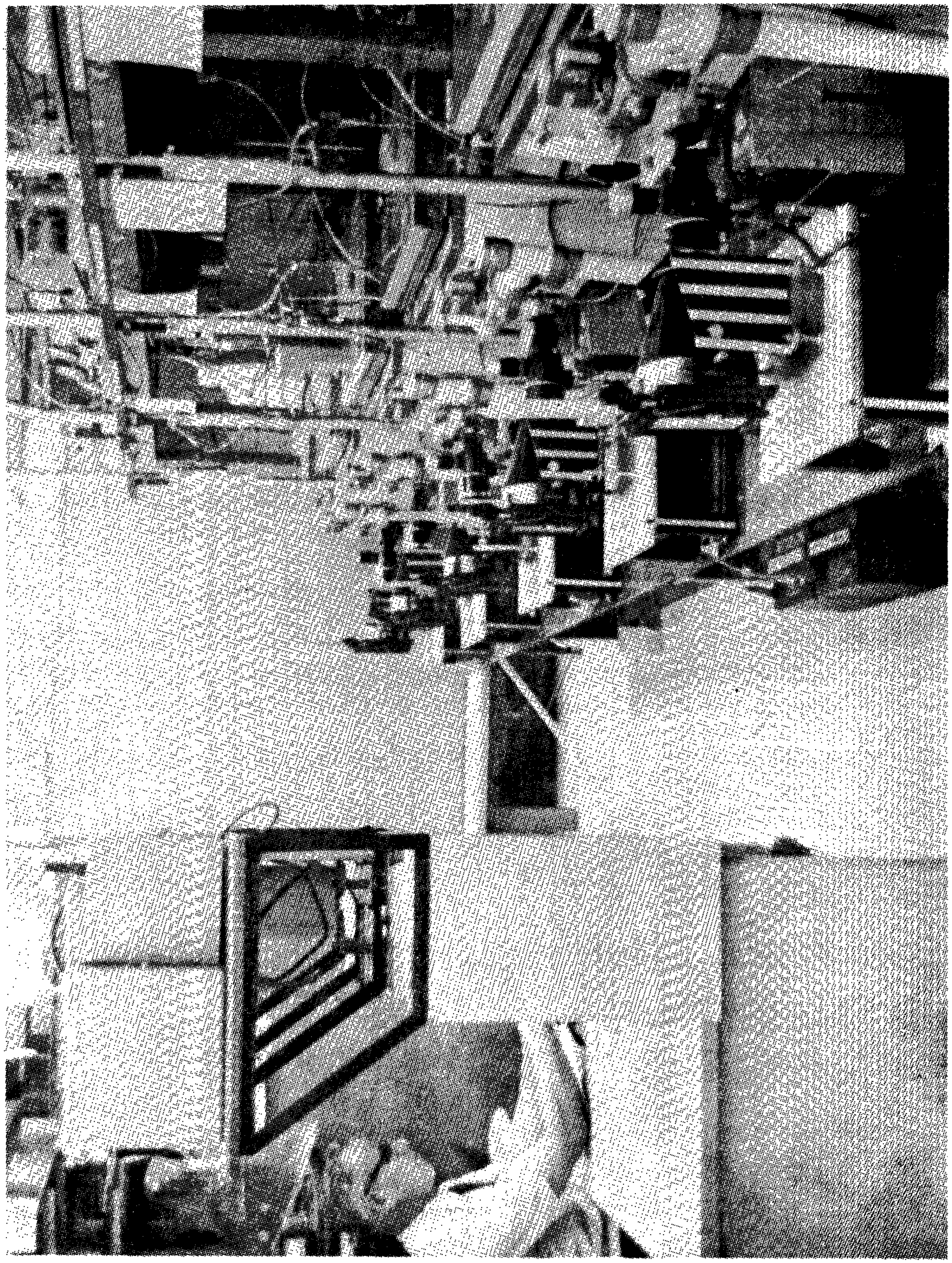
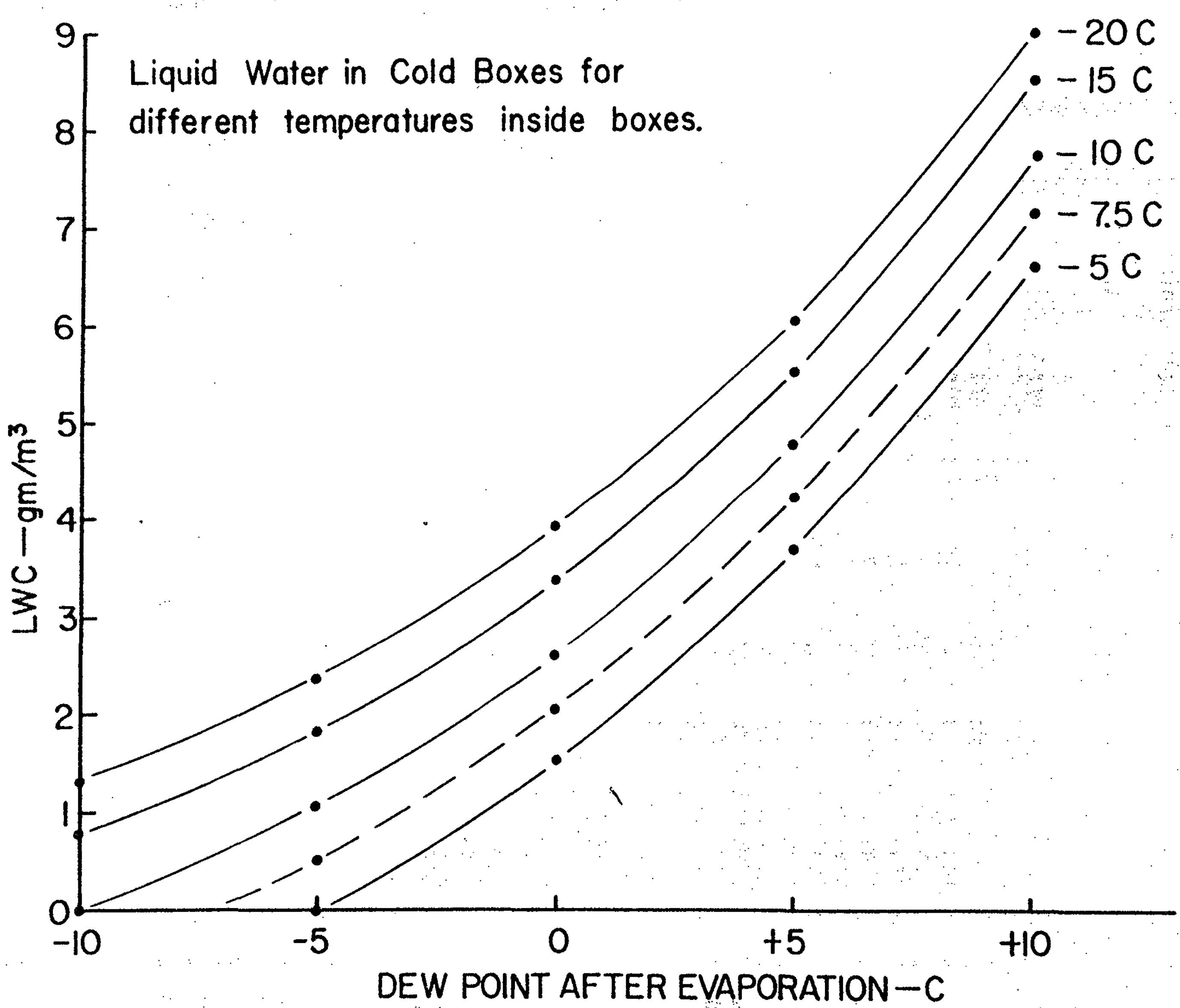


Fig. 3. Photograph of the cloud chamber complex.



Fix. 4. Curves for determination of the liquid water content of the atmosphere inside the cold boxes.

NOZZLES FOR SPRAYING WARM FOGS

John Carroz

Naval Weapons Center
China Lake, California

ABSTRACT

Attempts were made to develop nozzles for spraying hygroscopic liquids into fogs at 400 gallons per minute from aircraft traveling from 100 to 200 knots. The spray was to have droplet diameters all between 10 and 100 microns. Of the commercial nozzles tested, all sprayed similarly when used in a 100 to 133 knot airflow. These sprays came close to meeting the requirements. Two efforts to design new nozzles to meet the objectives were unsuccessful.

SUMMARY AND CONCLUSIONS

According to theory, liquids atomized in an airstream will be broken up into very small particles (see Tables 4 and 5). Measurements taken by White (Ref. 1) (see Table 6) in a wind tunnel gave parametric values intermediate between the values listed in Tables 4 and 5, and not in good agreement with either. This was no surprise due to the crude form of the equations and to measuring techniques. White's experimental method for impacting the droplets probably favored catching the 4 to 10 micron diameter drops and shifted the size distribution of the drops counted. For measuring droplets smaller than 4 microns in a high velocity airstream the measuring method used was inadequate; so droplets of this size and smaller, if there were any present, were not counted in the calculations of the median diameters. Using state-of-the-art methods, better measurements are not possible.

TABLE 6*
Test Results

Nozzle (Catalog No)	Orifice diameter (inches)	Air velocity (knots)	Sauter mean diameter (microns)
2515	.100	100	32 - 42
2515	.100	125	11 - 26
2515	.100	136	22

*Data taken from W. White, Table 3 (Ref. 1)

The most important parameters affecting particle size were airspeed and nozzle orientation. The droplet sizes were smaller at the faster air velocities with the nozzle exist perpendicular to the airflow. The type of nozzle had a minor effect on particle sizes. The measurements taken from aircraft mounted slides were in good agreement with the equivalent measurements taken in the wind tunnel.

Attempts at designing a 20-gallon per minute nozzle which would produce a very small, uniform drop size were unsuccessful. Having a uniform drop size of fog treating agent is very important. The droplets over 100 microns diameter are ineffective (Ref. 2) and consume a lot of agent (weight is proportional to volume which is proportional to the radius cubed!) The droplets under 8 microns do not settle fast enough, even after diffusional growth. By the time these small droplets settle out, the treated fog would likely be off the target. Sub-micron particles may stabilize a fog (Ref. 3, p. 20 and 21). At this time there is no known way of atomizing high volume flow rates of small diameter particles without producing, with a small part of the volume, large numbers of droplets with diameters less than 10 microns.

INTRODUCTION

Hygroscopic solutions have been sprayed from aircraft onto warm fogs for the purpose of dissipating the treated sections of fog. Experience has shown a need for disseminating large quantities of solution to effect dissipation. Also, the solution droplets must be small to stay within the fog a sufficient amount of time to grow and remove liquid water and water vapor. Computer modeling of fog modification has shown increased effectiveness with treatment droplets of approximately 17 microns in diameter (Ref.2). However, there is a problem of generating high spray rates of small droplet sizes with present day off-the-shelf equipment. Fog clearing requires high dosage rates of uniformly sized droplets which are under 100 microns in diameter for optimum results. Table 1 shows the effect of atomization on coverage.

TABLE 1.—EFFECT OF ATOMIZATION ON COVERAGE.[a]

Droplet Diam (μ)	No. spray droplets/in.[b]
10	1,148,000
20	143,000
50	9,224
100	1,164
200	142
300	43
500	9

[a]Data from Brown 1951.

[b]Rate of application: 1 gal/acre.

OPERATIONAL OBJECTIVES

The objective is to spray the fog-clearing liquids into a narrower particle size range while maintaining the high flow rates and small particle sizes.

The nozzles are the critical part of the total spraying system. In so much as they effect the total system, the objectives of the spraying system are:

1) Spray liquids from aircraft traveling from 100 to 200 knots.
2) Spraying into the fog with rates in excess of 400 gallons per minute.

Since the larger particles in the spray (over 100 microns) have high weights and contribute little to clearing the fog, the objective is to eliminate them. Elimination of the larger particles will reduce the mass median droplet size.

OPERATIONAL BASIS

Nozzles are mounted on booms on the wings of the spraying aircraft. Up to 100 nozzles have been used on each aircraft, depending on each nozzle's flow rate. Liquid at elevated pressure flows in the booms to the nozzles. The liquid is atomized by the nozzles and by the air flowing past the aircraft. The location of the nozzles on the aircraft determines the droplet density across the aircraft flight path. The type of nozzle determines the flow rate per nozzle and the droplet size distribution.

TECHNICAL BASIS

The fog droplets which reduce visibility are below 20 microns in diameter. They are the ones which must be removed to "clear" the fog. When a large droplet of hygroscopic solution (over 100 microns in diameter) is sprayed into the fog, it is ineffective for two reasons. First, because the droplet's fall velocity is too large and the droplet does not remain airborne in thc fog for a long enough time to absorb its full share of moisture to "dry out the fog". The droplets fall through the fog too rapidly for much condensation to occur (Table 3). Secondly the larger droplets have poorer collision efficiencies with the smaller fog droplets. Not only are there fewer droplets to "sweep out a path" when the drops are larger (Table 1), but when the droplets are larger, the small fog droplets follow the stream-lines around the large droplets. Collision coalescence is more likely between droplets of approximately equal sizes. For example, if the droplet size distribution were as poor as that shown in Table 2, 93% of the material being sprayed into a fog would be practically wasted. When the optimum size particles are sprayed, less material is required for the same effect.

Most people in the field believe atomization produces sprays which have a log normal distribution. Professor Chester Himel, who is an expert in agricultural spraying, stated: ".....distribution is important because it means that all sprays, no matter how 'coarse', will be accompanied by a significant number and volume of spray droplets in the 1-50 micron size range. There is no commercial spray device available which produces spray droplets in a small range of sizes."[1]

It is unfortunate that state-of-the-art nozzles are unable to spray a uniform drop size and, in particular, inevitably spray some of the liquid into droplets under 8 microns in diameter.

TABLE 2.—DROPLET SIZES OF OIL SPRAY EMMITTED FROM FAN-TYPE NOZZLES AT 45 PSI ON A BOOM ON THE STINSON L-5 AIRCRAFT.[a]

Size class (μ)	% of droplets in each size class (by mass)
10-40	0.14
41-100	7.20
101-160	27.20
161-220	35.90
221-280	10.70
281-340	18.50

[a]From Sebora (1946).

Calculations performed by Paul Tag and others (Ref. 2) showed 70 micron diameter (median-mass) and above of 9:1 solution (urea-ammonium nitrate and water) droplets were too large to produce improvements in visibility. The computer calculations indicated that 35 microns diameter (median-mass) droplets of 9:1 solution were sufficienctly small to produce good results under the conditions at which the calculations were computed. The effectiveness depends on the droplet sizes. Droplets 8 microns and under were calculated too small for fog treatment.

[1]Himel, C.M. (1969) "The Physics and Biology of the Control of Cotton Insect Populations with Insecticide Sprays." J. Georgia Entomol. Soc., Vol. 4, No. 2, April 1969.

TABLE 3.—RATES OF FALL OF SPERICAL DROPLETS
(sp. gr. = 1) IN STILL AIR*

Diameter (microns)	Velocity: (ft/sec)	(cm/sec)
200	2.3	70
100	.8	24.4
80	.55	16.8
50	.25	7.6
40	.15	4.6
20	.04	1.22
10	.01	.348
5	.0027	.082
1	10^{-4}	.00348

*Taken from Figure 114A, Terminal Velocities of Spherical Particles settling in air at 70°F, Chemical Engineers Handbook, 3rd Ed. P. 1021.

An empirical equation for the drop size produced by a gas-atomizing nozzle has been given by Nukiyama and Tanasawa (Ref. 5).

$$D_o = \frac{585\sqrt{\sigma}}{V\sqrt{\rho}} + 597\left(\frac{\mu}{\sigma\rho}\right)^{0.45}\left(\frac{1000\ Q_1}{Q_2}\right)^{1.5} \qquad (1)$$

where D_o = diameter of a single drop with same ratios of surface to volume as total sum of drops, microns (also known as Sauter mean diameter).

V = relative velocity between the airstream and the liquid stream, meters per second

Q_1/Q_2 = ratio of the volume flow rate of liquid to volume flow rate of air at vena contracta

ρ = liquid density, grams per cc

μ = liquid viscosity, poises

σ = liquid surface tension, dynes per cm

The equation is not dimensionally consistent.

The equation is applicable to solutions whose density is between 0.7 and 1.2 grams per cc and whose surface tension is between 19 and 73 dynes per cm.

When the constants for the 9:1 solution are put into the equation:

$$D_o = \frac{4570}{V} + 80.5 \left(\frac{1000\ Q_1}{Q_2}\right)^{1.5}$$

When Q_1 is 400 gallons per minute and the aircraft velocity 100 knots (169 ft/sec) it would be necessary to spray in a cross sectional area of 63 sq ft to cause the second term of the equation to be less than 2 microns. Since this is reasonable, the second term is dropped.

So when $Q_2 > 63\ ft^2$ we drop the 2nd term, then $D_o = \left(\frac{4570}{V}\right)$ microns

TABLE 4, Calculated Diameters as a Function of Air Velocity per (eqn 1)

V knots	V ft/sec	V meters/sec	D_o microns
100	169	51.5	89
120	203	65	74
150	253	77	59
200	338	103	44
300	507	155	30

When the ratio of air flow to liquid flow drops, the second term of the equation rises rapidly and thus the calculated droplet diameter.

ATOMIZATION MODEL

A model was developed with the appropriate equations to help describe the breakup of a liquid being sprayed from an aircraft.

The minimum energy required to form droplets would be the energy required to form the new surface.

$$E_{min} = \frac{3\ \sigma}{r\ \rho} W(\text{dyne-cm})$$

$$= 3.06 \times 10^{-3} \frac{\sigma\ W}{r\ \rho} \quad (\text{gram - cm})$$

where σ = liquid surface tension (dynes/cm)

r = drop radius (cm)

ρ = liquid density $g/(cm)^2$

w = liquid weight (grams)

The liquid is broken up by its kinetic energy relative to the air it impacts.

$$KE = 1/2\ mV^2 = 1/2\frac{W}{g}V^2$$

where V = velocity difference between liquid and air (cm/sec)

g = acceleration of gravity 980.6 cm/sec^2

By equating the above two equations we obtain

$$1/2\ \frac{W}{g}\ V^2 = 3.06 \times 10^{-3}\ \frac{\sigma W}{r\rho}$$

or

$$r = \frac{2(3.06)\sigma\ g(10^{-3})}{\rho}\left(\frac{1}{V^2}\right)$$

for 9/1 solution

$$r = 353\ (\frac{1}{V^2})\ \text{cm}$$

TABLE 5

The Minimum Velocity Necessary to Break up
9/1 Solution into Droplets of Radius, r, per (eqn. 2)

V	r	V	r
cm/sec	cm	ft/sec	μ
595	10^{-3}	19.5	10
1880	10^{-4}	61.6	1
5950	10^{-5}	195.0	0.1

The equation Nukiyama and Tanasawa developed (equation 1) and the equation developed based on minimum energy, equation 2,

$$d = 2r = \frac{129}{V^2} \frac{\sigma}{\rho} \qquad (2)$$

can be compared. σ, V, ρ and diameter are expressed in the same units in both equations (dynes/cm, meters/sec, g/cc, and microns). The lack of agreement of these two equations may be due to the experimental error most investigators have made in the past. Nukiyama and Tamasawa, as well as others, probably did not measure many, if any, of the particles under 4 microns in diameter which were present (Ref. 4). Furthermore, it is even more probable they did not measure proportionate amount of the smallest particles since small particle collection efficiencies on flat plates are so small relative to the larger drops.

It is of interest to think what the forces are on the liquid as it breaks up. Initially the liquid is moving at high speed relative to the air it is in. (For example, a 1 mm drop at 100 knots has an initial Reynolds number of 3500). The liquid is drawn out into ligaments which break and then collapse into droplets. The forces on the drop are a function of time and of velocity.

The instantaneous forces deforming and atomizing the liquid are:

Drag Force:

$$F_D = 1/2 \rho C_D S (\Delta V)^2$$

where

ρ = density of air

C_D = drag coefficient

= ϕ (Reynold's number and Mach No.)

s = cross sectional area

s = ϕ (liquid shape & orientation to ΔV)

(ΔV) = difference in velocity of liquid and air

= ϕ (time and the sum total of what the liquid has done since t_o)

Stagnation Pressure: $F_p = 1/2 \rho (\Delta V)^2$

The forces opposing deformation and atomization are:

liquid surface tension: $F\sigma = C\sigma$

where C = length along which the surface force acts

= ϕ (shape and orientation)

σ = surface tension of the liquid

liquid inertia: $F_i = ma$

where $a = \phi$ (shape and rate of change of shape)

liquid viscosity: $F\mu = \rho Ah$

where the head loss, h, is $h = \left[\phi^{1}(Re)\right] \frac{L}{d} \frac{V2}{2g}$

where ρ = liquid density

A = cross sectional area of flow

$Re = \frac{Vd\rho}{9\mu}$ = Reynold's number

L/d = length to diameter of flow

V = velocity of flow

g = gravity constant

Since these forces are not coaxial, they should be added vectorally.

Complications which will effect atomization are turbulent motion and wake eddies which introduce uncertainty to the (ΔV) and fluctuating pressure, coagulation of the droplets, electrostatic charging of the droplets, and high frequency oscillations of ligaments prior to breaking. Due to these complications the above equations would, at best, only give a rough description of the atomization process.

MEASURING TECHNIQUE

The measuring technique used in the wind tunnel is explained in detail by W. White (Ref. 1). The drops were caught in an eddy behind a barrier. The eddy deposited the drops onto gelatin coated glass slides. The imprints in the gelatin were measured with a microscope.

The second series of tests in the wind tunnel and the airborne tests used a similar method. A baffle was mounted on a platform (See plates 1-3). The slides were exposed by moving them to an open window in the baseplate. Before and after exposure the slides are covered by the baseplate. The slides were moved by an electric motor, thus the operator can change slides remotely. The baffle is 10-inches wide and 4-1/8 inches high; it is three inches in front of the open window.

TEST RESULTS. The most important tests were conducted by W. White in the NWC wind tunnel (Ref. 1). The droplet sizes were smaller than anticipated. The measured sizes were over an order magnitude less than those of drops collected on the ground under an aircraft spray (10 μ's vs 300 μ's). The gelatin coated slides, used to collect the droplet imprints for later measurement, do not record droplets smaller than 4 microns, thus droplets smaller than 4 microns would not be measured (Ref. 4). Neither nozzle orifice size nor nozzle pressure have as much effect on the droplet size as does the air velocity (Ref. 1).

Photographs of the tests of nozzles in the wind tunnel apparently show fewer droplets at equal flow rates (thus larger droplets) when the liquid is sprayed out into the air in the same direction as the airflow. When the liquid is sprayed normal to, or into, the airflow smaller droplets are formed (see Plates 4-6). This observation is no surprise since the droplet size should be inversely proportional to the relative velocity between the liquid and air. At 100 psi the velocity of the solution at the nozzle throat is about 90 ft/sec; at 150 psi, about 110 ft/sec (100 knots is 169 ft/sec). There should be less coagulation of droplets when the nozzle exist is normal to the airflow compared to when it is into the airflow. Minimum droplet size will probably be in this position.

Aircraft tests were conducted. The slides were exposed by a remotely operated device (see Plates 1-3) which was mounted on the side of a B-26 fuselage, about five feet behind the spray boom. The airborne test was conducted with water at an indicated airspeed of 130 knots. The median number drop size of the airborne test was 10.9 microns. The manufacturer advertises that the nozzle will spray a median volume diameter in excess of 1000 microns (Spraying Systems C., Nozzle 2515, 250 Series). The median volume diameter for the wind tunnel and aircraft tests was not determinable because too few droplets were counted. It would have taken a few thousand droplet measurements for each test condition.

NEW NOZZLES

Special Venturi. A Venturi shape was used in order to increase the kinetic energy of the airstream. The nozzle exist was at the location of maximum kinetic energy. Increasing the kinetic energy of the airstream makes more energy available to break up the liquid. When the air velocity is sonic, good atomization results in very small droplets. A 1-inch diameter throat was used with a 4-inch diameter entrance (see Plate 7). This small sized Venturi had a high ratio of wall area to cross-sectional area; therefore the energy loss due to wall friction was a large value relative to the total energy. As a result, a large percentage of the air spilled around the outside of the Venturi. The Venturi was in an open airstream. When the air spilled around, the velocity did not increase enough in the throat. As a result, the desired sonic velocity and atomization were not achieved.

High Speed Chopper. A high speed chopper was designed and built. It was designed to cut off a slice of liquid thin enough so that it would fracture into uniform particles about 30 microns in diameter. The outer drum was designed to rotate at 30,000 RPM (see Plate 8). The matching holes of the inner and outer drums allow a small column of liquid to advance before it is chopped off. The volume of liquid which advances is a function of the liquid velocity, the length of time the hole is opened, and the number of holes. If there are twelve rows, each with three holes, and the RPM is 30,000, then the hole is open for 1/6000 sec. The flow rate per hole per second is then chopped into 6,000 pieces. The piece then may or may not fracture further into smaller pieces. The device was designed for 20 gal/hr flow rate.

The volume chopped off is:

$$V = \phi\left(\frac{Q}{A_t},\ \Delta t,\ A\right)$$

where A_t = 36A = total outlet area (in^2)

A = hole cross sectional area (in^2)

Q = total flow rate (in^3/sec)

Δt = time the hole is open (sec)

$$V = \frac{Q}{A_t}\Delta tA$$

$$= \frac{Q\Delta t}{36}$$

$$= \frac{1.285}{36}\ \frac{1}{6000}$$

$$= 5.94 \times 10^{-6} in^3$$

$$4/3\pi r^3 = 5.94 \times 10^{-6} in^3$$

Therefore $r = 1.12 \times 10^{-2}$ in

$= 1.12 \times 10^{-2}(25,400)$ microns

$= 285$ microns

It is interesting that the hole size is not important, only the total number of holes and the number of rows.

Patent Search. A patent search did not find many promising designs. The most promising design was Patent 3,250,473, 1966, Mr. Hermann Hege, Thalhausenstranse 40, Freising, Germany. This invention relates to the atomization of a liquid film at a free edge of a rotating body, and more particularly, to a method of atomization which yields droplets of substantially uniform size. To date efforts to contact Mr. Hege have not been successful.

REFERENCES

1. W. C. White, "Wind Tunnel Tests of Aircraft Spray Nozzles" NWC TP Jan. 1971.

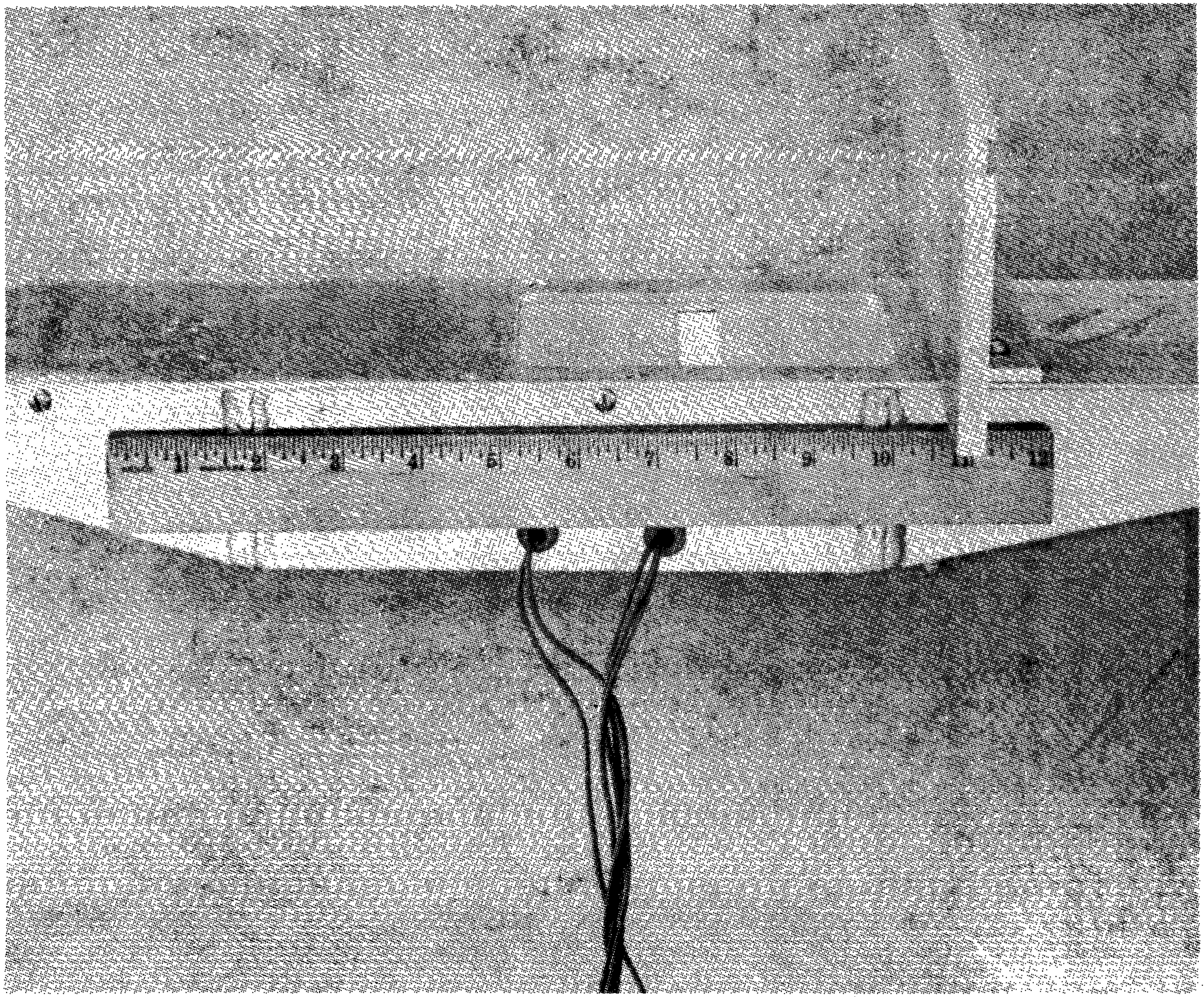
2. Paul Tag, David Johnson, Edward Hindman, "Engineering Fog Modification Experiments by Computer Modelling", Navy WEather Research Facility, TP No. 1-70, Jan. 1970.

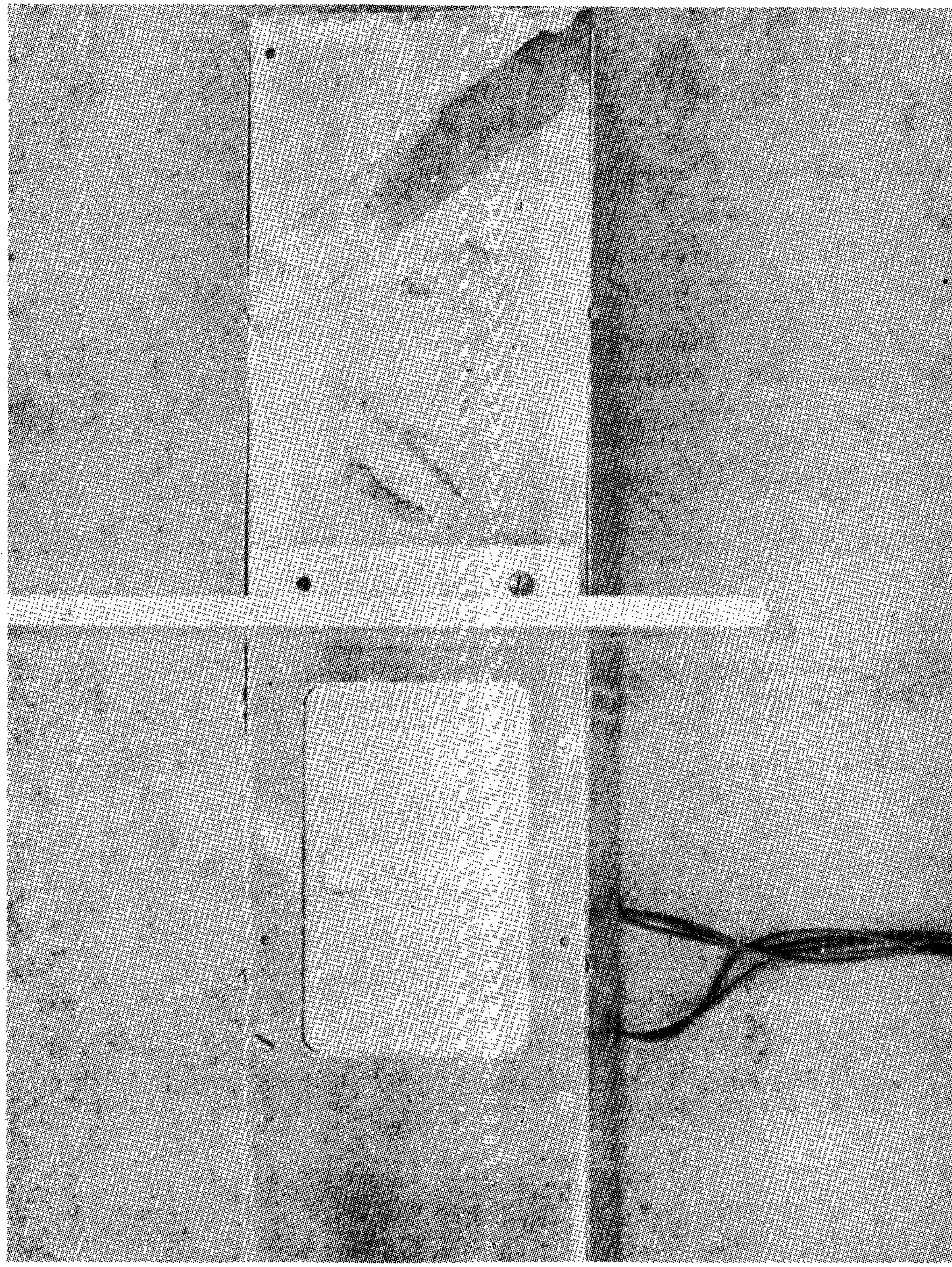
3. E. Blomerth, R. Clark, et al., "Project Foggy Cloud I", Naval Weapons Center, NWC-TP-4929, Aug. 1970.

4. Gale S. Rinehart, "Fog Drop Size Distributions - Measurement Methods and Evaluation", White Sands Missile Range, ECOM-5247, April 1969.

5. Nukiyama and Tanasawa, Trans. Soc. Mech. Engrs. (Japan), 5, 18, 63, 1939.

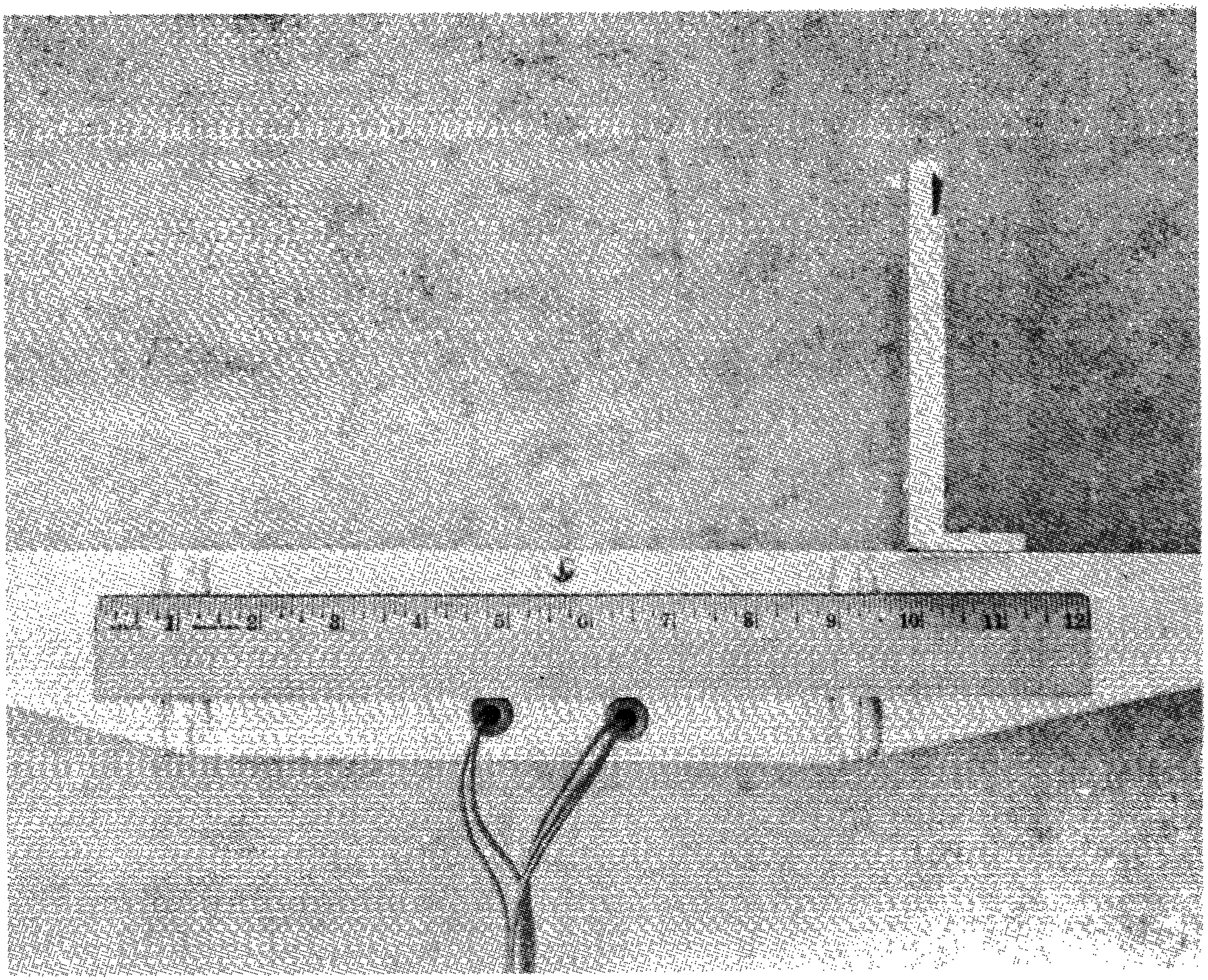
Plate 1: Airborne Droplet Measuring Device





Airborne Droplet Measuring Device

Plate 3: Airborne Droplet Measuring Device



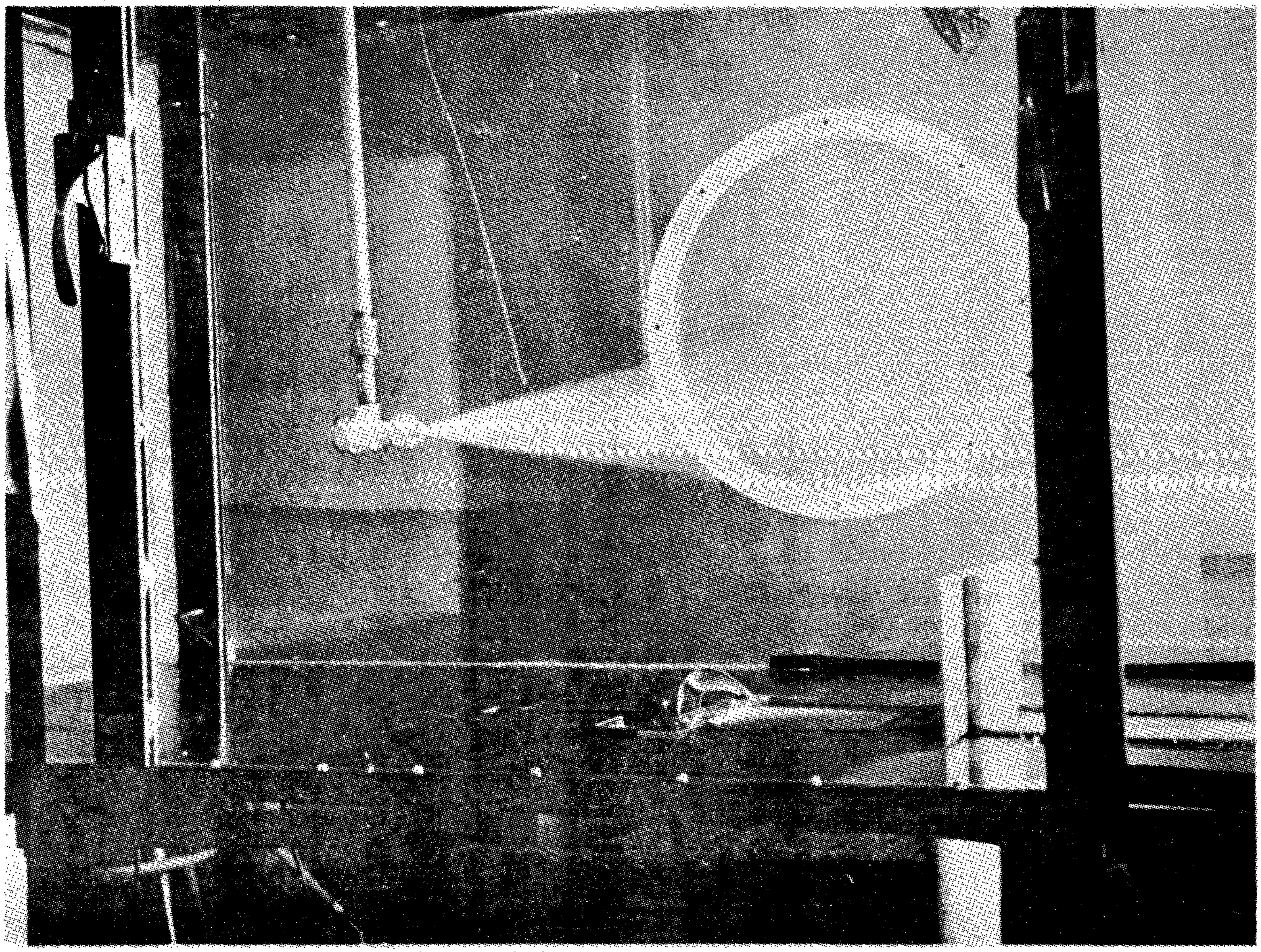
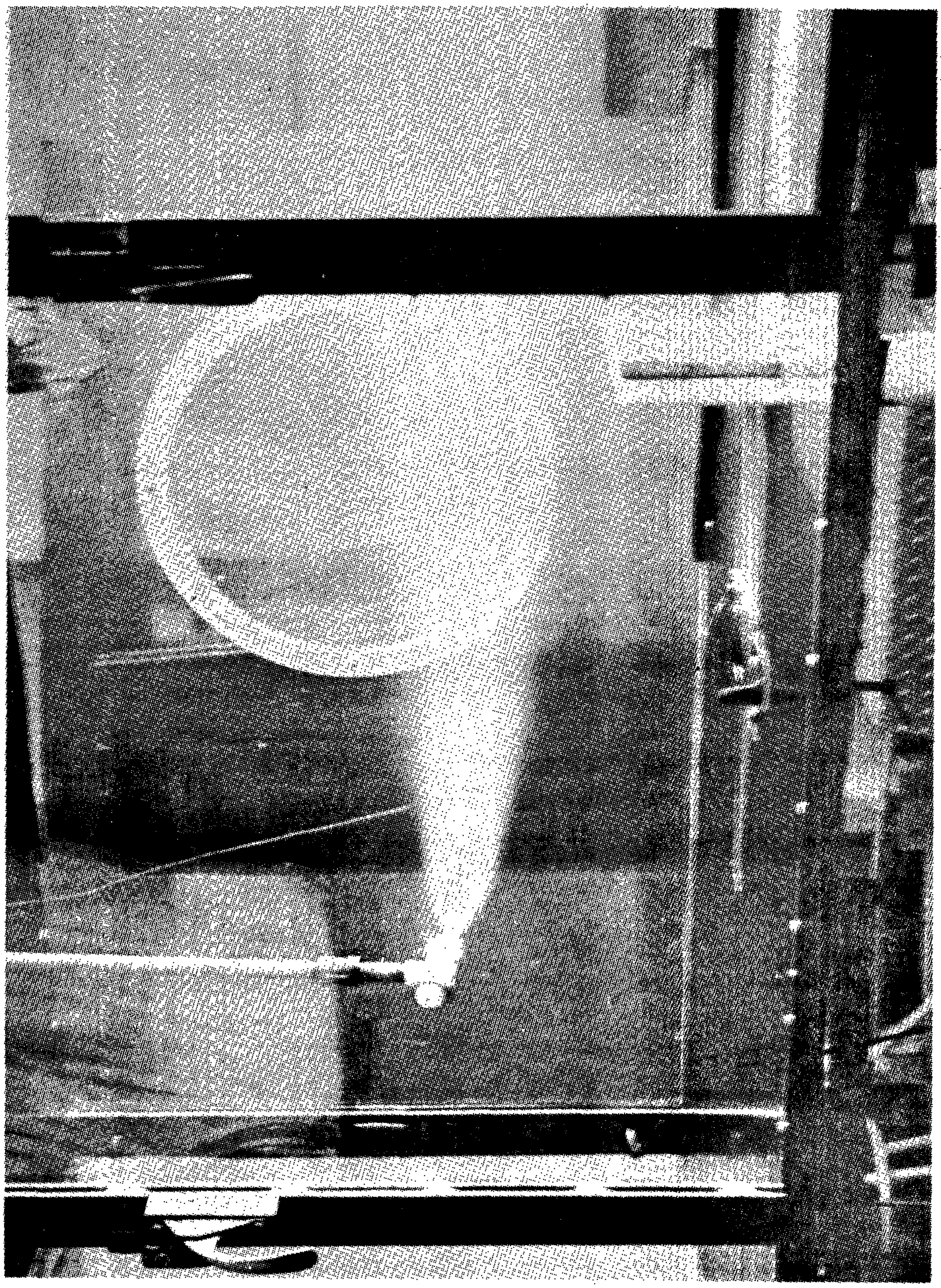


Plate 4: Spraying in Same Direction as Airflow

Plate 5: Spraying Normal to Airflow



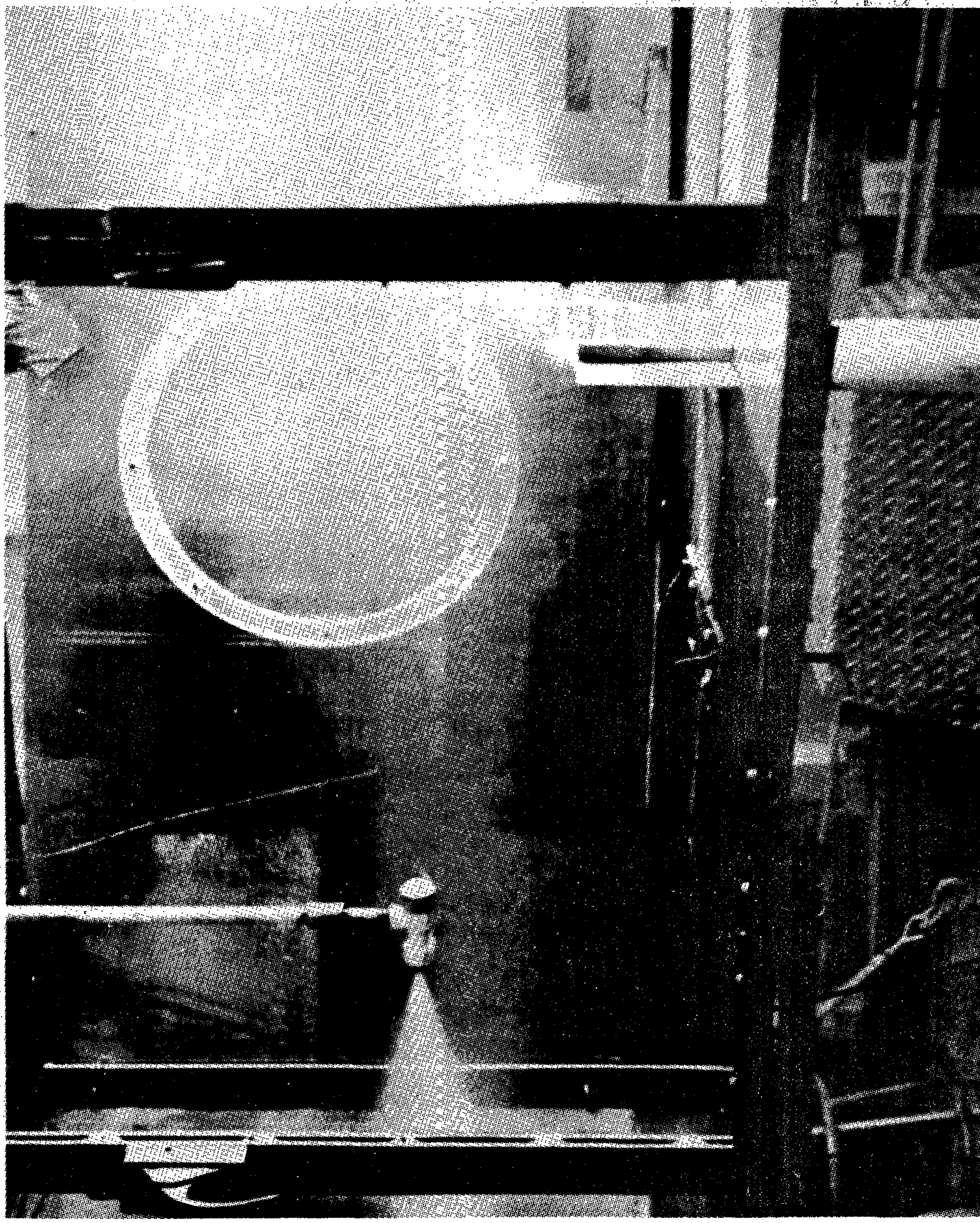
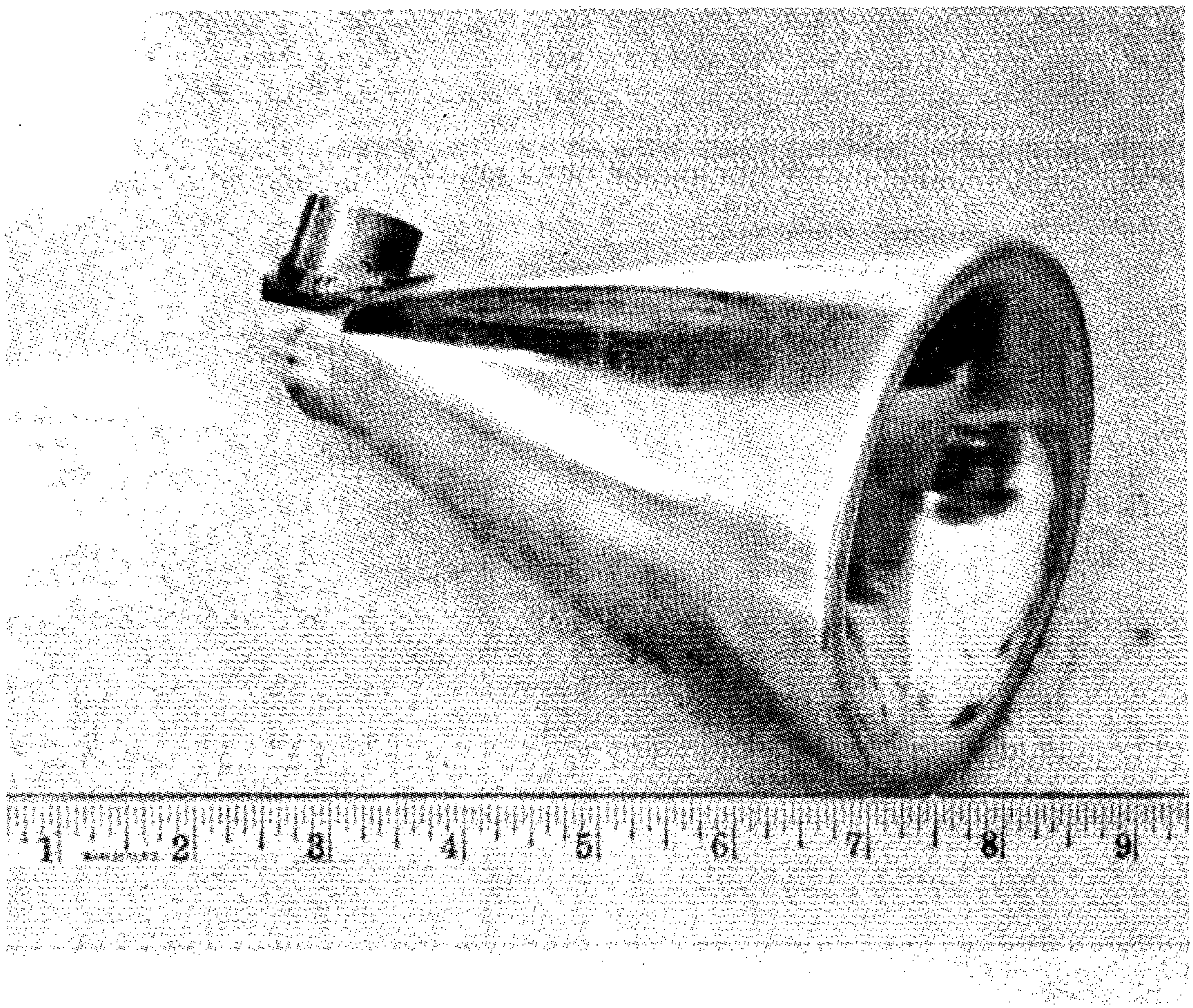


Plate 6: Spraying into the Airflow

Plate 7: Special Venturi



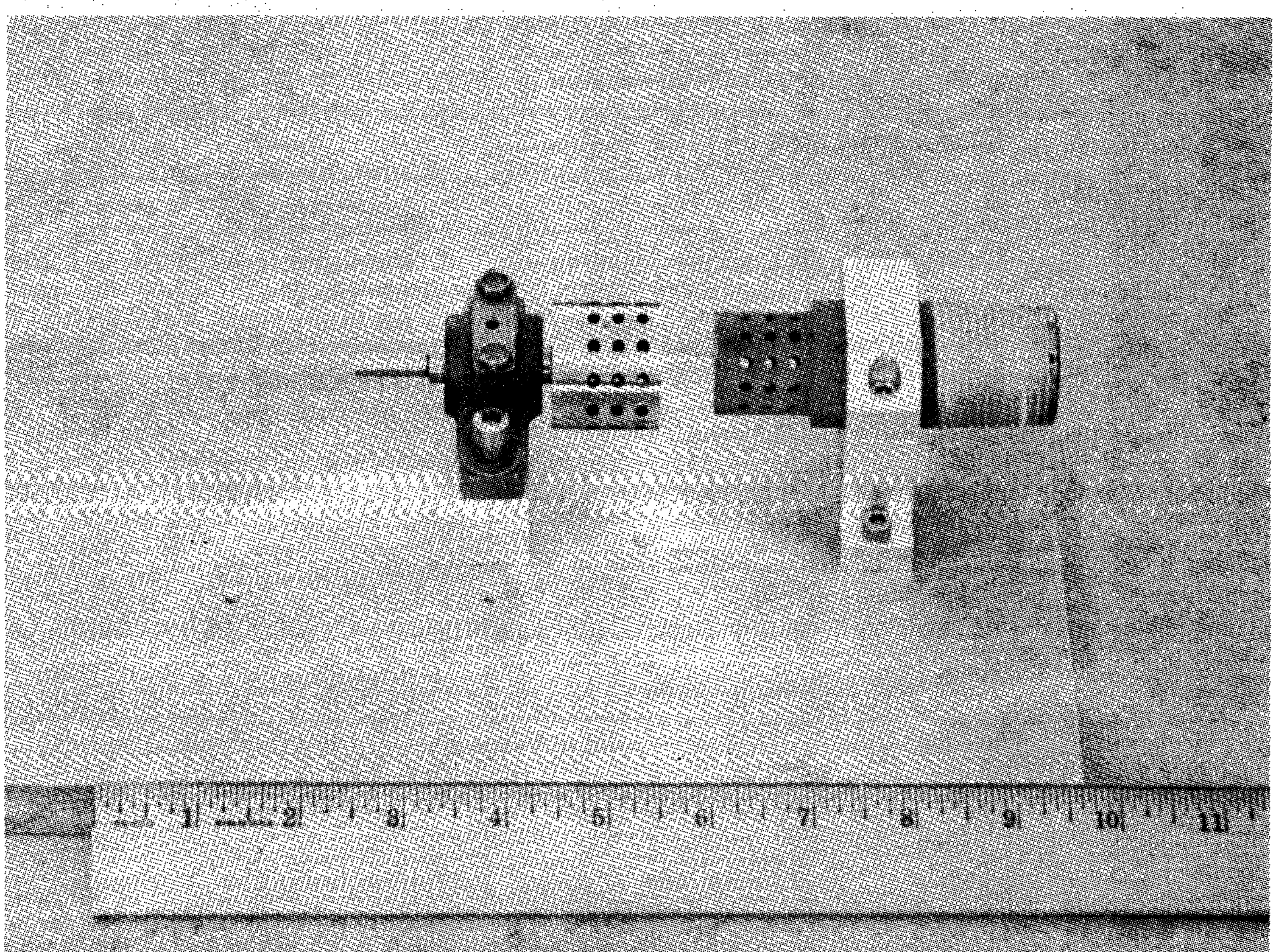


Plate 8: High Speed Chopper

MEANS FOR ESTIMATING AREAL HAIL-DAY FREQUENCIES

Stanley A. Changnon, Jr.
Illinois State Water Survey
Urbana, Illinois

INTRODUCTION

The design of experimental and operational hail suppression projects is faced with several obvious questions (Changnon, 1970b). An obvious planning problem concerns the need to know the number of hail days expected over the area where a suppression project is to occur. Point averages and extremes of hail days, based on long records at first-order stations of the U.S. Weather Bureau, were made available in 1947 (U.S. Department of Commerce, 1947), and more recently average monthly hail-day patterns, based on point values at 1,285 stations including first-order and cooperative substations in the central United States, were published (Stout and Changnon, 1968). However, there is very little data available to derive accurate areal frequencies of hail days.

The purpose of this paper was to assemble available point-area data, to examine the relationships between the average area and point hail-day frequencies, and to express these relationships in a format useful for determining areal frequencies, given point frequencies.

A hail day was defined as a date with hail at an observing point (station). This hail may or may not be damaging, may be small or large, and may have resulted from more than one hailstorm during the day. Nevertheless, hail-day frequencies are useful in planning operational day frequencies regardless of whether the project is based on a continuous or a random-choice seeding mode.

DATA

Operations of dense hail networks of varying sizes in Illinois and one in Colorado during the past 22 years have furnished useful data on areal-point relationships for areas ranging from 1 to 56,400 mi^2. Other studies involving data from the standard climatic (substation) network of the Weather Bureau that operates throughout the Nation also were employed to develop area-point values. The sources of data utilized in this study are described in Table 1, illustrating for each network-area or region its size, number of observation points, years of data collection, study period used per year, the average point hail-day value (based on all point data in the area), the average area hail-day frequencies, and the area-point ratio. Also noted are the sources of the data.

Comparison of the area size and the values for number of stations there are two station-density classes. All data sources, excluding those five labeled as regions, were essentially from dense hail networks often supplemented by extensive crop-hail insurance data. For instance, the data for the state of Illinois consisted of that from 119 Weather Bureau stations plus all available insurance hail-day data which involves 10% coverage (liability) of the entire state, or approximately one large sampling point per 10 mi^2. Thus, in all ten network-area cases shown in Table 1, the density of observations points exceeded one per 15 mi^2. The five geoegraphical study areas labeled as "regions" in Table 1 represent data sources where the sampling density was markedly less, ranging from 1 per 200 mi^2 to 1 per 560 mi^2.

RESULTS

The 10 "higher density" network values for area size were correlated with their area-point ratios, giving a coefficient of +0.99. This excellent relationship between the area-point ratios and the area size is also impressive because it is based on data from various study periods ranging from annual to summer season only and because the Colorado network data agree with that from Illinois. These 10 network values were also used to derive an equation, $\ln R = .058 + .313 \ln A$, where A represents area size (mi^2) and R = the area-point ratio. The five regional values also were used to derive an equation, $\ln R = -1.795 + .421 \ln A$.

The area size and area-point ratios of the 15 data sources were plotted (Fig. 1), and the best-fit lines from the predictive equations were constructed. The excellent agreement of the ratios with area for the 10 dense network values suggests this recent relationship is well established and appropriate to use in the Midwest and Great Plains to estimate area average hail-day frequencies where the average point values are known. For instance, in a 3,000 mi^2 area the area-point ratio would be 11.0, as determined from Fig. 1, and if the average point frequency was 5 hail days for the year (or for any given season of the year), the area average number of hail days would be 55.

Because of inadequate point sampling, the 5 regional values in Table 1 obviously (Fig. 1) do not provide accurate estimates of the areal hail day frequencies within them. However, since such data are the only "area" results available in these geographical areas, it is useful to compare their size-ratio relationships with those predicted by the curve based on the 10 dense networks to examine the degree of underestimate in the regional values. Furthermore, the extremely good fit of the 5 regional values (Fig. 1), 3 of which are from different climatic areas (Colorado, Illinois-Iowa, and South Dakota) indicates that although point frequencies may vary geographically and seasonally, there is little geographical variation in the area-point day-day relationships.

Table 1. Average Point and Areal Frequencies of Hail Days and Their Ratios for Various Sized Hail Networks and Regions

Network and/or Area	Area Size mi^2	No. of Sta. (pts.)	Length of Record	Study Period	Pt. Ave. Hail Days	Area Ave. Hail Days	Area-Pt. Ave.
Illinois Airport Network (1)	1	6	1967-70	Annual	3.3	4.0	1.2
Ill. Champaign Network (1)	10	13	1960-70	Annual	3.2	6.5	2.1
Illinois Dense Network (2)	100	108	1968-69	June-Sept	1.7	7.0	4.1
Colorado(Denv.)Network (3)	160	40	1949-55	Annual	5.0	23.0	4.6
Ill. Kankakee Network (4)	300	30*	1966-67	Apr. April-Sept	2.4	14.0	5.9
E. Central Ill.Network (5)	400	135	1967-69	April-Oct.	2.6	17.0	6.5
Southern Ill. Network (6)	550	49	1958-62	Annual	1.2	9.0	7.5
Central Ill. Network (7)	1,600	550	1968-69	April-Sept.	2.3	24.0	10.4
Ill. Observer Network (1)	18,440	1325*	1959-62) 1967-68)	April-Sept.	1.3	34.0	26.1
Illinois (8)	56,400	119*	1951-60	March-Oct.	2.4	84.0	35.0
Regions							
Southwestern Illinois (9)	1,000	5	1934-63	Annual	2.8	8.6	3.0
Southwest S. Dakota (10)	1,000	5	1910-64	June	0.86	2.9	3.3
Western Illinois (9)	3,000	8	1934-63	Annual	2.5	11.0	4.4
Northeast Colorado (11)	4,500	8	1946-69	15 May-31 July	2.3	13.0	5.7
Iowa (12)	56,200	150	1916-40	Annual	3.6	58.0	17.0

Sources

1) File data at Illinois State Water Survey
2) Changnon (1969a), Changnon (1970a)
3) Beckwith (1957)
4) Changnon (1969b)
5) Changnon (1969a), Changnon (1969b)
6) Changnon (1963)
7) Changnon (1969a), Changnon (1970a)
8) Changnon (1962)
9) Changnon and Schickedanz (1969)
10) Schleusener (1966)
11) Schickedanz and Changnon (1970)
12) Shands (1944)

*Point hail-day data supplemented by hail dates from paid crop-hail insurance claims. In the Kankakee and Observer Network 50% of the area insured, and 10% of Illinois was insured.

SUMMARY

The results presented in this paper offer a means for estimating the area average hail days for widely ranging sizes of areas. This should be of value in planning experimental and operational hail suppression projects and it will also be useful operational information to crop-hail insurance interests. The area-point relationships appear to have little geographical difference within the United States, but they do vary directly with area size. As expected, they also vary if the density of the observing points in the area is something less than 1 per 15 mi^2.

ACKNOWLEDGEMENTS

This research was supported by the Atmospheric Sciences Section of the National Science Foundation under NFS Grant GA-16917. Credit is due to the many individuals who processed the voluminous data required to derive these results, and the Crop-Hail Insurance Actuarial Association of Chicago is acknowledged for furnishing extensive insurance data. The comments of Mr. Thomas Henderson of Atmospherics Incorporated were extremely useful in the preparation of this paper.

REFERENCES

Beckwith, W. B., 1957: Characteristics of Denver Hailstorms. Bulletin American Meteorological Society, 38, 20-30.

Changnon, S. A., 1962: Areal Frequencies of Hail and Thunderstorm Days in Illinois. Mo. Wea. Rev., 90, 519-524.

Changnon, S. A. 1963: Precipitation in a 550-Square-Mile Area of Southern Illinois. Trans. Ill. and Science, 56, 165-187.

Changnon, S. A., 1969a: Insurance-Related Hail Research In Illinois During 1968, Res. Report 40, Crop-Hail Insurance Actuarial Assoc., Chicago, 46 pp.

Changnon, S. A., 1969b: Hail Evaluation Techniques. Final Report (Part 1) NSF 6A-482, Urbana, 97 pp.

Changnon, S. A., 1970a: Hailstreaks. J. Atmos. Sci., 27, 109-125.

Changnon, S. A., 1970b: Design Factors of a Hail Suppression Experiment in Illinois. Reprints Second National Conference on Weather Modification, 150-155.

Changnon, S. A., and P. T. Schickedanz, 1969: Utilization of Hail-Day Data in Designing and Evaluating Hail Suppression Projects, Mo. Wea. Rev., 97, 95-102.

Shands, A. L., 1944: The Hail-Thunderstorm Ratio. Mo. Wea. Rev., 72, 71-72.

Schickendanz, P. T. and S. A. Changnon, 1970: A Study of Crop-Hail Insurance Records for Northeastern Colorado with Respect to the Design of the National Hail Experiment. Final Report NCAR 155-170, Urbana, 85 pp.

Schleusener, R. A., 1966: Reports from Participating Groups. Project Hailswath Trial Report, Volume 11, NSF-C461.

Stout, G. E., and S. A. Changnon, 1968: Climatography of Hail in the Central United States. Res. Report 38, Crop-Hail Insurance Actuarial Assoc., Chicago, 49 pp.

U. S. Dept. of Commerce, 1947: Thunderstorm Rainfall. Hydrometeorological Report 5, Weather Bureau, 331 pp.

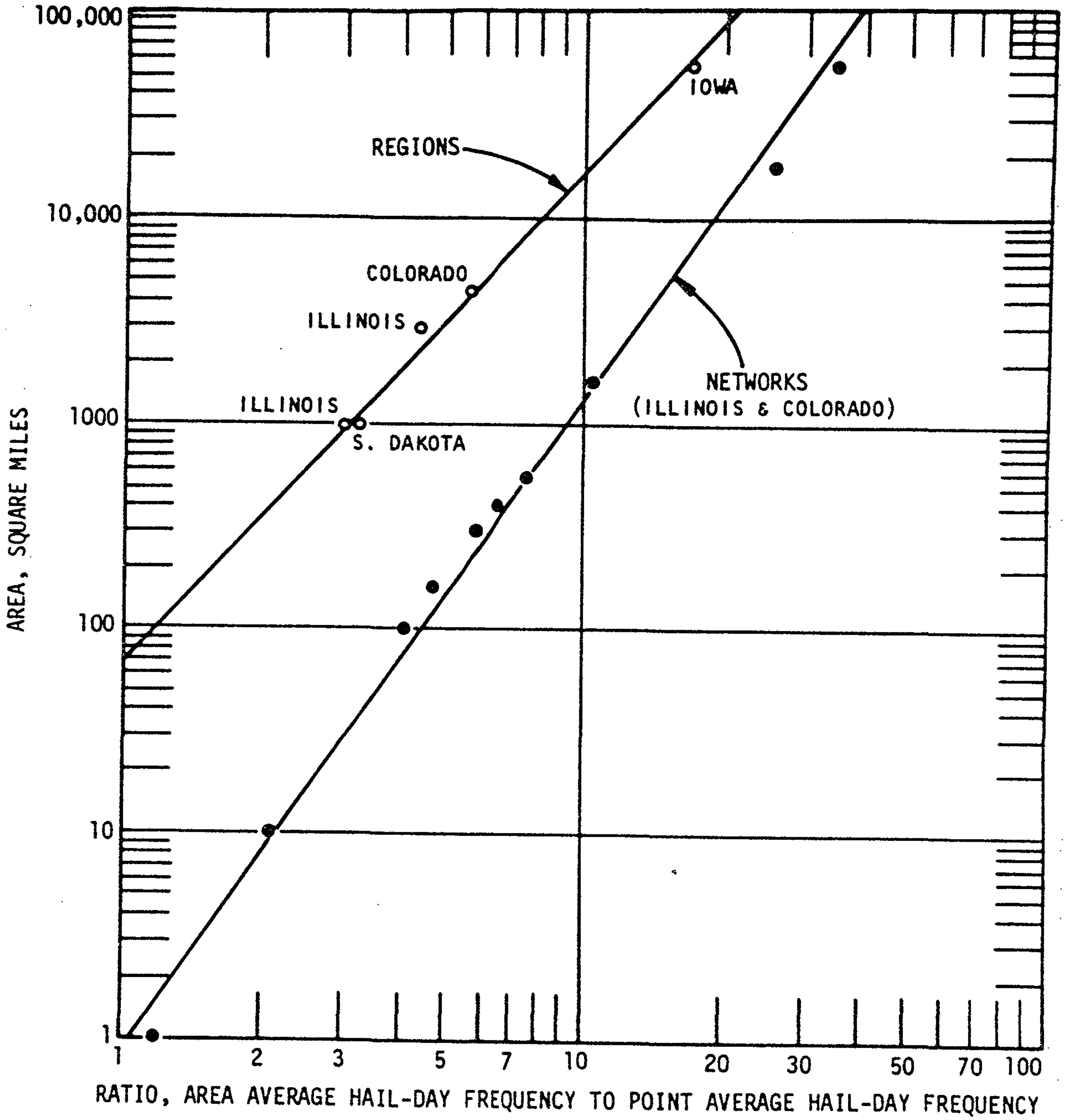


Figure 1. Area-Point Hail-Day Ratios as a Function of Area Size

THE DESIGN AND EVALUATION
OF
THE NATIONAL HAIL RESEARCH EXPERIMENT
IN NORTHEAST COLORADO

Paul T. Schickedanz
and
Stanley A. Changnon, Jr.

Illinois State Water Survey
Urbana, Illinois

ABSTRACT

Historical crop-hail insurance data were used to evaluate selected tests and experimental designs related to the daily experimental unit in a single target area. The random-experimental design (days are randomized over a single target area into seeded and non-seeded days with the non-seeded days being the control) was recommended for the design and eventual evaluation of the National Hail Research Experiment. Using percent loss insurance data, it is estimated that there is a 70 percent chance of detecting a 40 percent decrease in hail damage in 7 years and a 50 percent chance in 4 years. If a "combined" test is made when a 40 percent reduction in damage is accompanied by a 40 percent reduction in the probability of hail, there is a 70 percent chance of detection in 4 years and a 50 percent chance in 2 years.

1 INTRODUCTION

In the early planning stages of the National Hail Research Experiment (NHRE), a decision was made by NCAR officials that project seeding operations would involve the daily unit in a single target area. The Illinois State Water Survey which has made extensive studies of hail climatology and the design aspects for hail suppression experiments in Illinois (Changnon, 1969; Changnon and Schickedanz, 1969; Schickedanz and Changnon, 1970a) then made a study of the various aspects of the random daily design with the use of crop-hail insurance records from Colorado (Schickedanz and Changnon, 1970b). Other possibilities, such as a cross-over, target-control, and paired or individual storm designs were not included in the study because the choice of seeding on a random daily basis in a single target area had already been made by NCAR officials. Thus, the specific purpose of the study was to evaluate selected statistical tests and experimental designs through the use of historical crop-hail loss data in order to estimate the time required for the detection of possible reductions in hail damage and in the probability of hail. This paper presents the most pertinent findings and recommendations resulting from that study.

2. BASIC DATA

a. Source and description of data

Location of the three counties supplying basic data for the various studies in this research are shown in Fig. 1. The largest area was Weld County with 4004 mi^2, and the smallest was Morgan County with 1282 mi^2. Logan County consisted of 1827 mi^2, and the study area consisted of 600 mi^2. In addition to these three counties, an area which incorporated all three counties combined is often used and is designated as the Tri-County area. The Tri-County area consisted of 7113 mi^2 and was used along with the other three areas to provide information concerning the effect of areal size upon the sample requirements for the proposed hail experiment.

The basic insurance data used in this report were supplied at no cost by the Crop-Hail Insurance Actuarial Association. Data from the association of insurance companies were used to develop the areal pattern of total township insurance liability for the 1931-1962 period in the Tri-County area (Fig. 2). The pattern of total township liability for 1931-1962 in the three counties is shown in Fig. 2. This figure shows that the southeastern half of the proposed study area had more than $25,000 (cumulative) liability per township while the extreme northwestern area had liability that totaled less than $5,000 per township. Among the individual counties, Logan County appears to have had the greatest areal coverage of liability with cumulative amounts greater than $25,000 (cumulative) liability for most of its area. Morgan and Weld Counties have approximately half of their areas with more than $25,000 (cumulative) liability per township. The scarcity of liability in the northwestern part of the study area illustrates the need of a relationship between the hailfall parameters (energy, volume, of ice, and momentum) from detection devices and the crop-insurance damage in order to utilize the crop-insurance data to its fullest in the research area and to estimate crop losses with these devices during the experiment.

The daily insurance records were available for the period May-October, 1957-1969. Fig. 3 is a plot of the yearly dollar loss, number of damage hail days, and the annual amount of liability in the various areas. This figure implies that as the number of damaging hail days increases, the amount of damage also increases. This gives credibility to the hypothesis that if damage is decreased by seeding, there might also be a corresponding decrease in the number of hail days. Thus, in the statistical analyses considerable attention is given to this hypothesis. There is a tendency for the county liability to be less during the mid-60's than during any other time in the 13-year period (Fig. 3). Some of the years with high losses near the end of the 13-year period are partially explained by the greater amount of liability at those times. The reasons for the flutations involve crop successes and variation in

Figure 1: Location of the study area and the counties used in the study of the crop-hail insurance records.

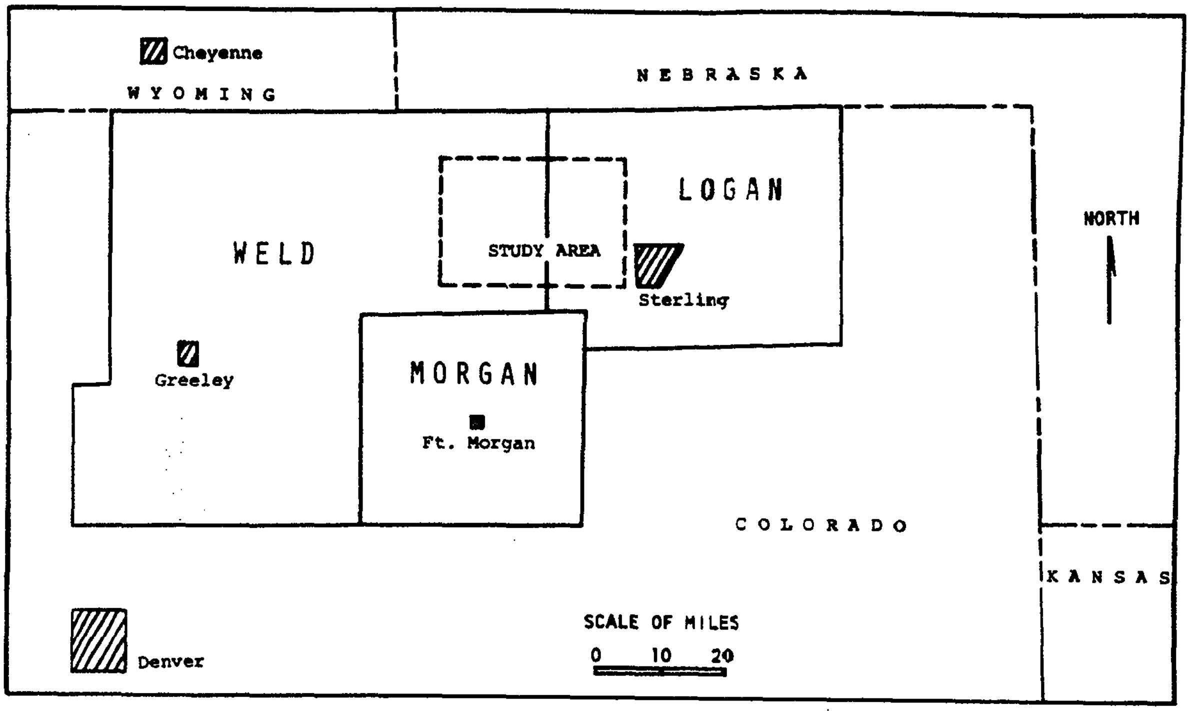
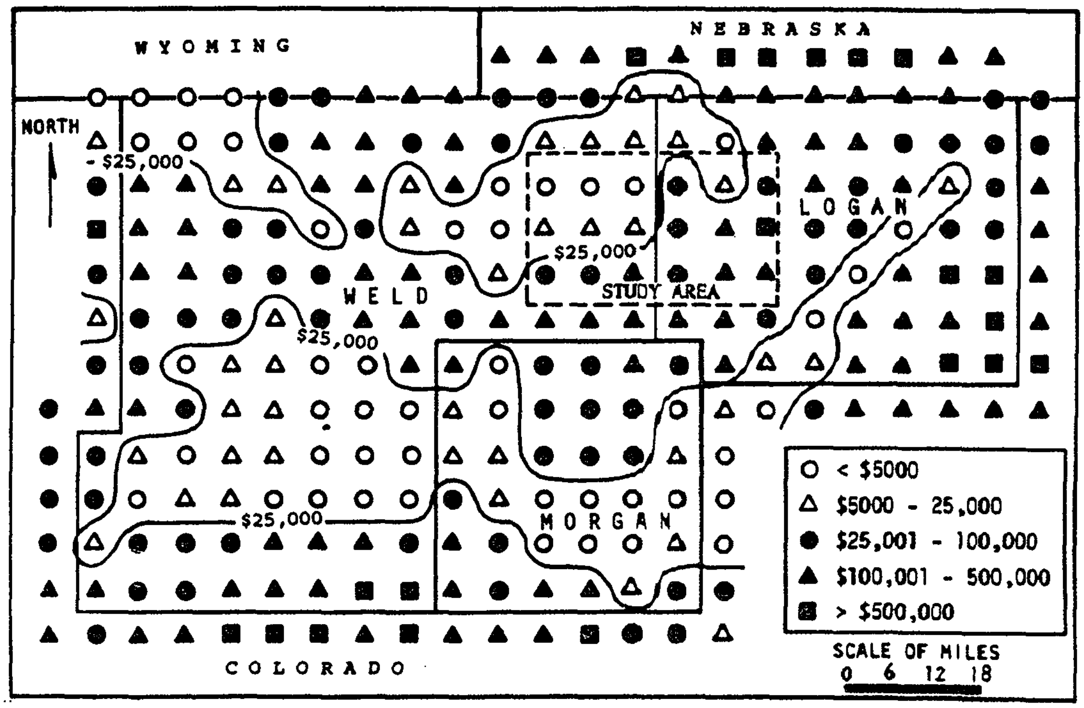


Figure 2: Pattern of total township liability (1931-62) in Weld, Morgan, and Logan counties and environs in Colorado. Township value plotted in center of township (6x6 miles).



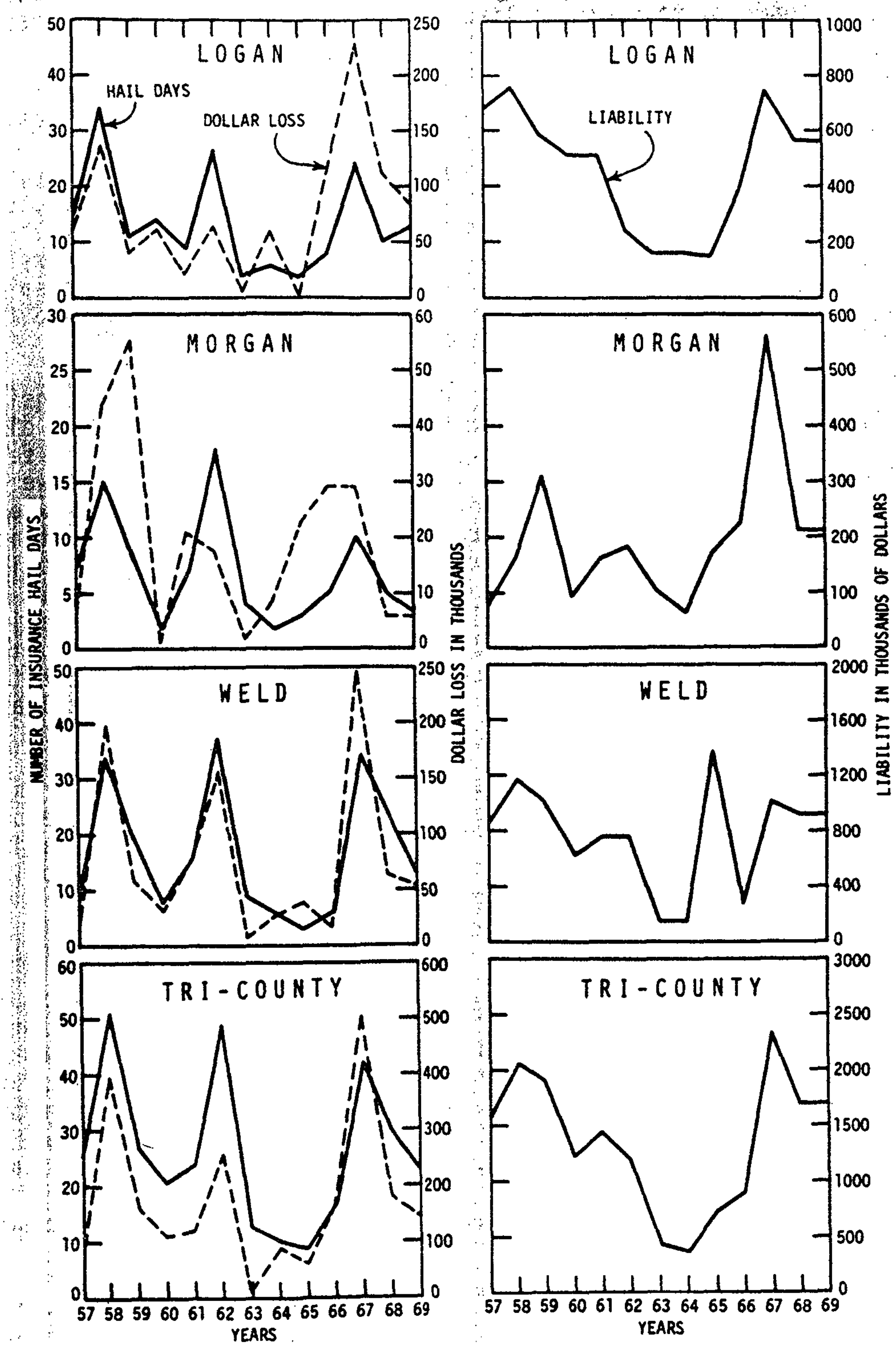


Figure 3: Yearly amounts of hail days, dollar loss, and liability for the four areas.

hail losses. That is, there is a tendency to decrease liability after periods of good yields and low hail losses and a strong tendency to increase liability after periods of heavy losses.

b. Adjustment indices and hail parameters

Although hail insurance data are a realistic practical measure for the evaulation of hail suppression activities, direct comparison of the loss in one month with that of another, or the comparison of the data in one year with that of another, cannot be accomplished without certain adjustments to the data. These problems of change during a crop season and between years involve factors such as susceptibility, liability, price and area size. Several adjustment indices were developed and applied to the hail insurance data (Schickedanz and Changnon, 1970b).

In order to obtain a further refinement of the intensity of the hailstorm, and to circumvent some of the adjustment difficulties inherent in the adjustment process, two additional sets of data were derived. First, the dollar and acreage data were expressed in terms of the average dollar loss per claim (dollar extent), and the average number of acres damaged per claim (areal extent). It was believed that these would be relatively free of liability and area size variability problems since the number of claims should vary with the amount of liability and the amount of area in crop land. Secondly, the dollar loss was divided by the amount of liability to obtain the percent loss. It was believed that this expression would be relatively free of liability, changing dollar, and areal size problems.

The end result of the adjustment procedures was the establishment of seven hail parameters which were used in this report. These were the dollar loss, the number of acres damaged, the adjusted dollar loss, the adjusted number of acres damaged, the dollar extent, the areal extent, and the percent loss.

c. Definitions

Operational days. These are defined in this research as those days on which hail is forecasted to occur. This definition was chosen because it was believed that the design of the experiment should be such that the forecasting scheme would be sure to include all of the hail days. Thus, it is a foregone conclusion that forecasting will be imperfect and some of the operational days will in fact have no hail. For computational purposes it was assumed that 62% of the operational days will have hail. This number is based on the 1969 and 1970 forecasting experience in the region of NHRE (Goyer*).

Seeded and non-seeded days. These days were designated to be seeded or non-seeded by random choice. It was assumed that random choices will be made from the operational days after they are

*Personal communication

chosen by the forecasting scheme. In this report sample sizes are computed for varying ratios of seeded to non-seeded days.

The *experimental unit* is defined to be an operational day in the experiment. Thus, the term "hail damage per experimental unit" refers to both the hail and non-hail days. *Liability* is defined to be the total dollar amount of insurance in force. Daily liability refers to only the liability on claims which had actual damage, whereas the total county liability for the year refers to the total amount of insurance in force irrespective of the amount of loss.

Alpha (α) is the probability of asserting that there is a seeding effect present, when in fact, there is not. *Beta* (β) is the probability of asserting that there is no seeding effect present, when in fact there is a seeding effect. The *power* ($1-\beta$) of the test refers to the probability of detecting a seeding effect when the effect is present.

d. Limitations

The reason for examination of historical climatological data in the area of the proposed hail experiment is to help establish useful elements of the design such as the observational unit, desirable predictor variables, the hail parameter, the size of target, and the proper tests to use in the evaluation. The method essentially is one of trial and error (Neyman and Scott, 1967a). The analyses then provide information on the expected duration for a specified precision level and particular test under the assumption that the future experiment will be performed in conditions like those reflected in the historical data. Thus, one limitation is that the weather conditions of the historical period will not necessarily be duplicated during the period of the experiment, and thus, sample size will not be exactly as estimated. But, projection of past experience into the future is still the best estimate available.

The use of results presented in this paper to evaluate actual results from the field experiment is affected by the fact that only half of the proposed study area has cumulative liability greater than $25,000 for the period 1931-1962. To translate the field project results into crop-loss values applicable to those in this report some degree of correlation must exist between the hail sensing devices to be installed and the damage to crops. Another alternative might be to use Logan County as the study area, since it has widespread liability and is larger than the present study area.

All results are presented under the assumption that 62% of the forecasted hail days will in fact have hail. In the analyses, this value was used for all three counties and the Tri-County area.

It is very likely that the forecast accuracy will vary according to the size of the area involved, but this factor was not considered in the analyses.

Also, the results in this paper concern only the daily experimental unit and designs involving that unit over a single area. Other designs are possible, including 2-area crossover, target-control, or random individual storm, but the design was chosen by NCAR before this study began and the sole purpose was to evaluate the various ramifications of using the daily experimental unit and a single target area.

3. Analytical procedures

a. Theoretical frequency distributions.

The log-normal distribution and gamma distributions were fitted to the seven hail insurance parameters derived. These distributions were then tested for "goodness of fit" by methods employed by Schickedanz and Changnon (1970a). The distributional parameters for the log-normal and gamma distributions along with sample sizes, means, standard deviations and the probabilities of the goodness of fit tests are given by Schickedanz and Changnon (1970b).

The testing of the hail distributions for goodness of fit was performed for the purpose of determining how well the log-normal and gamma probability models will describe the data because resulting computations of sample size are based upon the assumption that one or both are reasonable models. The conclusion reached here is that at least one and in some cases both of the models can be used with the seven hail parameters.

b. Design considerations

Only designs involving the daily experimental unit were investigated. This was based on a decision by the NCAR authorities regarding NHRE to use the daily unit as the experimental unit for the experiment. Thus, only four designs are considered. These designs are based first of all, upon the use of data during the experimental period and secondly based upon the use of historical data in the design. These designs are as follows: 1) random-experimental which involves randomization of days over a single target area and into seeded and non-seeded days with the non-seeded days being the control; 2) random-experimental with predictor variables which includes independent meteorological predictor variables incorporated into the random-experimental design for the purpose of increasing the precision and power of the experiment; 3) random-historical in which a random choice is made of days to be seeded over a single target area with the historical record as the control; 4) continuous-historical in which all hail days within a given stratification are seeded with the historical record as the control.

Designs based on historical data alone tend to produce smaller sample sizes than those based solely on experimental data. However, these designs depend upon certain assumptions in order for them to be valid. Historical designs involve the use of non-seeded hail amounts observed during a historical period preceding the actual experiment. Thus, it is possible that the historical method may bias the evaluation by the fact that the historical period may be dominated by storms either favorable or unfavorable to seeding. However, during the seeding period the opposite storm type may prevail. In the Colorado experiment, historical data for the hail sensing devices will not be available and unless a good relation is established between damage and sensing data, the designs involving historical data can not be used. To circumvent the difficulty of bias, Schickedanz and Changnon (1970a) suggested the use of a random-historical design where the random non-seeded days would be used as a control to check on trends during the experimental period. Subsequently, Huff and Schickedanz (1970) have suggested the use of the historical comparison as supplementary data to the random-experimental design. The merit of this type of combination is that one can take advantage of the lack of assumptions and the more stringent requirements involved in the random-experimental design and yet at the same time make use of the smaller sample sizes possible although less valid in regard to assumptions involved in the historical design. In the continuous-historical design, there is simply no way to insure that the seeding period does not have a different weather regime than during the historical control period. If the regime is different a fictitious seeding effect may be created because there are no control days in the experimental period. Thus, it appears that some randomization must be employed.

In this research, the number of years required to obtain significance for all four designs listed above are computed. However, the major emphasis is placed upon sample sizes obtained with the random-experimental design.

c. Tests of hypotheses and computation of sample sizes

Several assumptions were made in regard to various hypotheses and tests of hypotheses employed in this study. These assumptions are 1) whether seeded or not, to each experimental unit there is a positive probability of no hail, and that this probability may be affected by seeding. That is, seeding may stop hail damage that would have occurred on a given day or seeding may create hail damage on a day where it would normally not have occurred; 2) when hail damage does occur, the daily hail damage is distributed according to either the log-normal or gamma probability distributions. In the case of predictor variables, it is assumed that the hail damage has a linear regression on the predictors; and 3) the effect of seeding is to produce a scale change in the hail damage distributions. No other change is assumed, with the exception of complete elimination or creation of hail as in assumption 2 above.

Thus, the shape parameters of seeded distributions are assumed to be the same as the shape parameters of the non-seeded distributions.

These assumptions led to the formulation of three hypotheses which are likely to be tested during the hail reduction experiment. Under these hypotheses, various tests and designs involving the daily experimental unit over a single target area were considered as probable during the operational period. The number of experimental units necessary to insure detection of various assumed seeding decreases for a given precision level were then determined. The following null hypotheses were formulated.

Hypothesis A, seeding does not affect the conditional distribution of hail damage, given that hail occurs. For this hypothesis several tests were used in conjunction with the three designs. An optimal $C(\alpha)$ test was used which assumed that the hail-damage was gamma distributed or that there was a linear regression on the predictor variables. The test involved two cases: one without predictor variables and one with predictor variables. Formulas for these cases are given by Neyman and Scott (1967b) and were only employed with the random-experimental design. The log-normal (non-sequential) tests were used with all three designs without predictor variables. The log-normal sequential test was used with the random-historical and continuous-historical designs without predictor variables. Relationships necessary for the computation of sample size for the log-normal tests are given by Schickedanz and Changnon (1970b).

Hypothesis B, seeding does not affect the probability of hail in the target. For this hypothesis an optimal $C(\alpha)$ test which is a modification of the classical X (Neyman and Scott, 1967b) was used. Formulas for the number of experimental units with hail necessary to insure a given power and significance level are given by Neyman and Scott (1967b). This hypothesis by itself, is simply a test of whether the hail damage is eliminated completely or is created on a particular day, and it was considered only for the random-experimental design. The reduction in hail damage on a particular day is not considered under this hypothesis.

Hypothesis C, seeding does not affect the hail damage averaged per experimental unit. This is a combined test of whether the probability of hail on a given day and the amount of hail damage is reduced simultaneously. A $C(\alpha)$ optimum test was used which assumed that the distribution of hail damage is either gamma disdistributed or has a linear regression on the predictor variables. The test involved two cases: one without predictor variables and and one with predictor variables. Formulas for these cases were given by Neyman and Scott (1967b) and were only used with the random-experimental design.

In the computation of sample sizes the "single-sided" alternative hypothesis was used throughout the paper.

4. Results

The optimal $C(\alpha)$ test produced smaller sample sizes than the log-normal test for the insurance data employed in this study. Thus, with the exception of the historical designs, the results are only presented for the optimal $C(\alpha)$ test.

To convert the number of experimental units to the number of years required for detection of a given decrease under the Hypothesis A, the number of units expected per year per area and the ability of the forecast scheme to predict hail must be considered. Based on prior experience, it is expected that 62% of the operational days (forecasted hail days) will in fact have hail. It will be assumed that this figure will be applicable to all four areas. From the historical insurance records, there was an average of 6.9 hail days per year for Morgan County, 13.6 for Logan County, 16.7 for Weld County, and 26.3 in the Tri-County area (an obvious reflection of the areal size of the four areas). Furthermore, it is assumed that the forecasting scheme will be such that all hail days will be forecasted although 38 percent of these days will, in fact, have no hail. Thus, it is assumed that the number of operational days required per year would be 11.2, 22.0, 26.9, and 42.4 days, respectively, for the four areas. In order to convert the number of hail days required for a given decrease to the number of years required for Hypothesis A, it is sufficient to divide the number of hail days by the expected (average) number of hail days per year. For Hypotheses A and B, it is sufficient to divide the number of hail days by the expected (average) number of operational days per year.

The number of years required to detect various decreases in the study area were estimated for Morgan and Logan Counties. To obtain the estimates, the trend between Morgan County and Logan County was extrapolated linearly into a 600 mi^2 area (size of study area).

a. Overall results

Overall results for various combinations of hypothesis testing and designs are shown in Table 1 where all values are based on 50-50 randomization. The random-historical design (sequential test) requires the shortest length of time for detection (2 years for a 70 percent chance of detecting a 40 percent decrease in the study area). Of the three tests involving the random-experimental design, that combining the test of damage and hail days is the most powerful. There is a 70 percent chance of detecting a 40 percent decrease in damage in 4 years when combined with a 40 percent decrease in the number of hail days. Since it is believed that the historical design should not be the principle verifying technique, the random-experimental design is deemed necessary.

If hail damage in the study area is decreased 40 percent and no allowance is made for a possible reduction in hail days (random-experimental reduction in damage), there is a 70 percent chance of detecting the effect in 7 years, and a 50 percent chance in 4 years. If the frequency of hail days in the study area is reduced 40 percent and no allowance is made for a possible corresponding reduction in damage, there is a 70 percent chance of detection in 10 years and a 50 percent chance in 6 years.

It is natural to consider the effect of using a lower significance level. If the level of significance is reduced, thereby increasing the chance of asserting that a seeding effect exists when in fact it doesn't (Type 1 error, α), there is a corresponding increase in the probability of detecting the seeding effect, Thus, if α is chosen to be .10, (1 chance in 10 of wrongly asserting the existence of an effect) there is a corresponding decrease in the number of years required. Table 2 is a comparison of the number of years required for 3 different levels of α for the random-experimental design. With a 1-tail α level of .025 (2-tail, α = .05) test, 6 years would be required in the study area for a 70 percent chance of detecting a 40 percent decrease in damage when combined with a 40 percent decrease in the number of hail days. With a 1-tail α level of .10 (2-tail, α = .20), only 3 years would be required to have a 70 percent chance of detecting the same decrease combination.

It should be noted that when the percentage reduction in hail damage is equal to the reduction in the frequency of hail days (in the combined test), an optimum condition for detecting a decrease exists. If the percentage differences are not the same, the sample sizes for the combined test will be larger than when they are equal. If the percentage differences are greatly different, the values presented for reduction in damage alone or reduction in frequency of hail days alone may be smaller than that required by the combined test. More detailed information concerning sample sizes when the percentage differences of damage and hail probability are different, can be gleaned from the report by Schickedanz and Changnon (1970b).

b. Conclusions

If a test is made on damage reduction using the random-experimental design and no allowance is made for the probability of hail on a particular day, there is a 70 percent chance of detecting a 40 percent decrease in hail damage in 7 years and a 50 percent chance in 4 years (percent loss data).

If a test is made for reduction in the probability of hail on a day (reduction in hail days) there is a 70 percent chance of detecting a 40 percent decrease in 10 years, and a 50 percent chance in 6 years.

There are possibilities of decreasing these required sample sizes. If one uses a non-sequential test with historical data and all assumptions are satisfied, there would be a 70 percent chance of detecting a 40 percent decrease in 4 years, and a 50 percent

Table 1. The number of years required to detect 20-, 40-, and 60-% decreases for various combinations of hypothesis testing and designs, using percent loss and probability of hail data (1-tail test, α = .05, randomization = 1/2).

		Random-Experimental Reduction in Damage			Random-Historical (Non-Sequential) Reduction in Damage			Random-Historical (Sequential) Reduction in Damage			Random-Experimental Reduction in Hail Days			Random-Experimental Reduction in Damage and Hail Days*		
Decrease	Power	Logan	Morgan	Study Area	Logan	Morgan	Study Area	Logan	Morgan	Study Area	Logan	Morgan	Study Area	Logan	Morgan	Study Area
20	.70	12	23	36	8	14	21	3	6	9	13	26	42	6	11	17
	.50	7	13	21	5	8	12	2	3	5	8	15	24	3	6	10
40	.70	2	4	7	2	3	4	1	1	2	3	6	10	1	3	4
	.50	1	2	4	1	2	2	<1	1	1	2	4	6	1	2	2
60	.70	1	1	2	<1	1	1	<1	<1	1	1	3	5	1	1	2
	.50	<1	1	1	<1	1	1	<1	<1	<1	1	2	3	<1	1	1

* Reduction in probability of hail are the same as those specified for reductions in damage

Table 2. A comparison of the number of years required for detection of varying decreases and varying significance levels for the random-experimental design using percent loss data (randomization = 1/2).

		Reduction in Damage			Reduction in Hail Days			Reduction in Damage and Hail Days*		
Decrease	Power	Logan	Morgan	Study Area	Logan	Morgan	Study Area	Logan	Morgan	Study Area
		1-tail α = 0.025, 2-tail α = 0.05								
20	.70	16	30	47	17	34	54	7	14	23
	.50	10	18	29	11	21	34	4	9	14
40	.70	3	6	9	4	8	14	2	4	6
	.50	2	3	6	3	5	8	1	2	3
60	.70	1	2	3	2	4	6	1	2	3
	.50	1	1	2	1	2	4	1	1	2
		1-tail α = 0.05, 2-tail α = 0.10								
20	.70	12	23	36	13	26	42	6	11	17
	.50	7	13	21	8	15	24	3	6	10
40	.70	2	4	7	3	6	10	1	3	4
	.50	1	2	4	2	4	6	1	2	2
60	.70	1	1	2	1	3	5	1	1	2
	.50	<1	1	1	1	2	3	<1	1	1
		1-tail α = 0.10, 2-tail α = 0.20								
20	.70	8	16	25	9	17	29	4	8	13
	.50	4	8	13	5	9	14	2	4	6
40	.70	2	3	5	2	4	7	1	2	3
	.50	1	1	2	1	2	4	<1	1	2
60	.70	<1	1	1	1	2	3	<1	1	1
	.50	<1	<1	1	1	1	2	<1	<1	1

* Reductions in probability of hail are the same as those specified for reductions in damage

chance of detection in 2 years. Using the sequential test, there would be a 70 percent chance of damage detection in 2 years. However, there are many limitations associated with utilizing the sequential procedure based on historical data alone.

If a "combined" test is made on reduction in damage and reduction in probability of hail treated together, there is a 70 percent chance of detecting a 40 percent decrease in 4 years (50 percent in 2 years), if the percentage reduction in the damage is the same as the percentage reduction in probability of hail.

Certain combinations of predictor variables can be obtained which will yield some predictive power. Under Hypothesis A, a substantial reduction of sample sizes was obtained, but a very small reduction was obtained for Hypothesis C. Indeed, the sample sizes for Hypothesis A using predictor variables were approximately the same as the sample sizes affected by using Hypothesis C without predictor variables.

The results stated above are all based on a 1-tail, α = .05 (2-tail, α = .10) test of significance. This indicates that there is a 5 percent chance of wrongly asserting the existence of an effect when it in fact does not exist (1-tail test). In order to have the same precision with a 2-tail, α .05 test, sample sizes would be 9 years for a 40 percent decrease in damage alone, and 6 years when a 40 percent decrease in damage and probability are combined in the same test (70 percent chance of detection).

On the other hand, if one is willing to risk a 10 percent chance (1-tail, α = .10) of wrongly asserting an effect when in fact it does not exist, then the number of years required would be 5 for damage alone, and 3 years for the combined test (70 percent chance of detection).

It was found that percent crop loss, dollar extent, and areal extent were the most efficient hail parameters to use to detect decreases in the region of NHRE.

All results are based on an assumed ability to forecast the hail days with 62 percent accuracy. It is further assumed that the forecasting capability will be such that all hail days will be included in the operational days with 38 percent of the operational days having no hail.

The above results are also based on an assumed randomization factor of 50-50. For the random-experimental design it was found that the randomization factor could be 60-40 and the sample size would be nearly the same. However, if the randomization factor were 20-80 there would be an appreciable increase in sample size.

It is important to note that the "study area" estimates represent an extrapolation of required sample sizes from values of larger areas, and thus these study area estimates may be subject to some error.

c. Recommendations

It would seem that the use of the historical data in conjunction with the random-experimental design is the best choice for the design and eventual evaluation of NHRE results. Based on the established relationship between crop damages and the hail pad data (Changnon, 1970), the following design-evaluation procedure is recommended.

The seeding design should be developed in the context of the random-experimental design with a randomization factor of either 50-50 or 60-40. The data from the non-seeded days could then be compared with the historical record as a test in addition to the usual test comparison between random samples of the seeded and non-seeded data during the experimental period. Also, the non-seeded data could be compared with the historical data to test for a trend and the representativeness of the historical record in relation to the experimental period. It is also possible that the historical non-seeded comparison could be used to remove any serious trend found in the data. Thus, even though the recommended experiment would not be conducted in a sequential manner, there is no reason why the sequential test could not be monitored along with the other tests. The authors believe this would enhance the chance of detecting a seeding effect, over the sole use of the random-experimental design, and at the same time utilize the statistical advantage of the random-experimental design. In this manner, allowances could also be made to include any useful predictor variables since they would provide detection of lesser decreases in the 5-year NHRE period.

The Denver upper air data were the only data used in the study of predictor variables. A study of predictor variables using synoptic data collected by NCAR in the immediate study area would be worthwhile. For example, a study should be made of the sounding data collected by NCAR in 1969 and 1970 to determine if regression equations can be established between these data and the hailfall within the study area.

There would be a much better chance of detecting seeding effects if a larger area were used and, if possible, it is recommended that a larger study area be used, particularly if the random-experimental design is used.

Since the study area has very little insurance coverage, the recommended evaluation procedure described above requires that a relationship between the hail parameters (as detected by surface hail sensors) and crop damage (percent loss) for the study area and environs is established _if_ the insurance data are used for the basic source of data. Therefore, to supplement the Illinois crop-hail relationships and to check for regional differences, surface hail sensing devices should be operated adjacent to crops in Northeast Colorado prior to the beginning of the experiment.

The insurance data were used to provide estimates of sampling sizes required because they were the only historical data available to make such estimates. The recommended design evaluation procedure allows for other parameters (echo coverage), echo heights and other physical parameters) which may be measured during the experiment to be used in the evaluation. However, prior estimates of sampling size cannot be provided for such parameters.

ACKNOWLEDGEMENTS

This research was supported by Subcontract No. NCAR 155-70 with University Corporation for Atmospheric Research under Prime Contract No. NSF-C460. The detailed crop-hail insurance data for Colorado were supplied at no cost by Crop-Hail Insurance Actuarial Association of Chicago. The assistance of this Association and the interest and advice of Mr. E. Ray Fosse, Manager of the Association, are greatly appreciated.

The work was done under the general supervision of Dr. William C. Ackermann, Chief of the Illinois State Water Survey. Although much of this research was supported by funds from NCAR, some support came from the State of Illinois.

Appreciation is expressed to Marion Busch who supervised the computations involved in this research effort.

REFERENCES

Changnon, S. A., 1969: Hail evaluation techniques, Urbana, Part I, Final report, NSF GA-482, 97 pp.

Changnon, S. A., and P. T. Schickedanz, 1969: "Utilization of hail-day data in designing hail suppression projects, Mo. Wea. Rev., 97-95-102.

Changnon, S. A., 1970: "Hail fall characteristics related to crop damages," to be published in J. of Appl. Meteor.

Huff, F. A., W. L. Shipp, and P. T. Schickedanz, 1969: Evaluation of precipitation modification experiments from precipitation rate measurements, Urbana Final Report Contract INT 14-06-D-6575, 122 pp.

Huff, F. A., and P. T. Schickedanz, 1970: Rainfall evaluation studies, Part II, Description of individual studies, Urbana, Final Report, NSF GA-1360, 224 pp.

Neyman, J., and E. L. Scott, 1967a: "Some outstanding problems relating to rain modification," Proceedings of the Fifth Berkeley Symposium on Mathematical Statistics and Probability, 293-350.

Neyman, J., and E. L. Scott, 1967b: "Note on techniques of evaluation of single rain stimulation experiments, "Proceedings of the Fifth Berkeley Symposium on Mathematical Statistics and Probability, 371-384.

Schickedanz, P.T., S. A. Changnon, and C.G. Lonnquist, 1969: A Statistical methodology for the planning and evaluation of hail suppression experiments in Illinois, Urbana, Part II, Final Report NSF GA-482, 140 pp.

Schickedanz, P. T. and S. A. Changnon, 1970: "The design and evaluation of hail suppression experiments, "Mo. Wea. Rev., 98:242-252.

Schickedanz, P. T. and S. A. Changnon, Jr., 1970: A study of crop-hail insurance records with respect to the National Hail Experiment, Urbana, Final Report NCAR 155-70, Prime Contract NSF-C460, 98 pp.

APPLICATION OF NUMERICAL MODELS TO CUMULUS CLOUD MODIFICATION[1]

A. I. Weinstein

Meteorology Research, Inc.
Altadena, California 91001

ABSTRACT

The strong and weak points of the three classes of existing cumulus cloud models are reviewed in order to determine the most appropriate application of each class. One-dimensional steady-state models are best used for routine analysis of thermodynamic instability potential. One-dimensional time-dependent models are most applicable to warm cloud and/or hailstorm simulation, where the interaction between cumulus dynamics and microphysics is most important. Two-dimensional models hold great potential for accurately simulating the development of meso-scale storm systems and for parameterizing the important aspects of two dimensions in one-dimensional models. The one-dimensional time-dependent and two-dimensional models need verification against observed data to build confidence in their simulations.

I INTRODUCTION

The advances made in weather modification in recent years are due in large part to the widespread use of numerical model simulation. As research tools, models focus attention on the processes that are most important in the phenomenon being modified and indicate the observations that have to be made to understand these processes. As operational tools, models can be instrumental in developing routine decision-making techniques.

The level of sophistication required in a model is largely a function of its intended use. The range of sophistication of existing cumulus models extends from one-dimensional steady-state models for use largely as thermodynamic instability indicators, to two-dimensional time-dependent models for use principally as research tools. In this paper existing cumulus cloud models will be compared in order to elaborate on the theme of sophistication vs. application.

1 Support provided by U.S. Dept. of the Interior, Bureau of Reclamation, Division of Atmospheric Water Resources Management, under Contract No. 14-06-D-6581.

II ONE-DIMENSIONAL, STEADY-STATE, LAGRANGIAN (SSL) MODELS

One-dimensional, steady-state, Lagrangian models, as described by Weinstein and Davis (1968), Simpson and Wiggert (1969), and others, carry out the same calculations that are done graphically on thermodynamic diagrams by the classical parcel method with three major improvements:

1) incorporation of the effects of the horizontal dimension of the clouds through the use of the entrainment concept originally proposed by Stommel (1947);
2) calculation of the release of latent heat of fusion; and
3) calculation of the development of precipitation.

Recently, each of these improvements has come under critical fire. The entrainment calculation using the inverse radius relationship is based upon laboratory experimentation (Woodward, 1959) and the theory of dry jets (Levine, 1959). There have been few direct verifications in actual clouds. Most of the cloud verification has been indirect using computed and observed cloud-top heights. In fact, there have been some in-cloud observations that tend to discredit the inverse radius relationship (i.e., Warner, 1970, and Sloss, 1967). The accuracy (Simpson, 1971) and/or representativeness (Weinstein, 1971) of the direct evidence, however, has been questioned.

The realism of the inverse radius relationship is presently unresolved. The important point to be made here is that for certain applications the realism is sufficient for the desired application, that application being to predict the thermodynamic and dynamic reactions of isolated cumuli. Just as the undilute parcel method produced better cloud-top height predictions than its predecessor, the inverse radius relationship produces better predictions than the undilute parcel method.

The other calculation features of the SSL models, the freezing calculations and the precipitation formation simulations, have been improved by Takeuchi (1969), Cotton (1970), and Howell and Lopez (1968). In the first two cases, the improvements have been made in simulating the freezing more realistically, but at least in the case of Takeuchi, there was no improvement in the accuracy of the primary model calculation, the cloud-top height.

The precipitation distribution improvement made by Howell and Lopez resulted in a significant improvement in the model caclulation accuracy, at least the liquid water content is high and the environmental sounding is close to the moist adiabatic lapse rate.

A major positive feature of the SSL models is their rapid calculation speed and/or small computer core storage requirements. The calculations that they do best are the thermodynamic and dynamic simulations. The results of horizontal mixing and freezing are fairly well simulated although, in some cases, the actual details of these processes are not well handled. The microphysical processes of precipitation development and redistribution, in general, are not well simulated. Since the precipitation redistribution usually has only a secondary effect on the cloud dynamics (a notable exception here is when the environmental lapse rate is close to the moist adiabatic value through a deep layer as described by Weinstein and MacCready, 1969), one-dimensional steady-state models simulate the cloud dynamics rather well. It is not surprising to find that their cloud-top height predictions are generally accurate as found by Weinstein and MacCready (1969), Sac (1969), Simpson and Wiggert (1969), Marwitz et al. (1970), and others. Since the details of the precipitation distribution do not always affect the total rain coming out of the cloud, the one-dimensional steady-state models also have some power in predicting total rainfall, as shown by Weinstein and MacCready (1969). Recent improvements to the PSU model by Cotton (1970) offer further hope for even better precipitation predictions from SSL models.

The primary application of SSL models is in the anlysis of the convective potential of different atmospheric thermal and moisture structures.

In research the first such application can be found in a report soon to be published by the Panel on Weather Modification of the National Academy of Sciences.[2] It will be stated in that report that it is no longer a question of <u>can</u> precipitation be augmented by seeding, but rather <u>when</u> can it be accomplished. The Panel report will recommend studies to determine when this augmentation can be accomplished in different geographical and/or climatological areas. The speed and accuracy of one-dimensional models make them ideally suited to carry out such seedability climatology studies with respect to isolated supercooled cumuli.

The originally intended use, and probably still the most appropriate utilization of the SSL models, is in conjunction with field programs. It is now clear that the precipitation from isolated cumuli can be materially augmented under clearly identifiable synoptic conditions. Essentially, the favorable conditions are when the heat released by artifically induced freezing stimulates the clouds' dynamics so as to significantly increase the clouds' dimensions. There are some synoptic conditions under which a wide range of cloud sizes are susceptible to such stimulation. Under other conditions, only a narrow range of cloud sizes will respond. The one-dimensional steady-state models have shown skill at identifying these synoptic conditions. They should be used as an integral part of operational decision-making in the field. One example of this application was the program run for the Bureau of Land Management in Alaska in 1970 (see Staff of MRI, 1970).

Another worthwhile application of SSL models in conjunction with field programs is in the evaluation of the seeding. Often on operational

[2]As reported by L. J. Battan at the 51st Annual Meeting of the AMS, 11-14 January 1971, San Francisco, Calif.

programs economic considerations preclude the use of elavorate instrumentation and detailed data reduction. Under these conditions, sufficient data need only be taken to verify and/or calibrate the steady-state model. After this, a fair degree of confidence can be placed in the model calculations as simulating control conditions, and thus the model calculations can be used for evaluation.

Other applications of the SSL models that have been proposed include forecasting of cumulus intensity and in hail research. The former has never actually been demonstrated but, in principle, the model's sounding diagnostic capability makes it appropriate for forecasting whenever the undilute parcel method is presently used. The calculation speed of the model makes it economically feasible to use it operationally in forecasting of cumulus intensity.

In hail research, it has been applied by Marwitz et al, (1970) to infer characteristics of updraft vaults in hailstorms. One could use the model calculations of updraft and liquid water content as the framework within which one could grow hailstones from various size kernels and thus evaluate potential hail suppression techniques. Others have used the adiabatic profile for this purpose, but the model calculation profiles should be more realistic as they can account for the size of the cloud.

In summary, one-dimensional steady-state models have fulfilled their primary research function of identifying the broad conditions under which ice phase seeding should produce dynamic response in isolated cumuli. The primary remaining function of these models is in using their predictive ability on a routine basis.

III ONE-DIMENSIONAL, TIME-DEPENDENT, EULERIAN (TDE) MODELS

A prime drawback of steady-state models is their inability to simulate the interaction between cumulus dynamics and microphysics. Time-dependent Eulerian formulations of the same equations as used in the one-dimensional steady-state models overcome this drawback. The TDE models consume more computer time and space than do their SSL counterparts, but their speed is still approximately one order of magnitude faster than real time.

The prime advantage of the TDE models over the SSL ones, their simulation of the vertical redistribution of liquid water, is most important in warm cloud and/or hailstorm simulation, for it is in these two phenomena that water loading might play the greatest role.

The formulation described by Srivastava (1967) and Weinstein (1970) can be used in the initial stages of studies of either type of cloud. The microphysical processes in this simulation are parameterized as described by Kessler (1969), and the same constants are used for liquid and solid hydrometeors. The initial calculations with this model indicated that the warm cloud microphysics in their parameterized form interact with the dynamics in the expected manner to produce the periodic oscillations and rain gushes often observed in cumuli. The parameterization of the microphysics, however, makes this model of only limited value for detailed warm cloud modification experimentation.

The TDE model as described by Weinstein (1970) is presently being modified to replace the parameterized microphysical calculations with more realistic simulations as suggested by Kovitz and Olund (1969) and applied by Nelson (1970). With this improved TDE model, it will be possible to carry out the sensitivity analyses needed to access the modification potential of warm clouds. Whereas the conditions under which the dynamics of supercooled cumuli can be profitably modified are well known from many SSL model calculation and field programs, the conditions under which warm cloud techniques can profitably be employed are only qualitatively understood.

A major application of TDE models is in determining the conditions under which clouds can be modified by warm cloud seeding techniques. A second application is in pointing out the observations that need be taken to better understand the process active in warm clouds. The parallel application of TDE models to warm clouds, as SSL models are applied to supercooled cumuli, is apparent.

The major difference between the application of the two classes of models is that the SSL models have been widely verified as to their ability to accurately simulate the cloud dynamics reactions to seeding, whereas the TDE models have not shown similar veritifcation with respect to the cloud microphysics. It is mandatory that before the TDE models are used routinely they be applied in conjunction with field observation to improve them and build confidence in their simulations. Once this verification phase has been completed, the TDE models should be used for the same type of studies of warm clouds as the SSL models are used for supercooled cumuli. The TDE models require more computer time than do their SSL counterparts, so most of the studies will have to be confined to fewer size clouds. This is not expected to be a major drawback since the process best simulated by the TDE models, the microphysical process, is probably not seriously affected by the size of the cloud.

The application of the TDE models to hail research has been done by Wisner (1970) and Danielson (private communication). The primary improvement of these formulations over the model of Weinstein (1970) is the more realistic parameterization of the ice hydrometeor development. As is the case with the TDE models as applied to warm clouds, the routine hail application of the TDE models must await the model improvements that come with full-scale field program verification.

IV. TWO-DIMENSIONAL (2-D) MODELS

The second dimension of two-dimensional models allows for the elimination of the prime objection to one-dimensional models, the treatment of horizontal mixing. In 2-D models, the mixing is simulated by Fickian diffusion. There are some objections to this approach on physical grounds and some numerical problems, but the approach is probably more realistic than the entrainment concept using the inverse radius relationship.

The two most noteworthy recent efforts in this area are described by Orville and Sloan (1970) and Murray (1970). Due to their time interation and the second dimension, the 2-D models can be orders of magnitude slower than real time and usually require computers of the size of a CDC 6600. Some impressive results have been presented by Orville and Sloan showing that their model is capable of predicting motion on a scale of less than 1 km. Many of the characteristics of a hailstorm, not realistically predicted in one-dimensional models, are simulated in the model results of Orville and Sloan (i.e., indications of an echo-free vault).

Their requirements of large computers and major amounts of computer time severely limit the number of cases that can be run with 2-D models. It is the author's opinion that the major contribution of these models will be in the indication of ways to parameterize the major effects of two-dimensions in one-dimensional models. Although the 2-D models require a major investment in computer costs, these costs are far less than the costs of observing the parameters in the detail that the models predict. Thus, once one has sufficient observational verification of the model's predictions (such verification presently only exists in a very few cases and is still very qualitative), it will be far more economical to parameterize some processes from the model calculations than from actual observations.

Some of the processes, in addition to horizontal mixing, that cannot be simulated on one-dimensional models but can be modeled in two-dimensions are those that go into the production of mesoscale storm systems. Such systems can develop through the interaction and subsequent merger of previously isolated clouds, or through the steady intensification and growth of a single cloud. In either case, simple one-dimensional models cannot simulate the development nor the organized inflow into a mesoscale storm. Such simulation must remain in the domain of two-dimensional models.

Because the 2-D models require such a large commitment of computer time for each case, one must be willing to commit a like amount of time (of the order of months) to the thorough analysis of each case.

V. SUMMARY

A brief summary has been presented of the three classes of cumulus cloud models presently being used.

The one-dimensional steady-state models utilize the inverse radius relationship for the entrainment rate and parameterized cloud microphysics, along with detailed thermodynamics and dynamics, to produce cloud-top height and precipitation predictions on small computers in seconds. The principal application for these models is as a sounding analyzer for dynamic response of isolated cumuli to the release of latent heat. Some suggested uses include ice phase seedability potential climatologies, real-time operational tools in the field, use as seeding evaluation controls, and in forecasting of convection intensity.

The one-dimensional time-dependent models maintain the inverse radius relationship for the entrainment rate, but utilize more realistic precipitation development simulations and allow for a more realistic simulation of the interaction between cumulus dynamics, thermodynamics, and microphysics. These models require larger computers and more computer time than do their steady-state counterparts, but can still be programmed on most available computers and run at least as fast as real time. The primary application of these models is in studying clouds in which the interaction of the cumulus dynamics and microphysics is important. Warm clouds and hailstorms are examples of such clouds. Some prospective uses for one-dimensional time-dependent models with respect to warm clouds include identification of opportunities for profitable warm cloud seeding, determination of the scale of magnitude of the precipitation augmentation that could be derived from warm cloud seeding and, ultimately, for the evaluation of operational warm cloud seeding programs. Before most of these applications can be fully realized, considerable field observations must be obtained to verify the time-dependent model predictions.

The two-dimensional time-dependent models are potentially the most realisitic of all existing cumulus cloud models. These models require a large amount of time on large computers and, consequently, enough cases have not yet been run to verify the model calculations and realize the potential. The results to date are qualitatively verified in that they appear realistic. A series of model calculations using real sounding and making detailed comparisons with a set of specially made observations are needed to lend confidence to the two-dimensional model simulations. Two applications of these models, once they have been verified, are (1) to study possible parameterizations that can be made to streamline the models for operational use, and (2) to experiment on the formation and persistence of mesoscale storm systems through judicious seeding.

Numerical model simulation has progressed to the point where the models can no longer be considered ends in themselves. It is the responsibility of the authors of models, who best know their models' approximations, to point out the best applications of their products as well as presenting some model calculation results.

REFERENCES

Cotton, W. R., 1970: A numerical simulation of precipitation development in supercooled cumuli. PhD. Disertation, 5 September 1970, Dept. of Meteor., Penn State University, University Park, Pa., 179 pp.

Howell, W. F., and M. Lopez, 1968: Project Rainstart. Interim Report to NSF, Cont. No. NSF-C453, E. Bollay Associates of EG&G, 66 pp.

Kessler, E., 1969: On the distribution and continuity of water substance in atmospheric circulations. Meteor. Monogr., 10, 32, 84 pp.

Kovitz, A., and B. Olund, 1969: The effect of coalescence and condensation on rain formation in a cloud of finite vertical extent. J. Atmos. Sci., 26, 1060-1065.

Levine, J., 1959: Spherical vortex theory of bubble-like motion in cumulus clouds. J. Meteor., 16, 653-662.

Marwitz, J. D., J. R. Middleton, A. H. Auer, Jr., and D. L. Veal, 1970; The dynamics of updraft vaults in hailstorms as inferred from the entraining jet model. J. Atmos. Sci., 27, 1099-1102.

Meteorology Research, Inc., Staff of, 1970: Cloud seeding operations on wildfires, Alaska - 1970 (Project WOOO). Final Report to Bureau of Land Management, U.S. Dept. of the Interior, Washington, D.C., Cont. No. 08550-CTD-5.

Murray, F. W., 1970: Numerical models of a tropical cumulus cloud with bilateral and axial symmetry. Mon. Wea. Rev., 98, 14-28.

Nelson, L. D. 1970: A numerical simulation of the effects of water-spray seeding on the warm rain process. Preprints of Papers Presented at Second Natl. Conf. on Weather Modification, Santa Barbara, Calif. 6-9 April 1970, 18-21.

Orville, H. D., and L. J. Sloan, 1970: A numerical simulation of the life history of a rainstorm. J. Atmos. Sci., 27, 1148-1159.

Sax, R. I., 1969: The importance of natural glaciation on the modification of tropical martime cumuli by silver iodide seeding. J. Appl. Meteor., 8, 92-104.

Simpson, J., and V. Wiggert, 1969: Models of precipitating cumulus towers. Mon. Wea. Rev., 97, 471-489.

________, 1971: On cumulus entrainment and one-dimensional models. Submitted to J. Atmos. Sci.

Sloss, P. W., 1967: An empirical examination of cumulus entrainment. J. Appl. Meteor., 6, 878-881.

Srivastava, R. C., 1967: A study of the effects of precipitation on cumulus dynamics. J. Atmos. Sci., 24, 36-45.

Stommel, H., 1947: Entrainment of air into a cumulus cloud. J. Meteor. 4, 91-94.

Takeuchi, D. M., 1969: Ice phase cumulus cloud model. Research Report II in Annual Report. FY 1969 to Bureau of Reclamation, Meteorology Research, Inc., Altadena, Calif., Cont. 14-06-D-6581.

Warner, J., 1970: On steady-state one-dimensional models of cumulus convection. J. Atmos. Sci., 27, 1035-1040.

Weinstein, A. I., and L. G. Davis, 1968: A parameterized numerical model of cumulus convection. Penn State Univ., Dept. of Meteor., Report No. 11 to National Science Foundation, NSF GA-777, 43 pp.

__________, and P. B. MacCready, Jr., 1969: An isolated cumulus cloud modification project. J. Appl. Meteor., 8, 936-947.

__________, 1970: A numerical model of cumulus dynamics and microphysics. J. Atmos. Sci., 27, 246-255.

__________, 1971: On steady-state one-dimensional models of cumulus convection. Submitted to J. Atmos. Sci.

Wisner, C., 1970: A numerical model of a cumulus cloud. M. S. Dissertation, Dept. of Meteor., South Dakota School of Mines and Technology, Rapid City, S. D., 92 pp.

Woodward, E. B., 1959: The motion in and around isolated thermals. Quart. J. Roy. Meteor. Soc., 85, 144-151.

A UNIFIED THEORY FOR AEROSOL PHENOMENOLOGY

Ira Kohlberg

Analytical Systems Corporation
Burlington, Massachusettes

ABSTRACT

An approach to aerosol behavior is presented which is based upon a transport equation that simultaneously takes into account all aspects of aerosol phenomenology. Basically, this viewpoint permits one to regard the specialized branches of aerosol behavior from a common origin and thus assess the limitations implicit in the present theories. Mathematical techniques are discussed and evaluated which can be used to solve the aerosol transport equation, and thus predict aerosol behavior under very complex situations which heretofore has not been possible.

1. INTRODUCTION

The study of aerosols has become increasingly important in recent years, particularly in the areas of propagation of electromagnetic waves through the atmosphere, weather modification, cloud physics, nuclear fallout, air pollution and defoliation. In each application the fundamental problem boils down to the determination of the space, time and size distribution of the aerosols under various physical conditions. For example, one might be interested in determining the cloud formation time following seeding, or the optical transmission in a vertical convective smoggy environment, or perhaps the close-by aerosol distribution following a chemical release.

Several texts have appeared since 1962 which in their own domain cover a certain, and at times limited, portion of aerosol phenomenology. Pasquill,[1] for example, is concerned with describing the transport of individual aerosol particles following their release into the atmosphere (including the effect of turbulence) but is not concerned with coagulation during this motion. On the other hand, Green and Lane[2] principally discuss the growth, stability, optical properties, and measurement of aerosols, with only limited involvement in convective problems. A rather comprehensive treatment on aerosols can, however, be found in Fuch's[3] book on the mechanics of aerosols, where one finds separate discussions on "seemingly" unrelated areas of aerosol phenomenology. While it is sometimes possible to isolate the various aspects of aerosol phenomenology, it is very often not physically meaningful to do so. For example, Smoluchowski's theory[4-7] of coagulation of Brownian particles including the effects of foreign vapors[8], Van der Walls forces[9, 10] and electrical charges[11] has been studied extensively in a spatially homogeneous region but to our knowledge has not been considered in the region of concentration gradients. This letter case is more likely to be encountered in the real-world environment.

The fundamental theoretical-analytical problem related to predicting aerosol behavior is the difficulty of simultaneously considering growth (due to coagulation, nucleation, etc.) in the presence of external forces such as gravity and electric field, spatial concentration gradients, collisions, and statistically defined random forces attributable to turbulence. In this investigation we are proposing to study the behavior of aerosols via a new unified theoretical approach which by its mathematical nature is constructed to simultaneously handle the aforementioned aspects. Accordingly, the various isolated aspects of aerosol behavior appear as special cases. The advantages of this approach are (1) the conceptual cohesiveness it brings to the general treatment of aerosols and (2) the ability to investige interdisciplinary aspects of aerosol phenomenology previously reserved for separate consideration.

The basis idea of our method is the development of an aerosol transport equation, not unlike the Boltzmann transport equation[12, 13] of kinetic theory, or the more general Master equation[14]. Specifically we construct a mathematical function $F(\vec{r}, \vec{v}, M, t)$ such that $F\, d\vec{r}\, d\vec{v}\, dM$ ($d\vec{r} = dxdydz$; $dv = dv_x dv_y dv_z$) is the number of aerosol particles at the point $\vec{r}$ in the volume element $d\vec{r}$, with velocity $\vec{v}$ in the range $d\vec{v}$ and with mass M in the range dM at time t, and then proceed to determine its temporal behavior. Since $F(\vec{r}, \vec{v}, M, t)$, and its obvious generalization to include chemical designation, contains all the information about the aerosols, its determination suffices to completely define all aspects of the aerosol distribution such as the average mass, average velocity, etc. We shall now discuss the equation which determines $F(\vec{r}, \vec{v}, M, t)$.

2. A TRANSPORT EQUATION FOR AEROSOLS

Consider a group of aerosol particles of mass M, and velocity $\vec{v}$, located at position $\vec{r}$, moving through a non-turbulent environment in which field-derived forces (such as gravity and electric fields) exist, and where interaction with the medium can occur. Under these conditions the temporal behavior of $F(\vec{r}, \vec{v}, M, t)$ will be given by:

$$\frac{\partial F}{\partial t} = -\nabla_r \cdot (\vec{v}F) - \nabla_v \cdot (\frac{\vec{X}}{M}F) + (\frac{\partial F}{\partial t})_e + (\frac{\partial F}{\partial t})_m \qquad (2.1)$$

where $\nabla_r = \vec{i}\,(\partial/\partial x) + \vec{j}(\partial/\partial y) + \vec{k}(\partial/\partial_Z)$ is the gradient in coordinate space, $\nabla_v = \vec{i}(\partial/\partial v_x) + \vec{j}(\partial/\partial v_y) + \vec{k}(\partial/\partial v_z)$ is the gradient in velocity space, $\vec{X}$ is applied force, $(\partial F/\partial t)$ is the change in F attributable to the environment, and lastly $(\partial F/\partial t)_m$ is the change in F due to mass changing effects such as condensation, evaporation and coagulation.

Since all the terms, except $(\partial F/\partial t)_m$, are precisely the same as those in the Boltzmann equation, our proposed method for treating aerosols is recognized to be a generalization of the Boltzmann equation to include mass changing.

The first term on the right hand side of Eq. (2.1) is independent of M, the second depends on M through the ratio $\vec{X}/M$, while the last two can assume widely different functional dependences on the mass. When aerosol particles of molecular dimension interact through binary encounters with an ambient environment consisting of molecules of comparable mass the collision term will be the Boltzmann integral. However, when the ratio of the aerosol particle mass, M, to the gas mass, m, becomes very large the Boltzmann collision integral can be approximated by the Fokker-Planck expression[15-18]. That is, in this limit we have:

$$(\frac{\partial F}{\partial t})_e \simeq \beta \nabla_v . (\vec{v}F) + \frac{\beta kT}{M} \nabla_v^2 F \qquad (2.2)$$

where, as shown in reference 18, β, the so-called coefficient of dynamical friction equals $(M/M)\tau^{-1}$ where τ is the average time between collisions. (It is anticipated that aerosol particles of this size will be infrequently considered.) Interestingly enough, when the size of the aerosol particle increases farther, becoming large compared to the mean free path (but not large enough so that any effects that arise from the inertia of the displaced gas have to be considered) so that Stokes' law applies, the Fokker-Planck expression is still valid except that β is given by

$$\beta = (3\pi\eta d/M) \qquad (2.3)$$

where η is the viscosity of the gas, and d the aerosol diameter.[6] The non-random frictional force, $\vec{K}_e$, on the particle is given by the well-known expression

$$\vec{K}_e = -M\beta\vec{v} = -3\pi\eta d\vec{v} \qquad (2.4)$$

In this Brownian motion regime the statistical (or random) aspect of the force manifests itself through the term $(\beta kT/M)\nabla_v^2 F$ which assures us that the aerosol particle will eventually be brought into thermal equilibrium with the medium.

As the aerosol particle becomes larger the complexion of its interaction with the environment changes: first there is a sharp decrease in the purely random force on the particle, and secondly the non-random or average force will be given by an expression of the type

$$\vec{K}_e = -M\phi_f(|\vec{V}|)\vec{V} \tag{2.5}$$

where $\phi_f(|\vec{V}|)$ is a function of the fluid properties, and the particle's shape and mass. In this regime K_e is usually found via hydrodynamics; for example, by determining the drag coefficient. The corresponding interaction term becomes simply[6]

$$\left(\frac{\partial F}{\partial t}\right)_e = \nabla_v \cdot \left[\phi_f\,(|\vec{V}|)\vec{V}\right] \tag{2.6}$$

The last term in Eq (2.1) actually consists of two terms, the first one being due to condensation and evaporation

$$\left(\frac{\partial F}{\partial t}\right)_{m_1} = -\frac{\partial}{\partial m}\,(\dot{m}F) \tag{2.7}$$

where $\dot{m}$ is the evaporation rate per particle. For spherical particles at temperature T being bombarded by molecules at temperature T′, the evaporation rate will be given by:[19]

$$\dot{m} = A(M)\; m_o n_e\sqrt{\frac{kT'}{2\pi m_o}} - A(M)M\alpha(T), \tag{2.8}$$

where $A(M) = (6\sqrt{\pi}M/\rho)^{2/3}$ is the area of the aerosol, m_o, the molecular mass, n_e the ambient density, and $\alpha(T)$ the evaporation rate per unit area, which is generally of the form

$$\alpha(T) = C\,\exp(-V_o/kT) \tag{2.9}$$

where C is a constant and U_o is the heat of vaporation.

The coagulation term, denoted by $(\partial F/\partial t)_{m2}$ can be constructed in the following way: When two particles of masses M_1 and M_2 and velocities $\vec{V}_1$ and $\vec{V}_2$ respectively, collide, a new particle is formed with mass $M = M_1 + M_2$ and with a momentum $\vec{P} = M\vec{V} = M_1\vec{V}_1 + M_2\vec{V}_2$. Equivalently, the velocity of the newly-formed particle will be $\vec{V} = M^{-1}\,(M_1\vec{V}_1 + M_2\vec{V}_2)$. If $\sigma(M_1\vec{V}_1\,|M_2, \vec{V}_2)$ denotes the microscopic cross section for the reaction then the number of particles of mass M and velocity $\vec{V}$ formed per unit volume per second, denoted by $(\partial F/\partial t)^+_{m_2}$ is given by:

$$\left(\frac{\partial F}{\partial t}\right)^{+}_{m2} = \frac{1}{2} \int_{\vec{V}_1}\int_{\vec{V}_2}\int_{M_1}\int_{M_2} K(M_1,\vec{V}_1 \,|M_2, \vec{V}_2)\, F(\vec{V}_1, M_1)\, F(\vec{V}_2, M_2)\,\delta\left[M-(M_1+M_2)\right] \tag{2.10}$$

$$\times\delta\left[\vec{V} - (M_1/M)\vec{V} - (M_2/M)\vec{V}_2\right] dM_1\, dM_2\, d\vec{v}_1 d\vec{v}_2$$

where

$$K(M_1, \vec{V}_1 \,|M_2, \vec{V}_2) = E\,\sigma(M_1\, \vec{V}_1 \,|M_2, \vec{V}_2)\, |\vec{V}_1 - \vec{V}_2| \tag{2.11}$$

is the reaction rate and equals the product of sticking efficiency, E, times the cross section σ $(M_1, \vec{V}_1 \,|M_2, \vec{V}_2)$ multiplied by the relative velocity of the two particles. The "(1/2)" appearing in front of the integral sign is due to the fact that there is a reduction of one particle when two particles coagulate. The total number of new particles formed per second is given by

$$\frac{1}{2} \int_{\vec{V}_1}\int_{\vec{V}_2}\int_{M_1}\int_{M_2} K(M_1, \vec{V}_1 \,| M_2, \vec{V}_2)\, F(\vec{V}_1, M_1)\, F(\vec{V}_2, M_2)\, dM_1\, dM_2\, d\vec{V}_1\, d\vec{V}_2 \tag{2.12}$$

and consequently, the Dirac delta functions,

$$\delta\left[M - (M_1+M_2)\right]\delta\left[\vec{V}-(M_1/M)\vec{V}_1 -(M_2/M)\vec{V}_2\right]$$

select only those which end up with velocity $\vec{V}$ in the range $d\vec{v}$ and with mass M in the range dM.

Similarly, the number of particles lost per second per unit volume is given by:

$$\left(\frac{\partial F}{\partial t}\right)^{-}_{m2} = F(\vec{V}, M)\int_{\vec{V}_1}\int_{M_1} K(M, \vec{V}|M, \vec{V}_1)\, F(\vec{V}_1, M_1)\, dM_1\, d\vec{v}_1 \tag{2.13}$$

The total change in F due to coagulation is therefore:

$$\left(\frac{\partial F}{\partial t}\right)_{m2} = \left(\frac{\partial F}{\partial t}\right)^{+}_{m2} - \left(\frac{\partial F}{\partial t}\right)^{-}_{m2} \tag{2.14}$$

while the total change due to mass changing is

$$\left(\frac{\partial F}{\partial t}\right)_{m} = \left(\frac{\partial F}{\partial t}\right)_{m1} + \left(\frac{\partial F}{\partial t}\right)_{m2} \tag{2.15}$$

3. PROPOSED METHODS OF SOLUTION

Because of the intrinsic relationship between the aerosol transport equation and the Boltzmann equation (the former being a generalization of the latter to include mass changing) it is not surprising that the proposed methods of solution will be very similar.

Mathematically speaking, the aerosol transport equation is non-linear because the coagulation terms involve the distribution function to the "squared" power (cf., Eqs (2.10) and (2.13). Consequently, like most non-linear differential and integral equations, the road to solution is not always clear, although our approaches should yield meaningful results. These methods are essentially generalizations of the perturbation-theoretical techniques which have been used so successfully in transport theory to now include mass-changing effects.

As a matter of illustration, let us first consider the case in which the density of aerosol particles is low enough so that the coagulation term can be considered "small"; the question of "how small?" is relevant only to specific problems and will be investigated during the proposed work period. Now further assume that evaporation-condensation effects affect only the size of the aerosol particles but not the total number (viz., evaporation never proceeds to the point of completely destroying the particle). Under these conditions the coagulation term, however small, is the only one which accounts for the decrease in the number of particles and hence can never be neglected.

The mathematical structure of the aerosol equation under the aforementioned conditions can be described by the equation

$$\frac{\partial F}{\partial t} = B_o F + B_1(F,F) \tag{3.1}$$

where B_0 is the linear operator

$$B_o F = -\nabla_v \cdot \left(\frac{\vec{X}}{M} F\right) + \left(\frac{\partial F}{\partial t}\right)_e + \left(\frac{\partial F}{\partial t}\right)_{m1} \tag{3.2}$$

and B_1 is the non-linear coagulation operator

$$B_1(F,F) = \left(\frac{\partial F}{\partial t}\right)_{m2} \tag{3.3}$$

(In this illustrative example we are not considering spatial variations, although it will certainly be investigated during the course of the proposed investigation.) When coagulation is neglected altogether it is easy to show that the total number of aerosol particles per unit volume, given by

$$N(t) = \int \int F(\vec{v}, M, t)\, dM d\vec{v} \tag{3.4}$$

remains constant in time. This is readily deduced by integrating Eq (3.1), i.e.,

$$\frac{\partial}{\partial t} \int \int F(\vec{v},M,t)dMd\vec{v} = \int \int B_o F(\vec{v},M,t)dMd\vec{v} \tag{3.5}$$

and subsequently noting that the term-by-term integration of the right hand side equals zero. This fact leads us to interpret B_o as an operator which affects only the velocity-mass shape of the aerosol distribution function, in constract to the operator B_1 which affects not only the shape but the overall number of particles.

The distinction between B_o and B_1, coupled with the assumption that B_1 is "small" permits a perturbation-type solution to (3.1) according to the following scheme. Let us write

$$F(\vec{v},M,t) \equiv N(t)\, \phi(\vec{v},M,t) \tag{3.6}$$

where N (t) is the density given by Eq. (3.4) and $\phi(\vec{v},M,t)$ is a normalized spectrum.

$$\int \int \phi(\vec{v},M,t)\, dMd\vec{v} = 1 \tag{3.7}$$

Substituting Eq (3.6) into Eq (3.1) and subsequently integrating over $dMd\vec{v}$ gives

$$\frac{\partial N}{\partial t} = - \tfrac{1}{2} N^2 g(t) \tag{3.8}$$

where

$$g(t) = \int_{\vec{v}_1} \int_{\vec{v}_2} \int_{m_1} \int_{m_2} K(M_1, \vec{v}_1|M_2,\vec{v}_2)\, \phi(\vec{v}_1,M_1,t)\, \phi(\vec{v}_2,M_2 t)dM_1 dM_2 d\vec{v}_1 d\vec{v}_2 \tag{3.9}$$

Eq (3.8) gives the rate of decrease in density N (t) as a function of g (t), which is itself dependent upon the shape function $\phi(\vec{v}, M, t)$ as can be seen by examining the integral in Eq (3.9). Although Eq (3.8) is exact, the solution to the problem is not complete since $\phi(\vec{v}, M, t)$ is not known. At this point the physical assumption that the non-linear operator is small is used. Once again substituting Eq (3.6) into Eq (3.1), leads to the following equation for $\phi(\vec{v}, M, t)$.

$$\frac{\partial \phi}{\partial t} = B_o \phi + N\{ \tfrac{1}{2} \int_{\vec{v}_1} \int_{\vec{v}_2} \int_{m_1} \int_{m_2} \delta\left[M-(M_1+M_2)\right] \delta\left[\vec{v}-(M_1\vec{v}_1+M_2\vec{v}_2)/M\right] K(M_1,\vec{v}_1|M_2,\vec{v}_2) \times \phi(\vec{v}_1,M_1,t)\, \phi(\vec{v}_2,M_2,t)dM_1 dM_2 d\vec{v}_1 d\vec{v}_2 \tag{3.10}$$

$$+\tfrac{1}{2}\phi(\vec{v},M,t)g(t) - \phi(\vec{v},M,t) \int_{\vec{v}_1} \int_{M_1} K(M,\vec{v}|M_1,\vec{v}_1)\, \phi(\vec{v}_1,M_1,t)dM_1 d\vec{v}_1 \}$$

As observed, $\phi(\vec{v},M,t)$ is determined in part by the linear operator B_0 and also by a term which is proportional to the aerosol density N, multiplied by functions which depend non-linearly on ϕ. If the density N is "low enough" the N {} term in Eq. (3.10) can be neglected (at least in the first approximation) with ϕ $(\vec{v},M,t)$ determined by the equation

$$\frac{\partial \phi^{(o)}}{\partial t} = B_o \phi^{(o)} \qquad (3.11)$$

The corresponding decay of aerosol density is then given by

$$\frac{\partial N^{(o)}}{\partial t} = -\tfrac{1}{2}(N^{(o)})^2 g^{(o)}(t) \qquad (3.12)$$

where

$$g^{(o)} = \int_{\vec{v}_1}\int_{\vec{v}_2}\int_{m_1}\int_{m_2} K(M_1,\vec{v}_1|M_2,\vec{v}_2)\,\phi^{(o)}(\vec{v}_1,M_1,t)\,\phi^{(o)}(\vec{v}_2,M_2,t)\,dM_1dM_2dv_1dv_2 \qquad (3.13)$$

Consequently, if the solution of the linear equation (3.11) could be found, then in the first approximation the mass-velocity shape and number of aerosol particles could be determined. In a similar way it can be shown that a complete perturbation method can be achieved which is correct to all powers of the density. This is accomplished by writing ϕ in the form

$$\phi(\vec{v}, M, t) = \sum_{l=o}^{\infty} N^{(l)}(t)\,\phi^{(l)}(\vec{v}, M, t) \qquad (3.12)$$

and g(t) in the form

$$g(t) = \sum_{l-o}^{\infty} N^{(l)}(t) g^{(l)}(t) \qquad (3.15)$$

both being power series of the density N(t). Inserting Eqs (3.14) and (3.15) into Eqs (3.9) and (3.10), and subsequently equating equal-exponent coefficients of $N^{(m)}$ then gives the following coupled equations

$$\underline{L = o} \quad \frac{\partial}{\partial t}\phi^{(o)} - B_o\phi^{(o)} = o \qquad (3.16)$$

$$\underline{L \geq o} \quad \frac{\partial}{\partial t}\phi^{(l+1)} - B_o\phi^{(l+1)} = \sum_{n=o}^{l} \{\tfrac{1}{2}\int_{\vec{v}_1}\int_{\vec{v}_2}\int_{m_1}\int_{m_2} \delta\left[M-(M_1+M_2)\right]\delta\left[\vec{v}-(M_1\vec{v}_1+M_2\vec{v}_2)/M\right] x \qquad (3.17)$$

$$K(M_1,\vec{v}_1|M_2,\vec{v}_2)\phi^{(l-n)}(\vec{v}_1,M_1)\phi^{(n)}(\vec{v}_2,M_2)dM_1dM_2d\vec{v}_1d\vec{v}_2$$

$$+\tfrac{1}{2}\phi^{(l-n)}g^{(n)} - \phi^{(l-n)}\int_{\vec{v}_1}\int_{m_1} K(M,\vec{v}|M_1,\vec{v}_1)\phi^{(n)}(\vec{v}_1,M_1)dM_1d\vec{v}_1,$$

where

$$g^{(1)} = \sum_{n=o}^{1} \int_{\vec{v}_1}\int_{\vec{v}_2}\int_{m_1}\int_{m_2} K(M_1,\vec{v}_1|M_2,\vec{v}_2)\,\phi^{(n)}(\vec{v}_1,M_1)\,\phi^{1-n}(\vec{v}_2,M_2)\,dM_1 dM_2 d\vec{v}_1 d\vec{v}_2 \tag{3.18}$$

Careful examination of Eqs (3.16)-(3.18) shows that $\phi^{(L+1)}$ is related only to its predecessors so that if the solution to $\phi^{(0)}$ could be found the whole problem would be determined by standard perturbation means; namely the determination of the inhomogeneous solution from the homogeneous solution via operator technique (cf., chapter (9) in reference 20).

At present the cases under which $\phi^{(0)}$ can be found by well-known methods include (1) those for which B_0 does not include evaporation or condensation (in which case B_0 reduces to the usual Boltzmann operator) and (2) those for which simple evaporation models describe the growth.

During the course of the proposed work period we plan to extend the technique just outlined to include spatial variations, and carefully examine the conditions under which this is feasible and meaningful.

In addition, we shall explore, through independent avenues of attack, possibilities of achieving exact closed-form special case solutions, as for example has been shown to be feasible in coagulation processes when the reaction rate is mass-independent.*

Another approach to the solution of the aerosol equation, which can be used in conjunction with the perturbation method, is based upon the expansion of the distribution function $F(\vec{r},\vec{v},M,t)$ in a complete set of orthogonal functions in velocity space and mass space. While expansions of the distribution function in velocity space have been used successfully in conjunction with the Boltzmann equation, the extension to mass space is novel. Briefly, we expand the distribution function in the form--

$$F(\vec{r},\vec{v},M,t) = \sum_{n,k} \Gamma_{n,k}^{\rightarrow} (\vec{r},t)\, W_n(\vec{v})\, L_k(M) \tag{3.19}$$

---

* In this case one deals with the equation

$$\frac{\partial F(M)}{\partial t} = \frac{K}{2} \int_0^m F(M-M')F(M')dM' - KF(M) \int_0^\infty F(M')dM'$$

where K is the reaction rate. The solution of this equation is given in closed form, as discussed in reference 3.

where $W_n(\vec{v})$ and $L_k(M)$ are independent orthogonal sets of functions which satisfy the equations

$$\int W_n(\vec{v})W_1(\vec{v})d\vec{v} = \delta_{ne} \tag{3.20}$$

$$\int L_k(M)L_s(M)dM = \delta_{ks} \tag{3.21}$$

where δ_{α_β} is the Kronecker delta function. Possible choices of orthogonal sets of velocity space orthogonal functions are discussed in references 12 and 13 while a suitable orthogonal set of $L_k(M)$ can be constructed from the Laguerre polynomials which cover the range $o \leq M \leq \infty$.

Upon inserting Eq (3.19) into the aerosol equation as given by Eq. (2.1), multiplying the resulting equation on the left by a specific choice of $W_r(\vec{v})L_s(M)$, and then integrating the resulting expressions over velocity space and mass space yields a system of coupled space-time differential equations for the $\Gamma_{n,k}(\vec{r},t)$. Without going into details it can be shown that the following system of equations results:

$$\frac{\partial \Gamma_{r,s}}{\partial t} + \nabla_r \cdot \sum_n \Gamma_{n,s}\vec{V}_{r,n} + \sum_{n,k} \Gamma_{n,k} f_{r,s,n,k} = \sum_{n,k} \Gamma_{n,k} a_{n,k,r,s} + \sum_{n,k} \Gamma_{n,k} b_{n,k,r,s} + \sum_{n,k} \sum_{n',k'} \Gamma_{n,k}\Gamma_{n',k'} c_{n,n',k,k',s,s'} \tag{3.22}$$

where

$$\vec{V}_{r,n} = \int_{\vec{v}} W_r(\vec{v})\vec{V}\, W_n(\vec{v})\, d\vec{v} \tag{3.23}$$

$$f_{r,s,n,k} = \int_{\vec{v}} \int_m W_r(\vec{v})L_s(M)\left[\nabla_V \cdot \frac{\vec{X}}{M} W_n(\vec{v})L_k(M)\right] dMd\vec{v} \tag{3.24}$$

the $a_{n,k,r,s,}$'s constants resulting from mass-velocity space integration of the Boltzmann collision kernel, the $b_{n,k,r,s,}$'s constants resulting from integration of the condensation (evaporation) term, and the $c_{n,n',k,k',r,s,}$'s constants resulting from integration over the coagulation operator.

Although the foregoing system of equations may be exact, it will in general be extremely difficult to solve unless some simplifying assumptions can be made which will permit truncation. This will be investigated.

REFERENCES

1. F. Pasquill, "Atmospheric Diffusion", D. Van Nostrand Company, (New York, 1962)

2. H. S. Green and W. R. Lane, "Particulate Clouds: Dusts, Smokes and Mists", 2nd Edition, D. Van Nostrand Company, (New York, 1964)

3. N. A. Fuchs, "The Mechanics of Aerosols", The Macmillan Company, (New York, 1964).

4. M. von Smoluchowski, Z. Physik. 17, 557, 585 (1916).

5. M. von Smoluckowski, Z. Physik. Chem. 92, 129 (1917).

6. S. Chandrasekhar, Rev. Mod. Phys. 15, 1 (1943).

7. N. Fuchs, Z. Physik. Chem. A 171, 199 (1934).

8. L. V. Radushkevich and O. K. Chugunova J. Phys. Chem. U.S.S.R. 15, 811 (1941).

9. I. V. Petrjanov, M. V. Tikhomirov, and N. Tunitskii, Acta Phys. Chem. U.S.S.R. 17, 185 (1942).

10. R. S. Bradley, Trans. Faraday Soc. 32, 1088 (1936).

11. N. Fuchs, Z. Phys. 89, 736 (1934)

12. S. Chapman and T. Cowling, "The Mathematical Theory of Non-Uniform Gases", Cambridge University Press. (Cambridge, 1958).

13. H. Grad, "Principles of the Kinetic Theory of Gases", Handbuch der Physik, XII (Berlin, Germany: Springer-Verlag, 1958).

14. M. Kac, "Probability and Related Topics in Physical Sciences", Interscience Publishers, (New York, 1959).

15. G. E. Uhlenbeck and L. S. Ornstein, Phys. Rev. 36, 551 (1961).

16. S. Chandrasekhar, Rev. Mod., Phys. 15, 1 (1943).

17. M. C. Wang and G. E. Uhlenbeck, Rev. Mod. Phys. 17, 323 (1945).

18. H. Akama and A. Siegel, Phys. Flüids 8, 1218 (1965).

19. J. Frenkel, "Kinetic Theory of Liquids", Dover Publications, Inc., (New York, 1955).

20. P. Morse and H. Fesbach, "Methods of Theoretical Physics". McGraw-Hill, Inc., (New York, 1953).

21. E. Ikenberry, J. Math. Analysis and Appl. 3, 355 (1961).

22. E. Ikenberry, Arch. for Ratl. Mech. Anal. 9, 255 (1962).

OBSERVATIONS OF SIERRA NEVADA SNOW STORMS WITH AN MTI-EQUIPPED RADAR

Robert L. Peace, Jr.

Fresno State College Foundation
Atmospheric Water Resources Research

I. INTRODUCTION

A major aspect of the Fresno State College Foundation, Atmospheric Water Resources Research activity in weather modification is concerned with the detailed observations of winter storms in the central Sierra Nevada and their meteorological environments. These observations play a vital role both in guiding the seeding operations and in post-storm analysis designed to increase understanding of storm character. One potentially fruitful type of observation instrument is weather radar, but the terrain poses serious problems in its use. From the AWRR radar site at the Amador County Airport (Jackson, California), the radar ground return extends from the crest of the Sierra to the east to the eastern slopes of the Coastal Range to the west (Figure 1).

The high intensity of the radar echoes observed in the central California region that result from substantial ice-water contents in winter storms precludes the use of an X-band (3 cm wavelength) radar. An S-band (10 cm wavelength) radar would require a very large antenna and high-elevation angle to reduce ground return sufficiently to observe weather echoes in the same area. (The WSR-57 radar at the NOAA office in Sacramento has a 12-foot diameter antenna and experiences serious ground return at elevation angles of less than 2.5°). The solution adapted to the problem was the use of a radar equipped with Moving Target Indicator (MTI) capability.

II. RADAR CHARACTERISTICS

The radar used by AWRR for weather target observations is an MSG-3A, 1 megawatt, S-band radar with a 1.4° horizontal and a 5.5° vertical beam width and a range capability of 120 n mi. This radar is a more recent version of the M-33 radar often used for weather studies and in many ways, such as antenna and transmitter, is similar to the M-33. The antenna pedestal has been tilted 2° toward the west (where the radar site's 1700 foot MSL elevation provides an unobstructed line-of-sight to the Coastal Range) and is operated at 2° elevation angle to provide a 4° elevated view up the slope of the Sierra to the east (mean terrain slope about 3°).

The use of MTI to suppress coherent targets (most ground return) results in a sensitivity loss of about 15 db when the set is well adjusted

However, observations and documentations of weather echo configuration, location and movement are now possible and have revealed a number of distintive features of Central Sierra storms that were formerly unknown or only suspected. Foremost among these are the apparent prevalence of small-scale precipitation lines and the movement of precipitation cells very nearly with the winds at 8 - 10,000 feet MSL.

III. THE BANDED STRUCTURE OF MANY PRECIPITATION PATTERNS

The MTI on the MSG-3A radar has been functioning well for only six weeks at this time and already multiple lines of echoes have been observed on five separate storm situations. The occurrence of lines, belts or bands of precipitation have been detected, studied and reported on by several authors, such as Boucher and Wexler [1], Elliott and Hovind [2], Harper and Biemers [3], Higuchi [4], Kuettner [5], Malkus [6], Peace [7,8], Plank [9], and Woodcock and Wyman [10]. With the findings of these authors in mind, a study of PPI scope film from the Sacramento WSR-57 for two winter seasons revealed few instances of mesoscale echo lines or bands. The explanation for the high frequency of multiple, mesoscale, echo-line observations over the short period between November 30, 1970 and January 11, 1971 is unknown. It may be a coincidence, but the existence of such lines has been indicated by the intermittent nature of precipitation recorded at lower altitude project sites over the past two years. It appears more likely that the explanation lies in the location and characteristics of the MSG-3A radar (1700 feet MSL site, wide vertical beam width, and 15 db attentuation due to the MTI). In either case, these echo lines are pertinent to those persons controlling cloud seeding activities because they represent convergence lines.

Figures 2, 4, 5, 7, and 8 are samples of the PPI scope observations made when multiple, small-scale echo lines were evident on the scope. Figure 2 shows the second and third of four echo lines observed on November 30, 1970. The range marks on the scope represent the 40 and 57 n mi ranges from the radar. At this time (1842P) the echo lines were about 51 mi apart at their southern ends and about 35 mi apart north of Jackson. (They had been almost parallel until the eastern line entered the foothills to the north-northwest of Jackson when the northern end slowed and became wider.) Two hours later when the last of the four lines lay over the radar the two bands in Figure 2 were separated by only 11 miles. Each line decreased speed as passed over the foothills and moved eastward. The surface maps for 0400P on November 30 and December 1, 1970, appear in Figure 3. The precipitation echoes were associated with the passage of the weak, cold front shown entering California on the 30th and over southern California on the 1st.

Figure 4 shows a pair of echo lines that were observed at 1957P on December 15, 1970. At 0530P on December 16, 1970 the echo lines in Fig. 5 were observed. Figure 6 shows the surface maps for 0400P on both December 15 and 16, 1970. While the echo lines in Figure 5 were associated with a distinct cold front, those in Figure 4 do not appear to have been associated with any specific macroscale meteorological feature.

In Figure 7 and 8 are two more examples of multiple precipitation bands. Both of these systems produced light precipitation and the bands were correspondingly faint on the scope. Figure 7 is a pair of closely spaced echo lines observed at 2127P on December 28, 1970 while Figure 8 shows three closely spaced echo lines observed at 0537P on January 11, 1971. Both sets of echo lines are more distinguishable in the series of PPI scope photographs taken than in any single photograph.

IV ECHO MOVEMENTS AND THEIR RELATIONSHIP TO WINDS AND LINE MOVEMENT

The orientation of the echo lines studied appeared to bear little relationship to the winds aloft, but their movement is related to the winds aloft through the motion of individual cells within the lines. The trajectories of the cells studied very closely paralleled the winds aloft at 8 to 10,000 feet MSL and the lines of echoes in turn moved with the component of echo movement normal to the line of which they were a part.

Part of the AWRR observing program consists of making rawinsonde releases at Jackson, Calif. at 1700 ft. MSL in the Sierra foothills and at the Bear Valley Ski Area (1968-1969 and 1969-1970 seasons) or Tamara (1970-1971 season) at the 6,000 foot MSL level in the Sierra. When winds from these soundings have been compared with each other and with the Oakland, California rawinsonde there is a persistent 10° to 60° backing of the winds from Oakland to Jackson to Bear Valley or Tamarack (cyclonic streamline curvature). When the individual radar echoes are traced from a sequence of PPI scope photographs, they too generally show a cyclonically curved trajectory. In addition, when a number of individual radar echoes are traced for the same time period, the eastern-most ones tend to have more southerly trajectories than those further west.

V. EXAMPLES OF ECHO MOVEMENT

Tracings of individual radar echoes have been made from two to three minute interval PPI scope photographs taken during the November 30, 1970 storm. Figures 2 and 9 show the locations of the two echo lines that were observable at 1842P when a radiosonde was released at Jackson, and tracked by an X-band tracking radar. Four echo cell tracings in Fig. 9 for 1840P are shaded and a second position is shown for each. These represent the location of each of these four cells 15 minutes after the echo line tracings. The fifteen minute average direction and speed of each is entered adjacent to the echoes' location at 1840P. The echo directions in the two lines differ by 35° (southern end) to 55° (northern end) and the speeds are greater in the eastern line.

The Jackson rawinsonde was released into the eastern-most line shown in Figure 9. The balloon apparently collected a coating of ice and it began to descend from 6,000 feet MSL. Twenty-five minutes after release the balloon had descended to 3,000 feet MSL and was located 19 mi north of Jackson. At that time the western edge of the radar echo passed Jackson and the rain stopped. The balloon subsequently ascended continuously to 19,000 feet MSL where the radar track was lost at a range of 42 mi to the north-northeast. The winds for this run are tabulated in Table I. These winds represent the conditions along the western edge of the rain area very near the echo that moved from 205° at 43 knots. This is in close agreement with the 8,000 foot MSL winds in Table I.

About three hours after the time of Figure 9, the western band was located just east of Jackson, the remnant of the eastern band had moved to about 20 mi east of the radar and a third band had formed behind the western band, and was located over the station. Tracings of the three echo lines are shown in Figure 10 for 2221P. Also shown in Figure 10 are three 15-minute interval positions of one cell in the echo line just east of Jackson (positions at 2154, 2209, and 2221P), and the 2209 and 2221P positions for two echoes in the newly formed echo line over Jackson. The echo for which 30 minutes of continuity are shown moved from 225° for the first 15 minutes and from 210° for the second with a half-hour average movement from 215°. The other two echoes both moved from 215°. This direction of movement from 210° to 215° at 24 to 25 knots is typical of cell movements in both western lines (clear movement could not be acquired for cells in the remnant of the eastern line). The 210° movement of the eastern cell is 35° more southerly than the movement observed at the southern end of this same line at 1840 and 1855P, and almost parallel to the echo line. The echo line, in turn, moved at only about 2 to 5 knots after 2230P. The cell movement in Fig. 10 is also in close agreement with the winds acquired by radar track at Jackson (Table II).

The storm of December 16, 1970 provided the opportunity to track a single, continuously identifiable cell for over two hours while the band of echoes of which it was a part, moved across the Sacramento and San Joaquin Valleys and into the Sierra foothills. Figure 11 shows tracings of the echo line at 0530P and 0731P on December 16, 1970. The shaded echoes are the tracings of a single echo at 30 minute intervals for the 0530P and 0731P period. While the echo line appeared to move from about 305° at 10 knots the echo moved along a cyclonically curved path that varied from 240° at 28 knots to 225° at 30 knots. The winds aloft for Oakland, Lodi and Jackson for about 0400P are shown in Table III. The backing of the winds-aloft from west to east is similar to that commonly observed and in close agreement to the cyclonically curved cell trajectory. The average of the 8000 foot MSL winds at Oakland and Lodi was 245° at 30 to 31 knots and the average of the 10,000 foot MSL winds was 240° at 34 or 35 knots. This compares closely to the 240° at 28 knots movement of a cell about half-way between. When the cell was due north of Jackson (but closer to the mountains because of the orientation of the range) its 225° at 30 knot cell movement is comparable to the 8,000 and 10,000 foot winds at Jackson (see Table III) when the four-hour difference in time and 20 mi difference in locations are considered.

Echo motion studies have been performed on PPI scope film data from the WSR-57 at Sacramento, California for the 1968-1969 and 1969-1970 winter seasons. Figure 12 is an example of the results of such studies.

In Figure 12 a number of echoes are shown that were trackable for a 25-minute period on the PPI scope film for February 13, 1969. The 1017P and 1042P position of each echo is shown along with the 8,000 ft. MSL winds for Jackson and the 10,000 ft. MSL winds for Bear Valley station (Mt. Reba REB) observed at about 1000P. The west to east change in echo direction from 250° to 215° shows the same kind of cyclonic turning shown in Figures 9 and 11 and similar agreement with the 8 - 10,000 foot MSL winds aloft.

VI. CONCLUSIONS

The MTI equipped radar has proved to be a valuable observational tool in support of both seeding operations and post-analysis of storm characteristics. Careful post-storm analyses strongly indicate that radar echo movement is a good indicator of wind direction and a close approximation of wind speed at the 8 to 10,000 foot MSL level. These data should be usable for both streamline analyses over areas where no wind data is available and a real time indication of wind conditions for seeding operations.

The persistence of cyclonic curvature in radar echo tracks coupled with a similar west to east backing of the observed winds aloft indicates that a 10° to 60° corrections must be made in winds observed west of the foothills before they can be used to predict the drift of seeding material targeted for the mountains. It also indicates that echo extrapolation in support of seeding operations should be along a cyclonically curved trajectory.

Lastly, the banded structure of storms as reflected by radar echo lines on the attenuated MTI-equipped radar appear to agree with banded structures observed in other locations, and further study should provide insight into the dynamics of Central Sierra Nevada snow storms.

VIII ACKNOWLEDGEMENTS

This work was performed under Contract No. 14-06-D-6592 with the U. S. Department of the Interior, Bureau of Reclamation, as a part of Project Skywater. The radar and upper-air observations used in the analyses described in this report were made possible by the support of the Earth and Planetary Sciences Division of the Naval Weapons Center in the form of the long-term loan of the MSG-3A radar and the GMD-1 rawinsonde located at Jackson, California. Supplemental radar measurements with the WSR-57 radar during storm periods by Mr. Roger Pappus and staff at the NOAA office in Sacramento were also vital to the work.

REFERENCES

1. Boucher, R. J. and Wexler, R., 1961, "The Motion and Predictability of Precipitation Line," J. Meteor., 18, 160-171.

2. Elliott, R. D. and Hovind, E. L. 1964. "On Convection Bands Within Pacific Coast Storms and Their Relation to Storm Stucture," J. Appl. Meteor., 3, 143-154.

3. Harper, W. G. and Beimers, J.G.D., 1958, "Movement of Precipitation Belts", Quart. J. R. Meteor. Soc., 84, 242-249.

4. Higuchi, K., 1963, "The Band Structure of Snowfall," J. of Meteor. Soc. of Japan, Ser. II, 41, 53-70.

5. Kuettner, J., 1959, "The Banded Structure of the Atmosphere", Tellus, 11, 267-294.

6. Malkus, J. S. 1963, "Cloud Patterns Over Tropical Oceans", Science, 141, 767-778.

7. Peace, R. L. Jr. and Sykes, R. B., Jr., 1966, "Mesoscale Study of a Lake Effect Storm", Mon. Wea. Rev., 94, 495-507.

8. Peace, R. L. Jr., 1966, "Radar Characteristics of Lake Effect Storms," Proc. 12th Conf. on Radio Meteor., 454-460.

9. Plank, V. G., 1965, "The Cumulus and Meteorological Events of the Florida Peninsula During a Particular Summertime Period," AFCRL Environmental Research Paper No. 151, Bedford, Massachusettes, 156 pp.

10. Woodcock, A. H. and Wyman, J., 1947, "Convective Motion in Air Over the Sea," Ann. N. Y. Academy of Sci., 48, 749-776.

TABLE I

Winds Aloft for Jackson, California
November 30, 1970 - 1842 P release

Surface	180/18 kn	6000' MSL	205/46
2000' MSL	180/32	7	205/50
3	180/32	8	205/46
4	185/43	9	210/46
5	190/40	10	215/45
6	205/40	12	230/45
5	190/46	14	235/50
4	185/45	16	250/63
3	180/32	18	260/80
4	185/40	19	260/85
5	195/43		

TABLE II

Winds Aloft for Jackson, California
November 30, 1970 - 2324 P release

Surface	195/12 kn	9	215/30
2000 ' MSL	200/16	10	215/34
3	200/22	12	225/34
4	215/23	14	245/44
5	215/23	16	260/53
6	215/22	18	260/53
7	215/25	20	260/67
8	215/28		

TABLE III

Winds Aloft for December 16, 1970

Height	Jackson (0334 P)	Lodi (0407 P)	Oakland (0400 P)
2000' MSL	150/11 Kn	230/19 Kn	240/22 Kn
3	195/15	240/24	240/22
4	230/19	240/28	245/21
5	245/23	240/30	245/22
6	245/27	235/28	250/25
7	235/31	235/30	250/28
8	230/34	240/32	250/29
9	225/40	240/34	250/33
10	225/50	230/33	250/36
12	225/54	250/37	245/72
14	240/65	240/57	260/93
16	240/88	240/73	250/95
18	245/102	240/94	250/97
20	250/112		250/97

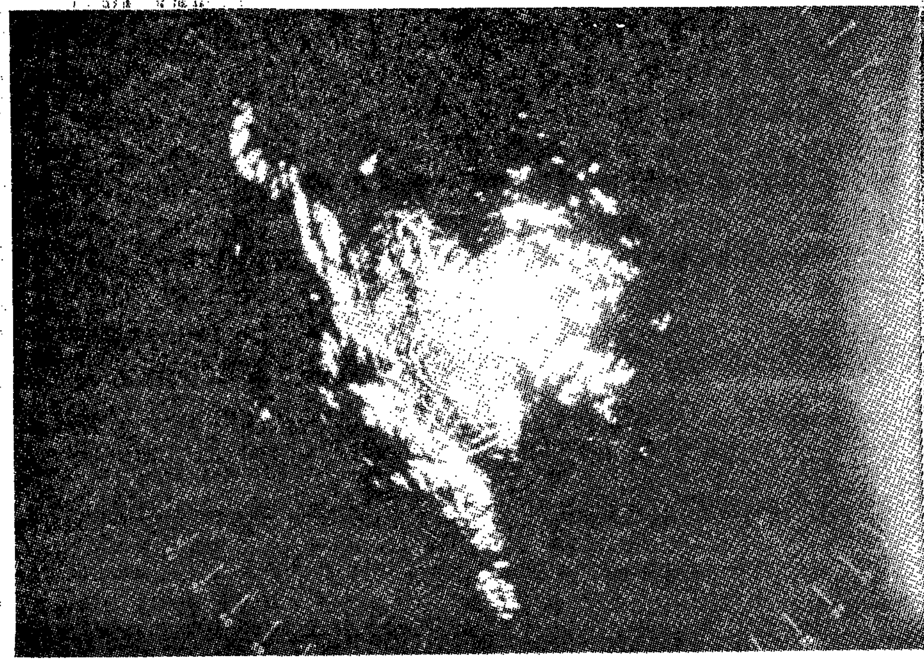


Figure 1. A PPI scope photograph of the ground return experienced by the AWRR radar at Jackson, California. The scope range is 120 n mi and the antenna elevation angle was 6° toward the west and 10° toward the east.

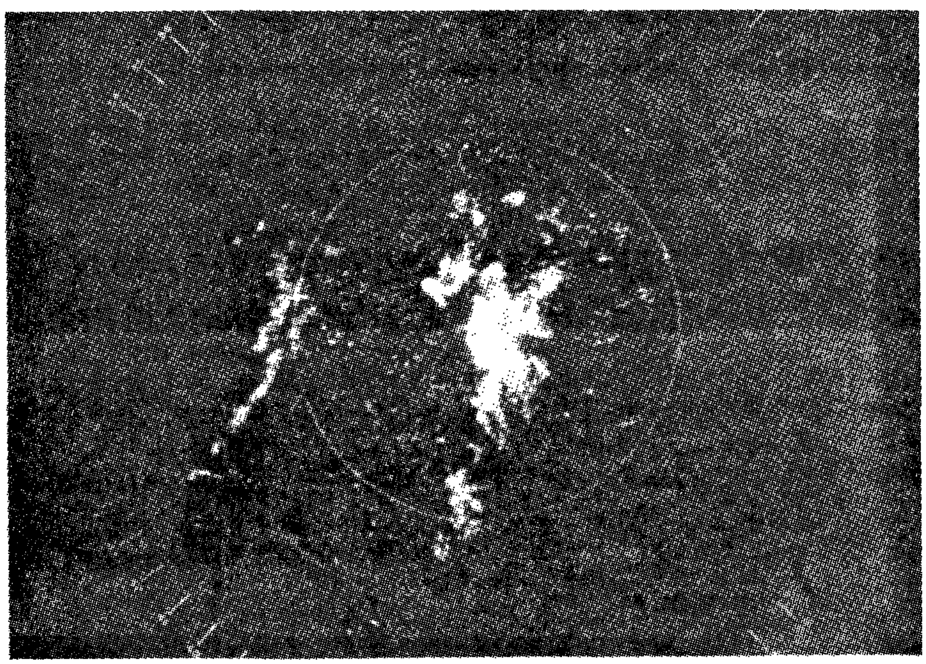


Figure 2. A photograph of the PPI scope of the Jackson, California radar taken at 1842P on November 30, 1970. The range marks represent 40 and 57 n mi ranges. The antenna elevation angle was 0° toward the west and 4° toward the east.

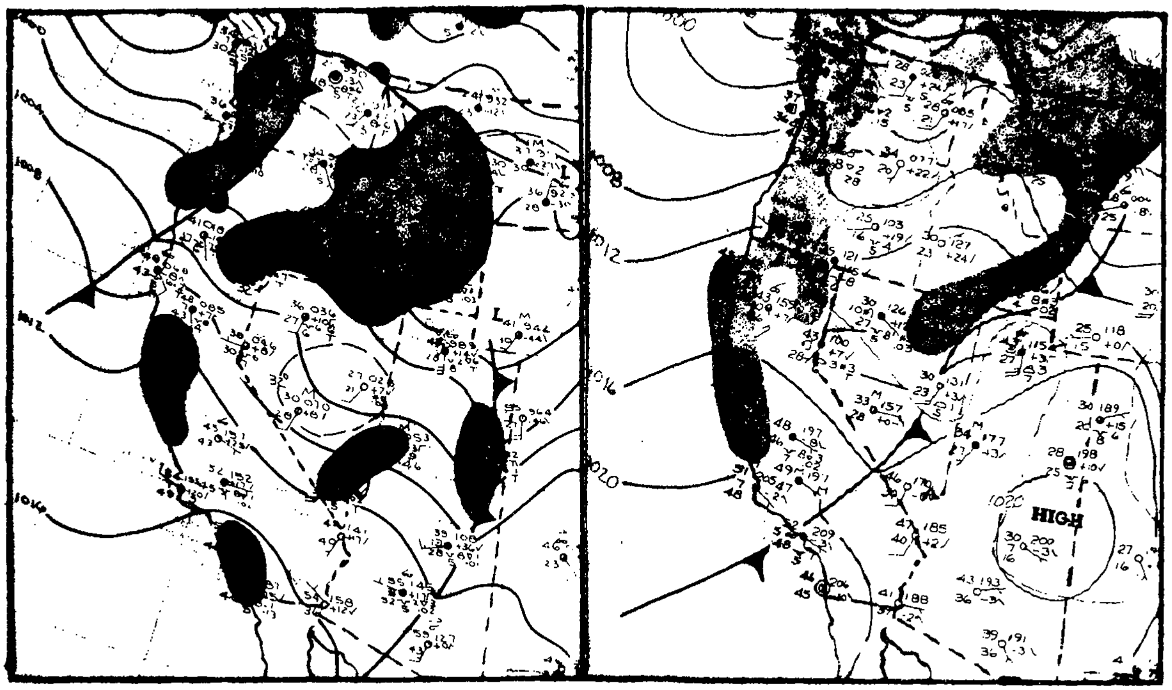


Figure 3. The western portion of the NOAA surface maps for 0400P on November 30 and December 1, 1970.

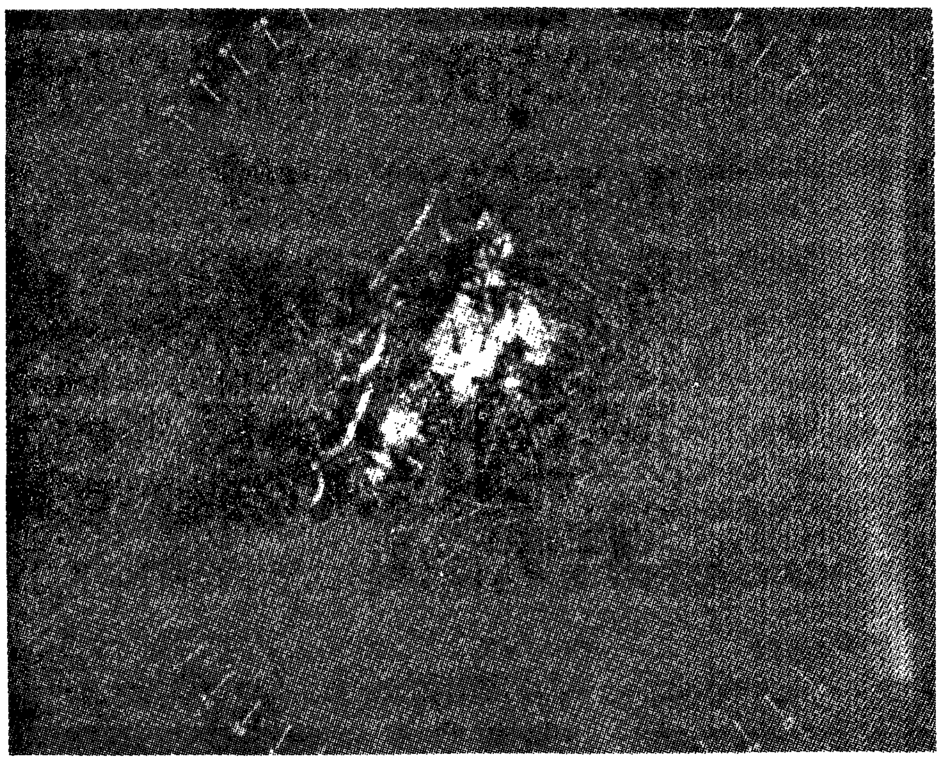


Figure 4. A photograph of the Jackson radar's PPI scope taken at 1957P on December 15, 1970. The range marks represent 40, 57, 73, and 106 n mi ranges. The antenna elevation angle was 0^{o} toward the west and 4^{o} toward the east.

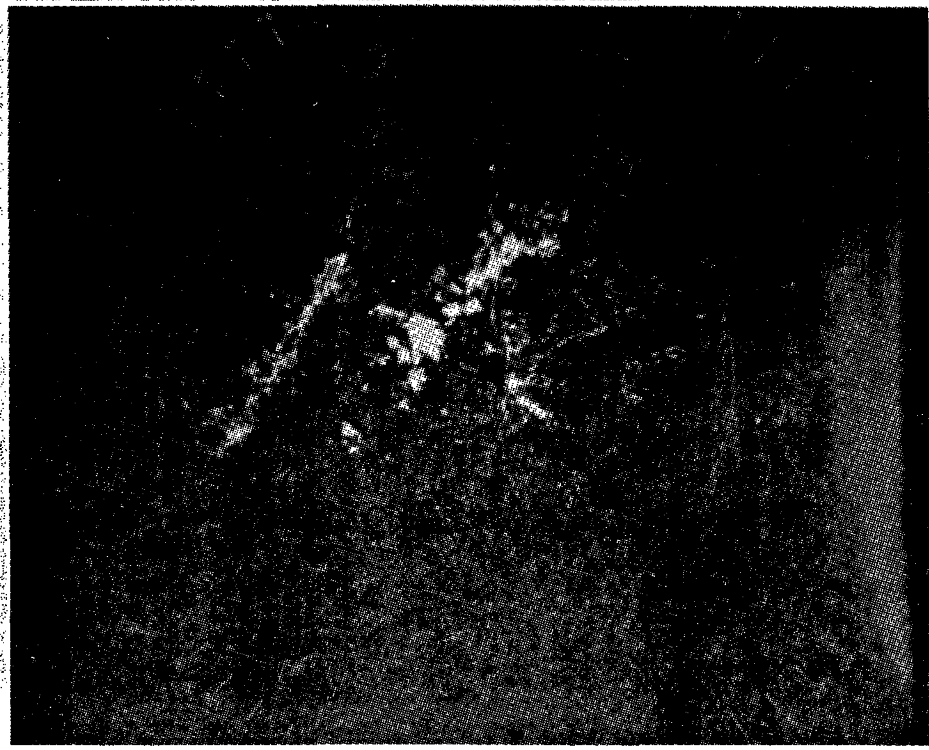


Figure 5. A photograph of the Jackson radar's PPI scope taken at 0530P on December 16, 1970. The range marks represent 40 and 57 n mi ranges. The antenna elevation angle was 0° toward the west and 4° toward the east.

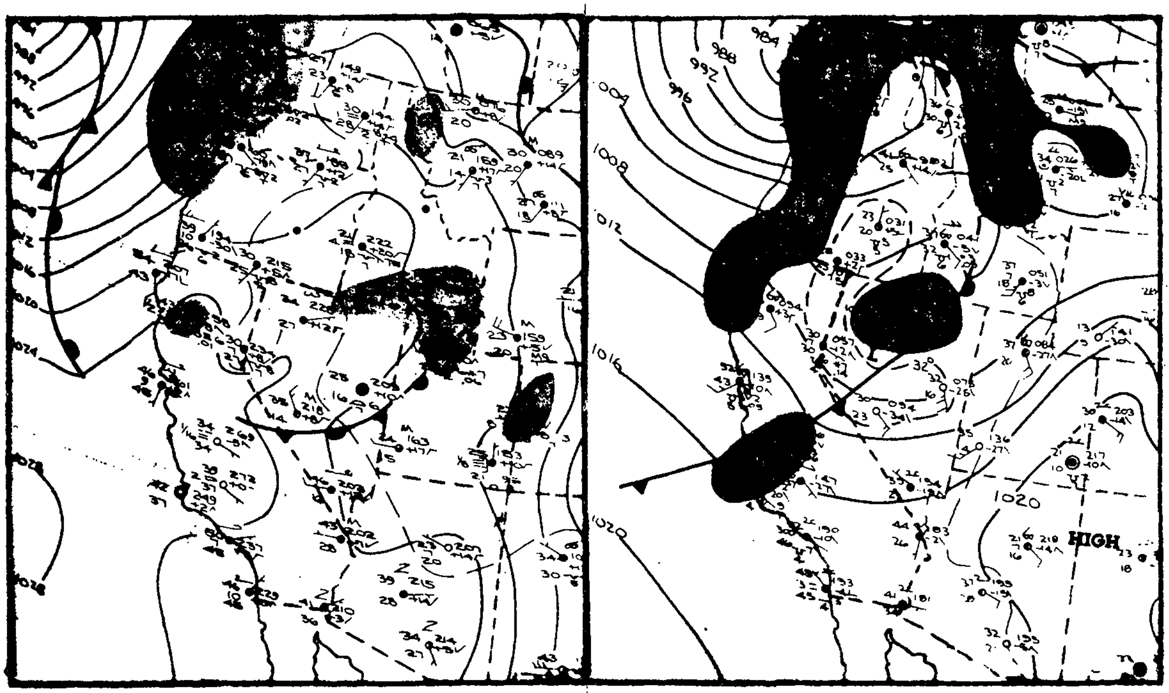


Figure 6. The western portion of the NOAA surface maps for 0400P on December 15 and 16, 1970.

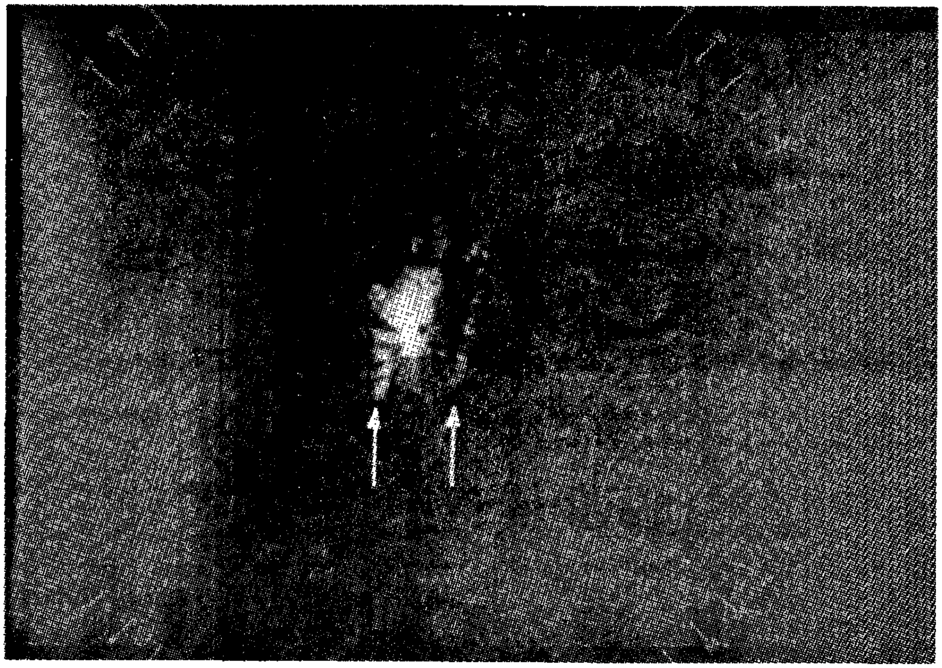


Figure 7. A photograph of the Jackson radar's PPI scope taken at 2127P on December 28, 1970. The range marks represent 40 and 57 n mi ranges. The antenna elevation angle was 0^{o} toward the west and 4^{o} toward the east. The white arrows indicate the two weak echo lines observed.

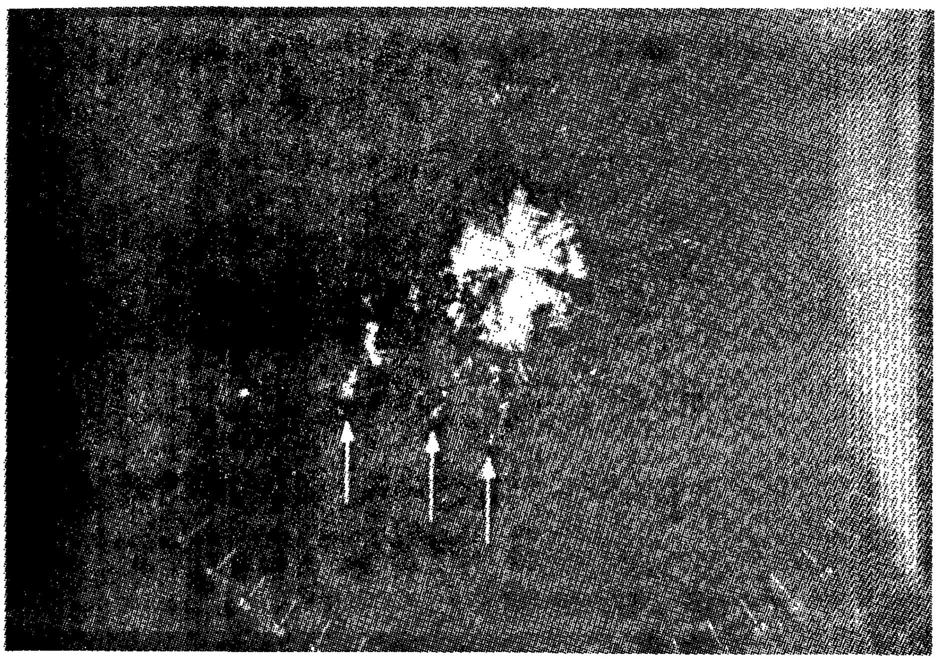


Figure 8. A photograph of the Jackson radar's PPI scope taken at 0537P on January 11, 1971. The range marks represent 40 and 57 n mi ranges. The antenna elevation angle was 0^{o} toward the west and 4^{o} toward the east. The white arrows indicate the locations of the three weak echo lines observed.

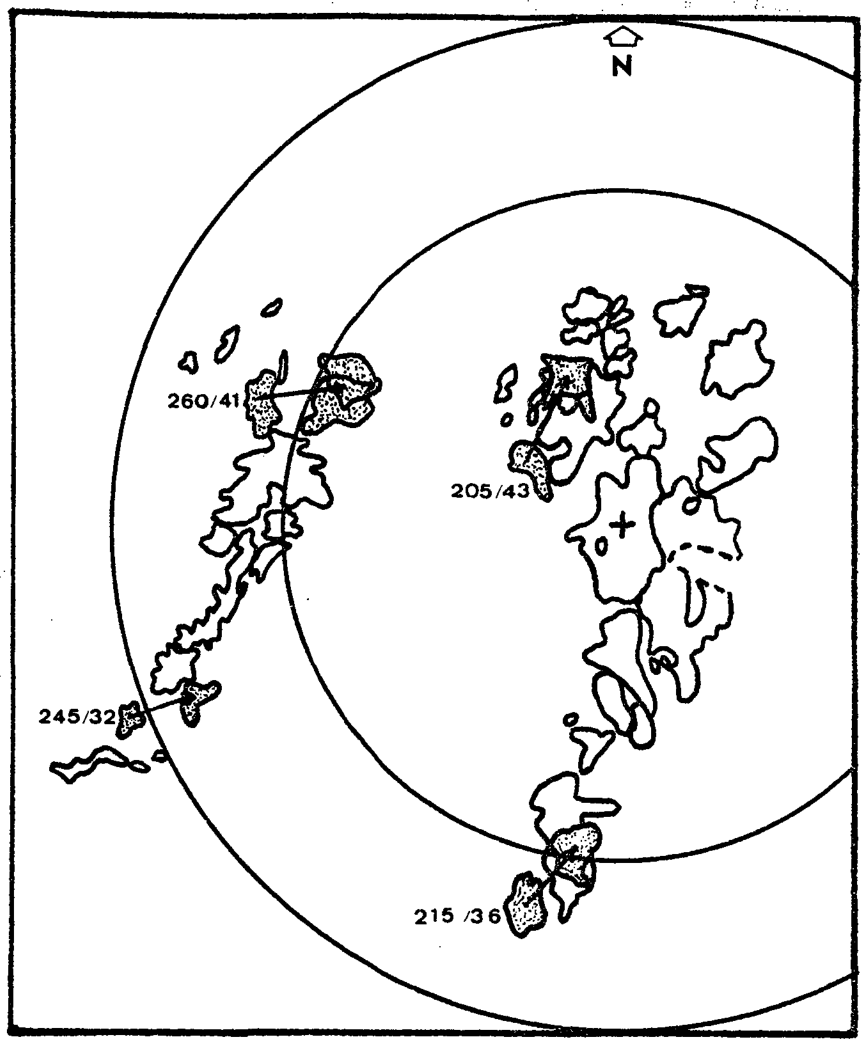


Figure 9. Tracings of the echo lines photographed on the Jackson radar's PPI scope at 1840P on November 30, 1970. The shaded echoes are tracings of four selected echoes as they appeared at 1840P and 1855P. The approximate mean speed and direction are entered adjacent to the 1840P position of each echo in degrees and knots. The range marks represent 40 and 57 n mi ranges.

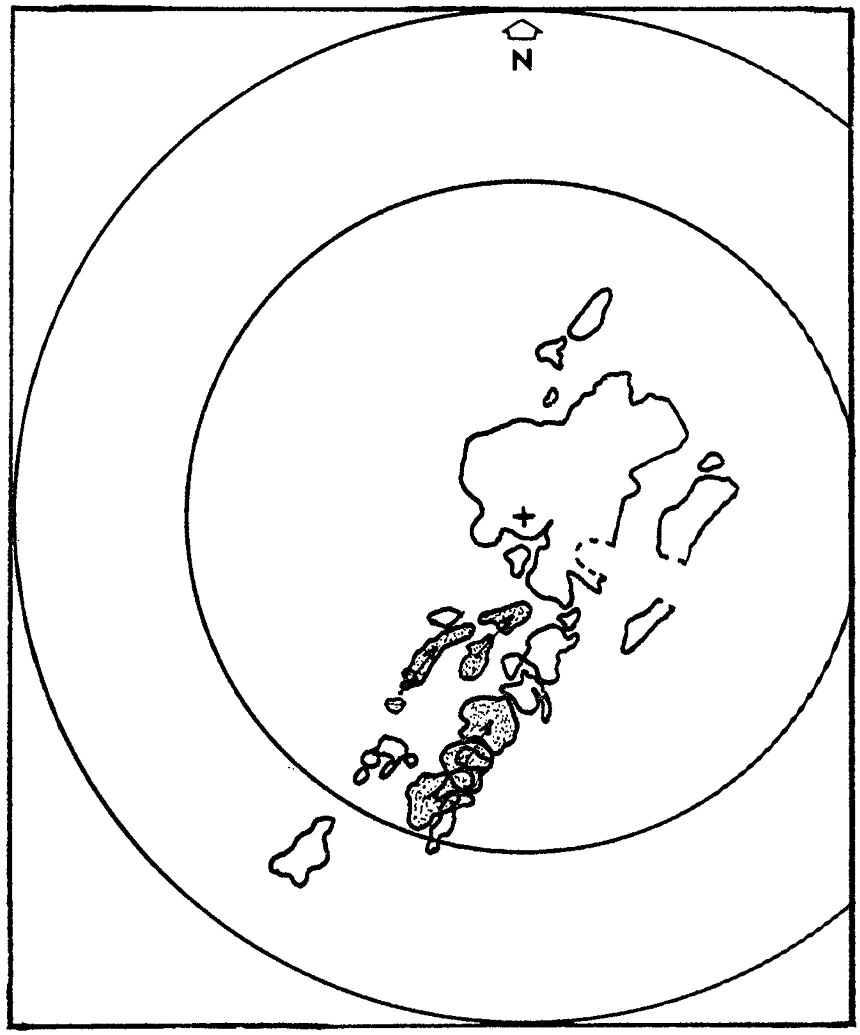


Figure 10. Tracings of three echo lines photographed on the Jackson radar's PPI scope at 2221P on November 30, 1970. One set of shaded echoes represents the 2154, 2209, and 2221P positions of a single echo in the same echo line that appeared as the western line in Figure 9. The two pairs of echoes represent the 2209 and 2221P positions of two echoes in the newly formed western line. These two echoes both moved from 215° and the speed of the right hand one was determined to be 25 knots. The cell represented by three positions moved from 225° at 24 knots for the first 15 minutes and from 210° at 25 knots for the last 15 minutes.

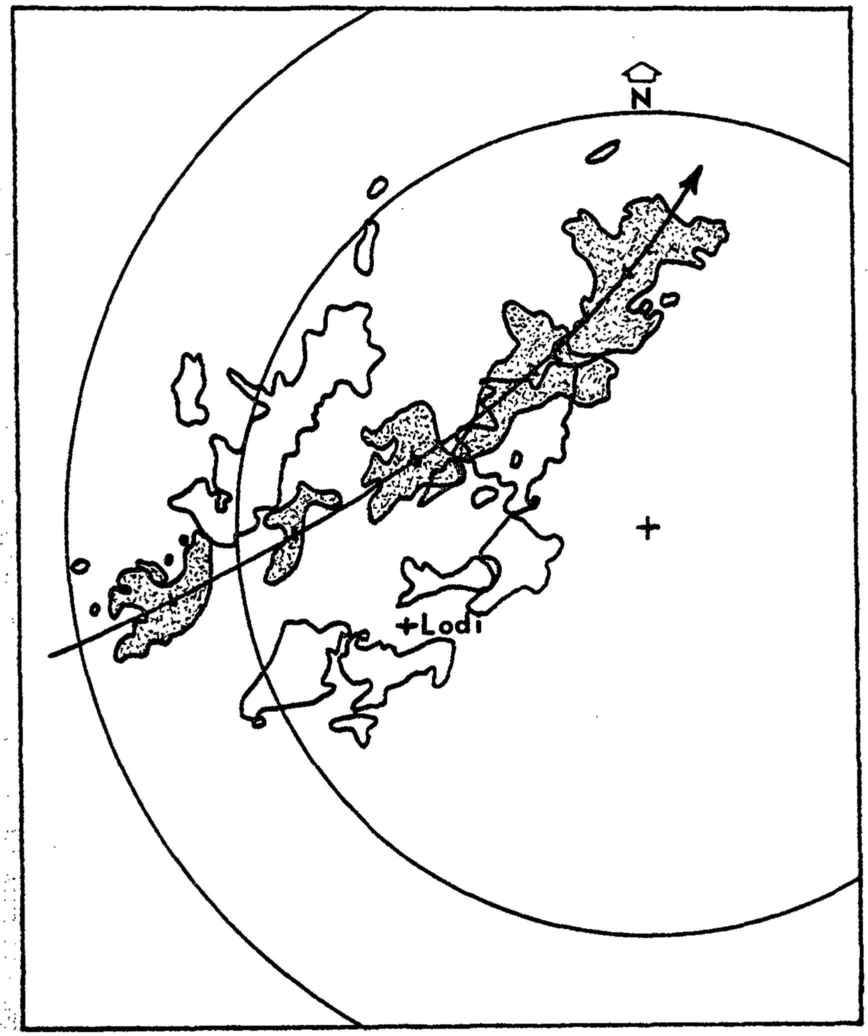


Figure 11. Tracings of a single echo line as it was photographed at 0530 and 0731P on December 16, 1970. The shaded echoes are the 30 minute interval locations of a single cell that formed the southern end of the echo line at 0530P and its northern end at 0731P. The line represents the echo's trajectory for the 2-hour period. The echo's movement for successive 30-minute intervals was 240° at 28 knots, 240° at 28 knots, 235° at 29 knots and 225° at 30 knots. The location of the Lodi balloon release site is indicated for reference.

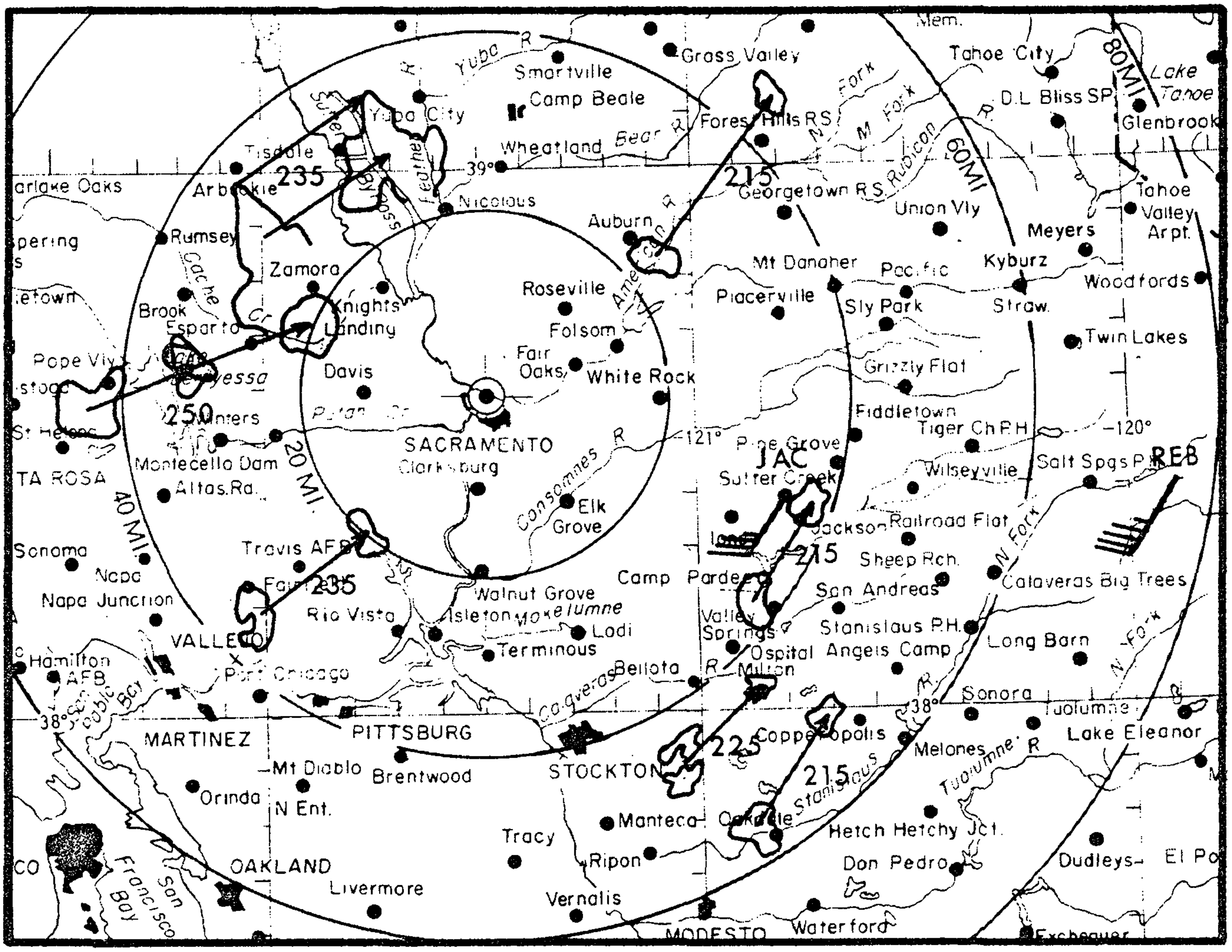


Figure 12. The locations of eight radar echoes observed by the NOAA WSR-57 radar at Sacramento, California at 1017 and 1042P on February 13, 1969. The antenna elevation was 3° and all photographs from which 3-minute interval echo positions were traced were made at full radar gain. The number adjacent to the arrow-connecting the two locations of each echo is the mean direction of echo movement during the 25-minute period. The two wind arrows are the 8000 foot MSL wind for Jackson and the 10,000 foot MSL wind for Bear Valley Ski Area (Mt. Reba).

SUMMER RUNOFF INCREASES BY WEATHER MODIFICATION

Donald E. Lehrman

Fresno State College Foundation
Atmospheric Water Resources Research

INTRODUCTION

The annual precipitation regime in the Sierra Nevada mountains of California is characterized by predominantly winter-time occurrences. Approximately 95 percent of the total annual precipitation occurs during the period October through April. Extensive agricultural development in the Central Valley of California has been based on the utilization of the water supplies represented by the accumulated high elevation precipitation. The occurrence of subnormal precipitation results in shortages in supply during the summer months when irrigation demands are at a maximum. During these periods, the degree of water resource development make it economically feasible to consider supplementing the natural supply through modification of the summer cloud systems despite their comparatively low runoff volume potential.

A project to investigate the potential for enhancing water supplies through seeding summer-time orographic cumuli over the high portions of the Sierra Nevada was developed in May 1966[1]. This project was designed in a step-wise manner to provide a tool for use by water resources groups within a period of three years. The four phases of the project consisted of: 1) hydrologic studies, 2) pair seeding, 3) quantiative determinations, and 4) forecast tools and physical models.

The project was conducted during the 1966, 1967, and 1968 summer seasons[2]. Support for the project was primarily by the U. S. Department of the Interior, Bureau of Reclamation, Office of Atmospheric Water Resources, under Contract Nos. 14-06-D-5819 and 14-06-D-6592, with supplementary support from the Kings River Conservation District for field operations during the 1968 season. Research operations were conducted by the staff of Atmospheric Water Resources Research, Fresno State College Foundation, with contract assistance for field operations from Atmospherics Incorporated, and hydrologic studies by Sierra Hydrotech.

This report will be concerned with the third phase of the Sierra Cumulus; the evaluation design and results of two years of cloud seeding over a high elevation target area.

THE DESIGN

Streamflow was selected as the evaluation parameter for several reasons. Historical precipitation data in the higher elevation watersheds in the Sierras are virtually non-existent, which precludes the use of any target-control storm precipitation amount relationships or comparisons of yearly or storm precipitation amount deviations from a mean. Additionally, since much of the higher elevation portions are designated wilderness areas, precipitation gages could not be installed in order to measure rainfall during the research period, thus a randomized project was not feasible. Furthermore, streamflow represents the quantities which are of interest to water users in terms of increases in usable water. Several factors influence the relationship between rainfall and runoff; among these are basin efficiency, basin priming, and the timing of the precipitation.

On the other hand, a substantial number of streamflow records have been maintained according to standardized methods and published by the United States Geological Survey(3) in the Southern Sierra for many years. Over the years the number of stations has increased, and the quality of the record is quite good.

In a preliminary study to investigate runoff as an index for evaluating a weather modification experiment(4), it was determined that a randomized project would be impractical because of the time-sequential relationship between runoff "occurrences" (in this report, an "occurrence" indicates a detectable increase in runoff over the established recession or base flow). It is quite possible that seeding on days which produced no appreciable runoff might well affect later runoff significantly. Additionally, randomization of basins on an annual basis is impractical because of the small portion of the total hydrograph under consideration. There is a large degree of variability between years and between storms in runoff produced from summer precipitation in any given basin. This almost precludes any method which would evaluate a change in means, as it would take an extremely large number of events (or extremely large increases in runoff) to obtain significant results. On the other hand, even though there is great variation in precipitation and runoff between basins in any individual storm period, most runoff production results from storms which cover a number of sub-basins. This makes it possible to achieve a fair degree of correlation between runoff occurring on various basins from the same storm or storm period.

Thus, the target basin/control basin approach was utilized in the evaluation design. A number of sub-basin hydrographs were prepared and analyzed for the period 1952 through 1964. Each "occurrence" of runoff attributable to summer precipitation was separated from the annual hydrographs for each basin and was assigned a volume and a date. Occurrences before the end of snowmelt could not be easily separated, thus only the period from the end of snowmelt (defined for this study to be when the discharge on the Kings River above North Fork dropped below 1,350 or approximately 1.4 cfs/square mile) through September 30 was considered in the design. The separation was made by computer from the daily hydrograph, anticipating the shape of systematic runoff (recession curve) by searching the record up to 30 days in advance of date of computation. It should be noted that this sytematic runoff was not always decreasing during the summer. It was found that the systematic flow tended to increase in the fall (September-October) even without significant precipitation, most probably as a result of decreased transpiration requirements at this time of the year. An example of a separated hydrograph is offered in Figure 1. The dashed line indicates the projected recession curve.

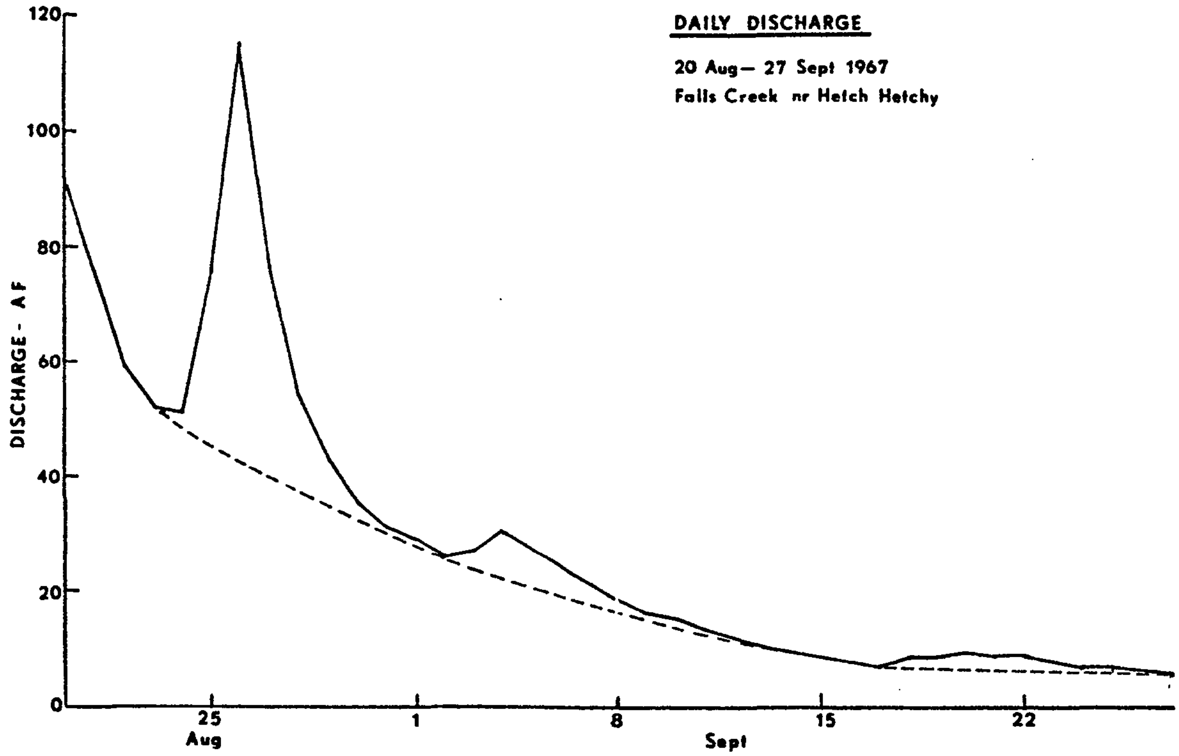


Figure 1. Example of Separated Hydrograph

On the basis of correlations achieved between different basins and occurrences on these hydrographs, and upon the physical characteristics of the watersheds; Bear Creek near Lake Thomas A. Edison was selected as the target basin. The control basins selected were Falls Creek near Hetch Hetchy, the Merced River at Happy Isles, the north fork of the San Joaquin below Iron Creek, and the Kings River above North Fork. The locations of the target and control basins are presented in Figure 2. Bear Creek basin is separated from the nearest control basin by approximately twenty miles, and is somewhat over 50 square miles in area. This basin has a mean elevation of 10,000 feet, and the distance from the basin centroid to the crest of the Sierra is four miles. Bear Creek basin has a vegetative cover of 48 percent and represents a fairly efficient basin in terms of runoff. A regression equation relating the volume of runoff from Bear Creek to the four control basins for an "occurrence" was developed.

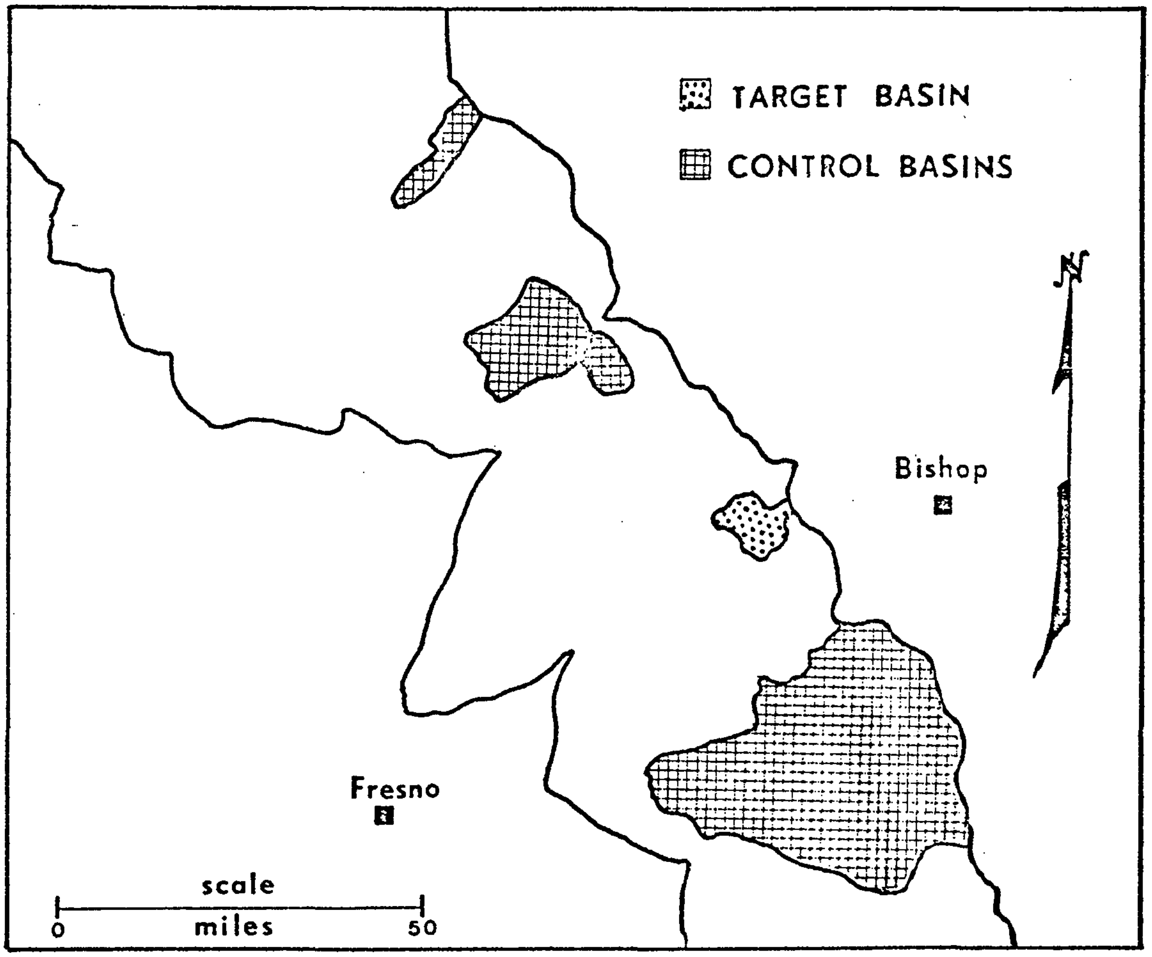


Figure 2. Location Map

Assuming a normal distribution of errors (a satisfactory assumption), estimates of the number of "occurrences" required on Bear Creek to detect various percentage increases in runoff attributable to seeding at various levels of significance were made. The average number of "occurrences" per year over the historical period was 7.2, varying from 4 to 10. It was concluded that at the .01 level, an average increase of 2 acre-feet per square mile would take two years to detect, while an increase of 1 acre-foot per square mile might take five years to detect.

The proceeding results then became the basis for the experiment design. During the summer periods of 1967 and 1968, all general storms (storms which were forecast to cover a number of sub-basins as opposed to isolated convective activity) occurring over the project area were seeded. The operating period was from the end of snowmelt (as defined by the streamflow volume of the Kings) through September. It was appreciated that because of the sort of statistical approach utilized, exact numbers of additional runoff produced through artificial enhancement of summer-time cloud systems could not be achieved. However, indication of the amounts (if any) could be resultant.

RESULTS

During the two years of operations the anticipated number of suitable storms did not occur. Due to an unusually late date of the end of snowmelt, only a limited period of time remained in which to conduct weather modification activities during the summer period of 1967. Although the end of snowmelt occurred quite early in 1968, very few storms which met the criteria for operations appeared. Table I presents a summary of the occurrences encountered during the two years, and the actual and predicted runoff, as well as the apparent increase (decrease) from the target and control basins. The dates shown indicate the day of peak runoff.

In 1967, the first storm period was the most productive and covered an extensive area in the Sierras, lasting for several days. Unfortunately, this storm was seeded only on September 3rd. Nevertheless the observed runoff was substantially greater than anticipated by the historical relationships. The second seeded occurrence was in the middle of September and lasted only one to two days. Although the storm was widespread, the activity was light in the Sierra. In this event, the volume of runoff from the target was less than anticipated by the regression equation prediction. The last storm of the 1967 season which could be separated as a storm occurrence, had a peak runoff on September 26th. This storm occurrence lasted approximately five days. The runoff produced was greater than predicted, although not significantly. Some additional storm activity took place before the end of September, but runoff characteristics of the various basins made it impossible to use data from this period. No seeding took place after September 25th.

During the dry 1968 season, only four suitable occurrences presented themselves, and all of those were seeded. Two of these occurrences lasted over a period of several days, and were seeded repeatedly throughout their duration. Seeding activities were conducted on four days on the occurrence which had its peak runoff on July 8th. The control watersheds had little runoff during this period. However, substantial amounts of runoff were observed from Bear Creek during the same period. The second extended duration storm, peaking on the hydrograph on July 30th, was seeded on three of five days. Again substantial apparent increases were noted from the runoff volumes. The third occurrence of the season, on August 13th, was seeded on that day but produced little volume of runoff on any of the basins. The observed and predicted amounts are in good agreement. The fourth storm period showed a peak runoff on August 21st. Major storm activity lasted only one day in the Bear Creek area, although significant runoff was noted in several basins to the north of Bear Creek, indicating that a major storm activity took place north of the area of interest. Predicted and observed amounts of runoff were negligible. After a lengthy, dry period, seeding took place on a short duration storm on September 29th; no runoff of consequence was noted on any of the basins precluding any evaluation of results. Because of the dry period most probably all the resulting storm precipitation was utilized in priming the basins.

Table I

Summary of 1967-1968 Events

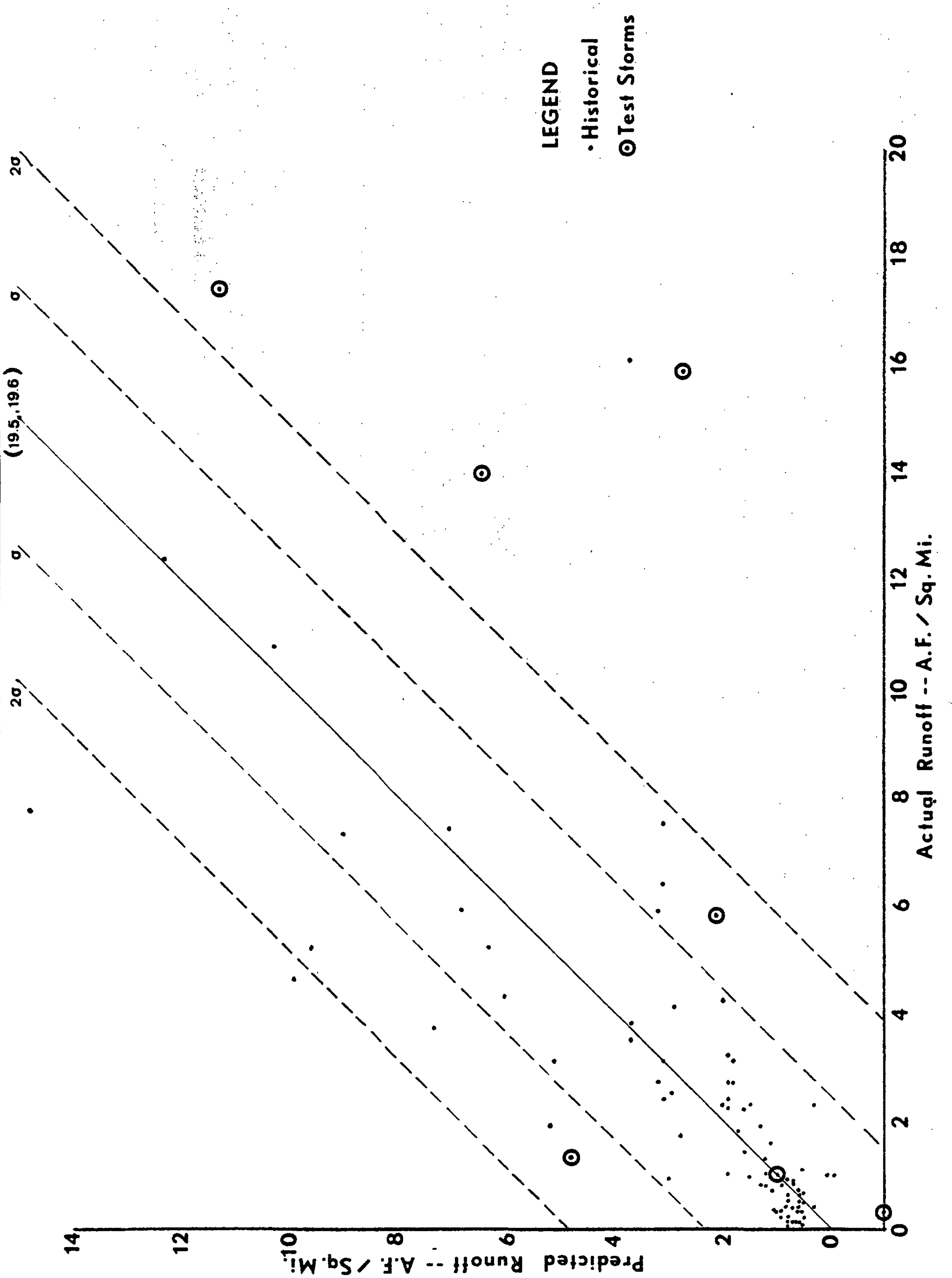
Date (of Peak Runoff)	Falls Creek	Merced River	North Fork San Joaquin	Kings River	Predicted Bear Creek	Actual Bear Creek	Actual - Predicted
1967							
9 - 4	.65	4.19	19.15	8.82	11.29	17.38	+6.09
9 - 19	0	1.77	4.78	2.83	4.81	1.31	-3.50
9 - 26	0	.66	3.94	1.26	2.13	5.79	+3.66
1968							
7 - 9	2.00	1.22	19.25	3.65	2.71	15.90	+13.19
7 - 30	.69	3.15	21.05	5.14	6.37	14.00	+7.64
8 - 13	0	.44	1.35	0	1.04	1.01	- .03
8 - 21	6.05	0	12.45	.19	-1.58	.28	+1.86
1967-1968					26.77	55.67	28.90

* Runoff in units of acre-feet per square mile

These results are summarized in Figures 3 and 4, which are scattergrams of the predicted and actual runoff volumes of the historical data as well as the occurrences during the operating period. Also included on the scattergrams, represented as dashed lines, are the standard error

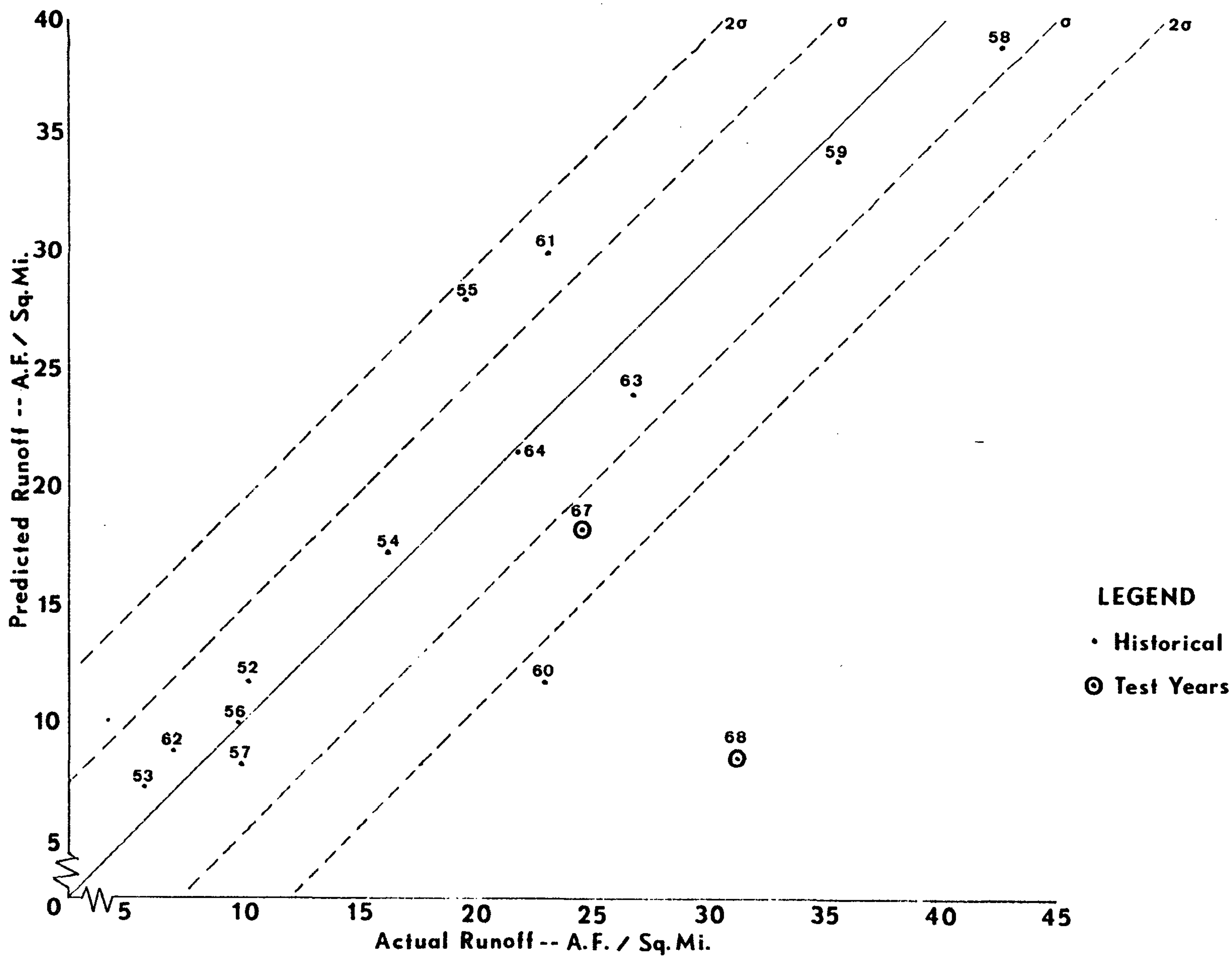
AWRR Sierra Cumulus

PREDICTED vs ACTUAL RUNOFF BY STORM



AWRR Sierra Cumulus

PREDICTED vs ACTUAL RUNOFF BY YEAR



of estimate, one and two-fold. Figure 3 presents the records on a storm basis while Figure 4 presents those results on an annual basis. On both sets of data the results are encouraging. The three storms lying outside 2σ were those which lasted several days. It is significant that the occurrence on July 9th, 1968 produced an observed/predicted ratio greater than any occurrence in the thirteen-year base period. Similarly, as can be noted on Figure 4, the observed/predicted ratio is greater for the yearly total 1968 than any other year.Combining the results of 1968 and 1969 yields an associated t-statistic indicating that there is a 0.5% chance that the null hypothesis - "no increase in runoff occurred from cloud seeding" - was incorrectly rejected.

SUMMARY AND CONCLUSIONS

The original project design anticipated a two year period in which an average of seven storm occurrences per year would be experienced. Because of a combination of circumstances, a total of only seven storm occurrences were recorded during the summers of 1968 and 1969. However, the apparent increases were considerably greater than anticipated in the project design. As a consequence, the experiments were able to demonstrate that there is considerable likelihood that weather modification was responsible for increasing runoff during summer-time storms over the Sierra, and that these increases can be of the order of 100 to 250 percent. It is equally apparent that significant increases are related to the duration of the storm. The short-duration storms (1 - 2 days), and at least the earlier portions of the longer storms, most probably succeed only in priming the basin and produce very little runoff.

Since the characteristics of a watershed determines its efficiency and thus the amount of runoff produced, the percentage increases experienced on Bear Creek may not necessarily be realized on another watershed. However, there appears to be little question that the target watershed produced more water from summer storms during the two year study period than might be reasonably anticipated based upon results from the thirteen year base period of analysis and the statistical evaluation tool used. It can only be assumed that the cloud seeding activities on the Bear Creek watershed during the project period were directly responsible for this increase in runoff.

It should be mentioned that the period before the end of the snowmelt offers a greater potential for supplemental water than the period considered in this study. In addition to the storms occurring early in the season being greater water producers, the basin losses are considerably less. However, because of the difficulties involved in separating the hydrograph, operations were restricted to that period after the end of the snowmelt. Nevertheless, it is generally felt that cloud seeding activities in the early season could yield even more economically beneficial results.

ACKNOWLEDGEMENTS

The preceding discussions of the hydrologic evaluation were derived from a series of in-house reports by Mr. Jack F. Hannaford of Sierra Hydrotech.

REFERENCES

Hannaford, J. F. and M. C. Williams, "Summer Hydrology of the High Sierra", Atmospheric Water Resources Research, Fresno State College Foundation, U. S. Bureau of REclamation Contract No. 14-06-D-5819, April 1967.

Williams, M. C. and D. E. Lehrman, "Sierra Cumulus", Atmospheric Water Resources Research, Fresno State College Foundation, U. S. Bureau of Reclamation Contract No. 14-06-D-6592, April 1970.

U. S. Geological Survey, "Surface Water Supply of the United States", Vols. 11-12, U. S. Government Printing Office, 1922-1964.

Hannaford, J. F., "Sierra Cumulus Evaluation Technique", Office Report to Atmospheric Water Resources Research, Fresno State College Foundation, Fresno, California, September 1967.

ANALYSIS OF FOUR WINTER STORMS

D. A. Griffith, G. L. Smith,
D. E. Lehrman, J. R. Vowell

Fresno State College Foundation
Atmospheric Water Resources Research

I INTRODUCTION

Research conducted by the Atmospheric Water Resources Research (AWRR), Fresno State College Foundation, is concerned with weather modification activities in winter-time storms occurring over the Stanislaus and Mokelumne River drainages located in the Central Sierra Nevada. A description of the project and its goals can be found in the Annual Report No. 2 submitted by AWRR to the U. S. Department of the Interior, Bureau of Reclamation, on September 1, 1970[1], under Contract No. 14-06-D-6592.

Analysis of data collected during field operations occupies a key position in this overall project. It is the goal in post-season analysis to obtain a thorough understanding of the precipitation processes in winter storms which cross the Sierra Nevada barrier in an aerological sense. Once these processes are understood, the overall goal of efficient and effective weather modification activities will be much closer to realization.

Data collected on four storm periods observed during the 1969-70 winter season were selected for detailed analysis. Each of these storm periods represented a different type of storm as classified by a storm typing system developed by AWRR[2]. The data collection, storm analysis, and storm typing work was all accomplished for the U. S. Department of the Interior, Bureau of Reclamation under Contract No. 14-06-D-6592.

II. DATA USED

Various sources of data were used in these analyses. Teletype data supplied by the National Weather Service served as the basic network to which special observations, unpublished data, and data collected by AWRR and its cooperators as a part of the research activities were added to give as complete a coverage as possible.

Some of these data required computer reduction and/or computation. This was especially true of the rawinsonde data collected by AWRR and the Fallon Naval Air Station in Nevada. Computer programs developed by AWRR were used to obtain potential temperature (θ) and equivalent potential temperature (θ_e) values for each 50 mb level.

The primary source of supplemental data was derived from upper-air stations manned by AWRR which were located at the Amador County Airport near Jackson, California, the Mt. Reba Ski Area, and the Fallon Naval Air Station. This third site was operated by Navy personnel, and was located near Fallon, Nevada. Figure 1 gives locations of these sites. Observations from these stations were made on a three-hourly basis during storms such that every fourth rawinsonde would coincide with the Weather Services' synoptic observation times of 0400 and 1600 PST.

III TYPES OF ANALYSIS

Several different types of analysis were attempted on each storm. One of the first of these was the analysis of three hourly, surface sectionals (California and Nevada) covering the storm period. These portrayals were informative in giving a general understanding of the storm pattern. However, frontal systems were often difficult to locate in the Sacramento and San Joaquin Valleys at the surface, since these surfaces tend to ride over the trapped air in these valleys as they move through the area.

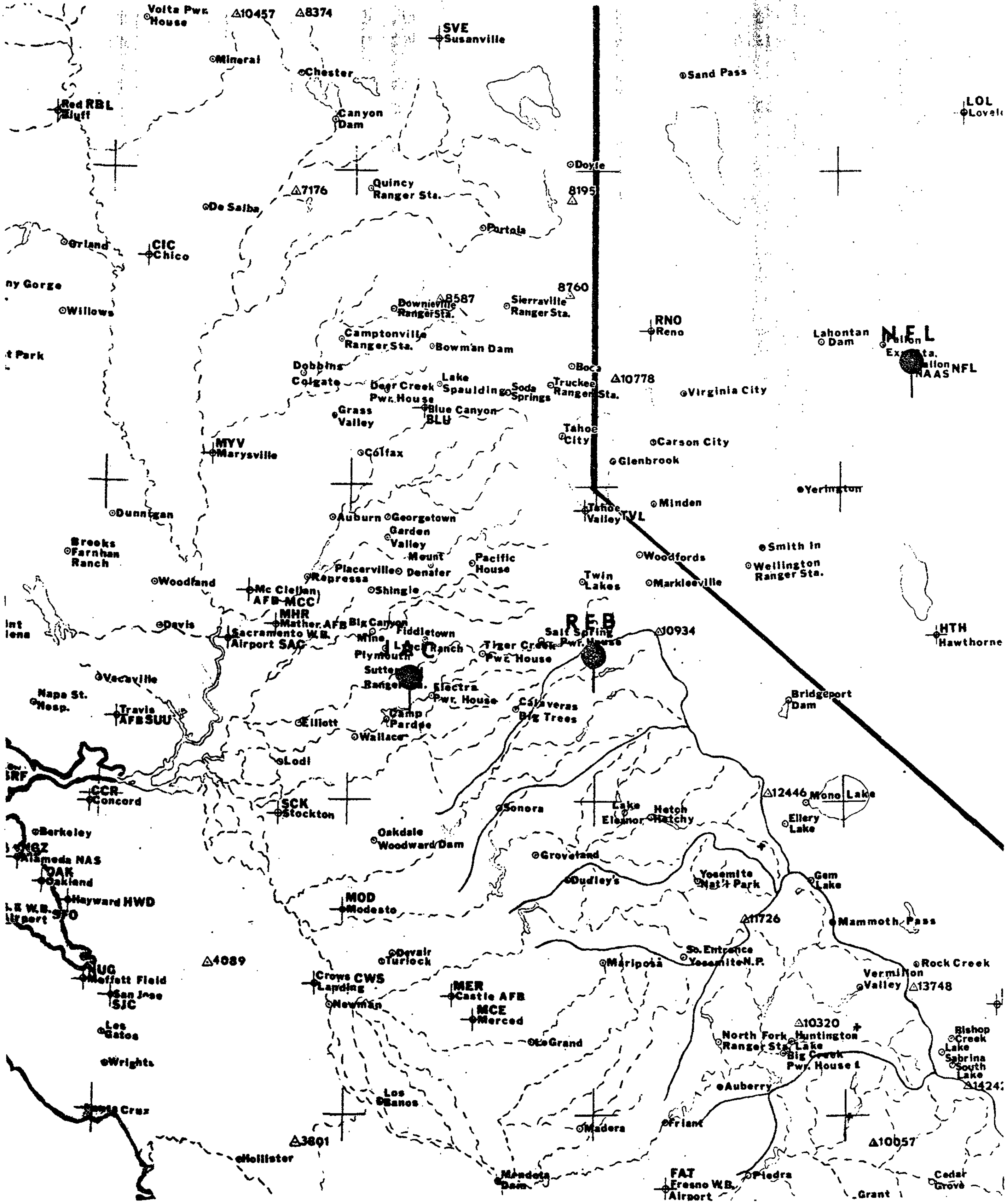
The next step was the analysis of time cross-sections for the Amador County Airport and/or Mt. Reba Ski Area observations. These analyses were based on all upper-air soundings for the total period of the storm. The data were plotted at 50 mb intervals and consisted of θ, θ_e, wind speed and direction. This type of analysis provided a more complete understanding of the behavior of these parameters during the storm. These analyses also enabled the analyst to select the most interesting time period for more detailed study in the form of space cross-sections.

Space cross-sections were the next type of analysis to be completed. There were several possible profiles for studying these cross-sections (i.e., Oakland-Jackson-Mt. Reba-Fallon NAS-Ely; Medford-Jackson-San Nicolas Island, etc.). The most useful ones proved to be from Oakland to Ely, Nevada and Oakland to Winnemucca since these lines represent traverses of the orographic barrier. The same information was plotted on these cross-sections as that plotted on the time cross-sections. In addition, surface observations from stations along the cross-section were plotted on these charts. A terrain profile was added to these charts to provide a visual representation of the orographic barrier. Several interesting features were observed on this type of analysis but these features will be discussed in Section IV.

Constant pressure maps (for 850, 700, 600, 500, 400 and 400 mb) covering the western United States were plotted and analyzed for θ and θ_e. This step was required in order to assure consistency between the various analyses. Thermal winds were checked along with the vertical stacking of the analyses to acquire this consistency. These analyses were also helpful in providing an idea of the vertical structure of the atmosphere between points of observation.

Sectional (California and Nevada) analyses of constant pressure

Figure 1. - AWRR Radiosonde Network



for θ and θ_e at 850, 700, 600, 500, 400 and 300 mb were then completed, using the preceding analysis to determine boundary conditions.

A separate isohyetal analysis of storm precipitation amounts was constructed for each of these storm periods.

An example of each type of analysis (except the western United States sectionals) for one storm period (January 13-14, 1970) have been included in Appendix A.

IV RESULTS

Results from the analysis of one of these storm periods is given below;

Storm No. 7, January 13-14, 1970 (Seed)

1. General Synoptic Description

This storm was classified according to "Storm Typing - California," Technical Report No. 10, as a "jet stream" (J) despite the fact that it did have some characteristics of a "southwest" (SW) near the end of the storm. At the upper levels, a closed low was embedded north-northeast of Hawaii near 40 N in a trough extending from the Gulf of Alaska to Hawaii. A weak flat ridge covered the western United States and extreme eastern Pacific Ocean. Disturbances moved out of the southern portion of the trough east-northeastward to California at about 36 hour intervals. These storms were relatively warm and quite moist due to their low latitude origin.

2. Sub-Synoptic Scale

A warm and very moist air mass, riding over the replacing a cooler air mass, moved over the project area with light to moderate precipitation starting near 1700 PST on 13 January 1970. An intense squall line with north-south orientation moved eastward over Jackson, California in the warm over-riding airmass near midnight producing thundershowers and moderate to heavy precipitation until approximately 0400 PST on the 14th. This line appears to have moved over Mt. Reba approximately one hour after passing Jackson.

The initial activity was followed by a short period of heavy precipitation (0700 - 1000 PST) caused by the intrusion of a cold air mass which first appeared at about the 600 mb level as a cold, dry air mass advected over warm, moist air on both the θ and θ_e vertical time cross-section (Figure A, Appendix) and the 0400 PST (14 January) vertical space cross-section (Figure A, Appendix) from OAK across the project area. The vertical time cross-section (θ, θ_e) analyzed for Jackson also shows the subsequent overturning of the air mass due to the instability created by the cold air advection followed by the intrusion of the cold air at the surface.

3. Seeding Potential

The layer of air that could be most affected by silver iodide nuclei for precipitation increase was between approximately 665 mb and 550 mb (-5°C to -14°C), or roughly 11,500 to 16,000 feet msl. The mean flow in that layer during the periods considered best for precipitation increase was approximately 245° at 35 to 40 knots. The airmass over the high altitude generator sites between the surface and the effective level was best represented by the Mt. Reba radiosondes observations. This layer was between 8,000 and 11,500 feet MSL or approximately 3,500 feet thick.

Stability, as indicated by θ_e during the period between approximately 2000 PST on the 13th and 0800 PST on the 14th, ranged between 1°K and 4°K (differences between θ_e values at the surface and the seeding level) for the 3,500 foot layer below the effective layer. The mean flow in that layer was approximately 230° at 35 knots. Assuming that lift provided by orographic conditions, convective cells and by the approach of the cold front was 100 to 1,000 feet per minute, the silver iodide nuclei would reach the effective temperature region approximately 20 minutes before moving out of the project area. In some cases, nuclei would not have reached the effective zone before passing outside the project area. The 20-minute period in the effective zone appears to be a marginal amount of time for nucleation, then fallout to occur in time to affect the target area.

A period occurred, after the beginning of the cold air advection aloft between 0700 and 1400 PST on the 14th, when unstable conditions existed along with reduced wind velocities such that the high elevation generator sites could have been used effectively. Better results could have been expected from the high altitude generators under these conditions.

Stabilities in the layer between the low-level generator sites and the effective nucleation level were considerably less favorable than those mentioned for the high altitude sites. Stabilities of 7°K to 9°K (θ_e differences between the surface and the seeding level) were present during the precipitation periods until the cold airmass had pushed over the area. Apparently only the activity associated with the cold air advection aloft (and cold air intrusion at the surface) could have been increased by the low-level ground generators. Overall, it is estimated that no more than approximately one-third of the precipitation volume from the storm could have been enhanced by seeding with ground generators. If the ground seeding had been conducted with maximum efficiency, the benefits would have been quite small when compared to potential benefits from aircraft seeding on a storm of this type.

Another interesting point noted in this study, which has been noted in other instances, is the possible existence of a permanent atmospheric pressure ridge (during storm periods) over the Sierra Nevada. This appears to be especially true over the windward slopes

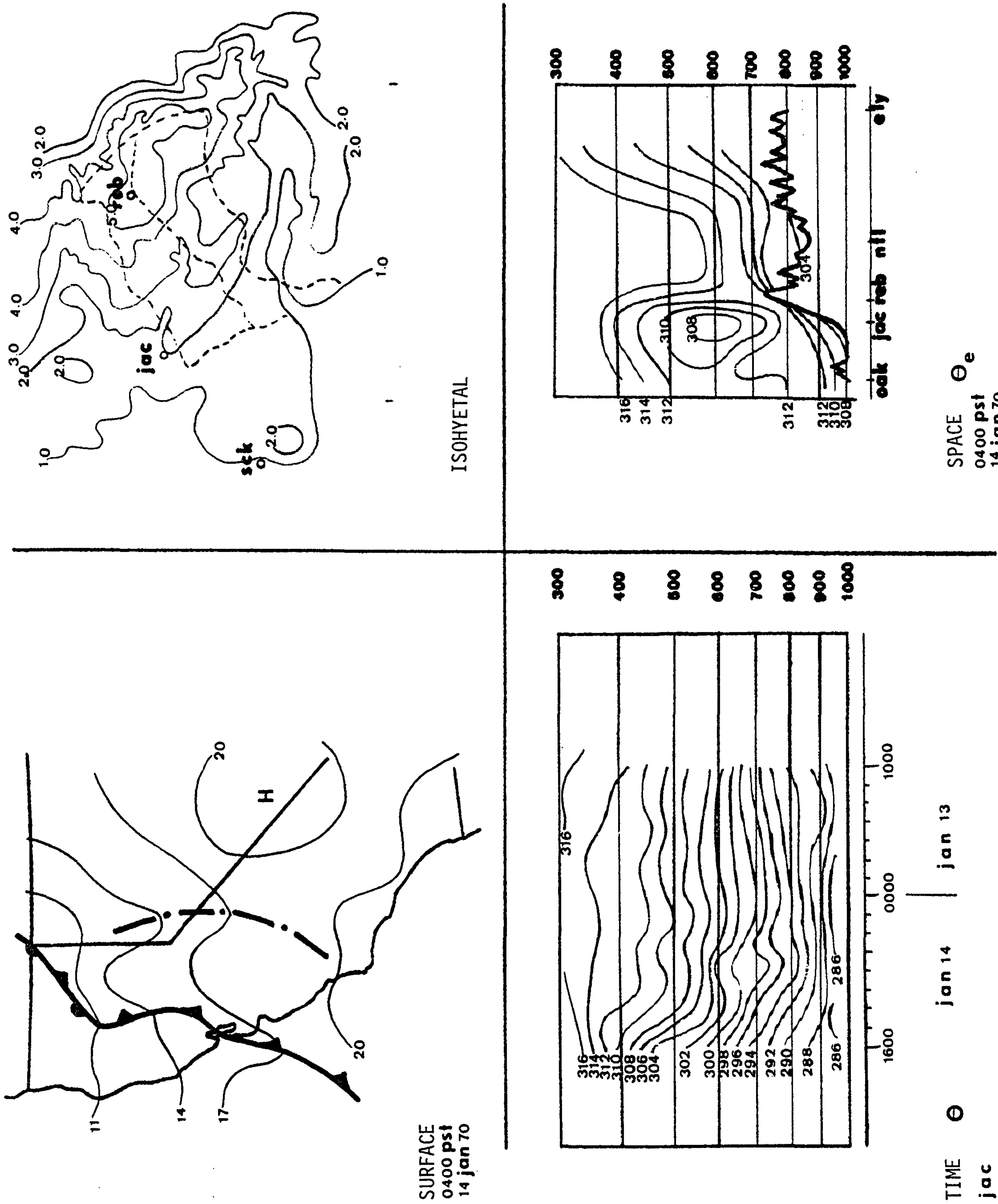


Figure A. Surface, Isohyetal, Time Cross-Section and Space Cross-Section Analyses January 13-14, 1970

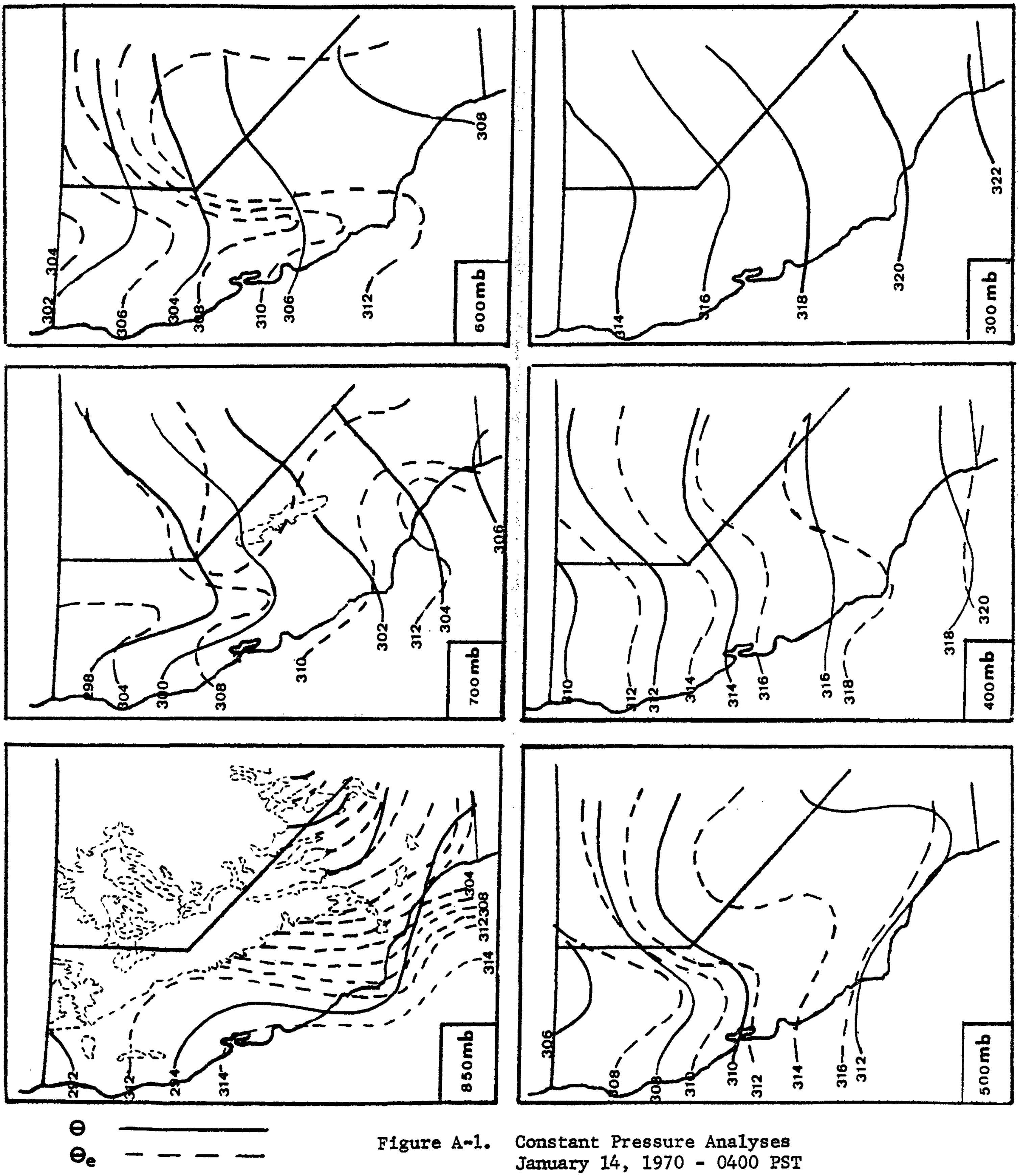


Figure A-1. Constant Pressure Analyses
January 14, 1970 - 0400 PST

during this study period. The ridge remains quite well defined even with the passage of the major trough associated with this storm, especially at the surface, 850 mb, and 700 mb levels but becomes practically nonexistent at the 500 mb level as the major trough passes over the area.

4. Precipitation

Precipitation from this storm was quite heavy. The foothills received from 1.5 to 2.5 inches of precipitation. The project area, which begins at the 5,000 foot MSL elevation, received three to four inches of precipitation during the period from 1600 PST January 13 to 1600 PST on January 14th.

VI CONCLUSIONS AND RECOMMENDATIONS

It is felt that these types of analyses are quite useful in obtaining a more thorough understanding of the winter storm systems which pass over the Sierra Nevada barrier. Knowledge of this type is a basic requirement for establishing a successful weather modification capability in this area.

These types of analyses will, therefore, be continued with a long-range goal of providing detailed understanding of these storms which will eventually lead to the development of a storm climatology for each type of storm observed. These analyses may very well establish the need to revise certain weather types, or else combine them to form a new system of typing the winter storms. The goal in this respect will be to formulate the minimum number of distinct weather types which are representative of all storms observed in the Sierra.

In the repetition of these types of analyses, other types of analyses techniques may be suggested by the previous studies or improvements in the same types of analyses may become evident. Evolution of this type should lead to the development of those analyses most suited to AWRR's requirements.

Completion of these analyses represents a first step in the understanding of the winter-time storm systems associated with the central Sierra Nevada. This understanding will be used to develop the optimized procedures and decision-making criteria by which future operational programs are conducted.

REFERENCES

Annual Report No. 2, 1 September 1970, Atmospheric Water Resources Research, Fresno State College Foundation, under Bureau of Reclamation Contract No. 14-06-D-6592.

Williams, M.C., D.A. Griffith, G.L. Smith, Storm Typing-California, Technical Report No. 10 AWRR, Fresno State College Foundation, under Bureau of Reclamation Contract No. 14-06-D-6592.

EVIDENCE OF MICROSTABILITY IN COLD OROGRAPHIC CLOUDS

Charles F. Chappell

Utah Water Research Laboratory
Utah State University
Logan, Utah

I INTRODUCTION

Bergeron (1933) suggested that ice crystals growing in supercooled clouds at the expense of evaporating cloud droplets might explain the genesis of most raindrops. He postulated the presence of ice nuclei, or sublimation nuclei in the atmosphere that are thermodynamically activated and then grow by vapor deposition into ice crystals. Measurements of the number of these primary ice nuclei indicate significant spatial and temporal variations in their concentrations (Kline, 1963). Moreover, the number of ice nuclei activating in the atmosphere is strongly dependent upon temperature. In general, most observations have indicated crudely an exponential rise in ice nuclei counts with decreasing temperature (Fletcher, 1962). Chappell (1970) found that a mean activation spectrum for the Climax, Colorado area on non-seeded experimental days was given by $N = (2.575)\ (10^{-4})\ \exp\ (-0.435T)$, where T is in degrees Centigrade and N is the number of effective ice nuclei per liter.

This expression indicates that the number of effective ice nuclei increases tenfold as environmental temperature decreases by about 5C. If ice nuclei are responsible for overcoming cloud microstability and releasing precipitation from cold orographic clouds, the probability of precipitation occurrence should be dependent upon the temperature of the cloud system. The design employed in the Climax, Colorado, experiment affords an excellent opportunity to investigate this possibility.

2. CLIMAX EXPERIMENTAL DESIGN

A study of the precipitation processes that accompany central Colorado mountain snowfall was begun in 1959 at Colorado State University. The Climax experimental design has been discussed by Grant (1960), Grant and Schleusener (1961), and Grant and Mielke (1967). Briefly summarized, the experiment was randomized with a 24-hour sampling unit. The criteria of an experimental day was that at least .01 inches of precipitation be forecasted during a 24-hour sampling unit at Leadville, Colorado, accompanied by a 500 mb wind direction between 210 degrees and 360 degrees, inclusive. This forecast was prepared by the United States Weather Bureau in Denver. The forecasters had no knowledge of the seeding decision. Six Colorado State University modified skyfire, needle-type ground generators were used for seeding employing a seeding rate of about 20 grams of AgI per hour. Sixty-five snowfall observation sites in the area comprised the precipitation measuring network and were read on a daily basis.

From the beginning of the Climax experiment in 1960 to the end of the initial experimental mode on January 31, 1970, there were 321 non-seeded experimental days with the prescribed 500 mb wind direction (the original Climax experimental design has since been altered). This sample is employed in the present study.

3. ANALYSIS PROCEDURES

The 321 non-seeded experimental days were investigated to determine whether snowfall occurred anywhere in the target area during the designated 24-hour sampling periods. If not, a 24-hour sampling period was considered to be a zero precipitation day. Experimental days, experimental precipitation days, and experimental zero precipitation days were then assembled into classes according to the concurrent 500 mb temperature. The percentage of experimental precipitation days to be experimental days was also computed for each temperature class.

4. RESULTS

Table 1 shows the distribution of experimental days, experimental precipitation days and experimental zero precipitation days distributed with the concurrent 500 mb temperature. The percentage of experimental precipitation days to experimental days shown in Table 1 is really a measure of the accuracy in forecasting measurable precipitation by present day forecasting techniques. As pointed out previously, experimental days of the Climax experiment were based upon a forecast of measurable precipitation at Leadville, Colorado, which lies within the target area.

Table 1. Distribution of experimental days, experimental precipitation days and experimental zero precipitation days with the concurrent 500 mb temperature for 321 non-seeded events of the Climax experiment (1960-1970).

Temperature Category (°C)	No. of Exp. Days	No. of Exp. Precip. Days	No. of Exp. Zero Days	Percentage of Exp. Precip. Days to Exp. Days
-40 to -36	2	2	0	100.0
-35 to -31	17	16	1	94.2
-30 to -26	64	60	4	93.8
-25 to -21	129	103	26	79.8
-20 to -16	88	50	38	56.8
-15 to -11	21	7	14	33.3
TOTAL	321	238	83	73.9

The decline in forecast accuracy as cloud temperatures become warmer is striking. The Weather Bureau Forecast Center in Denver was over 93 percent accurate in their forecasts of measurable precipitation when 500 mb temperatures were -26C and colder. Forecast accuracy declined steadily as 500 mb temperatures became warmer reaching a low of 33.3 percent for the warmest cloud systems. It seems reasonable to assume that the forecasters, using the usual forecast methods and procedures, found these events to be equally likely to produce measurable precipitation. It follows then, that the cause of the decline in forecast accuracy is probably due to the neglect of an important and temperature dependent factor. It is believed that current forecast procedures attempt to estimate condensation rates, but exclude ice growth rates from consideration, and the usual assumption that condensate will automatically be converted to precipitation does not hold for these warmer orographic cloud systems.

5. DISCUSSION AND CONCLUSIONS

Table 1 demonstrates dramatically the dependency of wintertime precipitation occurrence in the Climax area upon cloud system temperatures. This in turn, suggests that the concentration of effective ice nuclei in the cloud is vital to securing a precipitation release from these cloud systems. As cloud temperatures become warmer, the probability decreases that cloud microstability will be overcome naturally and a precipitation release obtained.

It is of considerable interest that Grant and Mielke (1967) and Chappell (1970) found slight negative seeding effects during the Climax experiment for events with 500 mb temperatures -26C and colder. A very small positive seeding effect was obtained for events with 500 mb temperatures -26C thru -21C, and large snowfall increases were observed when events with 500 mb temperatures -20C and warmer were seeded. These results suggest that seeding during the Climax experiment was effective in overcoming cloud microstability inherent in the warmer cloud systems. The dominant effect of seeding under these conditions may be the initiation of precipitation and consequently, the increase of its duration. If this is the case, emphasis should be placed on "cloud" seeding rather than on "precipitation" seeding for best results.

It also appears that considerable improvement in forecasting precipitation from cold orographic cloud systems could be brought about by employing current cloud physics knowledge to the problem. Attention needs to be directed toward estimating the strength of the ice growth process present in the cloud system, rather than limiting consideration to the condensation process alone.

ACKNOWLEDGEMENTS

This research was supported by the Atmospheric Sciences Section, National Science Foundation, under Grant GA-1553. Special acknowledgement is due the Denver Office of the U. S. Weather Bureau for its efforts in supplying special forecasts to define experimental days; the High Altitude Observatory, and the Climax Molybdenum Company for their extensive efforts in assisting with observations in the early stages of the experiment and their continuing assistance with facilities.

REFERENCES

Bergeron, T., 1933: On the physics of clouds and precipitation. Proc. 5th Assembly U.G.G.I., Lisbon, 156-178.

Chappell, C. F., 1970: Modification of cold orographic clouds. Ph. D. Dissertation, Dept. of Atmos. Sci., Colorado State University, Fort Collins, 196 pp.

Fletcher, N. H., 1962: The Physics of Rain Clouds., Cambridge University Press, 122-127.

Grant, L. O., 1960: Colorado State University Climax study of the effect of cloud seeding on snowfall, General Information Booklet supplied to all participants (February).

Grant, L. O. and R. A. Schleusener, 1961: Snowfall and snowfall accumulation near Climax, Colorado. Proc. 29th Annual Meeting Western Snow Conf., Spokane, 53-64.

Grant, L. O. and P. W. Mielke, Jr., 1967: A randomized cloud seeding experiment at Climax, Colorado, 1960-65. Proc. of the Fifth Berkeley Symposium on Math. Statis. and Prob., 5, 115-131.

Kline, D., 1963: Evidence of geographical differences in ice nuclei concentration. Mon. Wea. Rev., 91, 681-686.

A COMPUTERIZED METHOD OF TELEMETERED PRECIPITATION DATA QUALITY CONTROL

George W. Reynolds
Ronald H. Campbell

Utah State University
Logan, Utah

INTRODUCTION

The experimental design of the Wasatch Weather Modification Project* provided for the evaluation of cloud seeding activities through the comparison of precipitation patterns for seeded and unseeded periods. The primary source of data is the Utah State University telemetered precipitation data network (Israelson and Griffin, 1969).

Data quality control has been a consideration since the inception of the Project, and corrective steps have been taken to eliminate inaccuracies in the reported data. For example, data from the 1968-69 experimental season indicated that the springs of this weighing gage system were responding to temperature changes. Consequently, the transducers were all installed below ground level, where temperature changes are much slower and smaller. Two experimental units have been installed for the 1970-71 experimental season which have a protective shell around the collection cans. This shell will shield these cans from the wind and prevent the buildup of rime and snow on the sides of the cans which are to be weighed. Precipitation values from these units will be compared with those from the regular network at the same rate in order to evaluate this proposed improvement.

There are several possible factors which may cause reported values to be somewhat in error: wind forces transmitted to the springs; vibratory motions created by the wind; snow and ice temporarily sticking to the sides of collection cans; radio interference; friction between the cans and guides; etc.

An examination of numerous plots has strongly indicated that the telemetered precipitation data can be improved by thoughtful editing procedures. Since 75-100,000 data points are expected each year, a manageable data editing procedure must be computer compatible.

As a first step in the development of a computer editing procedure, the data from the 1968-69 experimental season were edited manually. Possible effects of concurrent environmental conditions were considered in an effort to define the causes of apparently anomalous precipitation data points. The ensuing information provided the basis for some of the corrective actions that have been taken. The reasoning behind each point

*This Project is funded by the Division of Atmospheric Water Resources Management of the Bureau of Reclamation under Contract #14-06-D-6820.

adjustment or discard was recorded, and a set of editing rules was eventually developed from these notes. These rules have been used as a guide in the development of a computer program which edits the raw data gives an accounting of the editing requirements, and extracts half-hourly values from the edited data. This program is operationa.

CATEGORIES OF QUESTIONABLE POINTS

Most questionable data points fall into three general categories.

1. Precipitation values so unreasonable that they are obviously unacceptable. These data points are called outliers, and the program simply discards them. Outliers seem to occur randomly and may be in either a positive or negative direction with respect to the preceding point.

2. Precipitation values exhibiting sudden, difficult-to-explain relatively small departures on the cumulative precipitation curve. These situations are usually the results of reports of negative precipitation rates between successive points. For these data points it is sometimes necessary to choose which point or points will be discarded or adjusted. The editing program must decide which of a series of points are the most meaningful for defining the true precipitation tendency. These departures seem to occur randomly and are referred to as "noise points".

3. Several precipitation values in succession indicating a negative trend in precipitation amounts. These apparent trends cannot be rationalized to be the results of evaporative losses or leaking cans. One possible explanation is the temporary accumulation of ice and/or snow on the outsides of the precipitation cans. Since this is a weighing system, both the accumulation on the outside and the subsequent melting would be reflected in the measurements. Temperature conditions observed during these negative trends in general, support this concept. This phenomenon occurs under a limited set of meteorological conditions. On one occasion, snow and ice scraped off of the outside of a 36-inch collection can and a 70" collection can yielded 0.3 inch and 1.7 inches of water equivalent respectively. Both cans had the standard 8" orifice. The 36" capacity can was straightsided and the 70" capacity can had sloping sides between the top and the storage portions.

THE EDITING PROGRAM

In general the data are handled in 10-12 hour blocks which coincide with experimental periods (Utah Water Research Laboratory, 1970). The telemetered precipitation data are reported by the Automatic Readout Console as a printout of electromagnetic periods on adding machine tape (Israelson and Griffin, 1969). There are 2 or 3 reports per station

/hour. The electromagnetic values are punched and converted to the inches of water equivalent of the contents in the can.* The basic data for the editing program are the precipitation values, but the data conversion, editing, and subsequent extraction of half-hourly amounts are accomplished in a continuous operation. The editing is accomplished in the following manner:

1. The data are processed one station at a time. At least eight data points are required for the application of the editing program.

2. The data are first checked for obvious key punch errors in the identification code, and appropriate action is taken. For example, data points are rejected if the values lie outside of the selected time period.

3. The data are ordered chronologically and converted to water content values. All negative water content values, indicating that the can is less than empty, are discarded.

4. The next step is to establish the validity of the first data point for the experimental period under study. "b_1" is defined as the precipitation rate (inches/hour) between the first and second points, and "b_2" as the rate between the second and third points. The first data point is temporarily accepted if:

$$b_1 < -1 \text{ and } b_2 > +1$$

or $$-1 < b_1 < +1, \text{ with no restrictions on } b_2$$

or $$b_1 > +1 \text{ and } b_2 < -1$$

If the first data point is rejected, the proce-s is repeated for the next points in succession until an acceptable starting point is obtained.

5. If the magnitude of the precipitation rate between successive points is greater than 0.2 inch per hour, the second of these two points is submitted to an additional test. Terms are defined as follows:

P_i is the point under test.

P_{i+1}, P_{i+2}, etc. are the successive precipitation points immediately following P_i.

* At the start of the season antifreeze equivalent to several inches of precipitation is put into each can after calibration.

P_{i-2}, P_{i-1}, etc. are the successive points immediately preceding P_i.

$$A = \frac{(P_{i-4} + P_{i-3} + P_{i-2} + P_{i-1})}{4}$$

$$B = \frac{(P_{i+1} + P_{i+2} + P_{i+3} + P_{i+4})}{4}$$

P_i is retained if $A \leq P_i \leq B$. Otherwise, the point is called an "outlier" and discarded.

6. In this program, by definition, a negative trend occurs if at least two successive data points exceed each of the three immediately following data points by more than 0.02 inch. As previously indicated, such a trend can be the result of a gradual loss of snow and ice which has accumulated on the outside of the collection cans. In such cases the values preceding the negative trend would be too high. In this event, a new point (M' in Figure 1) is generated which has a precipitation value equal to the lowest of all of the points of the negative trend and the time recorded for the immediately preceding high peak value (M). The computer then goes back to the last point (E) which has a precipitation value lower than point M' and interpolates linearly with respect to time between E and M'. The precipitation amount is then assumed to be zero from point M' to point C. Thus, the precipitation rate is assumed to be constant from E to M', with no precipitation occurring between M' and C.

7. After these point adjustments, a new calculation is made to assure that the starting point is equal to or less than the average of the immediately following four points. If it is not, the starting point is rejected and the second point is tested, and so on until an acceptable new starting point is reached.

8. The final step in the editing procedure is an adjustment of "noise" points which are within .03 inch of a so-called "reasonable" value, with a reasonable value, P_i, being defined by $P_{i-1} \leq P_i \leq B$. If $(P_{i-1} - .03") \leq P_i \leq (B + .03")$, P_i is retained for adjustment. Otherwise it is discarded. A retained P_i is adjusted by adding or subtracting, as appropriate, the smallest number required for the point to assume a reasonable value. No adjustment is required if $P_{i-1} \leq P_i \leq B$.

9. The editing proceeds point by point from the first data point after chronological ordering. Once a point has been adjusted, the adjusted value is then used to test the following points as appropriate. Since outliers have already been discarded, and the average of the next four unedited points are used in the editing of the ith point, the effects of using a spurious point in the editing procedure is usually held to an acceptable level.

10. In the editing of the first four points at the beginning and the last four points at the end of a data set, the averages A and B, are computed on the basis of the number of points available.

Once the data editing is completed, half-hourly precipitation amounts are computed by linear interpolation. These half-hourly values are the basic precipitation data for seeding evaluation and precipitation pattern analyses. After computing these values for a given station, the computer goes to the next station's data and repeats the process.

RESULTS

Some of these steps are obviously somewhat arbitrary, and further refinements are being considered. However, the quality of the results thus far suggest that the cost-benefit ratio of additional refinement may not justify the effort. Figure 2 demonstrates the usual compatibility of the original and edited curves. These curves were plotted by the USU hybrid computer (E.A.I. 640).

Tables 1 and 2 are sample printouts from the computer. The accounting table is useful in judging the quality of the original data. In general, the data are not graphed unless the accounting statistics indicate that the plotting would yield significant additional information. However, the original and edited data are occasionally plotted on the same graph for an entire experimental period in order to maintain a check on the program.

SUMMARY

The USU computer editing procedure for telemetered precipitation data is operational and is believed to offer significant improvement over the original raw data. The program is checked occasionally for unexpected malfunctions. Further editing refinements are being considered. Data quality has been and continues to be a primary consideration in the design and operation of the field system.

ACKNOWLEDGEMENTS

Mr. William McNeill took the lead in analyzing data point anomaly--environmental condition relationships. He was assisted by Mrs. Janet Cleary, who has since applied these rules to data for several experimental periods.

REFERENCES

Israelson, C. Earl and Son L. Griffin. 1969. USU telemetering precipitation gage network. Technical Report PRWG-30-7, Utah Water Research Laboratory, Utah State University, Logan, Utah. 39 pp.

Utah Water Research Laboratory. 1970. The development and integration of cold cloud seeding technology for use in water management systems. Annual report. Utah State University, Logan, Utah. 26 pp.

- - - - - - - - - - -

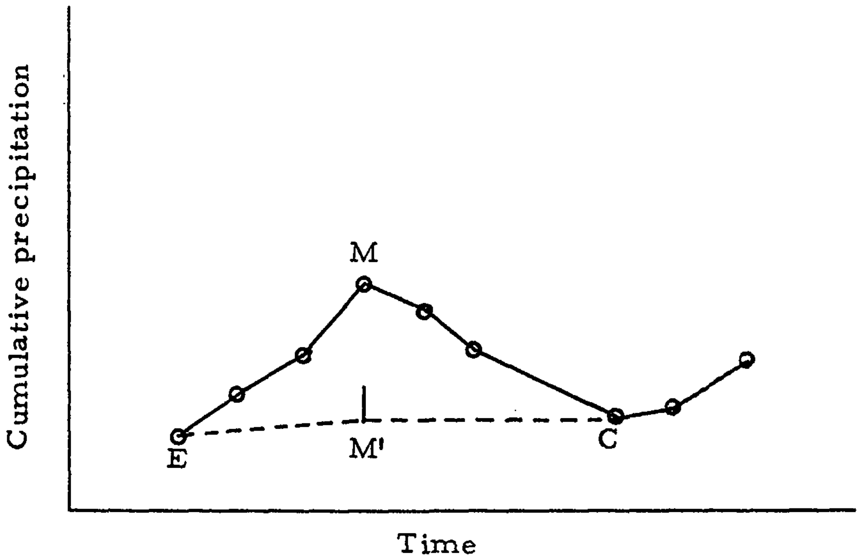


Figure 1. Example of a negative trend in the telemetered precipitation data.

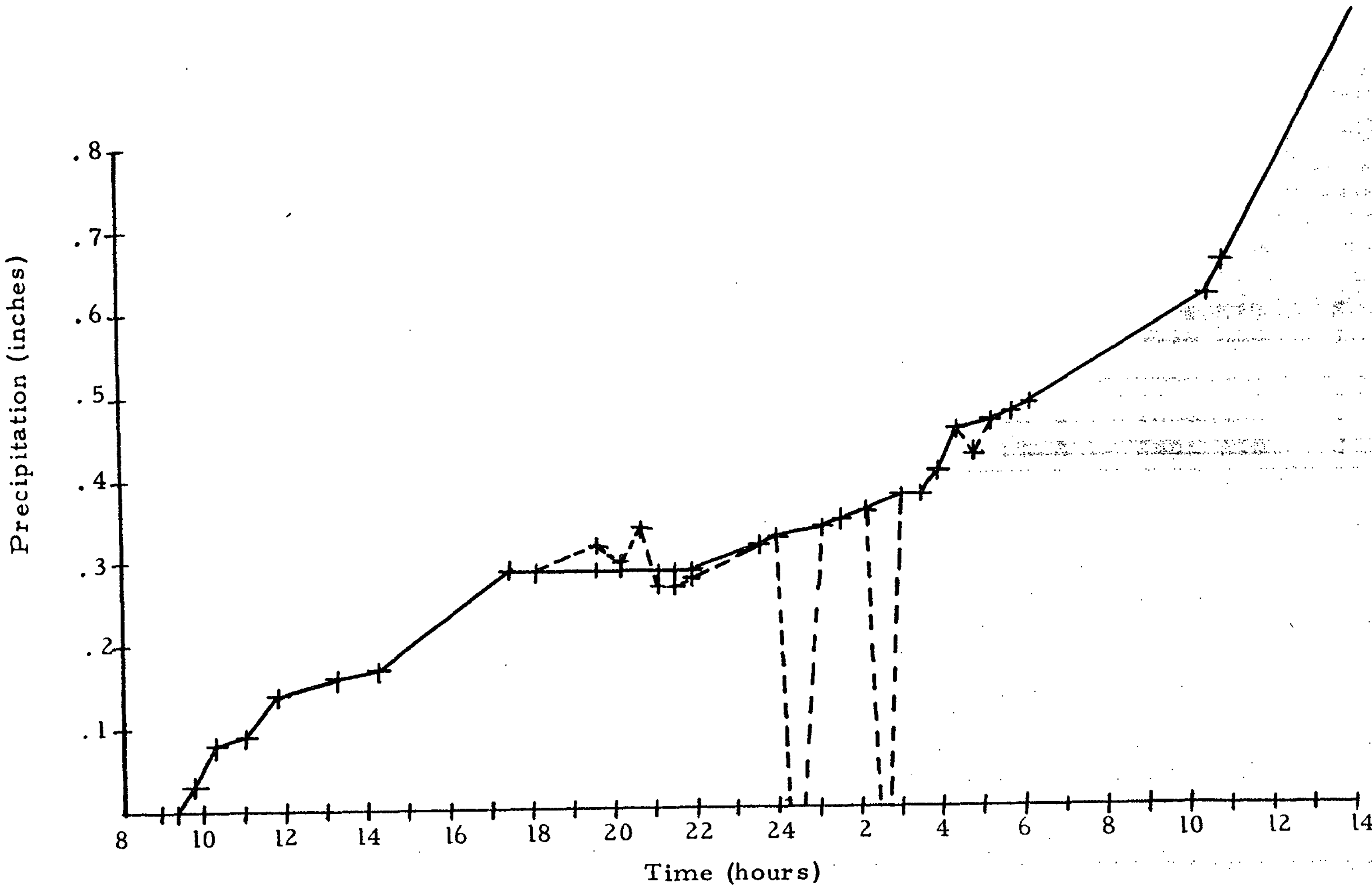


Figure 2. Unedited (-----) and computer edited (——) plots of cumulative precipitation. Monte Cristo (station 8003), January 11-12, 1971. (for portions with only one trace, the unedited and edited curves are identical)

```
************************************************************************
HALF HOURLY PRECIPITATION DATA FOR STATION  8003
ACCOUNTING DATA FOR STATION  8003
   34 POINTS ENTERED
    0 POINTS ADJUSTED FOR OVERSHOOT
    5 POINTS ADJUSTED FOR NOISE
    2 OUTLIERS REJECTED
    2 NOISE POINTS REJECTED
   25 ORIGINAL DATA POINTS LEFT UNTOUCHED
   30 POINTS ACCEPTED
    4 POINTS REJECTED
   88 PERCENT OF TOTAL POINTS INPUT WERE ACCEPTED
 800371 110 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0    2330 930    1
 800371 110 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 4    2330 930    2
 800371 111 3 1 3 2 1 1 0 1 1 2 2 1 2 2 2 0 0 0 0 0 0 0 0 0    2330 930    1
 800371 111 1 1 1 1 0 1 1 1 1 1 1 4 3 1 1 1 1 2 1 2 1 2 2 1    2330 930    2
 800371 112 3 5 4 5 5 4 5 4 5 5 4 5 5 4 5 4 5 4 0 0 0 0 0 0    2330 930    1
 800371 112 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0    2330 930    2

************************************************************************
```

Table 1. Accounting information for edited data set with half hourly interpolated precipitation amounts (hundredths of inches, two digits per data point).

Station 8001 Monte Cristo

	Original Data			Data After Editing		
1	2	3	4	5	6	7
1	10.973	26.72	.00	10.973	26.72	.00
2	10.991	26.75	.03	10.991	26.75	.03
3	11.012	26.80	.05	11.012	26.80	.05
4	11.043	26.81	.01	11.043	26.81	.01
5	11.075	26.86	.05	11.075	26.86	.05
6	11.134	26.88	.02	11.134	26.88	.02
7	11.176	26.89	.01	11.176	26.89	.01
8	11.308	27.01	.12	11.308	27.00	.11
9	11.337	27.01	.00	11.337	27.00	.00
10	11.398	27.04	.03	11.398	27.00	.00
11	11.425	27.02	-.02	11.425	27.00	.00
12	11.442	27.06	.04	11.460	27.01	.00
13	11.460	26.99	-.07	11.478	27.01	.00
14	11.478	26.99	.00	11.495	27.01	.00
15	11.495	27.00	.01	11.564	27.04	.03
16	11.564	27.04	.04	11.581	27.05	.01
17	11.581	27.05	.01	11.626	27.06	.01
18	11.599	21.48	-5.57	11.646	27.07	.01
19	11.626	27.06	5.58	11.671	27.08	.01
20	11.646	27.07	.01	11.706	27.10	.02
21	11.671	27.08	.01	11.724	27.10	.00
22	11.688	21.50	-5.58	11.744	27.13	.03
23	11.706	27.10	5.60	11.761	27.18	.05
24	11.724	27.10	.00	11.796	27.19	.01
25	11.744	27.13	.03	11.817	27.20	.01
26	11.761	27.18	.05	11.835	27.21	.01
27	11.779	27.15	-.03	12.011	27.34	.13
28	11.796	27.19	.04	12.029	27.38	.04
29	11.817	27.20	.01	12.345	28.08	.70
30	11.835	27.21	.01	12.375	28.14	.06
31	12.011	27.34	.13	12.375	28.14	.00
32	12.029	27.38	.04	12.375	28.14	.00
33	12.345	28.08	.70	12.375	28.14	.00
34	12.375	28.14	.06	12.375	28.14	.00

Table 2. Original and edited data sets. Column 1 is the data point number. Columns 2 and 5 give the times for data points in days. Columns 3 and 6 give the total water equivalence in the can. Columns 4 and 7 give changes in water content between successive readings.

WEATHER MODIFICATION - A FIRE CONTROL TOOL

James D. Harpster

Public Information Officer
Technical Services Branch

and

William J. Douglas

Research Meteorologist
Division of Atmospheric Water Resources Management
Bureau of Reclamation
United States Department of the Interior
Denver, Colorado

In years to come, it would be known as the "Year of the Big Burn". But there was little time to consider phrasemaking now, for Alaska was aflame. Towering columns of white smoke climbed the northern sky in every direction. Each day brought reports of new outbreaks as flames engulfed stands of virgin timber and surged mile after mile in areas so widespread that organized efforts at control were out of the question for all but the most threatening.

It was, in fact, the worst fire siege in Alaska's history. By conservative estimate, more than 3-1/2 million acres - an area larger than Delaware and Rhode Island combined - had been blackened.

Foresters blamed this 1969 holocuast on dry lightning storms, erratic winds, and the combination of an unprecedented dry season extending through two summers and the previous winter. In massive understatement, they termed it an extreme emergency.

Grasping at every means of halting the major fires, the Department of the Interior surveyed the capabilities of its various agencies and considered with new interest the Bureau of Reclamation and its Project Skywater - a scientific research program of cloud seeding to augment precipitation in water-short areas of the Western United States.

Could cloud seeding, the Department asked, be used to "douse" the worst of the Alaskan fires?

In posing the question, there was born the start of a new technology that may become a revolutionary weapon in the battle against forest fires that cause an estimated $50 million in timber losses each year in the United States alone. Two years now have passed since the concept, based upon up-to-date seeding techniques, was first voiced and scientists from Government and the private sector reserve judgement about its worth. There have been, in those two years, too few opportunities to test its application.

But from those opportunities has come evidence that cloud seeding may in fact provide a valuable and effective tool with which to combat this destructive member of mankind's natural enemies.

The proposal raised a wide range of questions to Bureau of Reclamation officials in that searing summer for Alaska. The Bureau's Project Skywater is directed to goals other than the suppression of fires.

But given certain conditions, Reclamation's scientists said, the scheme might be possible. Most important, there must be clouds of the proper character, with moisture and temperature regimes that meet established criteria, and in the proper place and at the proper time. Success would hinge on the willingness of nature to exactly aline a formidable number of variables, and the ability of the scientists to capitalize on those opportunities.

The Bureau of Land Management, which is responsible for administering much of the Alaskan interior, quickly arranged for a coordinated but brief field program in response to the Alaskan emergency. The Bureau of Reclamation would make available its expertise in precipitation management. Other participating agencies included the Environmental Science Service Administration (presently the National Oceanic and Atmospheric Administration), the U.S. Forest Service, the U.S. Air Force, and the University of Alaska. The overall effort was dubbed "Project MOD" (for modification), with headquarters at Fairbanks, Alaska.

Reclamation's representative on this task force responsible for the Weather Advisory Group of Project MOD was William J. Douglas of Denver, Research Meteorologist for the Division of Atmospheric Water Resources Management. Recruited as a technical consultant was Dr. Richard A. Schleusener, Director of the Institute of Atmospheric Sciences at South Dakota School of Mines and Technology at Rapid City. The institute has been conducting research for several years under contract to the Bureau of Reclamation's Project Skywater.

To locate and identify cloud cells suitable for seeding, the U.S. Air Force made available a radar unit whose data would be used by the Project MOD staff. The Forest Service provided an aircraft equipped with the infrared photo equipment whose pictures detect fire area "hot spots" invisible to the naked eye. The Bureau of Land Management offered a small jet aircraft for high-altitude observation, a helicopter, and a short-runway light aircraft. The University of Alaska extended the use of its computer center facilities for data analysis.

The seeding operations would be conducted under contract to BLM by a Boulder, Colorado weather research firm, EG&G, Inc., which had a twin-engine aircraft for field operations.

Once assembled at Fairbanks in mid-July, the immensity of the problem became clear to the Weather Advisory Group. There were, at this moment, 56 fires raging in the Alaskan interior. They had ravaged more than 3 million acres. Nearly 1,000 men were then engaged in battling 19 of the worst fires. The remaining 37 blazes were burning unchecked and untended.

The task force quickly set about establishing criteria for the program. Two scientific issues were paramount: Would seeding produce any rainfall that might reduce the intensity of the fires, and might there be any real potential for precipitation management in forest fire suppression?

The selection of fire areas to be treated would be governed in large part by the day-to-day availability of suitable clouds moving along the proper paths.

In the days that followed during the test period, the task force was destined to frustration. Only twice were there favorable days when clouds amenable to seeding appeared over fire areas, and even then, conditions were marginal. The first of these opportunities produced some dramatic results, however.

Near Fort Yukon, a series of fires was scrambling through heavy timber, two of them named "Fishhook" and "Rotten Fish Slough." The latter had blackened 15,000 acres and was moving on so broad a front that no effort had been made to put in ground crews.

On July 26, broken cumulus clouds with tops ranging to 15,500 feet blossomed upwind of Fort Yukon and seeding and observation aircraft went aloft. Because of the wind patterns that day, neither "Fishhook" nor "Rotten Fish Slough" was an intended target. The variables that alined themselves for the task force pointed to other fires instead.

Late in the afternoon, a test cloud was selected and seeded from above with pyrotechnic devices containing silver iodide. The cloud top grew, and a grey curtain of rain unfurled itself from the cloud base beginning some 30 minutes after seeding. From the air, it appeared the showers had some effect on scattered "hot spots" below.

The following day, however, there came a surprise. After the aircraft had left the seeding area, the test cloud had continued to intensify and had moved over a part of the Fishhook fire and directly over the Rotten Fish Slough fire. Bureau of Land Management fire control officials reported that: "The southern edge of the Fishhook fire had been 'knocked down' by rain and the Rotten Fish Slough fire had been hit by enough rain so that it was completely knocked out".

The Weather Advisory Group agreed: "The position of the (Rotten Fish Sough) fire with respect to the position of the seeded cloud upwind, and the timing of the observations with respect to the time of seeding indicate that the rain was probably initiated by the seeding. The effect of rain on this fire was very marked".

Even among those most enthusiastic over the prospects for Project MOD, the abbreviated 1969 effort suggested little more than that the scheme warranted further exploration. That recommendation of the Weather Advisory Group was made, and won prompt approval from the Department of the Interior. A second program was arranged accordingly for the 1970 summer, when fires continued to burn into the third year in the Alaskan interior.

The field site this time was Galena, a village some 225 miles west of Fairbanks, on the Yukon River. Accessible only by air and water, Galena offered housing, food, fuel facilities, and a paved runway.

The contractor to the Bureau of Land Management - Meteorology Research Inc., of Altadena, California, flew in a twin-engined seeding aircraft and a weather radar, and established a rawinsonde launch site to supplement meteorologic ascents from other Alaskan points. The radar was to be used for cloud targeting in a 50-mile radius and to assist in evaluating seeding results.

At Galena, too, the BLM stationed two helicopters, along with a twin-engine aircraft for low level observation. A high-level observation aircraft, pressurized and with turbo-prop engines, was based at Fairbanks. Ground crews comprised of BLM and Forest Service personnel were made available and equipped with portable gear that included rain gages, hand anemometers, sling psychrometers, time-lapse and still cameras.

As in the 1969 program, weather data were funneled to the headquarters facility at Fairbanks where the National Weather Service and the University of Alaska combined to provide teletype, facsimile, and computer support.

An important addition to the 1970 program, however, was the use of a computer model that projected minimum cloud parameters to indicate seeding potential. The cumulus model accepts radiosonde data and produces information to indicate cloud response to seeding - cloud top growth, expected increase in precipitation, and so on, as related to cloud diameter. The model was developed by Drs. Alan Weinstein and Larry Davis, then of Pennsylvania State University. It was based initially on theoretical equations, and later modified on the basis of field experience.

When the 1970 task force became operational on June 1, the fire situation in Alaska had greatly improved from the previous year. The winter had again been dry, but a number of fires had simply burned themselves out and others had been controlled by ground crews. In the vicinity of Galena, where fire incidence had been high in years previous, there were only occasional plumes of smoke on the mountainous horizon.

During June, there were 12 days with operational weather and cloud systems. But on 7 of those days, no fires burned anywhere in the area. Of the 5 days when operations were conducted, June 11 saw the most successful effort. A cloud about 8 miles upwind of the target fire was seeded with two vertical-fall silver iodide pyrotechnics, followed by one slow-burning flare.

The cloud top grew from 12,000 feet to 20,000 feet within 10 minutes, and observers both in the air and on the ground watched as generous showers developed. The raincloud passed directly over the fire, knocking it down and depositing - by official measurement - 0.25 inch of moisture.

July was the month of the "big bust," but again Nature conspired against the task force. A line of thunderstorms ripped through the area and, within 2 days, more than 100 new fire outbreaks were reported north and east of Galena. The largest of them destroyed some 60,000 acres. The others generally were controlled by ground crews before they reached such scale. Most were outside the radar's effective range, however, and too distant from the field site to permit use of the helicopters, whose 3-hour fuel supply gave them a safe, effective range of only 60-80 miles. The choppers were essential because only they could place crews on the ground to make the on-site observations and measurements required to scientifically document the results of seeding.

Only five operations were conducted during July, and the most successful was - appropriately - on July 4. Ninety-six fires were counted that day in the Galena area, not all of them within the operations area but nonetheless offering greater opportunities than at any previous time during the 1970 season.

Upwind of an active blaze near Daklia, the seeding aircraft treated three clouds and observed an 8,000-foot growth in the cloud tops. Precipitation followed, and tracked across the northern end of the fireline to suppress the blaze there. Returning to Galena from this mission, the seeding and observation aircraft spotted another blaze with suitable clouds immediately upwind. These, too, were seeded and responded dramatically. This fire, struck by rain, went from a fast-burning condition to smoke wisps within a few minutes.

During the 2-month operational period, the variables - the complex factors beyond scientific control that required alinement before any treatment could be considered - smiled on the task force on only three occasions.

These missions took place on 2 separate days and the resulting rains hit the target fires. Without exception, the results were impressive, the showers quenching active blazes that a few minutes before were roaring unchecked through stands of virgin timber.

By any scientific yardstick, however, the success ratio was unimpressive. A summary report on the 2 years' operations observed that: "Based on the low rate of success, it appears that cloud seeding <u>to a preselected target</u> is not a reliable fire suppression tool". (Emphasis added).

But to the two principal questions - that is, did seeding produce any rainfall that reduced the intensity of the fires, and is there real potential for weather modification in forest fire suppression - the task force members offered a qualified "yes".

The greatest prospect for this new scientific tool would appear to be in attacking what foresters call the "buildup index". This is a measure of cumulative dryness related to a regional fire hazard. At those times and in those places where a real forest fire hazard develops, selective cloud seeding might offer one means of reducing the potential for an outbreak of fires.

No decision has been made at this writing whether the fire suppression program will be extended, in Alaska or elsewhere. Should further testing and evaluation be recommended, however, members of the Weather Advisory Group concur that future programs should have greater flexibility than was possible in either the 1969 or 1970 Alaskan effort.

To permit as complete a test as possible, the aircraft-equipped seeding teams should have sufficient logistic support to attack fire situations anywhere they develop so long as suitable clouds are available. The experiences of Alaska indicate that too few opportunities are permitted when, even after the most careful consideration, a fixed base of operations is designated in a high fire incidence area; when limited-range radar support is provided; and when no more than a single seeding aircraft is available.

Under these conditions, far more suitable opportunities are destined to be lost than are identified and treated. Hence, a more widespread test on a broader front, over a longer time span, and with greater logistical support would appear to be desirable.

Whether in controlling the buildup index or in suppressing fires once they are in progress, the techniques of weather modification must be considered - at least at this point in their development - as a potential supplement to other fire control techniques already in use.

NON-SEEDABILITY OF HEXADECANOL-COATED MISTS

Thomas Y. Palmer

Forest Service, U.S. Department of Agriculture
Riverside, California

The effects of surface-active material on water drops and the resulting variation of fog and cloud parameters have recently been the subject of increased interest.[1-6]

In 1964, I demonstrated that super-cooled water fogs coated with the long-chain fatty alcohol hexadecanol were not seedable with dry ice at temperatures as low as -22°C.; in fact, splash marks were obtained from the impact of large drops (about 1 millimeter in diameter) on slides at this temperature.[7] But the results were never reported in literature.

Recently, I decided to do the experiment again--this time using recent developments in equipment for producing the coated fog drops. I found that a water drop fog coated with hexadecanol can be produced by boiling water covered with a layer of hexadecanol about one-half centimeter thick. The resulting drops are relatively small in radius (2.3 micrometers median size). The median size can be increased to 5-10 micrometers by causing the drops to condense in a region of electrical corona discharge.

The mixture of water and hexadecanol was either introduced directly into the cold box where condensation occurred, or coated fogs with larger drops were made by the electrical field process and then introduced into the cold chamber. Uncoated fogs were obtained by introducing steam from boiling distilled water into the cold box. The cold chamber I used can reach -54°C. The dry ice pellet for seeding the super-cooled fog was introduced on a long rod through a tube from the side of the cold box. Temperatures were monitored by a thermocouple.

The experiments were performed at a variety of temperatures between 0°C. and -38°C. (Fig. 1). The uncoated fog was nucleated as is illustrated by the clearing of the fog after dry ice was introduced[8]. The coated fog was not nucleated as is illustrated by the lack of change after dry ice was introduced.

Hexadecanol monolayers significantly affect parameters important in cloud physics, such as surface tension, dielectric constants, and evaporation rates. They also render fog and cloud nonseedable with dry ice. Only 1 to 10 kilograms per cubic kilometer[3] are required to coat the drops of most clouds. The nonseedability of super-cooled hexadecanol-coated water drops may offer a basis for work aimed at a better understanding of nucleation and other cloud processes.

I thank Dr. Vincent J. Schaefer, New York University, for suggesting this experiment; and Loyall Smith and Clifford Auvil, both of the Pacific Southwest Forest and Range Experiment Station, for their assistance in performing the experiments.

REFERENCES

1. C. C. Van Valin and P. A. Allee, Silanes as cloud stabilizers. Proc. 2nd Natl. Conf. on Weather Mod., Santa Barbara, Calif. April 6-9, 1970. Amer. Met. Soc., Boston, Mass. 129-133.

2. Robert T. Ryan, The possible modification of convective systems by the use of surface surfactants. Ibid, 393-396.

3. T. Y. Palmer, and R. A. Merriam, Cloud modification with electrically charged hexadecanol aerosol. Ibid, 401-405.

4. E. K. Bigg, J. L. Brownscombe, and W. J. Thomson, Fog modification with long-chain alcohols. Jour. Applied Met. 8, p. 75-82, 1969.

5. A. F. Wartburg, Louis Breydogle, and J. P. Lodge, Jr., Artificial stabilization of cloud droplets. Jour. of App. Met. 9, p. 830-831, 1970.

6. T. E. Hoffer and S. C. Mallen, Evaporation of contaminated and pure water drops in a wind tunnel. Jour. Atmos. Sci. 27, p. 914-918, 1970.

7. T. Y. Palmer, Development of an artificial stable water fog. Boeing Co. Doc. D2-90524, Seattle, Wash., 1964.

8. V. J. Schaefer, Simple experiments in atmospheric physics. Weatherwise, 8, p. 101-103, 1955.

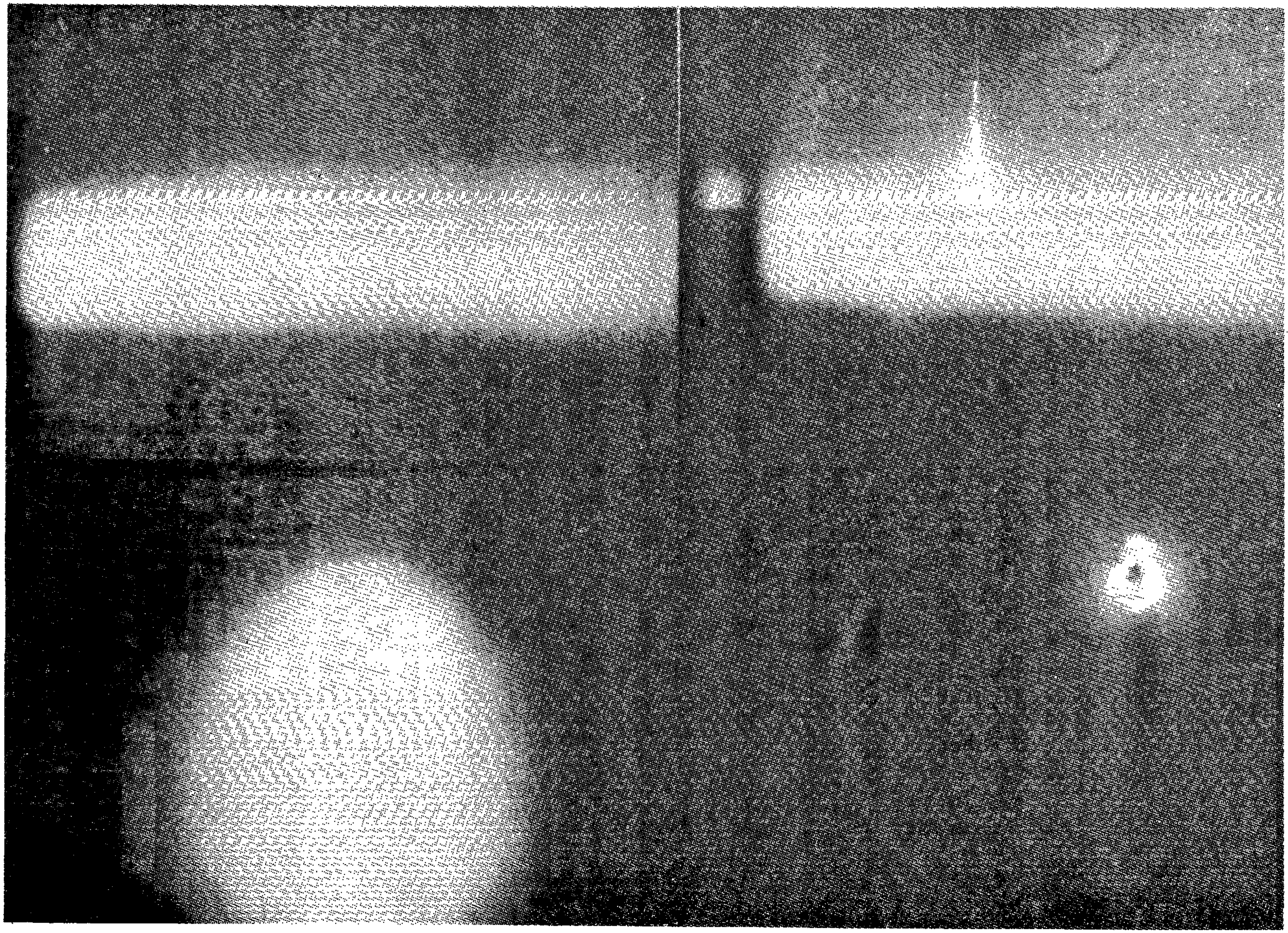


Fig. 1.--Supercooled water fogs seeded with a dry ice pellet are nucleated (bottom) while hexadecanol coated fogs are not. Pictures were taken three minutes after insertion of dry ice pellet.

A HISTORY OF CLOUD SEEDING IN THE WESTERN UNITED STATES

Keith J. Brown
Aerometric Research Inc.

INTRODUCTION

Since 1966, Aerometric Research Inc. (ARI) has been studying the Large Scale Effects of Cloud Seeding under Contract Nos. 14-06-D-5963 and 14-06-D-6841 with the U. S. Bureau of Reclamation, Division of Atmospheric Water Resources Management. The first phase of this study involved a statistical analysis of apparent seeding effects downwind from several long term operational cloud seeding projects. Since we were studying the precipitation patterns over large areas extending 200-300 miles downwind from the selected projects, it was necessary to determine whether any other seeding project was operated in the same general area during either the historical or operational period chosen.

This article will not go into the results of our study of downwind effects since this research is still in progress. Instead, we feel it is important that our research in locating records of past cloud seeding projects be documented in the literature for future reference.

PROCEDURE

Since the first phase of our study of Large Scale Effects of Cloud Seeding focused on long term operational projects, we restricted our search for historical records of past seeding projects to the eleven western states. It is in this area that all the long duration programs have been operated. The period covered by our investigation extended from 1948 through 1964.

The first step in our study was to contact the proper authorities in each of the eleven western states to determine what records on cloud seeding activity were collected and maintained by the state government. This produced the following results:

California - Starting in 1955, the California Department of Water Resources has published an annual summary of all cloud seeding activity within the state. These reports show the location of each project, the days of seeding operation, the operator and frequently the generator locations, output, etc. The first bulletin of this type was released in 1955 and summarized all past projects since the 1947-48 season. The California report could well be used as a model for other states to follow. While there are some omissions which are important to the scientist working in the field of weather modification, the availability of data is far superior to any other source found during this study.

Oregon - The Oregon Department of Agriculture, Chemical Applicator Supervisor maintained copies of reports from cloud seeding operators at the time of this study. They had reports from 1960. A trip was made to Salem, Oregon, to obtain copies of these reports.

Washington - The Washington Department of Conservation has a rather complete set of records of cloud seeding activity from 1957 on and a fair record prior to 1957. Mr. Stuart Shumway of the State Weather Modification Board maintained the records and was very helpful in providing information during our visit to Olympia.

Idaho - No records were available and according to the Idaho Secretary of State, they had no regulations on weather modification in 1966. Laws have been passed since that time however.

Nevada - Records were available from 1960 under the Department of Conservation and Natural Resources.

Utah - The University of Utah, Department of Meteorology, had records from 1962. A trip was made to Salt Lake City to obtain copies of these records.

Arizona - Records of cloud seeding activity were available since 1950 from the State Land Commissioner and copies of these were obtained.

New Mexico - The weather modification law in New Mexico was passed in 1965 and at the time of this study, no reports had been received by the designated agency, the New Mexico Institute of Mining and Technology.

Montana - In 1966 there was no weather modification regulation in Montana according to the Secretary of State. This has been changed since that time.

Colorado - The Division of Natural Resources in Denver had fairly complete records of cloud seeding activity dating back to 1959 with some data as far back as 1951. A trip was made to Denver to obtain these records.

Wyoming - The State Weather Modification Board in Cheyenne had only sketchy information on earlier projects.

A trip to Washington, D.C. to visit the National Science Foundation produced additional information on past projects including data from the President's Advisory Committee study in the mid-1950's. A visit to the National Archives failed to produce any new material.

The next source of information was a file of newspaper clippings regarding cloud seeding. This file was maintained by ARI's affiliated company, North American Weather Consultants (NAWC) and covered a period of about 15 years. This source provided considerable new information on past projects.

Finally, communications with some of the cloud seeding operators filled in a few gaps in our data.

The sum result of this effort is represented by the map shown as Figure 1 which outlines all the projects on which we were able to obtain data. Each state is given a two-letter designator and each project within the state an arbitrary number. Thus, project AR-1 is project number one in Arizona. A key for determining the area, operator and years of operation is shown in Table 1.

CONCLUSIONS

We recognize that the data presented in this article is certainly incomplete and possibly erroneous in some places. However, we feel it is important to have this information available in the literature as a reference and hopefully as a starting point for a more comprehensive study. It is essential that any statistical design utilizing historical precipitation records take into account the past seeding operations in that area to avoid contaminating the target or control area records.

This possibility of contamination is becoming increasingly important in light of the emerging evidence of significant effects of cloud seeding on precipitation patterns in areas over 100 miles downwind from the intended target area. One look at the extent of the land area in the western United States that has been involved in cloud seeding operations makes it abundantly clear that finding a completely virgin area for cloud seeding experiments is extremely difficult at least in the Western United States.

We cordially invite corrections and additions to this historical tabulation. We also would strongly encourage some agency or institution to expand this study in both area and scope to cover at least the entire United States and bring it up-to-date.

ACKNOWLEDGEMENTS

Aerometric Research Inc. would like to thank Dr. Archie Kahan and William Douglas of the Bureau of Reclamation, Division of Atmospheric Water Resources Management for their support and encouragement in this research. Our appreciation is also due to the various state officials, the National Science Foundation and the cloud seeding operators who provided the information used in this paper.

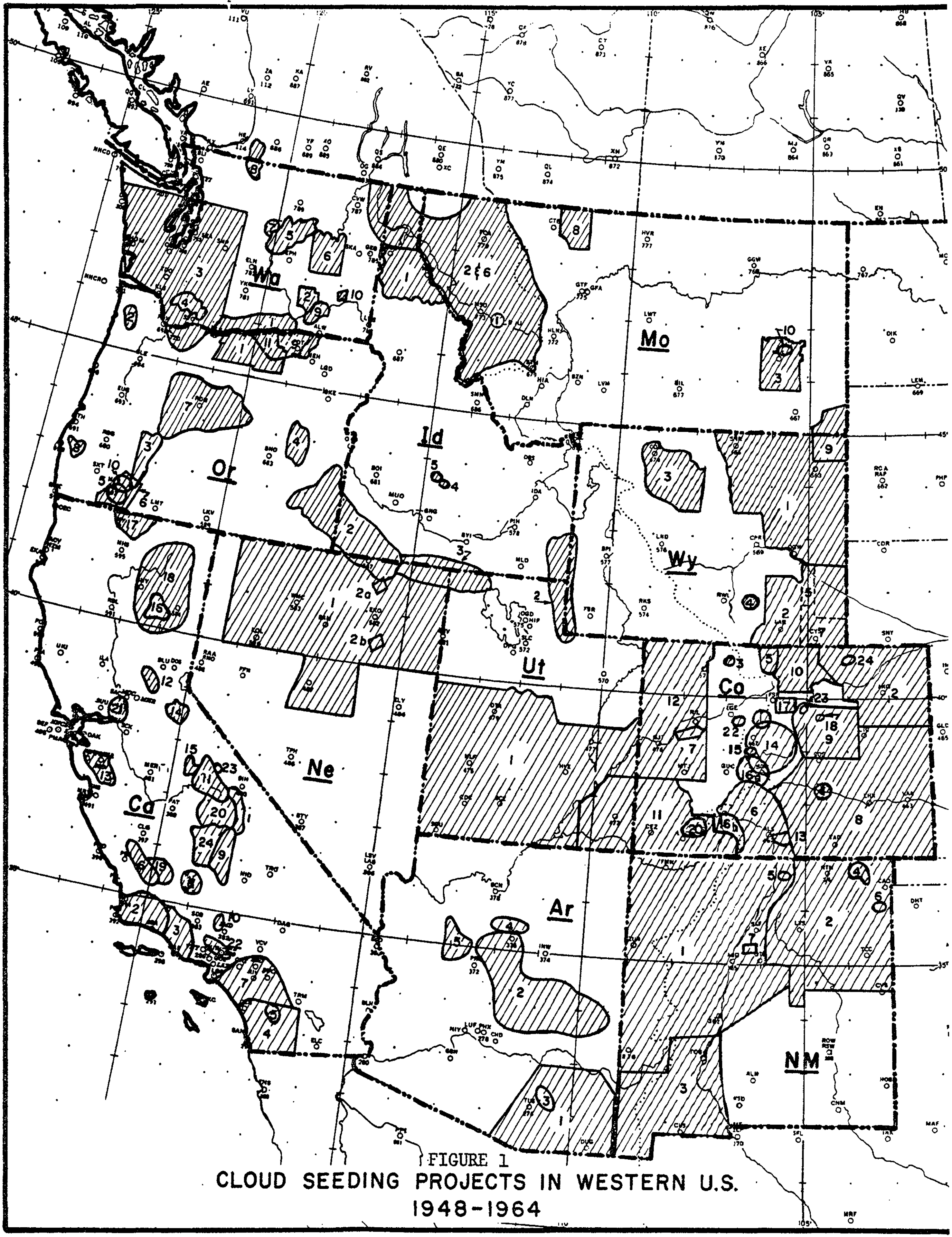


FIGURE 1
CLOUD SEEDING PROJECTS IN WESTERN U.S.
1948-1964

TABLE 1

CLOUD SEEDING PROJECTS IN THE ELEVEN WESTERN STATES - 1948-1964

ARIZONA

Project #	Area	Operator	Years
Ar 1	S. E. Arizona	W.R.D.C.	1950-52
2	Salt R. Valley	W.R.D.C. & S.R.V.W.U.A.	1949-51
3	Catalina Mts.	Univ. of Arizona	1957-62
4	Flagstaff - San Francisco Mts.	M.R.I. et al.	1958-date
5	Yavapai Land Co., N.W. Yavapai County	Individuals	1951-56

CALIFORNIA

Project #	Area	Operator	Years
Ca 1	Bishop Creek	Calif. Elec. Co. & Wx. Mod. Co.	1948-58
2	Santa Barbara County	N.A.W.C.	1948-60
3	Ventura County	N.A.W.C.	1948, 1952, 1957-60
4	San Diego City and County	N.A.W.C.	1948-56
5	Lake Henshaw - Vista Irrig. Dist.	W.R.D.C. and K.R.C.	1947-48, 1962-63
6	Carrizo Plain	Precip. Control Co.	1950-52
7	Santa Ana R. and Temescal R.	Wx. Mod. Co., W.R.D.C., N.A.W.C., & San Bernardino Mun. Water Co.	1950-date
8	Bakersfield Area	Precip. Control Co.	1948-52
9	Kern R. and Tule R.	Precip. Control Co.	1950-63
10	Antelope Valley	W.R.D.C. and N.A.W.C.	1951-52
11	San Joaquin R. So. Calif. Edison Co.	N.A.W.C.	1951-date
12	Placer County - Auburn	W.R.D.C.	1951-63
13	Santa Clara County	Wx. Mod. Co.	1951-date
14	Mokelumne & Stanislaus R. - P.G.&E.	N.A.W.C.	1953-61

CALIFORNIA (Cont'd)

Project #	Area	Operator	Years
Ca 15	Crane Valley - Bass Lake - P.G.&E.	N.A.W.C.	1954-59
16	Lake Alamanor - P.G.&E.	N.A.W.C. and P.G.&E.	1954-date
17	North Siskiyou County	W.R.D.C.	1954-58
18	N. E. California	Calif. Dept. of Forestry	1954-59
19	N. W. Kern County	Precip. Control Co.	1953-61
20	Kings River	Wx. Mod. Co. and Atmospherics Inc.	1955-date
21	Sacramento Area	W.R.D.C.	1958-59
22	San Gabriel Mts.	L.A.C.F.C.D. & N.A.W.C.	1958-date
23	Mammoth Lake	Wx. Mod. Co.	1958-60
24	Tulare County	Atmospherics Inc.	1962-date

Additional Projects of One Year Duration - Not Shown on Fig. 1

1947-48	E. Slope Coast Range West of Bakersfield	Individuals
	S. Slope Sierra Madre Mts. near Pasadena	Individuals
	Lake Elsinor Watershed	Individuals
	Big Bear Lake Watershed	Individuals
	San Jacinto Mts. near Riverside	Individuals
	Lower Salinas Valley	Individuals
1948-49	Colfax Divide	U.S.W.B.
1952-53	San Benito County*	Wx. Mod. Co.
	Crescent City - Eureka Area	Hubbard
1954-55	Tehachapi Valley	N.A.W.C.
1956-57	Lanfair Valley near Needles	O.X. Cattle Co.
1957-58	Trinity and Humboldt Counties	Calif. Dept. of Natural Resources
1959-60	Squaw Valley	W.R.D.C.

* San Benito County Project started again in 1964-65 and 1965-66 by N.A.W.C.

COLORADO

Project #	Area	Operator	Years
Co 1	Climax	Colo. State University	1960-date
2	N. E. Colorado	W.R.D.C.	1951, 1957-59
3	Park Range	E. Bollay Associates	1964-date
4	Pueblo	Steverson Atmospherics	1963
5	Little Laramie R.	W.R.D.C.	1951
6	Alamosa	W.R.D.C.	1951
7	Grand Mesa	W.R.D.C.	1961-date
8	S. E. Colorado	W.R.D.C.	1951
9	Denver Area	W.R.D.C.	1951
10	Fort Collins	W.R.D.C.	1951
11	S. W. Colorado	W.R.D.C.	1951
12	N. W. Colorado	W.R.D.C.	1951
13	Ft. Garland	Steverson Atmospherics	1964
14	Leadville Triangle	Steverson Atmospherics	1964
15	Antero Reservoir Watershed	Steverson Atmospherics	1963-64
16	Monarch Pass & Wolf Cr. Pass	W.R.D.C.	1964-date
17	Vail Ski Area	W.R.D.C.	1958, 1960-64
18	Box Elder Farms	W.R.D.C.	1953
19	Platte Valley #	W.R.D.C.	1954-55
20	San Juan Area	W.R.D.C.	1954-55
21	Mill Iron Ranches #	W.R.D.C.	1955-56
22	Aspen Ski Area	W.R.D.C.	1959-60, 1962-63
23	Loveland & Arapahoe Ski Areas	W.R.D.C.	1960-63
24	New Raymer	Colo. State University	1962-63

Exact target location unknown - not shown in Figure 1.

IDAHO

Project #		Area	Operator	Years
Id	1	Coeur d'Alene Lake Watershed (W.W.P.)	N.A.W.C.	1951-59
	2	Owyhee R. (Nev., Ore., Ida.)	N.A.W.C.	1955-56, 1959-62
	3	S. E. Idaho Canal Co.	N.A.W.C.	1954-55
	4	Sun Valley	W.R.D.C.	1958-60
	5	Camas Valley	W.R.D.C.	1950

MONTANA

Project #		Area	Operator	Years
Mo	1	Missoula Area (Project Skyfire)	U. S. Forest Service	1957-date
	2 & 6	Pend Oreille River Basin*	W.R.D.C.	1951, 1954-58
	3	Custer County	N.A.W.C.	1953
	4	Weather Improvement Corporation #	W.R.D.C.	1951
	5	Three Rivers Weather Research Co. #	W.R.D.C.	1951
	6	(See #2 above)		
	7	S. E. Montana (Dines Ranch) #	W.R.D.C.	1961-62
	8	Toole County	W.R.D.C.	1964-date
	9	Tri-Counties Rural Devel. Assoc. #	W.R.D.C.	1964-date
	10	Miles City Area - Wm. Tonn & Sons	W.R.D.C.	1961-62

NEVADA

Project #		Area	Operator	Years
Ne	1	Northern Nevada	W.R.D.C. and N.A.W.C.	1954-56
	2	Elko Area (Nev. Atmos. Res. Proj.)	N.A.W.C. and D.R.I.	1962-date

* Seeded in 1951 for Bonneville Power Admin. and in 1954-58 for Pacific Power & Light Co., Washington Water Power Co. and Kaiser Aluminum & Chemical Corp.

Exact target location unknown - not shown in Figure 1.

NEW MEXICO

Project #		Area	Operator	Years
NM	1	Western New Mexico-San Ysidro Range Imp.	W.R.D.C.	1951-52
	2	N. E. New Mexico	W.R.D.C.	1950-51
	3	Upper Rio Grande Basin	W.R.D.C.	1951
	4	Raton Area - T. O. Ranch	W.R.D.C.	1960-63
	5	Red River Ski Area	W.R.D.C.	1962-63
	6	Tequesquite Ranch	W.R.D.C.	1962-date
	7	Albuquerque Area - Campbell Farms	W.R.D.C.	1960

OREGON

Project #		Area	Operator	Years
Or	1	Tri-Counties (Sherman, Gilliam, Morrow)	W.R.D.C. & Wx. Mod. Co.	1950-54, 1957-61
	2	Tillamook Burn	N.A.W.C.	1951
	3	Southern Cascades (COPCO)	N.A.W.C.	1951-60
	4	Malheur R. Basin	W.R.D.C.	1950-51
	5	Medford Area	Wx. Control Inc.	1949-51
	6	Jackson County	N.A.W.C.	1955
	7	Deschutes R. Basin (Portland Gen. Elec.)	N.A.W.C.	1964
	8	Coquille R. Basin (Pacific Power & Light)	W.R.D.C.	1957-60
	9	Blue Mt.	W.R.D.C.	1952-53, 1955
	10	Rogue R. Valley	W.R.D.C.	1954-58
	11	Morrow County and Umatilla County	W.R.D.C.	1956-date

UTAH

Project #		Area	Operator	Years
Ut	1	Southern Utah	W.R.D.C.	1951-55
	2	Bear Lake Watershed (Utah Power & Light)	N.A.W.C.	1954-date

WASHINGTON

Project #	Area	Operator	Years
Wa 1	Horrigan Farms - Horse Heaven	W.R.D.C.	1950-date
2	Big Bend (Franklin, Adams, Lincoln, Doublas Counties)	W.R.D.C.	1950-date
3	Western Washington (A.C.N. Project)	U.S.W.B.	1953-54
4	Lewis R. Watershed (Pac. Power & Light)	W.R.D.C.	1952-60
5	No. Central Washington	W.R.D.C.	1951
6	Lincoln Water Development Corp.	W.R.D.C.	
7	Manson Area - Apple Weather Inc.	W.R.D.C.	1957, 1961-62
8	Skagit R. Basin	W.R.D.C. and Wash. Dept. of Nat. Res.	1958, 1963-64
9	N. and N.W. Walla Walla County (Eureka Wx. Mod. Corp)	W.R.D.C.	1959-64
10	McGregor Land Co.	W.R.D.C.	1963-64

WYOMING

Project #	Area	Operator	Years
Wy 1	N. E. Wyoming	W.R.D.C.	1951
2	S. E. Wyoming	W.R.D.C.	1951
3	N. W. Wyoming	W.R.D.C.	1951
4	Elk Mountain	Univ. of Wyoming	1963-date
5	Platte County	W.R.D.C.	1953
6	Wyoming Weather #	W.R.D.C.	1954
7	Laramie County	W.R.D.C.	1954
8	Wyo-Braska Weather #	W.R.D.C.	1954-55
9	N. E. Wyoming - S. E. Montana	Grazing Inc.	1961

Exact target location unknown - not shown in Figure 1.

Operator Abbreviations Used

W.R.D.C.	-	Water Resources Development Co.
S.R.V.W.U.A.	-	Salt River Valley Water Users Association
M.R.I.	-	Meteorology Research, Inc.
Wx. Mod. Co.	-	Weather Modification Co.
N.A.W.C.	-	North American Weather Consultants
L.A.C.F.C.D.	-	Los Angeles County Flood Control District
K.R.C.	-	K.R.C. Service Corp.
P. G. & E.	-	Pacific Gas & Electric Co.
U.S.W.B.	-	U. S. Weather Bureau

NEEDED - A BETTER ENERGY SINK

Dr. Vincent J. Schaefer, Director

Atmospheric Sciences Research Center
State University of New York at Albany

When I was eight years old, some strange things happened in Europe and within a few years I found myself knitting washcloths, using rainbow sugar on my breakfast cereal, saving tin foil, wandering around Camp Mohawk where our National Guard was training for combat, and then watching the tearful farewells as the troop trains pulled away for the first leg of the long trip to France. I remember too, the excitement at the end of the war, the false Armistice announcements and then the real one---and the sadness of neighbors whose sons didn't come home.

Then came the doldrums of the early thirties, a period of peace accompanied by one of the worst economic depressions of history.

Let's consider next the period of 1940 - 41. I was working hard on problems related to surface chemistry. --- I had just discovered a way to preserve snow crystals in plastic, and, incidentally, in the process, to more than double the brightness and contrast of the new television tubes.

One afternoon, "the boss" as we affectionately called Dr. Irving Langmuir, stopped by my laboratory bench, and asked me if I would like to tackle an interesting new problem---the search for a more efficient way to remove the particles from smokey air. There was a degree of urgency about the problem. Certain powers, who might some day be aligned against us in an aggressive war, were said to be feverishly producing poisonous smokes. At the time there was no protection available if such smokes were used against us or our friends.

Thus my second experience with war in 1940 - 45 was much closer to the realities of the terrible aspects of such conflicts. As defense preparedness accelerated, we became involved in such diverse activities as the production of artificial fogs for the screening of troops, beach landings, and warship protection; attempting to solve the problem of radio static which occurred when airplanes flew thru snowstorms; The detection of submarines using binaural sound detectors and finally the cause of aircraft icing and methods for preventing it.

During and after World War II and then the Korean troubles, it became increasingly apparent, that the so-called prosperity of America, and of the man on the street, was inextricably associated with threats of war, actual war, or the recovery from war. Once the threat, the reality or the effect of war became less immediate or urgent, we learned that it was now time to balance the budget, to inaugaurate economies <u>and</u>

insofar as possible, to reduce spending in Albany or Denver and Washington. I should emphasize at this point that I do not understand world economics (some of you perhaps, think you do at this stage of your career!) I merely know that economies, cut backs in spending, retrenchment and similar activities reduces the number of jobs available in the affected area. Perhaps I should hasten to add, that in retrospect, the depression years of 1930 - 1935 was an adventurous period for me. A sense of nostalgia glosses over the less attractive features of bread lines, soup kitchens, and other features which marked those days of despair, among many family men and women. I was not married at the time -- had no hungry youngsters and was protected from the grim realities of the period by a sense of idealism, optimism and other wonderful assets, which are the inheritance of the young at heart.

Now that I have a somewhat better appreciation of the responsibilities of a husband, father, and -- a grandfather (four times!), I have a somewhat more realistic impression of some of the things which seem to make our economy "go" -- whether I understand them or not.

It is quite apparent that as time goes on, and we learn to cope with the Cubas, the Viet Nams and the other brush wars which will probably plague us from time to time in this imperfect world of ours, we must come face to face with the urgent need of a substitute for the sense of urgency which is associated with war. This substitute is what I propose to call -- a better energy sink. Such a mechanism must have features not unlike those emergencies of the past which led to the boom economics related to the war periods of 1914 - 18 and 1940 - 45. They should feature however, a thorough understanding (insofar as is possible) of the long range objectives of the effort. What do I mean by an energy sink?

My concept of an "energy sink" is a procedure which effectively and efficiently utilizes assets of one kind or another, which accumulate or develop through the expenditure of energy. These assets might consist of a bale of hay, a deck of saw logs, a hundred million bushels of wheat, a freight train of coal or the gold reserves at Fort Knox. The very presence of such materials means that they exist as the result of hard work, sweat, the investment of money in tools, labor, administration and the like. By existing, such items are like money in the bank. Their existence represents the expenditure of some form of energy. In the ultimate view, their value can be assessed in terms of the effort that was expended in producing, synthesizing, accumulating or in other ways devleoping them as something having a tangible value. Like any investment, its very existence means in a sense, that so long as it is there, there is no need to replace it.

Thus, if it continually is accumulating there comes a time when more of the same is no longer needed. When that time comes, those who had been engaged in producing that particular item suddenly discover that their efforts are no longer needed, and they must find a new way to use their talents or abilities.

Obviously, the storage of "energy" whether in the form of water behind dams, gold at Fort Knox, stock piles of aluminum or coal -- all play a crucial role in the dynamic economy of the world.

Whenever an economy depends in some manner on the continuing utilization of natural or man made resources, it becomes necessary that such use continues without appreciable interruption. If use slows down or ends, the complex balance on which many other things depend is modified in a disconcerting way, and may lead to economic disaster. Business depressions, power blackouts, inflation of currency, the unexpected death of a world leader --- all play their role in such equilibria.

Suppose for example, that all the statesmen, the militarymen and the common people of the world were, by some modern miracle, to decide that at noon next Tuesday every effort presently devoted to the preparation for defense and aggression would suddenly end, (what a wonderful thought!). What would happen?

Overnight we would probably experience the beginning of the greatest monetary crises and economic depression the world has ever known! That activity which we and others refer to as defense spending, represents a massive energy sink --- one on which our current economy depends to a major degree.

While I am confident, that the intelligence, ingenuity and resourcefulness of our people could adjust to this utopian situation in time, just imagine the dislocation and change over equipment, talent and daily lives which would occur in the United States alone, if we suddenly were confronted, with the need to find new ways to spend the fifty-billion dollars now allocated yearly for defense activities.

While the current unrest in most parts of the world, accompanied by the present surgeof nationalism, makes me wonder if we will ever approach such a utopian ideal, I believe it is high time that we give serious thought to the development of new kinds and types of energy sinks. How many of us would know how to spend a billion dollars -- intelligently, usefully, and selflessly! I think I could probably find a good use for, maybe five million -- but what about two to five thousandtimes that amount -- every 12 months about $1500 a second -- $500,000 a half million an hour!

Some of you, I am sure are quite aware of our so-called "cold war". This was a rather cold-cold water until that historic day in October 1957 when the word flashed around the world that Sputnik I had been orbited. A day previous, I had assembled a small group of scientists friends at an isolated lodge in the Adirondack Mountains of upper New York State, to discuss informally, the basic properties of atmospheric particles. Even though this was not very long ago (many of you were just becoming aware of the fact that there was more to education than finishing the eighth grade at school), many of the "old ways" of looking at things have since disappeared.

I remember a sense of gloom among some of my associates as we assembled for the conference. Word had been "passed down the line" the previous week that the budgets for the next year would be drastically reduced. The discouragement was particularly noticeable among our friends from the military establishments. With this sense of frustration our discussions got underway. I remember, that one of the participants from the army was called to the phone (a concession allowed only for real emergencies!). When he returned it was with the electrifying news that Sputnik I was in orbit around the earth.

A dramatic change occurred in the attitude of our previously gloomy colleagues. We could all anticipate the reaction of the American public to this new development in the cold war. Thus arose the sudden urgency to "beat the Russians" in the space game.

How tragic that it required an intensification of the cold war to arouse us from a lethargic attitude! Since that time, the space program has become a substitute for war --- a very effective energy sink. The accomplishments in space since that time, is an intriguing demonstration of the truth of a saying of one of my revered friends, Dr. Willis R. Whitney -- originator and for many years, director of the famed G. E. Research Laboratory. "Doc Whitney" was a walking legend in our laboratory. He once told me --- "If many can imagine it -- he can do it!" This faith in man's ingenuity, inventiveness, creativity and resourcefulness paid off for General Electric many times, as Whitney guided his associates, in producing many profitable discoveries in what was once called "The House of Magic".

As evidence in favor of Doc Whitney's philosphy, witness the space accomplishments in the short period of time since Sputnik I orbited. In the past eight years the United States has successfully launched nearly three times as many satellites as other countries with an overall efficiency of 83%. What an exciting time to be alive!

I wish sometime you could accompany me on one of my consulting visits to a big space oriented industry. In some of their design engineering shops one can literally see acres of engineers, scientists and other professional people, all working on some aspect of our space program.

One can certainly argue, that it may be a questionable use of so many trained persons and so much money in a program, which in the long run, may show a rather low percentage "pay off" for the eventual welfare of mankind. My only comment is -- Thank the Good Lord for Space! How much better, to spend money for this rare adventure of the spirit than for a killing war. I would grant that some of the "space" money might better be spent for education or hospitals or pollution control -- or for relieving the poverty on the Reservations. I feel impelled however to say that those of us in education, medicine, sociology and meteorology, to mention a few, have not elevated our sights and our thinking to the point where we can usefully spend money in the large scale comparable to the defense effort or the conquest of space. This naturally conservative attitude was brought home to me very forcefully a few weeks ago, in Washington at a Senate hearing. A friend of mine who is associated with one of our large and very good Universities, expressed his dismay over the fact that a bill receiving preliminary hearings, proposed placing a ceiling of $35,000,000 for yearly spending in support of weather modification activities. His expression of dismay was not because of the ceiling, but of his sincere belief that it is not possible to spend a tenth of that amount without undue extravagance. I for one, do not believe this to be the case, but it illustrates my point rather well.

We in the sciences, education, engineering, sociology and the humanities <u>must</u> learn how to spend large amounts of money carefully, intelligently and with integrity.

I believe that whether we like it or not -- whether Democrats, Republicans or Independents, the "energy sink" is an integral part of our economy.

The near disaster which confronts us in virtually all of our cities, characterized by a combination of air and water pollution, urban sprawl, fabricated ugliness and man-made lava, as one of my friends recently described our proliferating highway system, --- all of these are very worthwhile subjects to be identified with an effective energy sink.

Unlike Outer Space which is such a good sink that many of the devices which are launched never even come back to haunt us, as do our derelict autos, the energy devoted to improving the world's living space is likely to produce tangible dividends of incalcuable value.

Two years ago after a slow and somewhat exhausting climb up the eastern slope of Mountain View Crest about 30 miles northwest of this Campus, I was privileged to gaze on one of the most spectacular wild mountain vistas in North America -- Pigeon, Turret, Aeolus, Windom, Sunlight -- what magnificént mountains! I wonder how many of your have taken advantage of this nearby opportunity -- or how many have been startled by the sudden flight of the snow white ptarmigan from the high mountain meadows near the Eldorado Lakes, or puzzled over the warm springs near Baker Bridge, or pondered about the remarkable manner in which the myriads of springs above West Virginia Gulch merge to become a crystal clear trout stream tumbling down the granite ledges on the way to Vellicitos Lake.

The San Juans, although' one of the best, is but one segment of the remarkable heritage which is our birthright in America. Up until now our parents and those before, have, too often, taken many of these resources for granted. The forests were hogged, the streams gutted and then polluted, and the air used as a sewer in the sky.

We, and this means everyone in this room, and in Durango; and Colorado; and the United States; the Planet Earth and its environs, can no longer consider these precious assets as "expendables".

It is past the time that all of us should pay heed, to the urgent need for assuming responsibility as intelligent individuals, for the proper treatment of our environment. We are all offenders -- whether it be in driving cars, burning trash, operating our furnaces, asking for electric power or permitting our industries and towns to use our streams and our air as areas for dumping debris.

What to do about it? Let's develop a new type of "energy sink"!

Its establishment will involve a more realistic attitude toward our individual and collective responsibilities concerning these problems needing immediate attention.

American ingenuity I am sure, can solve all of these problems, whether they involve science, engineering, sociology, the humanities or just plain responsible citizenship and voting activity.

The solution to such complex problems will not be easy --- many are far from glamorous. For this very reason, they pose a challenge of the first order. How do our political and social scientists solve the "people problem". How do we get the traveling public to use litter bags and to have a concept of proper behavior in our environment, whether it be a city street, a country lane or a mountain meadow?

I'll never forget the disgust I experienced as I listened to a so-called sportsman, proudly describe how he had killed a family of hoary marmots on the rocky slopes near Hunchback Pass on the upper reaches of Bear Creek. These fascinating creatures occupy an important niche in the ecosystem of the mountains --- and no one has the "right" to remove their cheery whistle from the high country! This person was not a mountain dweller seeking to eke out an austere diet with a hunter's stew - rather - he was a respected medical doctor from a large urban community. How much better --- if he had to satisfy some primitive urge, to use his discarded beer cans as targets so that they might rust away a little faster (if not made of aluminum).

How can we do things better! This can only be achieved with the realization --- your realization, that there is more to a college education than a means of making a "fast buck", followed by a life of ease. It is essential that the real joy of hard work be given new lustre, that we become more aware of the fact that true education has only just started as one leaves college, and that there is joy and privelege in helping the other fellow.

If these thoughts sound old fashioned --- so be it. However, I am continually thrilled, to witness the solid if slow progress being made by the Peace Corps, by Vista, Vita, Hope, Field Service and the myriads of other positive actions and attitudes of all types of Americans when there is a job that needs doing. While it sometimes takes a flood, a tornado, an ice storm or a blackout to bring these finer emotions to the surface, they are latent virtues in everyone. A sense of idealism or religion, or humanism --- whatever one wants to call it, is part of the innate characteristics of all mankind, or at least of those who have had the opportunity to sense these potential virtues. It is my wish and prayer that each of you, at this milestone in your intellectual life, give serious consideration to those truly immortal words of President Kennedy and each in his own way ponder the question, "What can I do for my country"?

We are experiencing the "Golden Age" of America. We still have an abundance of most raw materials, our economy continues to expand at a nearly frightening speed --- and there is so much to do! What is to be done about expanding populations, the proper feeding of it, the best treatment of our air, water and other natural resources, the peaceful utilization of atomic energy, the wise use of weather control? The development of a high degree of comptability by man of his environment and --- most urgent of all --- how to get along with the other fellow whether white, black, red, yellow or tan! These efforts can represent and form the energy sink which we all need to develop so it can be useful. Never has there been more or better opportunities for those with the "will to do".

I do not envy you your youth or your high potential since I am still having the time of my life! But I do wish to leave one parting thought with you, which my old friend Lomahaftewa of the Second Hopi Mesa conveyed in commenting on the weather research activities of friends of mine.

--- I hope you have --- a good time!!

(Editor's Note:

This address was presented at the commencement exercises, Ft. Lewis College, Durango, Colorado, on April 17, 1966. We asked several persons to read Dr. Schaefer's material and comment on its appropriativeness to the Journal of Weather Modification. All our reviewers felt that, while the material was not specifically directed to weather modification operations or research, the thoughts were as real and timely today as they were in 1966 and certainly could be applied to our present concern for national weather modification concepts. The editor thought so, too!!)