THE PHYSICS OF CLOUD MODIFICATION

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1. Introduction

During the decade that followed immediately upon cessation of World War II there occurred remarkably vigorous developments in almost all phases of meteorology. It would seem that no single factor suffices to account for this marked acceleration of meteorological research that began in the mid-forties; rather it appears in retrospect that a peculiar combination of factors joined to produce this phenomenon. First, a very great number of younger workers who had received meteorological training during their military service entered the field in many countries throughout the world. Second, the recognition of the many ways in which progress in geophysics relates to matters of military importance operated, by virtue of the circumstances of the decade, to stimulate support of geophysical research in general, and meteorological research in particular, at a level unparalleled in earlier years. Finally, a number of significant new techniques of observation and research which had been developed either directly or indirectly through the exigencies of wartime activities became available to the meteorologist just at a time when personnel and research support permitted their optimal exploitation.

One of the most exciting of these meteorological developments of the past decade has been the discovery of techniques for modifying, to a certain degree, the physical processes occurring within natural clouds. This development, abetted by all of the auspicious circumstances cited above, has given great impetus to advance in all of those portions of physical meteorology concerned with the physics of clouds and precipitation processes. As a result, one is tempted to say that there is an order-of-magnitude difference between what is known today about cloud physics and what was known a mere decade ago; but this statement, even if a fair one, tends to obscure what has become still more evident, namely, that our present knowledge of cloud physics is still an order-of-magnitude short of what it must be if we are ever to exploit optimally the recently recognized prospects of control of natural precipitation.

In the following discussion, the present position of cloud physics will be reviewed, and the progress that has been made toward the goal of controlling, or at least modifying significantly, the phenomena occurring within natural clouds will be summarized. A peculiar circumstance enters in the form of the severe difficulty of accurately assessing that progress as it may (or may not) be exhibited in actual field experiments. These difficulties, and their origin in the physical complexity of the problem, must be clearly understood by the geophysicist or other reader who wishes to understand the current status of cloud modification and the challenge that the goal of controlled cloud modification has placed before the meteorologist. My main objective will be to discuss the salient points of the physical theories of cloud and precipitation processes which have undergone such rapid evolution in the past few years, and to try to call attention to the principal implications which these theories have for prospects of control of certain links in the chain of events leading to precipitation.

2. Clouds and the Atmospheric Water Vapor Cycle

2.1 Terrestrial Clouds

The chief goal of man's efforts at cloud modification is simply this: to cause more precipitation to fall upon selected continental areas than would occur by wholly natural processes.

Before turning to an examination of microphysical phenomena in the cloud that enter into these precipitation processes, it is pertinent to look rather broadly at the basic question of how there happen to be clouds and rain processes to modify in the first place and to inspect the magnitudes of certain parameters of the hydrologic cycle that influence the scope of potential modification methods.

At the start it must be recognized that the clouds with which we are all so familiar and upon which we must depend as immediate sources of all but a small fraction of our continental water supplies are in many respects quite adventitious features of our planet's atmosphere. Even without exploring recent insights gained in theoretical studies of the geochemical evolution of planetary atmospheres, we need only turn to our closest neighbors in the solar system to see that water substance cannot be taken for granted as an inevitably abundant material on the surface of a planet of the earth's size, for neither of these neighbor-planets has spectroscopically observable quantities of water vapor in its atmosphere. But, even given the presence of an extensive layer of liquid water covering most of our planet's surface and noting that its physical and chemical properties are such as to make it almost uniquely suited to support the kind of plant and animal life in which we have so strong a practical interest, it is not immediately evident that it was inevitable that geophysical processes should unfold in such ways as to insure that significant amounts of this material would be transported to continental areas.

As a matter of fact, viewed in percentual terms, only a seemingly insignificant fraction of the oceanic water is thus transported in the hydrologic cycle. Of the total amount of terrestrial water variously estimated at from about 5×10^{23} to about 5×10^{24} gm (Hutchinson [1] gives 1.4×10^{24} gm, for example), only about 10^{20} gm, or a mere hundredth of one per cent of the total, is found at any time in the rivers and lakes of the continents. A very much larger fraction, perhaps four or five per cent in all [2], is present as comparatively immobile ground water and glacier ice, but these latter constitute quasi-steady-state storage reservoirs associated with processes having such slow replenishment rates as to be poor indicators of the rate of exchange of water between oceans and continents. Taking Bannon's recent results on world-wide distribution of atmospheric water vapor, as summarized by Sutcliffe [3], namely a value of 2.5 cm for the global yearly mean precipitable water-vapor content (integrated vapor content from surface to outer limits of the atmosphere, expressed in equivalent depth to which it would stand if entirely condensed and vertically precipitated), we find that there is only of the order of 10^{19} gm of H_2O present in our atmosphere at any one time as vapor, or a mere *thousandth of one per cent* of the terrestrial stock of water substance. That the total atmospheric water vapor content is so small despite presence of large bodies of liquid water is, of course, a consequence of the comparatively small saturation vapor pressure of water at terrestrial temperatures, which, in turn, results from the unusually high latent heat of vaporization of water. (The latter, finally, is a consequence of the molecular structure of water that permits strong association by hydrogen bonding.)

The one-thousandth of one per cent of the total terrestrial water that is, at any one time, found in the form of atmospheric water vapor is advected in the air movements of the general circulation from one part of the world to another and thereby occasionally reaches continental interiors where, if forced to ascend by any of several dynamical processes, it may condense to form a cloud. From that cloud there may, if a fairly complex series of events unfolds rapidly enough, fall some precipitation that reaches the continental surface. In the two following sections certain scale factors and characteristic magnitudes of these processes will be examined.

2.2 Scale Considerations

It is instructive to make the following order-of-magnitude calculations to clarify the quantitative aspects of the role of clouds in the hydrologic cycle. These calculations will serve to reveal the scope of the problem man encounters when he seeks to make any appreciable change in the planetary water budget. The immediate objective will be to attempt an estimate of the fraction of total instantaneous stock of water vapor which might be made to undergo artificially induced precipitation if a hypothetical world-wide program of continental rainmaking were operating at optimistic efficiencies. Such an estimate is surely of academic interest to the geophysicist and also sheds light on fears that have sometimes been raised concerning tampering with the natural hydrologic cycle.

Students of the global heat balance who have had to make climatological estimates of mean global cloudiness for use in evaluating the albedo term of the heat-budget have arrived at a figure of about 0.50 for the mean cloud cover for the year and the earth as a whole. Cloud depths will, in general, decrease with increasing latitude while the zonal mean cloudiness will tend to increase toward the latitude of the subpolar low pressure belt in such a way that it seems reasonable (though clearly very approximate) to assume for present purposes a mean cloud depth of about 2 km as a global average. Liquid water contents will tend to vary latitudinally in similar fashion, both because of the effect of the latitudinal temperature gradient and because of meridional variations in prevailing cloud types, and with these variations in mind, current data suggest that half a gram of cloud water per cubic meter of cloud space will be reasonable as an over-all average. Combining average cloudiness, mean depth, and mean liquid water content yields an average of a tenth of a gram of cloud liquid water per square centimeter of surface for the world as a whole. Bannon's data, cited above, show that the mean precipitable water vapor content is about 2.5 cm for the global annual mean. Hence, cloud liquid water content accounts for about 0.10/2.5, or only one twenty-fifth, of total atmospheric water vapor content, on an annual global average basis. Of this amount of water, roughly one-fourth will occur in clouds lying over continental areas, so only about 1/100th of the instantaneous store of atmospheric H_2O is in a form and location even remotely amenable to useful manipulation by cloud-modification techniques.

The next step in this estimate of the geophysical scale of our potential cloud-modification problem should consist in cutting down our last fraction, 1/100th, by some factor representing the portion of all continental cloud types that are brought by natural processes sufficiently near to the stage of precipitating so that one can begin to discuss manipulating their release of water. I know of no source of climatic data to assist in making a close estimate of this factor, for there is a glaring lack of good data on world-wide distribution of clouds reported in a manner that could be termed cloud-physically meaningful. In view of the nature of the mean cloudiness figure of 0.50 used above (applying as it does to all clouds without regard to their being of a type associated with precipitation), I should guess that not more than about one-tenth of the global mean cloud cover is modifiable in the sense of being near enough to undergoing natural release of precipitation that manipulation would hold any promise at all. It scarcely seems possible that such an estimate of one-tenth could be low by even a factor of two; it is much more likely to be unduly large. Hence, let us say, conservatively, that perhaps one-tenth of 1/100th or one-thousandth of the total instantaneous atmospheric H_2O is actually subject at any one time to modification techniques over continental areas.

Finally, uncertain as is the present state of evaluation of recent modification experiments, most meteorologists would now probably agree that prospective techniques cannot be expected to augment natural precipitation by much more than about ten per cent, as an optimistic upper limit. This means that about a tenth of a thousandth, *or of the order of only one ten-thousandth of the instantaneous world-wide stock of atmospheric water*

TABLE I. Estimated magnitudes of quantities controlling effects of world-wide cloud seeding.

Estimated fraction of total atmospheric H_2O condensed, at any one instant, into cloud liquid water (based on mean cloudiness of 0.5 and mean cloud depth of 2 km)	1/25
Same fraction for continental areas only	1/100
Estimated upper limit of fraction of all continental cloud cover amenable to treatment to induce artificial precipitation	1/10
Generally accepted (1958) order of upper limiting magnitude of obtainable increase in precipitation by cloud modification methods	1/10
Consequent upper limit to estimated fraction of total atmospheric H_2O which, at any instant, might be undergoing artificially stimulated precipitation over continents	1/10,000

substance might be undergoing artifically induced precipitation from favorable cloud types over continental areas if all such clouds throughout the world were continually being treated in a manner yielding ten per cent increases. The quantities leading to this conclusion are assembled in Table I. It bears repeating that the third and fourth fractions listed in Table I are believed to be too high but are used here to render the final conclusions conservative with respect to relative magnitude of the geophysical effects of cloud seeding at a maximum conceivable rate.

Two main conclusions may be drawn from the above estimate. First, it shows us that, as in so many other geophysical problems, man's efforts to alter the over-all course of geophysical events constitute a minuscule alteration of natural processes. Second, it shows fairly clearly that, notwithstanding the mounting evidence for the comparatively large role that latent heat exchanges play in the energetics of the general circulation of our atmosphere, even a very ambitious world-wide cloud-modification program can scarcely be a source of serious interference with the energy balance of the whole atmosphere—and the figures seem to speak directly for such a program being incapable of sensibly altering the prevailing water vapor content of the atmosphere as a whole. The above discussion glosses over a number of points wherein locally important disturbances might possibly occur, but serves to establish perspective concerning what has been the sometimes misconstrued geophysical scale of potential cloud-modification effects.

2.3. Precipitation Release Rates and Atmospheric Water Vapor Turnover Rates

In spite of the great practical importance of precipitation, there exists a surprising dearth of reliable figures on the efficiency with which cloud processes release water from our atmosphere. Braham [4] found in a detailed analysis of mid latitude thunderstorm clouds, that only about one-tenth of all water molecules entering such clouds as vapor ever reach the ground as precipitation; the remainder is evaporated in the downdraft or is expelled by the upper divergent portions of the cloud circulation. No comparable figure for other cloud types has come to my attention, but pertinent data of a somewhat different type have been presented by Huff and Stout [5] who found that about only 5% of all of the water flowing as vapor into the space above the state of Illinois over a three-year period was precipitated upon that area, the remaining 95% simply flowing out of the space in vapor (or cloud) form. The Illinois release rate exhibited a range of from slightly over 4% to slightly over 7% from one seasonal average to another, so the mean is reasonably representative for all seasons. Reitan [6], using a somewhat different approach, found that during the summer rainy season in Arizona, about 5% of the total precipitable water vapor overhead at any given time had precipitated during the ensuing twenty-four hours, with an observed range of from about three to thirteen per cent in this daily release rate. Inasmuch as the time-period used by Reitan for the Arizona case is close to the time required for air parcels to move across Arizona at normal summer wind speeds, and inasmuch as Illinois and Arizona have linear dimensions of comparable size, these two estimates can be seen to agree rather well. Both Braham's figure for a specific rain-producing cloud type and the other two figures of more general nature have this important implication for cloud modification studies: *The natural efficiencies of removal of water from the atmospheric water vapor stock seem rather low* (when defined as above), so low as to seem to offer some hope that artificially induced increases might be achieved if sufficiently complete knowledge of all pertinent processes were at hand. Specifically, these few efficiency indicators suggest that a cloud-modification technique that accomplished no more than a 10% relative increase of the natural precipitation rate in clouds over a given area would actually represent an absolute increase of release rate of rather less than about one precentage unit on an efficiency scale defined in either of the two basic ways used just above (fraction of vapor entering a cloud, or fraction of vapor flowing across a given geographical area), which does not seem an entirely hopelessly large improvement upon nature.

Further indication of the relatively small influence of cloud seeding on the hydrologic cycle, as well as further appreciation of the time-scale of the potentially modifiable meteorological processes operating in the hydrologic cycle, can be gained by considering the following argument (see, for example, Sutcliffe [3]): Define the "turnover time" of the atmospheric water vapor as the length of time required, on the average, for world-wide precipitation processes to extract from our atmosphere an amount of water equal to its mean instantaneous stock of water vapor. Now, as the average world-wide precipitable water vapor content of the atmosphere is close to 3 cm while the world-wide average annual precipitation is about 100 cm, approximately one-thirtieth of a year, or about *twelve days*, must be required, in the mean, for one complete turnover of the atmospheric water vapor content.

If this estimate of the water vapor turnover time is correct (and it is in very good agreement with results of an independent argument based on consideration of how solar radiation dictates the evaporative rate that constitutes the equilibrant of the above-considered precipitation rate), then, because this is from many points of view a quite short time interval, one is tempted to say that atmospheric precipitation processes are not of such low efficiency after all, despite the efficiency estimates made earlier in this section. But this, it will be seen, becomes simply a matter of definition. From the viewpoint of, say, the Arizonan, a mere 5% removal rate per day seems regrettably low and worth trying to increase (and this Arizona rate is, in fact, only about half the global average rate implied by a ten-day turnover time); but in ten to twenty days the air parcels that passed over Arizona with a daily loss thereto of only about 5% of their moisture will have suffered sufficient number of encounters with circulation systems of the cloud-generating type that the probability of their having lost all of their original water vapor content approaches unity. Briefly, the fact that the entire quantity of atmospheric water vapor (exclusive of the small quantities circulating above the tropopause) is effectively removed and replenished once every ten or twelve days must surely be admitted to imply a rapid exchange rate for the world as a whole (especially so when compared with exchange rates of all other atmospheric gases, of which CO_2 is one of the most rapidly exchanged yet has a turnover time now estimated at about ten *years*). Nevertheless, this inevitably means that, for any given geographic area, *the horizontal flux of vapor overhead averages more than an order-of-magnitude larger than the vertically downward flux of precipitation*, and this does make the latter seem to be governed by "inefficient" processes. To repeat, "efficiency" is here entirely a matter of definition and viewpoint.

From our estimate of the turnover time we may now draw two conclusions relevant to the general prospects of cloud modification.

(1) If the natural hydrologic cycle has a characteristic atmospheric turnover time of only about ten or twelve days for its evaporation-precipitation link, then the rate of recovery from the globally small effects of even a very ambitious continental seeding program will be quite rapid. Thus, we seem, on these grounds, still further justified in rejecting fears

that widespread seeding at rates now envisaged by the most ardent enthusiasts could so deplete the natural stock of atmospheric water vapor as to alter appreciably other meteorological phenomena on a world-wide scale.

(2) If the turnover time is of the order of ten days, then during each day, about one-tenth of the water vapor overhead must, on the average, be returned to the earth's surface by precipitation. But earlier we concluded that an average of only about one twenty-fifth of the water substance in the atmosphere is at any one time condensed into clouds and, of this, I guessed that only about one-tenth is of a type capable of yielding significant precipitation; so at any one moment only about $\frac{1}{250}$th of the total stock of atmospheric water vapor is instantaneously contained in clouds of a type that are at or near the precipitation stage. During the course of an average day, clouds in various parts of the world continually form and dissipate and a small portion of these release precipitation before dissipating. The $\frac{1}{250}$th of the total atmospheric water that is at one moment in a certain set of clouds scattered over the world will not be found in the same set of clouds, say, an hour or two later, for these have by then precipitated or evaporated; but what we can say on a statistical basis is that, if each day world-wide precipitation must account for an average removal of about one-tenth of the total stock of atmospheric water vapor, whereas at any one moment only $\frac{1}{250}$th of that total is locked up in clouds of precipitating type, then the process of formation of, and release of precipitation from, these clouds must repeat itself some twenty-five times per day, if we assume momentarily that all condensed water in these clouds falls out. But, as a matter of fact, two opposing characteristics of clouds actually enter. One must recognize, first, a tendency for clouds to have passing through their boundaries more total vapor than that equivalent to the liquid water content present at any one instant and, second, an opposite tendency for only a fraction of all of this vapor to be converted into particles capable of actually reaching the surface as precipitation. In ignorance of the quantitative balance drawn by actual cloud systems with respect to these two opposing tendencies, I shall here simply ignore both and draw only the less precise conclusion that the entire atmosphere must continually be going through the process of creating its precipitating-type clouds, releasing their total liquid water content, creating their replacements elsewhere in the atmosphere, and so on, for an *effective* total of about twenty-five repetitions per average day. This crude yet physically meaningful conclusion that has been drawn from foregoing estimates of scale factors and turnover rates of the atmospheric water economy is thus found capable of yielding an interesting estimate of the "effective lifetime" of precipitating-type clouds, namely one twenty-

fifth of a day or about *one hour*. That this effective lifetime comes out fairly close to actually observed lifetimes, at least in the case of convective-type clouds (order of tens of minutes), is indicative once more that all of the magnitudes being employed in the present discussion must be tolerably close to the true values and thus strengthens the claims to order-of-magnitude validity of my deductions concerning scale factors that must govern hydrologic effects and thermodynamic effects of currently envisaged cloud-modification programs.

The short lifetimes of clouds of most types (order of a few tens of minutes in agreement with the estimate just made) and the comparative infrequency of occurrence of the precipitating types, combined with the limited areas of direct influence of any known cloud-modification techniques (to be described in the following), can be used to support still further argument that no hazardous disturbance of natural events is imminent. If we take the global mean cloudiness fraction used above, namely one-half, and then reduce this by the estimated factor of one-tenth in order to dispose of the numerous nonprecipitating cloud types that are counted into the cloudiness fraction itself, we have a twentieth of the world covered at any one time by modifiable clouds. Taking as a seemingly generous figure 100 km^2 [1] as the area that might be influenced by, say, a single cloud-seeding generator of a generally used type operated in an area with potentially modifiable clouds, then no less than 4000 generators would be required to seed the modifiable clouds lying over an area equal to that of the United States at an average moment, and about twenty times that number, or some 100,000, would be required to insure that modification could be effected at any and all times and places at which treatable clouds happened to appear. It seems rather unlikely then that, in the near future, man's efforts at rainmaking will even approach the rather tiny upper limit of the maximum fraction (one ten-thousandth) of total atmospheric water vapor which he might be drawing upon on a continuing basis by known modification methods.

Do all of these considerations of the relatively minute scale of prospective tapping of the atmospheric water vapor reservoir therefore imply that there can be no practical significance in the recent development of cloud-modification techniques? Not at all. They indicate only that such efforts can scarcely be expected to interfere sensibly with the water- and energy-exchanges that our atmosphere is continually accomplishing on so vast a scale. That which is comparatively insignificant on the scale of geophysical magnitudes is quite often highly significant as judged by our own human standards. There appears, then, to be good reason to

[1] Mean drift rates plus photolytic decay rates (Section 4.2.1) combine to give an effective area of this order of magnitude.

pursue research on techniques of cloud modification for, although the complexity of the underlying problem has defied complete solution even after a decade of investigation, the ultimate prospects remain most intriguing.

A final order-of-magnitude calculation in this section devoted to the gross features of the atmospheric steps in the hydrologic cycle will help the reader unfamiliar with atmospheric energy-magnitudes to understand why, at present, there are no very bright prospects for modifying the *hydrodynamic motion fields* that are prerequisite to the very existence of clouds. The point here is that precipitation presupposes condensation, and condensation on a scale intense enough to form cloud masses of significant depth presupposes strong updrafts in which adiabatic cooling brings the vapor in the rising air to its condensation point. All meteorologists are familiar with one or another layman's scheme for creating clouds by some kind of forced upward motion, and in every case there appears the same failure to recognize the enormous amounts of energy nature shuffles about in everyday atmospheric processes. One simple means of estimating the magnitude of the energy involved in a particular case is very relevant here—namely, the calculation of the total latent heat released in a typical thunderstorm. It will be reasonable to consider a thunderstorm downdraft a kilometer in radius in which rain is falling at such a rate and for such a time that one centimeter of rain falls over all of the area of the downdraft during the lifetime of the storm (say, 30 min). A total mass of water amounting to 3×10^{10} gm will have undergone phase change in such a thunderstorm from vapor to liquid state, with about 600 cal released locally into the atmosphere for each gram condensed, or a total of approximately 2×10^{13} cal of released latent heat. The largest single energy source currently at man's disposal is the nuclear bomb, and for comparison, we may note that the energy released in a nominal atomic bomb of the fission type has been set [7] at 2×10^{13} cal, or exactly the magnitude of latent heat release calculated for the single, moderate thunderstorm considered above. Noting then that Braham [4] finds that the total condensation occurring in a typical thunderstorm may be about ten times greater than the amount indicated by just the precipitation reaching the ground, and realizing that latent heat release is but one part (though, to be sure, an important part) of the total energy exchange in a thunderstorm, it becomes clear that even small thunderstorms constitute mechanical systems in which energies in excess of the equivalent of ten World War II atom bombs are involved. This surprising magnitude points to the seeming futility of hoping to modify weather by methods directed toward the creation of clouds themselves.

There are, to be sure, possibilities of still unsuspected trigger mechanisms whereby expenditures of small amounts of energy might release large amounts of atmospheric potential energy in statically metastable air masses. One of these that has received more than passing consideration concerns alteration of the albedo of the ground over a large enough area to locally heat the atmosphere to the point of inducing convection and thence cloud formation, and another involves controlled fires set on a scale large enough to initiate updrafts in an unstable air column. At present these schemes appear to have only marginal interest. For the present, the only real hope, then, seems to be that of manipulating certain of the key microphysical processes occurring within clouds wherein relatively tiny expenditures of energy and material can conceivably alter the course of events set in motion by the tremendous natural energy exchanges that are prerequisite to cloud formation itself. A discussion of the physical details of these critically important microphysical processes that may be amenable to modification will be the subject of the next section.

3. Present Status of Cloud and Precipitation Physics[2]

3.1 Historical Remarks

One aspect of the history of the development of cloud physics which seems to me to be quite significant is its very slow development prior to about ten years ago. When one considers the great importance of precipitation in man's activities, above all in agricultural activities, when one considers in how many ways the past decade's research has been obstructed by sheer lack of many kinds of basic observational data on cloud and precipitation processes, and finally, when one considers the almost complete lack of any really exhaustive theoretical analyses of basic cloud-physical hypotheses carried out earlier than a dozen years ago, the historical background is rather disquieting.

The question that is of much more than historical interest is as follows. Why this comparative neglect of so fundamental a problem for so long a time? The answer seems to me to be that we face here one more of those many instances in the history of science where far too little research support was given to investigations while they were *apparently* of only academic interest. When, after 1946, there seemed to exist some prospect of control over a natural phenomenon whose economic value is so high, support of cloud physics research jumped by, what I would estimate must surely have been, a factor of two to three orders of magnitude, and total numbers of workers in the field must have increased by a factor of something like two orders of magnitude. Yet so complex are the phenomena

[2] A comprehensive book by B. J. Mason [7a] covering many aspects of cloud physics has recently appeared.

one encounters in attempting rational modification of precipitation, that even after a decade of investigations at these unprecedented levels of support, meteorologists still face many very fundamental questions not yet answered. Had fundamental meteorological research been sustained during earlier decades at a level in keeping with the importance of meteorological phenomena in all man's affairs, this embarrassing dearth of basic observational and theoretical background would not have so limited rapid progress toward evaluation of, and improvement upon, the discovery in 1946 of means of modifying cloud processes.

To illustrate the slow pace of progress in just the theoretical side of cloud physics in earlier years, no example seems more revealing, in my opinion, than the history of the ice-crystal hypothesis of precipitation, which will be examined in some detail later in this article. In 1911, A. Wegener, called attention to the difference in the vapor tensions (i.e., saturated vapor pressures) of subcooled water and ice at given subzero temperatures, yet twenty-two years elapsed before this observation was formally recalled by T. Bergeron who, in 1933, suggested that this vapor-tension difference might be responsible for the release of most, if not all, natural precipitation. And, even more curiously, an additional seventeen years then elapsed before a really adequate theoretical analysis of this hypothesis was given by Houghton [8]; yet the theoretical instruments that finally laid bare the relevant quantitative aspects of the Wegener-Bergeron hypothesis were, in all essential respects, already quite well developed in Wegener's day. This forty-year lag may be illuminatingly contrasted with what has often been no more than a lag of a few weeks or months between appearance of a new hypothesis in nuclear physics and its thorough theoretical and experimental testing. The difference almost certainly lies in the difference in total numbers of well-trained persons vigorously seeking to exploit every perceptible clue to the problems of these two fields. Since, even today, the field of cloud physics is not advancing at a pace that seems commensurate with the importance of its subject, the lesson of the past forty-odd years seems still to be quite timely: There is need for many more capable workers in this field.

3.2. The Condensation Process

The only physical process capable of cooling to the vapor saturation point large enough volumes of air to yield significant amounts of liquid water in droplet form is that of adiabatic expansion of ascending air, if by "significant" we understand "large enough to support natural precipitation processes." Air may ascend in a wind current impinging on a mountain slope or along a frontal discontinuity, it may be forced to ascend by dynamical convergence in a cyclonic circulation, or it may be caused to ascend as a result of buoyant forces. During any such ascent to levels of lower pressure, the air parcels perform expansion work against their surroundings, causing their temperature to decrease at the dry adiabatic rate of 9.8°C/km, and producing what should be regarded as a very odd phenomenon, namely, the tendency for any water vapor contained in the ascending air to approach and (with sufficient lifting) to reach vapor saturation. This is odd in the thermodynamic sense that whereas most vapors become *less* saturated upon undergoing expansion, water vapor, because of its anomalously high latent heat of vaporization, behaves oppositely; so we find clouds of drops of condensed water in our atmosphere occupying the *upper* limits of *updrafts*, rather than the lower limits of downdrafts. However, our familiar clouds must be regarded as curiosities not only on that score, but more so on the very fact that they occur at all, for phase transitions from vapor to liquid are always powerfully inhibited by the activation-energy barrier imposed by the appearance of surface free energy during formation of droplets of the liquid phase within an initially homogeneous vapor phase. Only with meteorologically unheard of supersaturations (relative humidities in excess of several hundred per cent) would droplets appear in an adiabatically cooling updraft if they had to be formed by homogenous nucleation, that is by passing through stages where polymers of two, three, four water molecules, and so on, grew by random collision processes. The classical problem of atmospheric condensation theory might well be said to be that of accounting for the improbable but observed behavior of terrestrial clouds which do invariably form as soon as rising air is cooled to just its *nominal* saturation state (i.e., to a state characterized by temperature and vapor pressure values which would insure molecular equilibrium with a plane surface of pure liquid water if such a surface were present). It has been known for a long time that the clue to this improbable behavior is that our atmosphere contains small but very significant concentrations of particles of such chemical composition as to be hygroscopic and of such size that, though submicroscopic, they are nevertheless large enough and have sufficient affinity for water that they can overcome the free-energy barrier that blocks the path of homogeneous nucleation of pure vapor. These particles are collectively termed *condensation nuclei*.

The basic thermodynamic principles of the meteorological condensation processes have been fairly well understood for over seventy years, since the days of Hertz and Neuhoff; the existence of the free-energy barrier has been known since 1870 when Kelvin derived its mathematical expression; and the inference that ordinary air must contain some kind of nuclei of condensation dates from work of Coulier in 1875. But the full elucidation of the true physical and chemical nature of the effective nuclei of condensation upon which cloud formation depends cannot yet be said to be achieved. Since a good summary of the status of the problem of the atmospheric condensation nuclei has been given recently by Junge [9] (who has also been the chief contributor to recent advances in our understanding of the condensation nuclei), this facet of cloud physics will be passed over here rather briefly, with only a few comments on those aspects of the condensation problem that influence precipitation and cloud-modification processes.

First, it is very important to stress, for the benefit of the nonspecialist, that all measurements of the populations of atmospheric nuclei are in essential agreement on one critical point: There appear at all times and in all parts of the world to be sufficient numbers of hygroscopic condensation nuclei that cloud growth begins at only an immeasurably higher altitude than the level of nominal saturation (i.e., at the "100% relative humidity" level) in an adiabatically cooling updraft of air. That this circumstance is not simply to be taken for granted is clearly shown by the equally well-established observation, to be discussed below, that the atmosphere invariably displays a quite significant *deficiency* of another class of nuclei, the so-called *ice nuclei*. These two very important observational facts can be restated, for emphasis, in the following way. The water substance in our atmosphere (at least in the troposphere) never exhibits marked vapor supersaturation, yet it characteristically displays very marked liquid subcooling. Cloud-modification possibilities, at least as far as concerns any techniques now seriously considered, are profoundly influenced by these two observational facts. The first precludes control of precipitation through any step that might be termed artificial cloud *formation*, inasmuch as wholly *natural processes are already entirely competent to produce clouds whenever adiabatic ascent occurs*. But the second fact implies, as will be elaborated below, that nature might occasionally profit from assistance in the form of modifications artificially made in the *ice nuclei* populations that play, in certain precipitation processes, a quite critical role. The all-important distinction here is the distinction between *condensation* nuclei and *ice* nuclei, a distinction which is too often overlooked by nonmeteorologists, especially in discussions of cloud-modification techniques.

Inasmuch as condensation from the vapor phase may involve growth of either liquid drops or solid crystals, it would be logically appropriate to discuss in this section the growth of ice particles by direct condensation onto types of nuclei called sublimation nuclei (and more fully discussed later in this article). Although the point is not yet fully settled, it appears likely that such a growth process may often account for production of cirrus clouds at high altitudes. It is also likely that such processes may be indirectly related to precipitation phenomena when those ice particles fall into subcooled clouds at lower altitudes. Since the growth of cirrus clouds themselves is beyond the scope of this paper and because the general role of ice crystals in precipitation processes will be discussed later in Section 3.3, no discussion of vapor-to-solid condensation will be given here.

A significant clarification of the details of cloud droplet growth in adiabatically cooling updrafts was accomplished by Howell [10], who assembled into a single system of differential equations a description of the numerous physical processes that govern particle growth from hygroscopic nuclei to full cloud-drop sizes. Prior to Howell's work, all of the contributing processes were individually understood reasonably well; his contribution was the mathematical synthesis of this information and the numerical integration of a small number of special cases which displayed quantitatively several highly important features of the condensational growth of drops in the lowest few hundred meters of a cloud. First, his results, even though they cannot be regarded as final and have been shown to contain a few small errors, show beyond doubt that the amount of vapor supersaturation that can develop near a cloud base, with normal numbers of hygroscopic nuclei present in the rising air, is of the order of only a *few tenths of a per cent* in all but the strongest of convective updrafts; and for the latter cases his results suggest, but do not show with finality, that supersaturations of perhaps no more than one or two per cent may be expected as upper limits. This low upper limit was anticipated by Houghton in 1938 [11] but is much more fully documented by Howell's results. It must not be thought that so low a peak supersaturation is also clearly deducible from direct cloud observation, for even a one per cent supersaturation represents an elevation increment of only a few meters in level of appearance[3] of cloud drops in a typical case, and so small a discrepancy can scarcely be detected observationally with presently available methods. Second, and more important for the theoretical clarification of cloud processes leading to precipitation, Howell's results indicate that only a surprisingly narrow range of cloud-drop sizes can be formed by solely condensational processes, acting upon commonly observed nuclei populations, for the diffusional growth law is of such functional form as to cause the smaller nuclei to catch up with the growth of larger nuclei once the former are activated for growth by the rising supersaturation, as can be seen in Fig. 1. This inherent tendency toward monodispersity of clouds produced solely by condensation processes, recognized and briefly discussed as early as 1933 by Houghton as a general characteristic of the growth of spheres by diffusion, was shown by Howell's analysis to be so strong as to demand greatly increased attention to the effects of all those collisional processes *not* taken into account in his growth equa-

[3] Or, more accurately, the level of sudden increase in drop growth-rate.

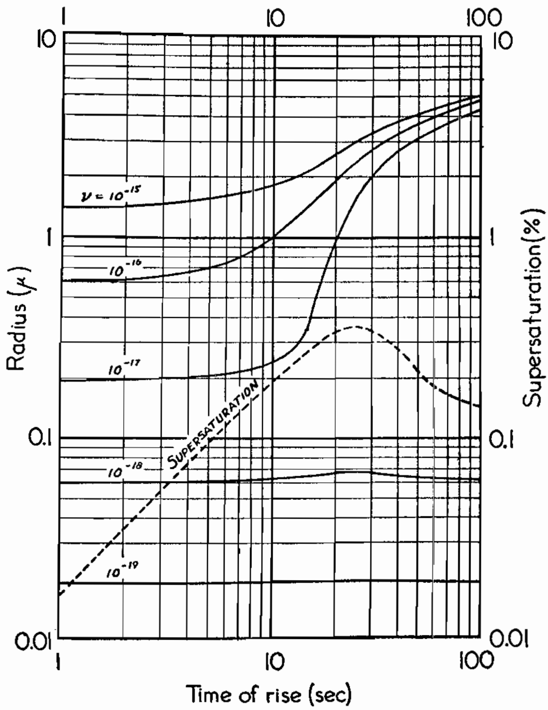

FIG. 1. Condensational growth curves for a weak updraft (0.6 m sec^{-1}) containing few nuclei (total 500 cm^{-3}). Abscissa is time since the air parcel passed the nominal saturation level. Values of ν are numbers of moles of salt per nucleus. Note that nuclei of 10^{-17} M are barely activated by the peak supersaturation, 0.35%, while those of 10^{-18} M or less are not activated at all in this updraft. The small range of drop sizes finally produced by purely condensational processes is clearly evident. From Howell [10], by courtesy of the American Meteorological Society.

tions—a case of a very significant insight being gained by a somewhat negative result, since observed drop-size distributions even for lower layers of clouds exhibit much broader ranges than seem obtainable by any purely condensational process, judging now on the basis of Howell's 1949 analysis.

Others have added to present knowledge of condensation physics. Kraus and Smith [12] carried out, almost simultaneously with Howell, a number of analyses of drop-growth, but their integral equation formulation omitted the quite important effects of the warming of growing cloud drops by release of latent heat of condensation, effects which Howell incorporated into his differential equation quite cleverly. Omission of this heating term might easily pass for a negligible error, since examination reveals that it accounts for a drop-warming effect of the order of only

0.01°C; yet so small are the vapor pressure gradients driving the diffusional growth of cloud drops that this tiny temperature increase raises the growing drop's vapor tension enough to roughly cut in half the mean vapor pressure difference from drop to ambient air! In another study, Squires [13] carried through a number of illuminating computations of factors involved in condensation growth, but unlike Howell, he did not integrate a complete system of equations depicting the interplay of processes actually found in the cloud. There remains great need for extension of Howell's work, using automatic computer methods to examine many more combinations of updraft speed, condensation level, and nuclei populations. There are also residual uncertainties in the adequacy of the type of diffusion law that underlies Howell's analysis, so this cannot yet be regarded a closed problem.

A final point to be considered under condensation physics as it bears on cloud modifications concerns the still somewhat controversial role of very large hygroscopic nuclei. It is necessary to explain here what is to be understood as a very large nucleus. The terminology, which has acquired standard usage is as follows. Particles with radii below about 0.1 μ are termed *Aitken nuclei* (after the developer of a simple expansion-chamber nuclei counter), those with radii between about 0.1 and 1.0 μ are called *large nuclei*, and those with radii in excess of about 1.0 μ are referred to as *giant nuclei*. Junge has gathered considerable evidence (see, for example, [14]) that the number concentration of the atmospheric aerosol rises very rapidly with decreasing size toward a maximum number in the Aitken range (the still smaller particles having such high mobility as to go over rather quickly by Brownian coagulation into Aitken nuclei). His evidence suggests that most of the so-called large nuclei are ammonium sulfate (or at least are rich in the ions of that salt), though the true nature of the source of such particles is still obscure. The Cl^- ion found abundantly in sea water is notably absent from the class of large nuclei, but makes up a substantial fraction of the giant nuclei. The Aitken nuclei are so small that they are not activated by the relatively small supersaturations now known to occur in cloud updrafts, while, on the other hand, the giant nuclei are less numerous by several orders of magnitude than the large nuclei; so there now remains little doubt that the bulk of cloud condensation takes place on the *large* nuclei, those with radii of from 0.1 to 1.0 μ.

However, despite the small numerical importance of the giant nuclei, considerable interest in this class of nuclei has recently developed as a result of the past few years' findings relative to the importance of collision and coalescence processes (accretion processes are discussed in Section 3.3.3) in producing precipitation. As will be elaborated below, collisions tend preferentially to occur between drops of dissimilar size. But Howell's theoretical results show that condensation on populations of large nuclei yield cloud drop-size distributions that are too narrow to yield appreciable collisional growth directly, so a number of investigators have stressed the potential importance of the comparatively few *giant* nuclei in starting the coalescence process. The work of Woodcock and his collaborators (see, for example, [15] and [16]) has been particularly important in the past ten years' developments in our knowledge of the distribution of these giant sea-salt nuclei which may play a critical role in at least some kinds of precipitation processes. Details must be passed over and the single point made that herein we find the one way in which cloud modification of a sort capable of influencing *precipitation* has been considered possible through manipulation of the population of *condensation* nuclei. For existing evidence suggests that continental air masses will frequently have too few giant nuclei of the size necessary (order of 10–20 μ in radius on entry into the cloud base) to start the accretion process immediately at a rate that is significant. Hence, the argument proceeds, one might artificially stimulate the accretion processes by adding to updraft air a rather modest number of very large salt particles. More will be said of this approach later (see Section 4.3), but before leaving this brief discussion of condensation as it bears on cloud modification, the reader must be cautioned to note that any such addition of a relatively small number of *giant* nuclei to existing natural populations of the much more numerous *large* nuclei cannot at all be construed as artificial *cloud formation*: the generalization still stands that no promising means are yet at hand for influencing the hydrologic cycle through anything that can properly be called artificial *production* of clouds. Nature, for the present, must supply the clouds.

3.3. The Precipitation Process

Condensation is necessary but not sufficient to account for precipitation on the scale characteristic of the earth's atmosphere, where some 10^{19} gm falls upon the entire world on an average day. But, despite its great importance, the mechanism by which the atmosphere accomplishes precipitation of portions of its total stock of water from time to time has long resisted satisfactory explanation. In the next four sections, the nature of the problem and some recent progress toward its solution will be briefly examined in order to delineate the background against which cloud-modification studies must be viewed.

3.3.1. The Central Problem of Precipitation Theory. Numerous observations, especially during recent years, from mountain observatories and from research aircraft, stand in good agreement with respect to the order of magnitude of the number-concentration of cloud drops: some hundreds per cubic centimeter. This concentration becomes higher in clouds of the convective type where strong updraft speeds lead to the higher peak supersaturations (order of a few per cent, as noted earlier) that permit activation of the more numerous smaller nuclei, and becomes lower in cloud types associated with slowly ascending air, where only the larger nuclei are activated; but 100 cm^{-3} to 1000 cm^{-3} would seem to represent reasonably well the lower and upper limits, respectively, to the commonly occurring range of cloud-drop concentrations.

It is necessary to distinguish, at least in a gross way, between cloud drops and raindrops. There is, of course, no absolute criterion, since there exists a continuous distribution of water drop-sizes from embryonic cloud drops of less than a micron in radius to the largest raindrops that are mechanically stable in normally turbulent air, namely, those with radii in the neighborhood of five millimeters. Nevertheless, the fact that drop radii are commonly of the order of 10 μ in clouds of nonprecipitating types whereas modal raindrop radii in most heavy showers are of the order of 1 mm, puts two orders-of-magnitude separation in radial dimensions between clearly cloud and clearly raindrops. Just one order of magnitude from each is the drop of 100 μ in radius, which makes this size a nicely symmetric dividing point between the two classes of particles. About the same division happens to be indicated by another, more physical, criterion: Whereas a 10-μ drop has a terminal falling speed of about 1 cm sec^{-1}, too low to permit it to fall through even very gentle cloud updrafts, and whereas an unstably large raindrop of 5 mm radius falls with a terminal velocity of about 9 m sec^{-1}, rather larger than most cloud updrafts, a drop of 100 μ radius falls at about 70 cm sec^{-1}, about equal to updraft speeds in weak convective clouds. For these and other reasons, it is common to take the 100-μ radius as the arbitrary point of separation, with smaller particles being regarded as large cloud drops, and larger particles being considered small drizzle drops.

Now, condensation processes alone will yield their largest cloud drops when acting with the fewest possible nuclei in a cloud whose base is as low in the atmosphere and at as high a temperature as is meteorologically attainable, and when the rising air parcels are carried by convection to the greatest possible altitudes. Hence, to place an upper limit on cloud-drop sizes attainable solely by condensation, we might imagine a tropical cumulus with a base at only about 500 meters altitude and base temperature of 30°C, which extends to the tropopause at about 16 km altitude, and we may take the drop concentration to be as low as 100 cm^{-3} near the base. By a simple calculation based on the thermodynamics of the saturated adiabatic process we find that by the time a given air parcel has been transported adiabatically to the cloud-top level in such an extremely favorable case, each of its drops will have grown, considering only the effects of condensation processes and assuming as a crude approximation to Howell's picture that all drops grew at equal rates, to a radius of *merely 40 μ*. A drop of this small size, falling back through the cloud at its terminal velocity of about 20 cm sec^{-1} would require almost exactly one day to reach the cloud base even if all updrafts could somehow be avoided, and it would then completely evaporate after falling only about a hundred meters in the nonsaturated air just under the cloud base. Thus, we find that such a process is quite incapable of producing precipitation in the ordinary sense of the term.

In contrast to the above hypothetical case of the most favored combination of circumstances dependent solely on condensational growth, one observes that *actual* clouds that yield heavy precipitation often have drop concentrations substantially larger, have bases much higher and colder, have tops much lower, have strong updrafts through which precipitation particles must descend; and yet release their precipitation, in the form of drops with radii as much as two orders-of-magnitude larger than in the above case, in times of the order of only an *hour* after first formation of the cloud.

Closer scrutiny of these two cases quickly reveals that the essential difference between actual precipitating clouds and our hypothetical most-favored cloud, supporting only purely condensational drop growth, is a difference in rapidity of development of *very large drops* (raindrops) whose terminal fall velocity is great enough to enable them to descend gravitationally through the often strong updrafts within the cloud and to pass quickly through the nonsaturated clear air between cloud and ground without wholly evaporating. This difference, quantitatively put, is the difference between condensationally produced *cloud drops* with radii of the order of 10 μ and observed *raindrops* with radii of the order of 1 mm. In these two magnitudes is implied the central problem of precipitation theory: *to account for the conversion of some million cloud drops into a single precipitation particle in a time of the order of only an hour after condensation starts at the base of an incipient cloud.* The comparative particle sizes, concentrations, and terminal falling speeds of the several particle types encountered in cloud physics are drawn to scale in Fig. 2 to display the important disparities in these magnitudes for cloud and precipitation particles.

Only within the past decade have meteorologists acquired what might justifiably be called a general understanding of the physics of these aggregation processes. Clearly, rational application of any prospective cloud-modification techniques aimed at augmenting precipitation will demand very detailed knowledge of these processes, and indeed it has been the recent discovery of conditionally effective modification techniques that

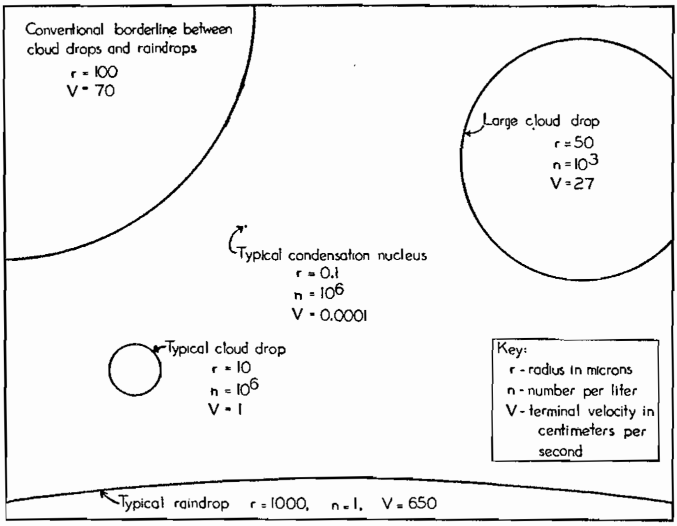

FIG. 2. Comparative sizes, concentrations, and terminal fall velocities of some particles involved in condensation and precipitation processes. Note particularly the great difference in radius of a typical cloud drop and of a typical raindrop.

has stimulated most of the research that underlies our present tentative answers to the central problem of precipitation theory. It now appears that there are just *two* essentially different aggregation processes, or more accurately, just two essentially different ways in which the aggregation process can be *initiated*. One of these is called the *ice-crystal process*, the other is the *accretion process*. Many other processes have been postulated in the past, but all other hypothetical processes have been winnowed out by the stringent rate requirements imposed by the surprisingly short lifetimes of individual clouds.

3.3.2. The Ice-Crystal Process. The ice-crystal process, also referred to frequently as the Bergeron process or the Bergeron-Findeisen process, hinges upon the coexistence of ice particles and subcooled liquid water drops within a cloud at temperatures below 0°C. That liquid water is quite frequently found in the subcooled state in clouds is not, I believe, common knowledge among nonmeteorologists. This fact certainly seems to contradict every-day observations of freezing phenomena one sees, for example, in winter in high latitudes. However, meteorologists have known for at least half a century that it is much more typical to find clouds at a temperature of, say, −5 to −10°C, containing only subcooled liquid drops than to find clouds in that temperature range comprised solely of ice crystals. Furthermore, about twenty years ago, difficulties with icing of aircraft (which can only occur in the presence of subcooled water and never in penetrating clouds comprised wholly of ice particles) had revealed that subcooling to at least as low a temperature as about −35°C occasionally occurs in clouds. (It deserves passing note here that the term "subcooling" is preferable to the commonly used "supercooling," and the latter should be abandoned.)

The reason why clouds are so susceptible to subcooling was not clarified until quite recently. Textbooks on meteorology written as little as half a dozen years ago still contained references to lack of mechanical vibration in clouds, or obscure references to suggested effects of surface tension on the freezing temperature, as possible explanations, and the general impression with which the student was left was that this common tendency toward marked subcooling in clouds was an anomaly not clearly explainable. Actually, subcooling in cloud drops is not anomalous at all and, in fact, the situation would only be truly thermodynamically anomalous if the reverse case were true, that is, if all clouds at subzero temperatures were pure ice clouds.

It is now known as a result of recent theoretical work (see, for example, Krastanow [17], Turnbull and Fisher [18], Mason [19], and McDonald [20], of which the last reference contains a general review of meteorological implications) that pure water (and any other pure liquid) undergoes subcooling for reasons associated with the kinetics and thermodynamics of phase change. Freezing in a mass of pure liquid water can only begin as a result of random collision processes that form an embryonic ice crystal one molecule at a time, and such a process is astonishingly improbable without appreciable subcooling. Thermodynamic analysis of this process reveals that there is a critical embryo size, at any given temperature, below which the embryo is unstable and tends to dissociate but above which spontaneous and rapid growth to a macroscopic ice crystal takes place. Throughout a mass of subcooled water, chance collisions tend continually to build up fractions of such embryos and thermal disgregation tends continually to dissociate them; but with increasing degree of subcooling, the later tendency is suppressed in such a way that the probability of molecular fluctuations forming an embryo of critical size in a given mass of subcooled water in a given time interval rises toward unity. Theoretical prediction of the temperature at which a cloud drop of given size should undergo this kind of *homogeneous nucleation*, as the above process is called, is at present blocked by ignorance of the exact value of the surface free energy of an ice-water interface, for this elusive parameter (whose value must be close to 15–20 ergs per square centimeter) plays a numerically decisive role (its cube appears in an exponent!) in the theoretical expression for the nucleation probabilities. Nevertheless, the theory has been quite enlightening in that it accounts for subcooling of pure water drops on general statistical and thermodynamic grounds and shows that whatever the actual ice-water surface energy is, the consequent nucleation rates will be extremely sensitive to temperature in such a way as to cause a cloud of the usual drop-size distribution to undergo homogeneous nucleation almost *en masse* once a certain critical temperature is approached. There is now considerable evidence (see, for example, Schaefer [21], Bigg [22], Jacobi [23], and Pound *et al.* [24]) for believing that with drops of the order of 10 μ in radius, the critical temperature required for homogeneous nucleation to occur within times of the order of seconds is very close to $-40°C$ (reported values range from about $-38°$ to $-41°C$).

The chief point to be stressed in explaining cloud subcooling is that a cloud is an unusual system in that its mass is distributed over a very large number of very small particles, each *one* of which must undergo a nucleation event somewhere within its boundaries before the cloud can become wholly frozen, and this is vastly different from the process required to freeze, say, a gallon of water in a single container. In the latter case, a single nucleation event taking place *anywhere* within the container suffices to cause the entire mass to freeze, hence the familiar resistance to subcooling in ordinary quantities of water.

Now, it is tempting to conclude at this point that subcooling to as much as about -35 to $-40°C$ in clouds is solely due to the effects of drop *size* on the probability of homogeneous nucleation in such tiny masses of water. But this view cannot be correct. So extremely rapidly does the homogeneous nucleation rate vary with temperature, that temperature effects completely dominate over volume effects. Thus, if it is indeed true (as one may now strongly suspect) that drops of about 10 μ radius will undergo homogenous nucleation in times of the order of seconds or minutes near $-40°C$, then raindrops of 1 mm radius will undergo homogeneous nucleation at a temperature only a few degrees warmer (somewhere near -36 to $-38°C$, the exact value depending on the still undetermined specific surface free energy of the ice-water interface). The lower degrees of subcooling actually observed in most laboratory studies with drops intermediate in size between cloud and raindrops must be due to some still undetermined effect of size on probability of containing or capturing a *freezing nucleus*, that is, a foreign particle that can induce so-called heterogeneous nucleation of the freezing process at only a modest degree of subcooling. The concept of the freezing nucleus is so important to both the basic theory of the ice-crystal process and the theory and practice of many types of cloud-modification techniques that it, and closely related concepts, must be examined in more detail here before returning to the elaboration of the ice-crystal process itself.

Terminology has at times, in the past years, been regrettably confused with respect to various types of nuclei, but the following nomenclature, first formally proposed by Fournier d'Albe [25], has acquired fairly general usage. A *freezing nucleus* is any small foreign particle whose size and crystalline structure permit it to serve as a growth center for an ice crystal when it is in contact with liquid water at subzero temperatures. A *sublimation nucleus* is any small foreign particle whose size and crystalline structure enable it to serve as a growth center for an ice crystal that is built up, not by solidification from the liquid phase, but by sublimation (the meteorologist's term for direct condensation to the solid state) from the vapor phase at subzero temperatures. A generic term embracing both of these two classes of nuclei is often convenient, and *ice nucleus* is the noncommittal term that subsumes both.

The term *sublimation* was first introduced into meteorological terminology in 1911 by Wegener and has acquired widespread usage in the sense employed in the term *sublimation nucleus*, yet this is a very misleading usage. Physicists always restrict the meaning of the term to the phase change from solid to vapor; so to speak of a *sublimation nucleus* is to the physicist a direct contradiction in terms. As a desirable substitute, the term *deposition* will be used here to denote the phase change from vapor to solid, so here *deposition nucleus* will be used in preference to "sublimation nucleus."

In both types of ice nuclei, size is important because the Kelvin effect inhibits growth of too-small crystallites in exactly the way it inhibits growth of too-small liquid drops (Section 3.2), and in general, ice nuclei should be larger than about 0.1 μ in radius to be effective, though even those an order-of-magnitude smaller can provide a substantial improvement over truly homogeneous nucleation. Crystal habit and lattice dimensions of the nucleant are now known to be very important, since a surface layer of water molecules will only be electrostatically bonded to the ice nucleus in a configuration compatible with the true ice lattice if the nucleus has a crystalline structure closely similar to that of ice. A mismatch of only a few per cent in any of the relevant lattice distances may prevent effective action as an ice nucleus except at appreciable degrees of subcooling, and this law of crystal physics has fundamental implications in the problem at hand. *It cuts down to virtually zero the number of naturally occurring crystalline atmospheric dusts with structural characteristics that permit heterogeneous nucleation of either the freezing or deposition processes at, or within a few degrees below, 0°C.* That is, it appears to be one of the geophysical facts of life that, though our atmosphere contains abundant numbers of foreign particles sufficiently hygroscopic to function very efficiently as *condensation* nuclei, it does not happen to satisfy nearly so well the very much more stringent demands of the *crystallization* process. As a result, we observe (as has been noted earlier here) that vapor supersaturation is almost a nonexistent state in our atmosphere whereas liquid subcooling in clouds is extremely common.

It would be logically next in order to discuss the exact chemical and physical nature of the principal freezing and deposition nuclei found in the atmosphere and their effective nucleation temperatures, but knowledge has not advanced far enough to permit this. There is not even agreement as to whether it is chiefly freezing nuclei or chiefly deposition nuclei that limit the commonly observed degrees of subcooling. Schaefer [26] has expressed doubt that significant numbers of *freezing* nuclei are operative in initiating ice-crystal growth in subcooled clouds, basing his view on the rarity with which microscopic examination of snowflakes discloses a frozen mass of cloud-drop size at the center, which is a reasonable, though as he points out, not decisive criterion. By testing in an experimental cold chamber a variety of natural desert and volcanic dusts of types that might become windborne in arid and semiarid regions, Schaefer [26] has found that there are many commonly occurring dusts that become fairly good deposition nuclei in clouds with subcooling to about −15 to −20°C. His results are reproduced in Fig. 3, which shows that at least one type of soil from the Northern Plains area exhibits a threshold of activity not far below that of the favored artificial cloud-seeding agent, silver iodide (which is discussed in Section 4.22). Isono [27] examined electron diffraction patterns of Formvar replicas of natural snowflakes and found evidence that many contained aluminum silicate materials of the clay type, tending to support Schaefer's suggestion concerning the role of silicaceous dusts. However, the technique is difficult and the identification somewhat indirect, so it is by no means decisive. Our knowledge of the chemical and physical characteristics of natural ice nuclei is, today, only about as well developed as was our knowledge of the nature of condensation nuclei some thirty years ago. It is certainly to be hoped that, just as surprising innovations in detection methods have recently been made in studies of condensation nuclei, so also will more delicate techniques soon be discovered for ascertaining the types and sizes of natural ice nuclei, since this kind of information would be extremely valuable in understanding how nature nucleates the ice-crystal processes which man now seeks to manipulate.

Observations such as those displayed in Fig. 3 are not in themselves sufficient to settle the question of the nature of the effective ice nuclei in the atmosphere, for one must also know the *concentrations* in which any of these dusts actually occur at cloud altitudes. Natural abrasion processes do not yield dusts with particles much under a few microns in diameter, and dust particles of that size will fall about a kilometer in a time of the order of ten days. Too little is known of the climatology of dust to enable

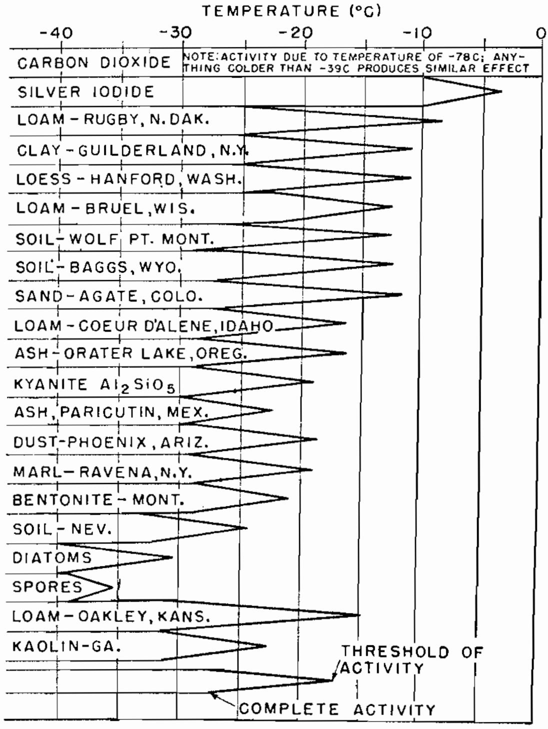

FIG. 3. Temperature range of nucleating activity of crystalline dusts from desert areas and volcanic deposits. From Schaefer [26], by courtesy of the American Meteorological Society.

one to decide how the dust addition rate would counteract that fallout rate, but since the fallout rate is not negligible, and since the altitude that the dusts of Fig. 3 must attain to become of cloud-physical importance is of the order of five kilometers, one is prepared to believe that the number concentration of ice nuclei at cloud altitudes will be rather low. Table II summarizes the results of a month of daily counts of ice nuclei made in the winter of 1955 at aircraft altitudes up to 15,000 ft in the free atmosphere over Tucson, Arizona, in prevailingly westerly flow [28]. These data for Arizona are sufficiently like corresponding data obtained elsewhere, as for example, the data for eastern Australia obtained by Smith and Heffernan [29] and still more recent nuclei counts taken over England by Murgatroyd and Garrod [30], to suggest that they may represent world averages tolerably well. Such magnitudes have close bearing on the principles of the ice-crystal process of precipitation and its artificial modification, and we are now able to return to the specific discussion of that process having exam-

TABLE II. Summary of the average temperatures (°C) at which the ice-crystal concentrations rose to specified values at three altitudes over southern Arizona during January 3–31, 1955. (From [28].)

Altitude (feet)	Ice-crystal concentrations			
	0.1/liter	1/liter	10/liter	100/liter
Sfc.	−25	−27	−28	−30
5000	−26	−29	−31	−33
15,000	−27	−29	−31	−33

ined sufficiently closely, for present purposes, the phenomenon of cloud subcooling and the phenomena of homogeneous and heterogeneous nucleation.

Given the existence of widespread tendency toward cloud subcooling, a question arises. What will occur when, in a cloud of metastable liquid water drops at subzero temperatures, a few ice crystals suddenly appear, formed by any conceivable process (homogeneous nucleation, natural heterogeneous nucleation, or artificial heterogeneous nucleation as in silver iodide cloud seeding)? It was this general question that Wegener posed and answered qualitatively in 1911, which Bergeron took up again, still qualitatively, in 1933, and which Houghton [8] treated quantitatively in 1950. Wegener observed that the vapor tension of subcooled water is slightly greater than that of ice at the same temperature, with a maximum excess of about 0.27 mb at −12°C, becoming zero at 0°C, where ice and water are in vapor equilibrium, and also approaching zero asymptotically for very large degrees of subcooling. In a cloud containing only subcooled drops, the actual vapor pressure will be, unless the cloud is growing very rapidly, essentially equal to the vapor tension of subcooled water at the cloud temperature. Hence, as soon as a few ice particles appear, they constitute vapor sinks and grow rapidly at the expense of the vapor which is supersaturated with respect to the ice particles by the initial amount of 0.27 mb. (This excess of 0.27 mb, it should be made clear, is a very large supersaturation by diffusional standards. For example, it can be shown to be about *ten times* greater than the vapor pressure difference from vapor to drops existing in the region of peak supersaturation at the base of a cloud in a strong updraft.) As the growing ice crystals consume water molecules from the vapor phase, the ambient vapor pressure is rather quickly reduced to a value less than that required for saturation with respect to the subcooled liquid water drops themselves, so the drops begin to suffer net evaporation. But the latter process merely serves temporarily to replenish the ambient vapor supply being heavily drawn upon by the ice crystals, and what was shortly before a metastable state (prior to appearance of any crystals) is rather quickly stabilized by distillation of all or most of the cloud water from liquid to solid state by diffusion processes. This is the ice-crystal process, and it was essentially the qualitative points of this paragraph that were made by Wegener in 1911.

Under what conditions, we must next ask, will the ice-crystal process provide a solution to the central problem of precipitation theory? That is, under what conditions will it produce a million-into-one type of transition in a time period of the order of observed cloud lifetimes, namely, in only tens of minutes? Clearly, only in circumstances where nucleation processes produce approximately one ice nucleus for every million subcooled water drops at temperatures where diffusion kinetics satisfy the rate requirements.

Cloud-drop concentrations in the upper portions of clouds are typically of the order of 100 cm^{-3}, so we must demand about one ice nucleus per 10 liters of cloud space if drops of large enough size to precipitate are to grow by the ice-crystal diffusion process. If a cloud contains too few nuclei, each of these few will quickly grow to such a large crystal (snow crystal) that all will tend to fall out of the subcooled region before they can extract much of the available water, which leads to a loss of precipitation efficiency. If, on the other hand, nucleation is too profuse, so many ice crystals may appear that competition among them for the total available water will prevent any of them from growing large enough to fall through the cloud updrafts, and then the requirements for precipitation are not satisfied at all. (It is this latter possibility that is referred to, in discussions of cloud modification, as "overseeding.")

The ice nuclei counts shown in Table II, though they cannot yet be assumed to be broadly representative, show in a general way how and why the ice-crystal process operates under natural conditions. In a cloud updraft over Arizona in January of 1955, subcooling would have persisted from an altitude of about 3 km, the 0°C level, to about 7 km, where the local degree of subcooling (to about −27°C) would reach a value great enough to "activate" ice nuclei in total concentrations of about one-tenth per liter. These would grow rapidly and would thus begin to lag behind the rising air, ultimately would cease rising and begin to fall, with respect to the ground, still growing by diffusion as they fell. Upon falling through the 0°C level, the snowflakes would melt and become raindrops under conditions typical of Arizona in winter, while under other climatic conditions they might reach the ground as snow.

Houghton's 1950 analysis [8] of the kinetics of this process brought out a number of interesting quantitative aspects of which only two will be mentioned here. First, his results constitute the first really adequate demonstration that Wegener's hypothesis is quantitatively important in cloud particle

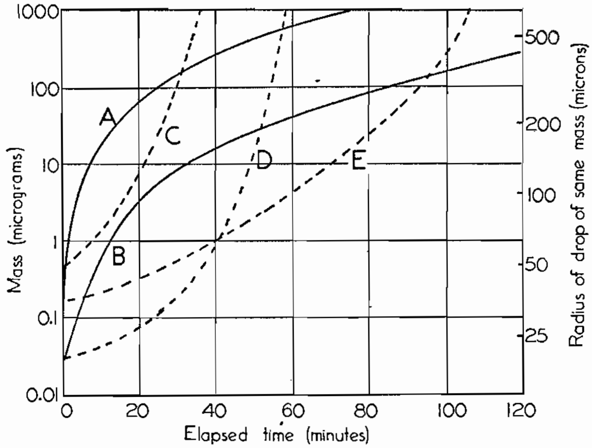

FIG. 4. Diffusional growth of ice crystals (solid curves) and accretional growth of water drops (dashed curves). *A*: plane-dendritic ice crystal, water saturation at $-15°C$; *B*: hexagonal plate, water saturation at $-5°C$; *C*: accretion in a cloud with $50\,\mu$ median diameter drops; *D*: accretion in a cloud with mean drop diameter of $24\,\mu$; *E*: accretion in a cloud with median drop diameter of $10\,\mu$. After Houghton [8].

growth. For example, plane-dendritic ice crystals, growing in a cloud region saturated with respect to water at $-15°C$, were found by Houghton to increase in mass by a factor of about 10^4 in only about 20 min of diffusional growth (corresponding to a flake-diameter change from a few tens of microns to about one millimeter). Second, the same analysis, which covered certain aspects of the accretion process as well, showed (see Fig. 4) that the ice-crystal process is likely to be significant chiefly in *initiating* growth of precipitation particles. For, by the time a snow crystal has attained by diffusion a mass of about 10 μgm (equivalent to a spherical diameter about 275 μ), accretion of cloud droplets and smaller crystals swept out by the falling flake has become equal in importance to diffusion, and shortly thereafter this accretion process completely dominates the growth process. That is, Houghton's results were particularly important in that they revealed that even in clouds of subcooled water whose precipitation history may *start* through the ice-crystal process, the later stages, wherein the bulk of the total liquid water is extracted from the cloud, are dominantly influenced by the second of the two main precipitation processes, that of collision and accretion by large particles falling through a cloud of smaller particles.

Snowflakes may grow by accretion of other snowflakes and also by accretion of subcooled water drops (riming). Different physical factors influence these two distinct accretion processes, but no discussion of the comparatively limited fund of present knowledge of such accretion details will be given here. This omission must not be construed as reflecting small importance of such details, for we need to know a great deal more about them than we now do.

It is interesting to note, as a final point in this summary of the physics of the ice-crystal process, that if our atmosphere happened to contain ice nuclei in number-concentrations and with nucleating efficiencies comparable with those of the abundant and efficient condensation nuclei, the ice-crystal process would not only be incapable of yielding precipitation in the ordinary sense of the term, but would positively *inhibit* precipitation processes by quickly converting every cloud drop to a tiny ice crystal as soon as the drop rose above the freezing level in a cloud updraft. Under such hypothetical conditions, subcooling would be unknown and no million-into-one transition could operate by the above-described diffusion process. It should be added, parenthetically, that this same principle yields the inverse implication that precipitation probability would, in at least one way, be enhanced if the terrestrial atmosphere contained only few and inefficient nuclei of condensation, for then large degrees of vapor supersaturation would develop before the liquid phase made its appearance; but once it appeared, the few drops would grow rapidly and could precipitate to the ground. The actually observed state of affairs with many condensation nuclei and relatively few ice nuclei is much the more probable one, simply because *there are many more salts that are merely hygroscopic than there are crystalline materials with lattice characteristics closely mimicking those of ice*. It is principally this latter deficit that leads to those cloud-modification possibilities which have received so much recent attention. These will be considered in Section 4.2.

3.3.3. The Accretion Process. A review of the meteorological literature, especially the textbook literature, of about fifteen to twenty years ago discloses a rather general, though certainly not unanimous, suspicion that most mid-latitude precipitation is at least initiated if not fully produced by the ice-crystal process. This view had been especially strongly defended by Findeisen (see, for example, [31]), whose name is therefore often linked with that of Bergeron in identifying the process. When, more recently, both theory and observation revealed increasing evidence that collision processes must at least rival the ice-crystal process, and possibly greatly exceed it in importance, there was some tendency to regard this as a distinctively new development in precipitation theory, particularly among those of us who had first come into contact with the field of meteorology in those World War II years when qualitative references to the Bergeron process had just become well diffused throughout the textbook literature.

However, the basic idea of collision and coalescence of drops as a mechanism of growth of precipitation particles is definitely not so new as that, and even quantitative analyses on a very limited scale can be found in the earlier literature. Simpson [32] was, in 1941, stressing the observations of heavy rainfall from tropical clouds with tops definitely known to lie below the level of the 0°C isotherm. Still earlier, in 1938, Houghton [8] had reported some simple computations of rates of particle growth by gravitationally induced collision, the numerical values of which justified his conclusion that a coalescence process "is capable of explaining the formation of rain drops once cloud particles of unequal size appear at the same level," though it is clear in retrospect that his results could only have corresponded rather roughly with actual rates inasmuch as collection efficiencies were not then known with any accuracy. Findeisen devoted considerable attention to collision processes but viewed them as only secondary in importance to the ice-crystal process. Possible effects of electrical charges on droplets were considered by Schmauss and Wiegand [33] as early as 1929; an early instance of attention to a factor that is currently receiving closer scrutiny. A short but historically interesting note on collisional growth by Humphreys in 1922 [34] shows that already by that early date calculations essentially similar to those Houghton discussed in 1938 had been made; but Humphreys dismisses their results on the curious ground that a drop, after falling 5000 feet within the cloud, sweeping out every drop in its path, would come out "only one-sixteenth of an inch in diameter." Theorists have grown easier to satisfy over the intervening years.

The same note also shows that at that date Brooks had clearly recognized the crucially important point that in the presence of moderate updrafts, the effective distance of fall of a drop undergoing collisional growth would exceed its actual distance of fall with respect to the earth in such a way that this growth process might be much more efficient than Humphreys' calculations suggested.

Milham's once widely used 1912 textbook [35] contains, on page 239, a paragraph on the mechanism of raindrop growth which describes, of course without quantitative defense, a statement almost identical with that which one would make today in explaining in general terms the collisional mode of growth. Whether or not still earlier speculations came equally close to the present point of view would be an interesting subject of consideration for the historian of meteorology. The above examples will suffice to show that the long delay in demonstrating conclusively the importance of collision and accretion processes in precipitation was not at all due to failure to recognize the possibility of these processes, but was due simply to failure to pursue the question through all of its quantitative ramifications.

Regrettably, a broad variety of terms has been applied at one time or another to the growth processes involving collision and aggregation, and some choice must be made for the usage to be employed here. Such terms as "coagulation," "aggregation," "accretion," and "coalescence" have been used, but not with uniform connotation. Here the term "accretion process" will be adopted as the term embracing all of those growth processes involving collision of particles (liquid or solid) with consequent aggregation of the two colliding particles into a single larger particle. There is need for precise formulation of definitions and uniform adoption of these definitions in this and many other areas of cloud physics.

The transition from the earlier period of rather cursory inspection of the gross magnitudes entering into accretion processes to the more recent period of detailed analysis of all facets of the problem seems to me to be quite clearly identifiable in a 1948 paper by Langmuir [36]. In that paper, Langmuir presented results of computations of aerodynamic collision efficiencies of large drops falling through a cloud of smaller drops, and discussed resulting collisional growth rates. Prior to this work, the rather complex aerodynamics of the collision process had never been quantitatively taken into account.

The concept of collision efficiency, and related concepts of coalescence efficiency, and accretion (collection) efficiency, are fundamentally important in precipitation processes. The *collision efficiency* of a drop of radius R falling at its terminal velocity through a cloud of smaller drops of radius r is the fraction of all of those smaller drops initially lying within the volume swept out by the larger drop which actually collide with the larger drop. Its upper limit, as so defined, is unity, and its lower limit is zero, the zero values occurring in those cases where a large drop falls through a cloud of such small drops that the aerodynamic pressure field set up by the large drop serves to deflect all of the small drops entirely out of its path as it descends upon them. The Langmuir theory shows that for any given R there is a critical value of r, greater than zero, below which the collision efficiency is zero. Also, for any given r less than about 10 μ, there exists a critical value of R such that zero collision efficiency will obtain for that R and all smaller R. In Table III is presented a list of critical values of R for specified small r, as found by Langmuir. The important implication of these results is that in clouds wherein condensation growth has yielded

TABLE III. Critical radii R_c of falling drops below which no collision occurs, for various cloud droplet radii r (radii in microns). (From [36].)

r	R_c	r	R_c
1.5	600	5	31
2	350	6	20
3	140	7	14
4	58	—	—

drops of only small radii (where "small" here means in the neighborhood of 10 μ), gravitational collisional growth cannot become important until a few drops somehow attain a size that marks them off distinctly from the bulk of the cloud-drop population. Much of the recent theoretical and observational work on liquid-phase accretion processes is primarily concerned with the difficult question of understanding how and when cloud processes do succeed in overcoming this principal obstacle to collisional growth.

The collision of two drops does not necessarily lead to their coalescence into a single larger drop (see, for example, Swinbank [37], and Blanchard [38]), though experimental cases of complete coalescence for certain pairs of drop radii have been reported (see, for example, Gunn and Hitschfeld [39]). That weak external electrostatic fields and net electric charges borne by the colliding drops certainly play a role has been known since the work of Rayleigh. It is impossible to avoid the conclusion that, at present, we scarcely understand the barest essentials of the highly complex physical phenomena controlling coalescence of colliding liquid drops. However, despite this ignorance, it is conceptually useful to define a *coalescence efficiency* for any given type of collision as the fraction of all such collisions which actually lead to coalescence. Finally, the product of the collision efficiency and the coalescence efficiency may be identified as the *accretion efficiency*. (The term "collection efficiency" would perhaps be preferable to "accretion efficiency," but it has already been used frequently as synonymous with the above concept of collision efficiency and also has distinct meaning in other problems of meteorological instrumentation.) Accretion efficiencies may, in particular cases, remain low because of very low coalescence efficiencies and despite high collision efficiencies, or may remain low because of low collision efficiencies and despite high coalescence efficiencies. The concept of accretion efficiency may be applied to collisional growth involving ice particles as well as liquid drops, but "coalescence efficiency" must then be replaced by "clumping efficiency."

Langmuir's 1948 work on the theory of collision efficiency represented an important contribution not only by virtue of the results Langmuir himself drew from it, but even more so from the fact that almost all subsequent theoretical analyses of growth of precipitation particles by collision processes have employed his collision efficiencies. Shortly after his results became available there appeared three different papers by Bowen [40], Houghton [8], and Ludlam [41] which, viewed jointly, clearly established for the first time that collision and coalescence processes must be of major importance in the precipitation problem. Bowen's theoretical results [40] revealed the critical role of the updraft velocity, for he found that the diameter of the raindrops falling out of the cloud base varies linearly with updraft

TABLE IV. Dependence of accretional growth on updraft speeds. (From [40].)*

w (cm sec^{-1})	Max height above base (m)	Final diameter (mm)	Growth time (min)
10	450	0.2	116
25	750	0.5	82
50	1200	0.9	69
100	2200	1.5	62
200	4000	3.0	60

* In a cloud of initially uniform radii of 20 μ and updraft speed w, two drops coalesce near the cloud base and begin accretional growth. Table entries show, for each of five values of w, the maximum height above the cloud base attained by the ccreting drop, the diameter it has on falling out of the cloud base, and time required to fall out of the base.

speed. Bowen assumed a uniform cloud of droplets of 20 μ radius in which two drops somehow coalesce to one of twice the normal mass and the latter then falls relative to the others (i.e., rises less rapidly than the others) and grows by accretion of small drops. In Table IV are shown Bowen's values of peak altitude above cloud base attained by this growing drop just before starting to return to the cloud base, the diameter it attains on falling out of the cloud base, and the total elapsed time for the process, for five different updraft speeds. Surprisingly, his calculations revealed that the time required for completion of the process varies inversely as the updraft speed (see Table IV) due chiefly to the extremely rapid final descent of the large drops formed in the stronger updrafts. I do not believe that this paradoxical result had ever been anticipated, even qualitatively. It seems correct to say that these findings of Bowen's marked the turning point in a redirection of attention to the hydrodynamic, as distinguished from microphysical, factors influencing precipitation processes. The calculated growth times which Bowen obtained were too great by a rather uncomfortable factor of two or three, as were also Houghton's; but, as will be explained below, this is now understood fairly well. Houghton's analysis [8] was chiefly aimed at comparing the relative rates of particle growth by ice-crystal and collision processes and gave the result, cited earlier here, that once particles reached a mass equivalent to that of a drop of about 275 μ diameter accretional growth dominates over diffusional growth regardless of how the early stages of the precipitation process might have been initiated. Ludlam [41] emphasized the critical nature of cloud depth, which is of course inherently closely related to updraft speed, and devoted considerable attention to the growth of just the rare large particles (giant nuclei) which might enter a cloud base with radii already as large as 20 to 40 μ. These large drops are of great interest for they have sufficiently high terminal fall velocities that they sweep out large volumes of cloud space and thus grow quickly. He concluded that if these rare large drops could attain radii of about 150 μ before reaching the cloud summit they were almost certain to enjoy continual growth to raindrop size, since a drop of such radius has a falling speed of about 1 m sec^{-1} and thus could scarcely be borne out of the top of the cloud by the air currents typically encountered in cloud tops. The main conclusion of these three studies may be qualitatively summarized as follows. Accretional growth is strongly enhanced by high updraft speeds and great cloud depths and by presence (or rapid formation) of at least a few drops with radii several times larger than the average for the condensationally produced drops; accretional growth outstrips diffusional growth when ice particles attain diameters of the order of a few hundred microns.

As a consequence of the above-mentioned work of Langmuir, Bowen, Houghton, and Ludlam, it had become clear by 1951 that much closer consideration was going to have to be given to all portions of the problem of accretion. Already by that time, the first rush of effort to exploit the newly found prospects of cloud seeding had begun to encounter difficulties inherent in trying to manipulate natural processes whose details were obviously demonstrating their complexity. Houghton's work showed the inseparability of the ice-crystal and accretion processes; and all of the accretion results combined to arouse suspicion that accretion processes alone might account for a larger fraction of all natural precipitation than had been previously recognized. There was a disturbing factor of about two or three in the excess of theoretical precipitation growth times over observed times in all of the work cited above, and there remained serious uncertainty as to how the indispensable larger-than-average drops might be formed within clouds. Finally, serious interest in the practicality of directly modifying the accretion process itself developed at about this same time. All of these research objectives have influenced the more recent investigations of the collision processes which have commanded steadily growing attention in cloud physics research.

Aircraft measurements, such as, for example, those of Weickmann and aufm Kampe [42], in convective clouds, were pointing more and more to the important role of collisions. The number of cloud drops per cubic centimeter reported by aufm Kampe and Weickmann fell off by as much as a factor of five from the base of the clouds to altitudes of only about 2000 meters above the base (specifically, from about 300 cm^{-3} to about 50 cm^{-3}), whereas liquid water content and mean drop size increased through the same layer. Each of these observations pointed to highly effective coalescence processes of some kind operative near the cloud bases.

Radar meteorological methods continued to shed a great deal of light on all parts of the precipitation problem (see general review in this series by Marshall *et al.* [43]), and have, in particular, documented the importance of accretional growth. Observations of precipitation radar-returns from nonfreezing clouds were reported from many areas, but in themselves only added weight to what were already fairly conclusive visual observations in the same category. More significantly, radar evidence for the dominance of accretional processes even in deep convective clouds extending to well above the freezing level in extratropical regions began to appear. Reynolds and Braham [44] pointed out that initial radar echoes appeared in the observations of the 1947 Thunderstorm Project at an average temperature level of about $-2°C$, and they gave radar meteorological reasons for believing that these echoes must have been produced by precipitation particles still too small to be falling with respect to the temperature field, thus implying that the earlier growth of these particles must have occurred at temperatures too warm to bring the ice-crystal process into consideration at all. This point was pursued still further by Battan [45], who used the same Thunderstorm Project data but reanalyzed it more completely. He concluded that fully 60 % of all first echoes were in convective cloud regions entirely below the freezing level (i.e., in regions warmer than 0°C) and hence must have been due to coalescence effects. Such applications of radar techniques seem historically especially interesting in that they were so illuminating in just those cases where earlier *visual* observations had given only the familiar chronological order of glaciation of cumuliform cloud tops followed shortly by precipitation, an order long regarded as one of the best arguments in support of dominance of the ice-crystal process, despite its *post hoc* nature.

The time-of-growth discrepancy of a factor of two or three between theory and observation that was apparent in Bowen's [40] calculations based on the Langmuir collision efficiencies now appears to be considerably clarified as a result of a most interesting series of observational, experimental, and theoretical investigations by the Sydney group. In 1953, Adderley [46], using a balloon-borne telemetering technique to measure cloud drop-size distribution, observed drop-size profiles strongly indicative of accretional growth rates some three to four times greater than could be accounted for on the basis of the Langmuir efficiencies, tending to cast doubt on the adequacy of the latter. Then, in 1955, Telford *et al.* [47] carried out some very cleverly planned laboratory experiments on collision and coalescence phenomena which were designed to yield collision efficiencies for pairs of colliding drops of nearly the same radii. The latter condition is that which obtains during the early stages of development of a rain cloud when condensational processes have created a rather narrow range of drop sizes, whereas previous experimental tests of the Langmuir efficiencies (for example [39]) had dealt with quite dissimilar dropsizes in collision. The average value of their experimentally determined accretion efficiencies was greater than unity, in fact exceeded 12, an astonishing result indeed. Scrutiny of photographic records of the collision kinematics disclosed to these investigators that the existence of collision cross sections over twelve times the geometric cross sections of the drops was due to a very remarkable phenomenon: Drop A can capture drop B (of radius, and hence fall velocity, close to A's) by drawing B *sidewise* into its (A's) low-pressure wake region, whereupon B quickly falls down from above A and coalesces with it! Langmuir's calculations dealt only with collisions in which the collected drop contacted the collector drop on the latter's *underside*, and furthermore Langmuir had assumed a type of flow regime incapable of predicting wake phenomena. The drops used in the experiments of Telford *et al.*, had radii of about 80 μ, and fell with Reynolds numbers of 20, so they cannot simply be extrapolated down to the case of cloud drops just formed by condensation (radii of the order of 10 to 20 μ at most and hence falling at Reynolds numbers well under unity); but clearly these results constitute a highly significant finding and one that helps to explain the over-all rapidity of natural collisional growth.

Two closely related studies in the same program of the Sydney group have added further valuable insights. Telford [48] carried out a careful theoretical analysis and numerical calculation of certain purely statistical aspects of the collision problem, somewhat similar to, but more extensive than some earlier work by Hitschfeld and Melzak. In brief, Telford considered the growth of the "fortunate" few drops which, as a result of purely random fluctuations of mean-free-times between collisions, enjoy a faster than average growth. Previous collision calculations had generally been based on the assumption of uniform mean-free-times (for simplicity and not because such fluctuations are absent). Telford found that in the very short time of 5 min, the favored few might grow to radii of over 20 μ from an initially homogeneous cloud of drops of 10 μ radius (as a result of some ten coalescenses, whereas under the standard assumption of continuous and uniform growth of all drops, over 30 min would be required to accomplish this same growth. Since there seems little doubt that Telford's model is more realistic than those previously used, and since the difference between 5 min and 30 min in any cloud processes is a highly significant difference in view of short cloud lifetimes, and finally, since all earlier collisional calculations have indicated that precipitation growth will accelerate rapidly once a few drops of about double the average size have appeared, one sees the key importance of this analysis. It must be carefully noted that no assumption was made by Telford concerning any turbulence-spectrum characteristics for the cloud; his fluctuations are statistical and not physical in origin.

The second of the more recent contributions of the Sydney group to be mentioned here has been a very extensive set of calculations carried out by Pearcey and Hill [49], using high-speed computer methods, to determine theoretically the flow patterns governing collision dynamics of cloud drops. This work constitutes a much more complete analysis of collision aerodynamics than went into Langmuir's 1948 results, and provides the first solutions (numerical, of necessity) for the pertinent flow equations at Reynolds numbers in the transition region between viscous and aerodynamic flow. The most interesting of many of their results is their theoretical affirmation of the 1955 experimental observations of Telford *et al.* [47] relative to the existence of effective collision efficiencies in excess of unity: Pearcey and Hill's calculated efficiencies range even as high as 100, and nicely confirm in theory the odd capture kinematics observed photographically by their colleagues. As of the time of this writing, no synthesis of all of these very recent developments in collision theory has been accomplished, but it would appear that, when undertaken, improved theory can now be expected to account for the observations substantially more closely than it could only a few years ago.

A number of other recent advances in collision-process theory can only be briefly mentioned. Efforts to get higher theoretical growth rates out of the Langmuir efficiencies by invoking cloud turbulence have been made by East and Marshall [50], their principal result being a required turbulence intensity rather too large to find support in any existing observational data (though this negation is far from fatal to their hypothesis since there is a serious dearth of relevant cloud turbulence data at present). It is of historical interest to note that, as early as 1939, Arenberg [51] had suggested, qualitatively, that microturbulence might play a significant role in cloud drop-growth, so one sees that fifteen years elapsed between qualitative suggestion and first quantitative analysis of this cloud-physical problem. An aspect of the interaction between condensation and collision processes that holds promise of accounting for another portion of the time discrepancy between theory and observation was treated by East [52]. He studied theoretically the consequences of continued condensational growth of cloud drops at the lower tail of the cloud drop-size distribution as air parcels are carried to levels of high liquid water content; and using only the Langmuir efficiencies he found interesting improvement in agreement between theory and radar observations of heights and times of first precipitation echoes.

Woodcock and Blanchard [16] have recently presented further evidence of indirect nature (rainwater salinity) tending to support Woodcock's earlier suggestions concerning the importance of giant sea-salt nuclei in starting accretional growth in maritime clouds. This work assumes particular interest in that it concerns natural processes which, if Woodcock's hypothesis is correct, might serve as models for artificial stimulation of collision growth of precipitation by addition of giant nuclei to updraft air entering cumuli in continental areas where droplets of sea-salt solution with radii of the order of 20 μ are probably present in deficient numbers at times (see Section 4.3). Numerous other studies have also contributed to the current progress in understanding accretion mechanisms of generating precipitation, but cannot be described in detail here, even though many are intimately related to the physics of precipitation.

Study of the accretional process of growth of precipitation particles has drawn increasing attention to the macroscopic dynamics of clouds, and this trend has been reinforced by growing awareness of the critical role of cloud lifetimes in all precipitation processes. Also, the question of entrainment of environmental air into clouds and all of the problems of momentum- , mass- , heat- , and vapor-exchange between cloud and environment have become somewhat more clearly appreciated as factors which place limits on precipitation release rates and which, in turn, must therefore limit cloud-modification possibilities. No attempt will be made here to outline the recent progress toward better understanding of cloud dynamics, for the slightly arbitrary reason that dynamic factors are not obviously amenable to modification, and here the physical basis of modification techniques is chiefly under discussion. Despite this omission, I must emphasize that there appears to be unanimous agreement among cloud physics workers that much more research on cloud dynamics must be undertaken before truly rational cloud-modification programs can be undertaken.

3.3.4. Summary. An attempt has been made in this section to outline the historical development and present status of knowledge of those aspects of cloud physics that have strong bearing on cloud-modification possibilities. Impressive recent gains have been made on almost all fronts. Within a relatively few years, questions that had been before meteorologists for many decades have been answered fairly satisfactorily. A great amount of sorting out of the important from the unimportant has gone on. Above all, the whole problem has been brought into much sharper focus for present and future studies.

The stimulus to cloud physics research that has been provided by prospects of cloud modification to increase natural precipitation has been, without doubt, the most important single source of impetus to recently accelerated research in this field. In the following sections, the scientific aspects of these newly developed cloud-modification techniques will be reviewed, and our present stock of fundamental knowledge will be compared with the knowledge required to exploit these techniques intelligently. It will be found that the above-reported progress which appears so dramatic when viewed in comparison with earlier historical growth of cloud physics falls, unfortunately, far short of what is required to modify clouds in a truly intelligent fashion.

4. Recent Developments in Cloud Modification Techniques

4.1. General

Pre-1946 "rainmaking" has a long and mostly dubious history. Putting aside the superstitious propitiations of early man and of contemporary primitive groups, there is still left a lengthy record of relatively recent rainmaking efforts, particularly in this country near the turn of the century, carried out, it would appear, sometimes in good faith, sometimes not, but certainly with little or no real scientific basis. During the serious American drought years of the mid-nineteen-thirties interest was stirred in what was to be an indirect means of rainmaking in the central United States by creating ponds and stock-watering tanks throughout the area, the evaporation from which was to augment natural rain. This idea, which reappears periodically in one form or another, is a measure of the general lack of appreciation of the enormous scale of operation of the atmospheric hydrologic cycle, a scale that is not to be sensibly altered by any such trifling additions of vapor to the huge stock existing in the atmosphere in even the most severe drought periods.

The current epoch of rainmaking is not in any way historically connected with those earlier attempts. One cannot, however, correctly say that the current period has exhibited none of the charlatanism of fifty years ago, for there have been, unfortunately, some patently fraudulent claims made by some persons and groups seeking to exploit commercially the recent interest in cloud-modification methods, and there has been a nearly continuous distribution of vigorously announced, but poorly substantiated, claims ranging from the seemingly preposterous to the merely enthusiastic. Personal experience leaves in my own mind no doubt that a most regrettable confusion developed after 1946 in the minds of laymen concerning the true scientific status of cloud-modification techniques. This confusion stemmed, I believe, from the fact that the often extravagant claims of some seeding operators were either made in pseudo-scientific form not readily evaluated by laymen or else were announced quite positively without attempt at clarification of the many existing scientific uncertainties not common knowledge among potential users of the operators' services. This confusion, it is important to note, never enjoyed the salutary influence of normal debate in the professional scientific literature; many who made strong claims simply did not publish, at least not in the regular technical periodicals, any results of their seeding operations that could be independently studied by disinterested meteorologists. And in those numerous instances where they published in nontechnical media, their reported data were completely inadequate to permit scientific evaluation. Although such confusion has had little reference to the pressing research problems of the field, it has had broader reference to the relations between science and the public which are very much the scientist's concern. Since geophysics is, for a number of self-evident reasons, somewhat prone to entanglement in such confusions, it seems very much in order to commend the history of the past dozen years' public relations in the area of weather modification to the general attention of geophysicists, though details are not relevant here.

4.2. Artificial Nucleation of the Ice-Crystal Process

4.2.1. The Dry-Ice Seeding Technique. It is not often that the exact start of a major development in a given research area can be identified; but in the case of current cloud-modification studies, the beginning is quite clearly identifiable with a July afternoon in 1946 when V. J. Schaefer, in the course of some investigations of aircraft icing at the General Electric Research Laboratories, introduced some dry ice (solid CO_2) into a deep-freeze chamber in which he had previously created a cloud of subcooled water drops. An immediate and striking transformation to a cloud of scintillating ice crystals occurred. Schaefer quickly ascertained that neither the chemical nature of the dry ice nor its sublimation temperature of $-78°C$ was essentially involved, but rather that any object sufficiently cold would, if merely waved through the cloud, induce freezing, the temperature threshold lying very near $-40°C$ [21]. Recognizing the potentialities for inducing the Bergeron ice-crystal process in natural subcooled clouds, Schaefer carried out, on November 16, 1946, a dry-ice seeding trial that was successful in causing streaks of snow to fall from the base of a cloud into which he had scattered crushed dry ice from an aircraft.

Although it took some years to clarify the point, if the point may indeed be claimed to be fully clarified yet, Schaefer's basic discovery was the detection of the temperature at which homogeneous nucleation of subcooled drops of size typical of natural clouds occurred. (More accurately, he found the temperature at which the time required for nucleation probability to approach unity in such drops was of the order of seconds, rather than of the order of hours as it is only a few degrees above the critical temperature.) On present theories, dry-ice seeding of a cloud amounts to dropping into a subcooled cloud a lot of pea-sized particles each much colder than the critical temperature for homogeneous nucleation and hence capable of cooling to below $-40°C$ the numerous droplets formed in the cold boundary layer where vapor is chilled to below the condensation point. That is, it seems probable that the cold dry-ice particle functions in two distinct and sequential ways: first, to condense vapor to droplets, and secondly, to induce homogeneous nucleation within those droplets which then leave the boundary layer as tiny ice crystals capable of serving as growth centers. The laboratory efficiency of this particular seeding process was reported to be phenomenal: Schaefer [21] estimated that a single pellet 1 cm in diameter can form, under optimal conditions, some 10^{16} ice crystals which, if each could grow to the size of small snow crystal, would amount to 300,000 tons of snow. Such efficiency is never attained in practice, but it remains true that a large magnification factor is available.

Although the recognition and exploitation of the opportunity of inducing the ice-crystal process in natural clouds was due to Schaefer, three other investigators had, within the preceding year or two, carried out experiments which had pointed to, but had not shown so decisively, the existence of a critical threshold near −40°C. These were Findeisen and Weickmann, both in Germany, and Cwilong in England. For general reviews of the work of these investigators see Houghton [53] and Ludlam [54]. There is difficulty in assigning credit for discovery of the critical threshold of homogeneous nucleation, for the results of all of these nearly synchronous studies contain internal contradictions that, in my opinion, have never been fully resolved. Schaefer's experiments, using a cold box rather than an expansion chamber, seem to have been the most clear-cut. It is of interest to note here that Findeisen had, in 1938 [31], predicted that interesting trigger effects might be placed at man's disposal if efficient ice nuclei could ever be added to metastable subcooled clouds at will.

The actual technique of seeding clouds with dry ice to promote the ice-crystal process is basically simple. Cakes of solid CO_2 are passed through a mechanical crusher in the seeding aircraft and dropped as fragments of about 5 to 15 mm in diameter. Although dry ice will start the ice-crystal process even at levels just slightly colder than 0°C, there is little chance for much of a seeding effect if released below about the −5°C level unless ice crystals will quickly be carried by updrafts to the −10°C to −15°C levels of most rapid diffusional growth; hence the technique requires use of aircraft capable of cruising readily at altitudes of at least about 3 km, and in many areas of the world and various seasons, altitudes of greater than 5 km must be reached (over 7 km in the southwestern United States in summer, for example). In view of these altitude requirements and considering space requirements for stowage of the dry-ice supply and operation of a crusher, it becomes clear that one must use multimotored aircraft if any significant seeding is to be performed.

What, then, *is* the present estimate of the efficiency of the dry-ice seeding technique? My own answer must be that I do not believe a really accurate estimate has yet been given. There was, of course, a great deal of quite nonscientific publicity given this technique some years ago, and commercial operators have at times made strong claims for the efficacy of the technique (before aircraft seeding with dry ice was abandoned as too expensive for most operators), but there have been too few carefully controlled dry-ice seeding experiments carried out under wholly satisfactory conditions to warrant any categorical statements.

Experiments made in Australia (see Smith [55], Squires and Smith [56], and Bowen [57]) gave favorable indications in the sense that seeding with dry ice was followed by observable streaks of precipitation leaving cloud bases in a number of instances wherein no other clouds for about 20 miles around were precipitating naturally. Since there is always a possibility of unconscious bias in selecting clouds for seeding, such a criterion cannot be accepted as a strong one, so these Australian tests, though interesting, cannot be regarded as giving conclusive evidence. Bowen [57] estimated that meterological circumstances offer possibilities for dry-ice seeding in southeastern Australia rarely enough that an increase of no more than 5 to 10%, at the upper limit, could ensue from this type of seeding.

A statistically much stronger experimental design was incorporated into dry-ice seeding tests carried out by the staff members of the Department of Meteorology of the University of Chicago [58]. In these experiments only an unfortunately small total number of treated clouds could be studied. Pairs of clouds were chosen by a flight controller who attempted to pick two cumuli as nearly alike as possible in all relevant respects, both of which were then penetrated, while only one was treated with dry ice. Only the operator of the dry-ice crusher in the rear of the B-17 aircraft knew which cloud was actually treated (this being decided by opening an envelope containing previously randomized instructions that dictated whether a given cloud was to be seeded or not) and the behavior of both members of each pair was observed by radar and visual techniques without conscious or unconscious bias based on knowledge of which member of each chosen pair had been treated. In this randomized-pair technique, a total of 27 pairs was studied in the summer of 1954 in the central United States. From this sample, the radar behavior gave statistical results such that the hypothesis that treatment had *no* effect could *not* be rejected, see Table V. For, of all 27 pairs, 7 cases showed that the treated cloud developed an echo while the untreated one did not, whereas 5 cases showed the treated pair developing no echo while the untreated member did. There was a theoretical probability of 0.39 of such a 7 to 5 split occurring purely by chance in the absence of real treatment effects, which is far too large a probability to warrant claims to artificial stimulation. The total sample was smaller than had been sought in this study, and the aircraft could not be operated high enough to optimize the dry-ice treatment, but the

TABLE V. Results of dry-ice seeding of 27 randomized pairs of cumulus congestus clouds in the central United States. Each table entry corresponds to one pair of cumuli. (From [58].)

		Treated cloud of pair	
		Echo	No echo
Untreated cloud of pair	Echo	1	5
	No echo	7	14

results seem to be chiefly significant in that they indicate that, contrary to the claims of a decade ago, dry-ice seeding is *not necessarily an efficient technique* for stimulating precipitation. Reports of still other dry-ice seeding projects, also less than conclusive in their finding, will be found in [58].

In the Chicago flight experiments, a notable lack of ice crystals was was observed by the flight controllers on repenetrating a cloud after treatment, so this matter was examined theoretically by Braham and Sievers [59], with special attention being given to the question of possible overseeding, since rates as high as 50 lb/mile had been used. They concluded that earlier estimates of numbers of ice nuclei generated by falling dry-ice pellets had been grossly overestimated, and even more significantly, they noted that crystal growth times lay so close to ordinary lifetimes of cumulus towers that there is both an inherent time limitation in dry-ice seeding and an inherently strong ambiguity in evaluation of seeding efficacy when *individual* cumuli are treated. It has to be recognized that the design of these particular experiments dictated that isolated cumuli be treated, and it remains possible that dry-ice seeding of clusters of convective towers or massive banks of such clouds might lead to different results.

In summary of the status of the dry-ice seeding technique, it can be said that, though there is no disagreement on the point that dry ice can indeed produce the Bergeron transition in subcooled clouds both in the laboratory and in nature, a variety of circumstances conspire to make this appear, at present, to be a less promising means of increasing natural precipitation in economically feasible ways than was hoped a decade ago.

4.2.2. The Silver Iodide Seeding Technique. Only a few months after Schaefer had made his first observations of laboratory effects of dry-ice treatment of subcooled clouds, his colleague Vonnegut made a very substantial contribution to cloud modification technology [60]. Reasoning that substances very similar to ice in crystalline structure might serve as ice nuclei (i.e., reasoning that nucleation should be dependent on epitaxy), he searched the literature of crystallography for materials closely resembling ice with respect to crystal system, space group, and lattice dimensions of the unit cell. Two substances out of a large total number examined appeared

TABLE VI. Comparative crystallography of ice, silver iodide, and lead iodide. (From [60].)

Substance	System	Space group	Lattice constants (Ångströms)	
			a	c
Ice	Hexagonal	D_{6h}^4	4.535	7.41
AgI	Hexagonal	C_{6v}^4	4.585	7.490
PbI_2	Hexagonal	D_{3d}^3	4.54	8.86

promising, silver iodide and lead iodide. In Table VI are shown the comparative data for ice, AgI, and PbI_2 which led Vonnegut to conduct experiments with the latter two materials. After some initially unsuccessful attempts, Vonnegut found means of generating large numbers of tiny particles of these materials, and both proved highly efficient as ice nuclei. Most of the subsequent consideration of such heterogeneous nucleants has been given to AgI, which Vonnegut showed to be effective at temperatures as warm as −4°C in subcooled clouds. Table VI shows that both lattice constants for AgI differ from the corresponding ice constants by only about one per cent, permitting growth of an ice lattice upon the AgI particle surfaces with very small mismatch, so that the AgI particle serves as just about as good a growth center as a true ice crystal. Despite the difference in space groups, the ice structure is almost identical with that of AgI, with the oxygen atoms occupying all of the lattice points occupied in AgI by silver and iodine atoms and with tetrahedral hydrogen bonding in the ice structure corresponding to tetrahedral silver-to-iodine bonding in AgI. Up to the present time, no naturally occurring dusts have been found to approach AgI or PbI in nucleating efficiency (cf. Fig. 3), and the observed tendency toward marked subcooling in clouds stands as incontrovertible evidence that even if any exist, they certainly are not swept up into the free atmosphere in significant concentrations.

All of the principal techniques subsequently used for generating AgI nuclei are derived from Vonnegut's early work [60, 61]. AgI can be coated on filaments which are then electrically heated to evaporate AgI smoke. It can be impregnated into string which is then fed into an oxyhydrogen flame. Silver electrodes can be sparked in the presence of iodine vapor. A solution of AgI and NaI in acetone can be prepared and charcoal or coke particles soaked in it and subsequently burned. Most commonly, the AgI-NaI-acetone solution (KI often replaces NaI) is sprayed into a hydrogen or hydrocarbon flame to form the nuclei. Solutions containing from 5 to 10% AgI by weight are typically used in the latter method.

All of the above nuclei generating techniques are essentially dependent upon vaporization of AgI followed rapidly by quenching in the ambient air to condense out very small AgI particles and these two processes may vary considerably in their effects from one type of generation to another. Vonnegut [60] found that the impregnated-string method produced particles with diameters of the order of 0.01 μ, while vaporization from a hot wire gave diameters close to 1 μ. Nucleating efficiency is size-dependent, partly by virtue of the Kelvin effect explained above and partly because of more subtle considerations of crystal nucleation, so it is not surprising that Vonnegut found that the 0.01-μ particles became effective nuclei only with subcooling to about $-8°C$, while the 1-μ particles were effective at $-4°C$. But, because of the generally small particle sizes for all methods of generation, extremely large absolute numbers of AgI particles are obtained per gram of AgI used, typical figures running to about 10^{14} to 10^{16} nuclei per gram of AgI.

The actual number of effective nuclei produced per second depends upon the rate at which AgI is consumed by a given type of generator and upon physicochemical details of the generation process. Also, mere particle-generation-rate is not a meaningful criterion, because some particles from a given generator are not effective (presumably for reasons of size and surface structure) until large degrees of subcooling are involved, while others are much more efficient. Smith and Boucher [62] have recently conducted comparison tests of three commonly used types of generators, and Fig. 5 summarizes their results. They give figures interpretable in terms of actual AgI consumption only for the propane-acetone generator. For reference these are given here. A solution of 16 lb of AgI and 4 lb of KI to 55 gal of acetone is burned in propane in this generator at a rate of just under 1 qt/hr. This amounts to a consumption of about 0.01 gm of AgI per second. The original reference must be consulted for experimental details and for data on the scatter about the curves reproduced in Fig. 5. The steep slope of the curve for the propane-acetone generator and its falloff at temperatures warmer than about $-10°C$ stand in contrast to the slopes of the other two generators, and it is evident that very different cloud seeding efficacy could result from use of these different generators at different cloud temperatures.

When it is desired to introduce AgI into clouds from aircraft flying in or near subcooled clouds, large nuclei generation rates are needed to compensate for aircraft speed, which rules out the impregnated string burner, while safety factors preclude use of coke. Hence some form of burner employing a solution of AgI is usually used in aircraft seeding. Warner and Twomey [63] describe a generator mounted externally on an aircraft to take advantage of ram pressure in sustaining a high combustion rate. An acetone solution of AgI, burned at a rate of about two gallons per hour (equivalent to about 1000 gm AgI per hour), gave a linear seeding rate of about 4

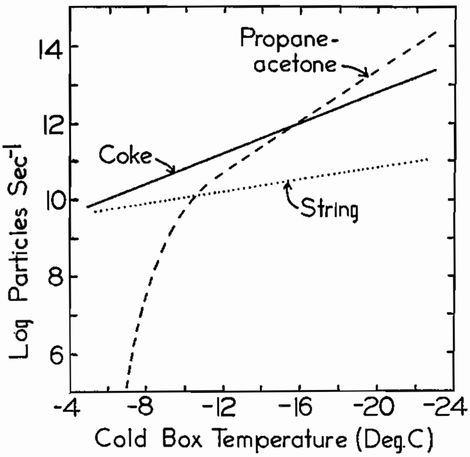

FIG. 5. Comparison of nuclei output of three types of silver iodide generators. Solid curve: coke particles impregnated with 40% AgI solution; dashed curve: acetone solution of AgI and KI burned in a propane flame; dotted curve: string impregnated with a 10% solution of AgI, burned in a propane flame. After Smith and Boucher in [62].

gm AgI per mile, a contribution of the order of 10^{12} to 10^{13} AgI nuclei per mile. As this line source of high nuclear density drifts downwind, turbulent diffusion expands the line into a cylindrical distribution. Estimates that I have made in conjunction with a similar seeding program conducted by the University of Chicago and the University of Arizona indicate that the diameter of the cylinder is of the order of 1 km after about 15 km downwind drift when released at an altitude of about 6 km. After such diffusive expansion of the trail of nuclei, the calculated average density is about 100 per liter if no photolytic decay occurs.

There is a most serious lack of truly critical studies of the physical processes occurring in generators and of the undoubtedly very complex surface physics and chemistry of the nuclei they produce. Nevertheless, a few very informative studies have been completed, and viewed optimistically, they suggest that there is probably much room for improvement in generator efficiencies. Among the results obtained from the rather small number of careful investigations of nuclei production and of nuclei characteristics that have already been carried out, a few will be summarized here for their bearing on cloud-modification physics.

One of the first and most important problems to receive attention was that of photolytic decay of AgI particles when released into the sunlit atmosphere, an obvious point of suspicion inasmuch as the silver halides are commonly used photosensitive agents in photographic film. The first published report on this problem [64] indicated that as a result of irradiation under bright noon-day summer sun in New Mexico the number of AgI particles possessing nucleating effectiveness fell off by a factor of about 100 in one hour. The nuclei were generated by burning an acetone solution. Since times of the order of an hour are frequently required for nuclei released from generators on the ground to be carried by diffusion and convection to cloud altitudes where they can be effective, this first-reported deactivation rate was quite disturbing. Equally discouraging were the results of Inn [65] who irradiated AgI (nuclei evaporated from a filament) with ultraviolet light of intensity roughly that of sea-level sunlight and found that complete photolytic deactivation occurred with wavelengths of less than 4300 Å units in about 20 min. Inn suggested that irradiation might irreversibly dissociate AgI with liberation of iodine and aggregation of silver atoms into surface clusters that spoiled the lattice compatibility between the AgI and the deposited water molecules. Vonnegut and Neubauer [66] reported, shortly after appearance of both of the above studies in 1951, that a deactivation rate of only about 50% per hour had been found in their laboratory tests with nuclei formed by burning impregnated charcoal. No completely satisfactory explanation was then given, or has ever been given, for these and other contradictory experimental results, but these authors proposed that trace impurities in the AgI might exert marked influences on deactivation rates and suggested that these impurities might vary from one generation method to another. This report was closely followed by another paper by Reynolds *et al.* [67] in which it was reported that samples of AgI nuclei whose ice-nucleating effectiveness was reduced by three to four orders of magnitude after irradiation by an ultraviolet source for only a few minutes were restored to high nucleating efficiency by treatment with traces of ammonia gas after deactivation. They were able to take account of Brownian precipitation of nuclei on the walls of their experimental chamber and thereby to provide indication that some of the disagreement between existing results of various investigators was a matter of chamber size. By analogy with the process of ammoniation of photographic plates, they hypothesized that ammoniation of a sample of nuclei might be chiefly a surface chemical effect acting upon the smaller AgI particles not ordinarily effective as nuclei and also not so susceptible to initial photolytic decay. No detailed treatment of this interesting hypothesis has yet been given, and the practical potentialities of ammoniation in cloud-seeding work would seem to warrant further study. Birstein [68] found that photolytic deactivation times were strongly dependent upon ambient relative humidity. Nuclei in an atmosphere of 100% relative humidity with the same ultraviolet source previously used by Inn [65] were found to lose completely their nucleation powers after 150 min of irradiation; at 60% this deactivation time fell to 60 min; and at zero per cent relative humidity the sample lost all its nucleating effectiveness in a mere 5 min. The normal tendency for relative humidity to increase from surface to cloud-base altitudes would, in view of Birstein's findings, imply that the most serious deactivation effects must occur while ground-released nuclei are drifting upward through the lowest kilometer or so. This suggests that generator sites should be located at highest available elevations for this reason, as well as for other more obvious meteorological reasons.

Two other more recent deactivation studies carried out in Australia seem to show conclusively that photolytic decay is a very real difficulty and that decay rates may depend considerably on generator characteristics in a way still not understood. Smith *et al.* [69] released a known flow of zinc sulfide powder, as a fluorescent tracer material, into the air from the site of their AgI generator and flew aircraft equipment at varying heights and downwind distances to measure concentrations of both of these particulate materials. Whereas earlier flight measurements of just AgI nuclei concentrations had been subject to uncertainty due to ignorance of exact relative importance of photolytic decay on the one hand, and of turbulent dilution on the other, in this study only the *ratio* of observed number of AgI particles to observed number of fluorescent ZnS particles was considered. This neatly obviated uncertain speculation as to the role of dilution itself. Smith *et al.* found that AgI nuclei from a kerosene-burning generator decayed in daylight at a rate of about tenfold per hour, while nuclei from a hydrogen-burning generator decayed very much faster, effective nuclei counts falling to one-tenth the initial value in only about eight minutes. Efforts to detect correlation between decay rates and amount and type of cloud cover were unsuccessful, a result that seems somewhat difficult to understand and that deserves further study. In a later investigation carried out in similar fashion, Smith and Heffernan [70] flying AgI and ZnS counting equipment under both daytime and nighttime conditions, found that whereas their daytime flights agreed closely with those of Smith, Heffernan, and Seely [69], their nighttime counts revealed no discernible decay in nucleating efficiency over times as long as 150 min, proving beyond reasonable doubt that the daytime decay is photolytic in nature. In Fig. 6 are shown their 1956 decay measurements for day and night for the case of just the output of a kerosene-burning generator. As pointed out by Smith and Heffernan, the very small spread of the nighttime counts about the zero-decay line is strong evidence that the inherent accuracy of the AgI-ZnS counting method is high and that the spread in the daytime counts is real and not just due to counting errors, though again efforts to correlate decay anomalies with cloud cover (and humidity) were

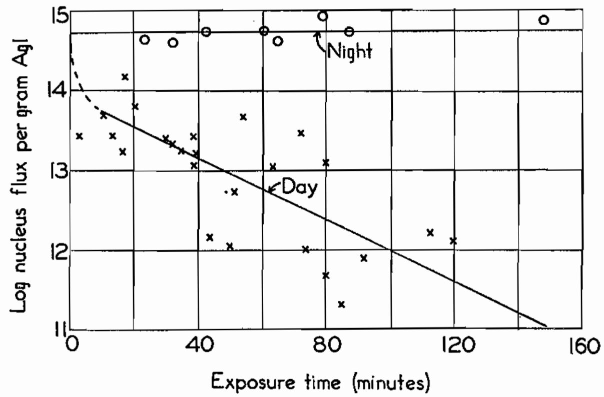

FIG. 6. Variation of flux of silver iodide nuclei with time of exposure in the free atmosphere under day *vs* night conditions. After Smith and Heffernan [70].

inconclusive. No dependence of decay rate on ambient air temperature could be found in this study. The reason for the much higher decay rate of nuclei obtained from a hydrogen generator as compared with a kerosene generator is not yet known. In consideration of all of these results on photolytic decay of AgI nuclei in sunlight there is now little basis for doubting that decay is a factor much too large to be overlooked in seeding operations and that amounts of AgI required to attain a given theoretical AgI nucleating rate in clouds are certainly considerably greater than had first been estimated. Concomitantly, these findings point to the pressing need for careful physical and chemical studies designed to find ways of suppressing photolytic decay or to find materials less subject to photolysis but equally efficient as nucleants.

More exact information is badly needed concerning the crystallographic properties of the surfaces on which the ice grows in the nucleation process and concerning the way in which high-temperature processes in nuclei generators influence these properties. A few significant experimental studies carried out in the past few years, chiefly on AgI, provide some initial information of this sort. To aid in the interpretation of these studies, a glossary of terms and crystal properties of AgI has been drawn up in Table VII. Since various authors have identified the three AgI crystal forms in confusingly different ways, this glossary may have reference value beyond its immediate use here. The information contained in it has been drawn from a variety of sources [71–73]. At room temperatures the hexagonal form is stable, but since all generators heat the AgI far above even the 146°C transition temperature, the question of which form will be found in nuclei after quenching is one that cannot be answered without informa-

TABLE VII. Crystallographic terminology and properties of silver iodide.*

Crystal system	Common name of arrangement	Temperature range of stability	Lattice constants (Ångströms)
Cubic (body-centered)	—	Above 146°C	$a = 5.03$
Cubic (face-centered)	Zincblende (ZnS)	135° to 146°C	$a = 6.47$
Hexagonal	Zincite (ZnO) or Wurtzite (ZnS)	Below 135°C	$a = 4.585$ $c = 7.490$

* Remarks: The hexagonal (zincite) modification of AgI is sometimes referred to as β-AgI, the cubic (zincblende) modification as γ-AgI. AgI melts at 552°C, decomposing upon melting into metallic silver and iodine vapor.

tion on the crystal physics of the condensation processes. There is some evidence that AgI produced by certain generators is principally in an amorphous form.

To determine the effects of generator temperature on AgI crystal form, Manson [72] passed the output of a temperature-controlled source through a thermal precipitator in such a way that a deposit of nuclei could be obtained in a form amenable to powder x-ray diffraction analysis. With the source operated at 650°C, 73% by volume were hexagonal crystals, while at both 800° and 1000°, 95% were hexagonal. The root-mean-cube particle diameter for the 800°C case was about 0.1 μ. The trend toward higher percentage of the hexagonal modification with higher source temperature Manson interprets, tentatively, as due to distillation of greater quantities of iodine vapor than of silver vapor from the high-temperature source to the quenching section, combined with a tendency (inferred from solution crystallization phenomena) for excess iodine to promote growth of the hexagonal modification rather than the cubic forms during condensation in the quenching section. Since Vonnegut originally chose AgI as a likely nucleant on grounds of structural similarity between ice and the hexagonal (wurtzite or zincite) modification of AgI, the above results seem to indicate that high source temperatures might be preferred.

However, this last inference is based on the premise that the hexagonal modification will be a better nucleant than the low-temperature cubic (zincblende) modification, and recent investigations cast some doubt on this premise. Manson [74] has called attention to the fact that low-temperature cubic modification has one plane, the diagonal (111) plane, in which the atomic spacing and pattern is almost identical with the basal (001) plane of ice and of hexagonal AgI. He succeeded in preparing samples with widely differing relative amounts of hexagonal and cubic AgI (the latter having strongly developed faces parallel to the (111) plane), and tested their ice-nucleating abilities. In the temperature range from $-9°$ to $-11°C$, he found no observable difference in nucleating ability of a sample containing 70% hexagonal crystals as compared with another containing 94% cubic (γ-AgI). Concluding from this that either nucleation occurs preferentially on (111) planes or else that nucleation is not related to surface atomic structure, he next devised a complementary test of the hypothesis that surface structure is unimportant. Calcite and vaterite are two modifications of $CaCO_3$, which can be prepared in such a way that the well-developed faces of the calcite have no hexagonal symmetry while those of the vaterite do have hexagonal symmetry with lattice spacing only 9% different from that of ice. If two such samples yielded equal nucleating effects, one might conclude that crystal structure of the nuclei faces is unimportant. However, Manson's tests showed that while the calcite crystals proved ineffective with subcooling to as low as $-25°C$, the vaterite was very effective at $-20°C$ and gave a few crystals even at temperatures as warm as $-13°C$. Hence, Manson's complementary experiment seems to stand as confirmation of the original premise on which AgI was selected by Vonnegut, namely, that nuclei operate through epitactic phenomena that demand lattice compatibility between nucleus and ice. One would hope that nucleating tests might soon be carried out on samples of the low-temperature cubic form of AgI so prepared as to have *minimal* development of (111) faces, a further test of the epitactial hypothesis. To my knowledge this sort of test has not yet been attempted, though it would complement Manson's reported study rather better and more conclusively than the calcite-vaterite test.

Almost concurrently with Manson's just-cited experiments, related studies were being carried on by Pruppacher and Sänger [75] in Switzerland that also gave the result that the cubic γ-AgI is about as good a nucleant as is the hexagonal β-AgI. Pruppacher and Sänger did not discuss the possibility of (111) faces controlling the nucleation in the cubic case, so Manson's work is for the moment the only evidence for (111)-plane action. On the other hand, Pruppacher and Sänger seem to have made a potentially very important discovery in their further work on nucleation. Among a very large number of substances which they tested as nuclei, cupric sulfide (CuS) appeared to rival AgI in nucleating efficiency; and to aggravate the general mystery, CuS, though hexagonal in crystal form, has lattice constants very different from those of ice (for CuS, $a = 3.80$ Å, $c = 16.43$ Å). Pruppacher and Sänger feel that epitaxy, though sufficient, cannot now be accepted as necessary for nucleation, and have as an alternative examined evidence bearing on Weyl's hypothesis that nucleation depends upon polarizability phenomena. On the latter hypothesis, nucleation of subcooled water drops is visualized as ensuing when a silver iodide (or other) crystal contacts a drop as a result of a random collision and thereupon locally alters the oriented dipole layer at the surface of the water drop in such a way as to lower the free energy barrier to freezing. No quantitative support for this hypothesis has yet been given and it implicitly depends upon the existence of essentially dry AgI particles reaching the subcooled region of clouds. Other recent work [76] strongly suggests (see below) that AgI nuclei never can remain *dry* during intracloud ascent to those levels where they can nucleate, so I have strong doubts that Weyl's hypothesis can be a correct description of the actual function of AgI or other similar nuclei in typical cloud-seeding operations. Other objections to Weyl's polarizability model can be raised, but these cannot put aside the now-puzzling and important problem of how CuS functions.

Many investigators have reported long lists of substances which they had found to serve as ice nuclei, and if all of these were correct, the accounting on theoretical grounds would indeed be difficult. However, many of these results are mutually contradictory, and since contamination is easy, above all in laboratories where AgI has been used without great care being taken to avoid casual contamination of room objects, one cannot take them all at face value. Mason and Hallet [73], after testing many of the reported nucleants under very carefully controlled conditions and after making various kinds of contamination tests, concluded that silver or iodine contamination has probably accounted for most of the observations of high nucleating efficiencies. They were also disposed to question Pruppacher and Sänger's results on CuS on grounds that CuS crystals themselves yield scintillations similar to those of true ice crystals, but in a subsequent report [77], they confirm the CuS observations, using a very pure and specially prepared sample. They found this sample of CuS capable of nucleating at $-6°C$.

Further insight into the critical question of the physicochemical nature of effective ice nuclei has come from studies [78] carried out by Birstein on the role of adsorption in heterogeneous nucleation. Samples of various nuclei loosely deposited in the small platinum bucket of a McBain adsorption balance were allowed to come to equilibrium with each of a series of successively higher vapor pressures, and the thickness (number of molecular layers) of the adsorbed water deposit was determined for each pressure by standard adsorption techniques. Birstein found that unusually large numbers of molecular layers of H_2O were adsorbed by both AgI and PbI_2. At $-20°C$, AgI nuclei of average radii of about 0.2 μ adsorbed approximately 200 molecular layers of H_2O near ice saturation, and PbI_2 nuclei were only slightly less extreme in their adsorption. Since adsorbed layers of this depth were built up with the vapor held at or below *ice* saturation, and since a nucleus with as many as 200 molecular layers is already essentially an ice crystal as far as further growth is concerned, Birstein drew the conclusion that both of these nucleants can undoubtedly act as deposition (sublimation) nuclei. He supported this conclusion by growing ice crystals on AgI nuclei within a cold chamber containing only vapor and no drops at −20°C (see also the 1954 work of Schaefer [76], cited later, for very strong evidence that AgI and PbI_2 can act as deposition nuclei). Subsequently, Birstein [79], repeated these adsorption experiments with *photolyzed* AgI nuclei and found a marked reduction in adsorption. For example, at −20°C, only about 25 molecular layers of H_2O were deposited at a vapor pressure corresponding to ice saturation, or only one-eighth of the deposit previously found on unphotolyzed AgI nuclei in the earlier study. This latter result shows so very specific a reduction in nucleating ability of AgI after exposure to ultraviolet light of about sunlight intensity that it plus all of the field and laboratory observations cited earlier here seem to close the discussion of whether AgI nuclei actually do undergo photolytic deactivation.

Since PbI_2 is not susceptible to photolysis in the way that AgI is, and since early cold-chamber experiments revealed that PbI_2 approaches AgI in nucleating efficacy, it would seem reasonable to look to PbI_2 as a possible substitute for AgI as a nucleant in field programs of cloud modification. An investigation conducted by Schaefer [76] in 1954 seems to me to imply that this is not an attractive possibility, although Schaefer did not himself draw this inference from his work. Using a continuous cloud chamber of the diffusion type, Schaefer performed experiments which seem to demonstrate rather neatly that, whereas both AgI and PbI can function as deposition nuclei at about −5°C, they are not equally effective as freezing nuclei. To test the latter point, Schaefer allowed all condensation nuclei ordinarily present in the air to rain out of the diffusion chamber and then introduced the AgI or PbI_2 nuclei into the upper part of the chamber where the temperature was about 5°C and where the vapor was supersaturated with respect to liquid water. Both the AgI and the PbI_2 nuclei then immediately functioned as *condensation* nuclei (a result consistent with Birstein's adsorption findings) and the resultant droplets fell through the diffusion chamber to subzero levels. But, whereas the droplets grown in this way on AgI froze reliably at about the −5°C level, those grown on the PbI_2 nuclei did not freeze until they fell to about the −20°C level. Schaefer suggested that this marked difference in behavior might be due to the fact that PbI_2 is some 10^5 times more soluble than is AgI. I feel sure that this does offer the clue to the explanation, because if one calculates the amount of water required just to dissolve a nucleus of PbI_2 of 0.1 μ radius, it proves to be about equal to that in a drop of 1 μ radius, while a nucleus of AgI of the same size will not be fully dissolved unless contained in a drop of about 100

μ radius. Now, in ordinary cloud-seeding operations with ground generators, the nuclei enter the clouds primarily through their bases; therefore, in almost all conceivable cases, the nuclei will then spend times of the order of minutes ascending through regions characterized by saturation with respect to liquid water. Hence, I think it is inevitable that such nuclei will always serve first as condensation nuclei and be enveloped by liquid water before they ever reach subzero levels where they can promote crystallization. But this means that whereas the highly insoluble AgI nuclei can survive as solid particles within their containing cloud drops during the ascent, the much more soluble PbI_2 nuclei will be wholly dissolved and will thereby have lost their effectiveness as ice nuclei.

It seems reasonably safe to generalize this behavior to all potential nucleants on the basis of Birstein's work. That is, those very crystalline particles whose structure promotes epitactic growth of ice crystals by deposition from the vapor phase should also be tolerably good condensation nuclei, so these nuclei will be *inside* water drops when they reach subcooled regions after ground release. (Aircraft seeding at or above the freezing level may present somewhat different conditions.) From this, I would draw three inferences. (1) Action of AgI or other similar nuclei can scarcely involve the sort of external-surface-contact dipole-reorientation effects which Pruppacher and Sänger [75] have suggested on the basis of Weyl's hypothesis. (2) Laboratory cold-chamber experiments and expansion-chamber experiments probably give a much less relevant measure of true cloud-seeding effectiveness of crystalline nuclei than has previously been appreciated; such experiments permit epitactic growth by deposition without competition from prior condensational growth, and thereby give over-optimistic results with moderately soluble nuclei such as PbI_2. (3) In the case of the latest addition to the small list of highly effective ice nuclei, cupric sulfide, one finds that its solubility happens to be such that a particle of 0.1 μ dry radius will just dissolve in a drop of about 10 μ radius, so there is only marginal possibility that CuS would actually work under typical ground-generator seeding conditions. This last case is, however, so much a borderline case that final conclusions as to its prospective effectiveness can only be drawn from much closer examination of the kinetics of condensational growth upon CuS particles. It should be noted that the Kelvin effect on solubility of small particles works against nucleation effectiveness in the case of submicron nuclei, though this will only become a serious factor for nuclei of radii well under 0.1 μ.

In addition to all of the microphysical questions concerning behavior of cloud-seeding nuclei, there have also been raised during the past few years some questions concerning whether ground-released nuclei can always be expected to be carried by diffusion processes to sufficiently high altitudes to enter clouds in the first place. In experiments in New Mexico [80, 81] and later in Australia [29], somewhat discouraging results were obtained, for the reported heights to which fluorescent zinc sulfide tracer particles reached after many tens of miles of downwind drift were rather less than typical cloud-base heights above terrain. This question is still open, though Smith and Heffernan, in a subsequent report [70], give data showing that their earlier results [29] had been obtained under conditions where eight of twelve cases were characterized by inversions low enough in the atmosphere to significantly influence diffusion, and the remaining cases were those in which the tracer plume was not explored out to more than 11 km from the source. Hence it would seem that their 1955 results must be interpreted in a restricted sense. Prior to this clarification, a tracer experiment to complement the experiments discussed in references 29, 80, and 81 was set up in Arizona by Kassander [82] for the chief purpose of examining vertical diffusion under such extremely favorable circumstances that *if* no particles had reached cloud-base altitudes the then extant pessimism would seem to have been fully confirmed. These favorable circumstances involved highly unstable air, namely, adiabatic lapse rates from terrain to about 3 km and a steep mountain slope rising to about 2.5 km just downwind from the release point. The ZnS tracer particles were actually found at altitudes as high as 4 to 5 km in this experiment, so the one-sided power of the test was not realized. Instead, the test showed that ground-released particles certainly attained cloud-base altitudes under at least these very favorable meteorological and topographic circumstances. It was repeatedly observed, during this series of experiments, that a very large degree of meandering of the plume occurred. Slight wind changes upwind of and over the mountainous terrain where this experiment was conducted made it extremely difficult to determine just where the tracer particles were going to drift, even though double-theodolite balloon observations and an auxiliary smoke plume were employed as aids. This experience tends to cast doubt on the frequently repeated assertion that one can "see" that a given generator is producing precipitation downwind, at least in rough terrain. From this, and from the few other diffusion experiments carried out to explore the drift of ground-released nuclei, it has become quite evident that much remains to be learned about diffusional ascent of nuclei. From the standpoint of optimizing cloud seeding, one wishes to be able to predict what flux of nuclei should be released at the generator site in order to insure a specified concentration at a given level within clouds at a roughly given distance. An error of much more than one order of magnitude in concentration may, on existing theory, spell the difference between seeding success and failure, yet such a precision places demands on turbulence theory that cannot at present be met, above all under conditions of airflow over orographic barriers of complex form. Since seeding of orographic clouds holds, in many ways, better promise than seeding of any other cloud type, this difficulty is not a trivial one.

Having now discussed the development of the silver iodide seeding technique and having summarized some of the most important work of the past few years on microphysical and macrophysical problems that have arisen in conjunction with this development, it is next in order to attempt a summary of the present state of the art of silver iodide seeding as a practical measure for increasing precipitation. It is, of course, exactly this kind of summary that the geographer, hydrologist, and ultimate water user all press the meteorologist to present. Readers in that category may not, therefore, be satisfied with what I have to say, for I simply do not believe that any concise statement can be given, unless it be the simple one that the practical efficacy of AgI seeding is still not adequately assessed.

In 1956 an international conference on the scientific status of cloud modification [83] was held for the chief purpose of taking stock of existing knowledge concerning cloud seeding of all kinds. I have summarized that conference elsewhere [84] in a form that seemed to represent the consensus of the participants. The essential conclusion of the conferees, as far as concerns AgI ground-generator seeding, was that there have not yet been reported any seeding experiments so designed and so evaluated as to permit either a clear-cut positive or clear-cut negative decision as to seeding efficacy. It seemed generally agreed by participants at this conference that the best prospects for success in silver iodide seeding with ground generators lie not in regions of level plains but in regions where marked orographic barriers block prevailing winds that tend to arrive with large moisture content.[4] In such areas, nuclei can be swept relatively rapidly into clouds that tend to be quasi-permanent, and existing evaluations for seeding projects in areas filling these requirements look promising. Above all, it seemed unanimously agreed by conferees that, although early enthusiastic claims now seem evidently to have been excessive, there still remain so many poorly understood factors, which may permit real improvements through future advances, that a certain optimism is in order. In the short

[4] Thom [85] has reported statistical evaluations of a number of commercial cloud-seeding programs from which he concludes that nonorographic projects exhibit no statistically significant increases, whereas the orographic projects, particularly on the West Coast of the United States, do indicate, according to his analyses, statistically significant increases averaging about 15%. Because he had available for analysis only results of nonrandomized seeding schemes, he urged that these findings be taken chiefly as guides to further theory and experiment. That orographic clouds offer optimum opportunity for augmenting natural precipitation has been stressed by others, for example, Bergeron [86] and Ludlam [87].

time that has since elapsed, I do not believe this picture has changed in any essential way.

This residual indeterminacy may seem odd to the reader who is aware of the very large number of AgI ground-seeding projects that have been carried on during the past decade by commercial operators throughout the United States and other countries, for it might be expected that more than ample field data would by now be available from such projects to permit drawing valid conclusions as to AgI seeding efficacy. This, however, is surprisingly far from being the case. Until quite recently, no such projects had ever been set up with a tightly controlled statistical design of the type demanded by the inherent variability of natural precipitation, so no very conclusive statistical inferences have been deducible from even the best of these many projects. Where such projects have been evaluated by disinterested meteorologists and statisticians, the results have fallen in about equal numbers on the side of positive and negative seeding effects, but it is principally to be stressed that little weight can safely be placed on either the optimistic or the pessimistic findings of analyses of improperly designed experiments.

This evaluation problem will be considered again in Section 5, where the severe difficulties that beset evaluation of cloud-modification programs will be briefly discussed. Here it need only be said that it has been most regrettable that despite the very large sums of money expended on commercial silver iodide seeding activities since 1946, very little reliable scientific information has been derived from these sources. One objective measure of the volume of sound information contributed from these sources is to be found in the total number of published papers in the American professional meteorological journals that have come from such seeding projects. Two issues of the American Meteorological Society's *Meteorological Abstracts and Bibliographies* [88, 89] have been devoted to cumulative abstracts of articles on cloud seeding appearing up to about mid-1955. Out of a total of 430 abstracts for all countries of the world and for all types of publication (professional and popular), I counted just four which had been contributed to any of the three United States professional journals covering the field of meteorology by workers associated with commercial seeding projects.

Research on silver iodide seeding has revealed a great deal that was not known in 1947 when Vonnegut discovered its nucleating capabilities in the laboratory. Properly designed field experiments continuing over a number of years are sorely needed, and much more extensive laboratory work is needed on nucleation physics in general and on generator technology in particular. It is still much too early to conclude that photolysis presents insuperable difficulties, despite recently established photolytic deactivation rates. Field trials of cupric sulfide (properly designed in the statistical sense) may now be in order, and certainly a continuing search for still unsuspected ice nucleants must be carried on as a matter of basic research. Finally, since there still remain many fundamental gaps in our knowledge of the actual world-wide importance of the ice-crystal process which we attempt to stimulate by heterogeneous nucleation, it is abundantly clear that progress will be seriously impeded unless basic research on cloud dynamics and on natural precipitation mechanisms is vigorously pursued. All of these efforts derive importance from the widely accepted view that some kind of ice-crystal nucleation probably affords greater probability of practical influence on precipitation processes than any other kind of cloud modification now accessible to man. The fact that early claims and hopes for very large increases in precipitation from AgI seeding have not been substantiated, and the fact that initially unrecognized difficulties have, through the research of the past dozen years, been identified and partially elucidated, simply do not justify pessimism as to long-term possibilities. The prospect of heterogeneous nucleation of the ice-crystal process, anticipated by Findeisen and brought to laboratory reality by Vonnegut, remains a challenge to meteorological and physical research whose importance demands, in my opinion, very much more over-all effort than it has yet received.

4.3. Artificial Stimulation of the Accretion Process

In this section, only those accretion processes occurring at temperatures above freezing, or at least involving liquid water rather than the ice phase, will be considered. As has been elaborated in Section 3.3.3, such accretion processes depend, in one way or another, on the existence of a broad range of cloud-drop sizes. But inasmuch as condensation alone yields only quite narrow drop-size distributions when operating on typical populations of condensation nuclei (Section 3.2), initiation of the liquid accretion process must depend upon the presence of a few very large nuclei in the air entering the cloud, upon some kind of random collision processes, upon microturbulence effects, or upon electrical effects still poorly understood. It bears repeating that, at present, there is nothing like definitive evidence as to which of these factors tends to be most significant in promoting accretion under natural conditions.

Quite probably the first listed factor (large nuclei) frequently plays a decisive role in convective clouds formed in fresh maritime air masses, especially when abundant production of sea-spray has occurred upwind of the area in question. Woodcock [90] and Ludlam [41] have considered the role of comparatively rare (order of one per liter) giant sea-salt nuclei in initiating accretion, and observation [15, 90, 92] of the concentrations of these giant nuclei (equivalent dry radii of the order of 10 μ) tend to support the view that they often occur in numbers adequate to account for observed rain from nonfreezing clouds in maritime air masses. Too few measurements have yet been made throughout the world to make it clear that maritime air *always* has ample giant salt nuclei (see [91] for some recent aircraft observations); in fact, existing data make that seem doubtful, since rather strong winds appear to be required to form the larger nuclei in appreciable numbers. Available data indicate that the giant nuclei are only produced at sea-surface wind speeds in excess of about 10 m sec^{-1}.

Turning to the case of continental interiors, one finds the picture even less clear. Woodcock found no appreciable reduction in total numbers of nuclei after passage of about 100 km over land. Twomey [92] found quite variable concentrations of large salt nuclei on flights extending for hundreds of kilometers across southeastern Australia, with low counts downwind of convective clouds and under post-frontal inversions. Mere length of overland air trajectory did not seem to be closely correlated with nuclei counts. Twomey noted that there were occasional situations, chiefly in summer, when high vapor contents occurred simultaneously with low counts of the oversize nuclei needed to start accretion processes. Those situations might, he felt, be susceptible to useful modification by seeding with water drops or large salt particles. Reitan and Braham [93] made measurements of large nuclei at the ground in Illinois and found surface concentrations of large particles too low to initiate accretion, but later aircraft measurements in the same area seem to suggest that *surface* nuclei counts are unrepresentatively low, a point that deserves consideration in evaluating all surface nuclei observations. One other set of salt-nuclei counts will be cited to indicate the type of data at hand. Measurements made in the Punjab in northwestern India [94], at a distance of about 1000 km from the Arabian Sea, showed that during the summer monsoon season, only about 5 to 10 particles per cubic meter with masses greater than about 10^{-9} gm occured in surface air, rather too few to serve in starting the accretion process.

In general, it must be recognized that far too few observational data on continental, and even maritime, counts of giant sea-salt particles have been gathered, and it is to be hoped that more such data will be obtained soon. Despite lack of positive evidence that significant deficiencies of giant nuclei occur, speculation and investigation concerning the feasibility of stimulating the collision-and-coalescence process have proceeded. Two chief modification methods have been proposed: seeding with a spray of large water drops released from aircraft, or seeding the updraft air of convective clouds with dry NaCl particles large enough (masses of order of 10^{-9} gm) to grow into oversize cloud drops quickly enough to start accretion ahead of the sometimes too-slow natural processes.

4.3.1. The Water-Spray Technique. In his 1948 paper on collisional growth, Langmuir [36] briefly suggested that useful stimulation of drop growth might be obtained by spraying water from an airplane by use of suitable nozzle arrangements. In a 1950 report [95], Langmuir proposed that water seeding might best be accomplished by dropping from an aircraft a quantity of the order of a gallon of water in a balloon attached to a string some 100 ft in length. As such a balloonful of water fell, it would lose much of its initial forward momentum so that on breaking open, after coming to the end of the string, the atomization due to first contact with the air would not break the water into drops too small to start what Langmuir anticipated might be a chain-reaction process (accretional growth followed by raindrop breakup and then more accretional growth). Later experience has made it virtually certain that a mere gallon of water could not significantly affect cloud precipitation processes but variants of this basic idea have produced observable cloud effects.

In 1948, Coons *et al.* [96] carried out some water-seeding trials on cumuliform clouds, using two spraying methods. A P-61 aircraft with two 165-gal wing tanks emptied its water load in about 2 min through 2-in. diam solenoid-operated valves. At an airspeed held down to 150 mph to minimize impact breakup, and with both valves open, this arrangement gave a seeding rate of about 50 gal per flight mile. An alternative scheme utilized a sprayer, mounted in the tail of a B-17, through which water could be pumped at a rate of about 1 gal/mi. The investigators estimated that the latter technique gave drops of about 0.5 mm diam, while the former method was thought to yield rather larger drops. This experiment is cited primarily to illustrate water dispersal techniques and because it appears to have been the first serious effort to carry out water seeding. The mode of evaluation of results in this experiment made it difficult to assign definite value to the water seeding, and a total of only eight clouds were seeded at the 50-gal/mi rate during the tests.

Bowen [97], in 1952, deduced from his previous calculations [40] concerning the collision and coalescence type of liquid accretion process that if drops with radii of about 25 μ could be sprayed into the bases of suitable convective clouds, rain should fall in a few tens of minutes. An aircraft was fitted with a 60-gal water supply tank and two spray-bars to permit release of water drops along a strip of air some 6 mi long, thus yielding a seeding rate of about 10 gal per flight mile. No experimental determination of drop size from the sprayer was made, but Bowen suggested that a mean drop-radius of about 25 μ was obtained, the range of sizes being broad. In eleven experimental spraying operations on cumuli, Bowen found four cases where rain or hail appeared shortly after spraying, four where only precipitation streaks (virgae) appeared, and only one of the eleven cases wherein no observable results of any kind could be discerned. These results seemed suggestive of real effects, but it deserves strong emphasis that such seeding designs where the clouds to be treated are selected subjectively without randomization render virtually impossible any quantitative applications of probability arguments in the evaluation of the efficacy of treatment. This difficulty will be discussed somewhat more fully in Section 5.

The most extensive experimental work on water seeding thus far performed has been done by Braham, Battan, and Byers [58], in 1954 in the vicinity of Puerto Rico and in the midwestern United States. A 400-gal water tank installed in the bomb-bay of a B-17 was equipped with a dump valve through which the water could be rapidly released through the bottom of the fuselage into the ambient air. When a circular valve 4 in. in diameter was employed, the 400 gal emptied in about 70 sec, giving a seeding rate of about 130 gal per flight mile at an airspeed of about 180 mph. The randomized-pairs method of seeding (described in Section 4.2.1) was used in all of these water-seeding trials. After treating over 25 pairs in the Puerto Rico area with the 4-in. valve, it appeared that no significant effects were being obtained with the seeding rate it gave, 130 gal/mi, and visual observations by flight observers tended to the same conclusion. Hence the small valve was replaced by a large dump valve, approximately 10 in.^{2}, which would release the 400 gal in only about 18 sec, thus giving a linear seeding rate of about 450 gal/mi at cruising speed of the B-17.

To determine the drop-size distribution produced by this massive dumping method, releases were made with the B-17 flying only a few tens of feet above an airport runway on which dye-impregnated filter papers had been laid out over a large area. The results showed that about 10% of all drops had radii under 150 μ, about 50% of all had radii under 250 μ, and about 90% of all had radii under about 500 μ. That is, the bulk of the drops sprayed into the clouds by the large valve were equivalent in size to typical drizzle drops. The question of how the spray output was distributed in a direction crosswise to the flight axis was of interest in understanding the seeding process, and efforts were made to determine this in approximate fashion by low-altitude drops over other arrays of dyed filter paper. In one test with the large valve flown 50 ft above the ground, the results showed that 90% of all of the water released was confined to a strip of 50 ft in width. Since downwash and tip-vortex effects are probably chiefly responsible for the lateral dispersion prior to control by purely cloud-turbulent effects, and since 50 ft of drop should, for a B-17, have given nearly full opportunity for these aerodynamic effects to act, these measurements are probably typical of initial lateral dispersion in actual cloud treatment. Radar tests were made to determine whether the cloud of drops emanating from the valve could be confused with cloud precipitation echoes themselves. It was not found possible to observe just the water plume itself with the radar set subsequently used in evaluating treatment efficacy.

Whereas Bowen [97] introduced his water spray into the bases of treated clouds, the Chicago group concluded, from theoretical considerations involving their measured drop-size distributions, that seeding near the cloud tops should be carried out in their flights. In the tradewind cumuli near Puerto Rico, where cloud bases lie near 2000 ft and tops typically range from 6000 to 10,000 ft altitude, the seeding runs were flown at from 5000 to 7000 ft altitude. In the midwestern summer cumuli of the United States, whose bases ranged from 5000 to 8000 ft and tops from 12,000 to about 20,000, the seedings were usually done in the layer from 10,000 to 15,000 ft altitude.

No observations of precipitation actually reaching the ground could be made under the experimental conditions of the trials in either geographical areas. Criterion of treatment effect was therefore necessarily limited to appearance or nonappearance of a radar echo in the two members of each pair. Using this radar-echo criterion, the Chicago group found that no significant effect was produced with either the small or large valve in seeding the cumuli of the Midwest, but only a small number of treatments was carried out, so those results are not conclusive for the Midwest. Using the small valve, they found no significant effect in tradewind cumuli near Puerto Rico, as stated earlier. But using the *large* valve (seeding rate 450 gal/mi) in Puerto Rico, they did find statistically significant evidence that water seeding was increasing the fraction of clouds that reached the stage of precipitation (formation of drops large enough to yield a radar echo). Specifically, statistical tests call for the *rejection*, at the 0.017-confidence level, of the hypothesis that seeding had *no* effect on the probability of precipitation when water was seeded at the 450-gal/mi rate into the trade cumuli. The seeding effects are somewhat more simply represented in terms of fractions of treated and untreated clouds that developed echoes, and this information is contained in Table VIII. An average of 48% of the trade cumuli seeded at 450 gal/mi gave echoes, while only 23% of untreated members of randomized pairs gave echoes, or less than half as many as in the treated population. The 95% confidence intervals for these two proportions barely overlap. Additional physical analysis of these experiments [98] lent further strength to these statistical conclusions that a real effect was produced by water seeding at the heavier rate.

The fact that no observable effects on stimulation of growth of drops to sizes yielding a radar echo were found in the Midwest at even the heavier

TABLE VIII. Observed proportion of spray-treated and untreated tradewind cumuli which developed radar echoes and the 95 and 99% confidence intervals for these proportions. (From [58].)

	Proportion of clouds with echoes		
	Observed	Confidence intervals	
		95%	99%
Untreated	0.23	0.14–0.34	0.12–0.37
Treated at rate of 130 gal/mi	0.19	0.07–0.36	0.05–0.42
Treated at rate of 450 gal/mi	0.48	0.33–0.63	0.31–0.69

seeding rate, and that a significant effect was obtainable in the Caribbean cumuli with the 450-gal/mi rate and not with the 130-gal/mi rate, raises questions concerning the earlier attempts of Coons *et al.* [96] whose seeding rate was either 1 or 50 gal/mi depending on which scheme was used, and the experiments carried out by Bowen [97] with a seeding rate of only about 10 gal/mi. However, the latter is not directly comparable with the Chicago seeding of large drops in cloud tops for Bowen's method involved seeding with smaller drops in cloud bases. Here, as in so many other places in the field of cloud modification, seemingly contradictory results cannot yet be adequately resolved for sheer lack of sufficient data.

From the data at hand, this method of spraying very *large* drops into the *tops* of convective clouds appears at present to be too costly to warrant large-scale application. However, since positively identified effects have at least been obtained by this method, since there may still be many unexplored possibilities for improving this particular technique, and since there may be better prospects in Bowen's less costly approach of spraying *much smaller* drops (though still large compared with cloud drops) into the *bases* of convective clouds, the water-seeding methods clearly warrant more careful investigation on a research basis. At this point, it should be almost unnecessary to comment on the indispensable role which fundamental knowledge concerning the entire accretion process must play in efforts to optimize this kind of cloud treatment.

4.3.2. The Salt-Seeding Technique. In Sections 3.2, 3.3.3, and 4.3, it has been pointed out that there may sometimes occur situations wherein addition of very large salt particles to natural nuclei populations entering cloud bases might accelerate the growth processes dependent upon accretion. There has been only a small number of attempts to test this scheme. Schaefer has reported [99] attempts to develop means of generating large numbers of particles of NaCl. A simple coke-burning furnace was built to melt and vaporize NaCl in a way that formed about 10^{11} particles per second with radii of the order of 1 μ. The physics of the evaporation-condensation technique of generating such particles limits the upper size readily obtainable, and for the particles with radii of the order of 10 μ or larger needed to stimulate accretion, Schaefer recommended some form of mechanical pulverization. No seeding experiments using NaCl or other hygroscopic salt were described by Schaefer.

In 1954, Davies [100] carried out experiments in East Africa in which finely ground salt was carried into clouds by means of balloons which were exploded by gunpowder at predetermined altitudes, showering the salt particles into the cloud. Of 64 cumuli so seeded, some form of precipitation was observed in 47 cases within a time lapse of from 7 to 35 min (average 24 min) after treatment. Of these 47 cases, 37 gave precipitation that actually reached the ground. These are statistics typical of those so frequently encountered in the field of cloud modification where the results seem tantalizing, but where lack of randomization stands firmly in the way of drawing any meaningful conclusions.

The cumuli of the summer monsoon of India have very warm base temperatures and often precipitate before their tops reach the rather high freezing level of that area and season, implying that collision and coalescence processes must be dominant in natural precipitation in such clouds. After making two years of counts of hygroscopic nuclei in Pakistan, and finding that the average surface-air content of giant nuclei with masses greater than 10^{-9} gm was about 5 to 10 particles per cubic meter (too few to be the chief agent in starting accretional growth, unless a process akin to Langmuir's chain-reaction process can occur), Fournier d'Albe *et al.* [94], set up a program of experimental salt seeding in the Punjab. Salt was ground to desired sizes, stored in sealed tins, and dispersed into the air from a hand-operated centrifugal hot-air blower of economical design. Seeding was restricted to days when the surface monsoonal flow from the east was present, and this occurred on 39 days during the two-month period in 1954 when operations were underway. The results of an analysis of rainfall in the area felt to be chiefly influenced by seeding, as contrasted with rainfall in adjoining control areas, indicated that the seeded area received more rainfall than could be expected on the basis of the concurrent amounts in the control areas. However, it was not possible to conclude with assurance that this was due to the seeding itself, for the same reasons that have arisen in most seeding projects involving only the target-and-control comparison scheme, namely, that this scheme does not safely overcome the subtle but all-important effects of natural rainfall variability.

Although a few other attempts at salt seeding have been made, these two are the only ones of which I now know wherein even approximate deductions as to seeding effect can be made. They appear to be favorable results but neither is conclusive, because of limitations in statistical design.

They scarcely exhaust the possibilities for research on this problem. Until better techniques for counting salt particles have been developed and widely used to gather now-rare data on averages and fluctuations in natural giant-nuclei counts in time and space especially in the free atmosphere rather than just at the surface, and until the relative importance of intrinsic coalescence processes as compared with effects of rare giant nuclei is ascertained, salt seeding will be pursued in about as dim a light as have many other seeding efforts. The alternative to hard-won physical insight is a sustained program of properly randomized seeding trials from which reliable deductions may ultimately be drawn solely from observed precipitation amounts.

4.4 Other Types of Cloud-Modification Techniques

All of the foregoing remarks have been concerned with efforts to induce *more precipitation to fall* from clouds by various artifices. There have been other objectives recognized in cloud-modification studies, including dissipation of cloud decks for aviation purposes, suppression of hail to reduce crop damage, suppression of thunderstorm electrical activity to prevent lightning fires over forested areas, and some thought has even been given to attempts at increasing cloud coverage indirectly in order to control solar radiation receipts at the earth's surface. None of these objectives has received as much research attention as that of stimulating precipitation, so only a few comments will be made on these other techniques.

Aufm Kampe, *et al.* [101, 101a] have seeded numerous stratus decks with both dry ice and silver iodide released from aircraft. Conversion of a subcooled cloud of high visual opacity to one composed of a much smaller number of large ice crystals quickly renders the ground visible from points above the deck, even when the latter is as much as 2000 ft thick, according to their reports. Similar effects have been found by earlier workers, and these results constitute particularly clear-cut evidence that artificial treatment can most assuredly induce the Bergeron transition in subcooled clouds. Aufm Kampe *et al.* have reported that dry ice is generally more effective than AgI in stratus-clearing, and that seeding rates of as much as 10 lb of dry ice per mile will not lead to overseeding in convective clouds (though overseeding in stratus-clearing has, of course, somewhat different meaning than in rain stimulation). Their experience indicates that normally present cloud turbulence diffuses the ice particles laterally at such a rate that the seeded strip grows to a width of two miles in about 30 min.

Suppression of hail and suppression of lightning are still only superficially understood [102, 105]. In each instance, the goal is that of so overseeding that the available subcooled water is converted into too many small ice particles to support hail and lightning development. Since hail damage is often a problem in areas where additional precipitation is beneficial, these hail-suppression efforts may work at cross-purposes with water requirements, and the same can be true in lightning suppression. Present knowledge does not enable a seeder to determine very precisely just what manipulations of the natural clouds will yield hail- and lightning-suppression without net sacrifice of precipitation. Whether or not future developments may make these and other miscellaneous modification techniques significant is impossible to predict. What can be stated with confidence is that here, as in all other applications, we see a pressing need for much more basic information as to physical processes in natural clouds.

5. The Evaluation of Modification Experiments

The problem of cloud modification to stimulate precipitation seems to have been an instructive one for meteorologists as a group in areas other than just physical meteorology. No other important meteorological research problem, other than a few on merely a laboratory scale, now has or ever before had, as an essential part of its rationale, the manipulation of natural processes as distinguished from the passive observation of those processes. In some experimental fields it is quite simple to apply treatment A and then to determine unambiguously whether effect X is or is not produced as a result of A. In other fields no such straightforward approach can be utilized because so many uncontrollable factors other than A operate concurrently to cause or to inhibit X that extreme danger of falling into the *post hoc* fallacy arises. Cloud modification is very much a field of this latter type. In the first few years of modification efforts there was a disconcertingly large amount of *post hoc* reasoning used. The admonitions of a relatively small number of statisticians seem to have been chiefly responsible for correcting, only a few years ago, the most extreme shortcomings of statistical logic applied to modification studies, though these shortcomings were certainly appreciated by many meteorologists at a much earlier date.

Although the subject of evaluation of cloud-seeding programs is largely outside the scope of this article, it is desirable to discuss evaluation difficulties just enough to see the important role they have played in the history of the past decade of modification efforts. Evaluation subtleties arise from the large inherent variability of natural precipitation, and from the meteorologists' present inability to predict cloud behavior with high enough accuracy to state what would have occurred in the absence of treatment. These circumstances have thrown the problem into the area of statistical inference, and there, broadly speaking, it must lie until substantial gains in physical meteorology have been achieved.

There is a bewildering degree of variability in condensation and precipitation phenomena, both in the small- and large-scale processes. At any instant on, say, a summer day, cumuli in different parts of the sky over a level plain may be evolving in strikingly different ways. In mountainous country, seemingly similar peaks only a few miles apart may, for subtle dynamic reasons, be causing updrafts of such different nature that one summit will be cloud-capped while the other is clear, and a short time later the situation may be reversed. One day's cloud history is often wholly dissimilar to the next. One summer's rain patterns in a given area may be different from those of the preceding summer to an extent not yet explicable in terms of any familiar principles of synoptic climatology. As one deals with increasingly longer time periods, this variability is suppressed more and more for simple probability reasons, but even when one considers precipitation totals for time periods of as long as one year, the historic variability is still uncomfortably large. To display this characteristic, for the case of United States stations, Table IX has been prepared. The annual coefficient of variation (standard deviation divided by mean) of even the least variable of the stations listed there, Iowa City, Iowa, is seen to amount to 0.14. This statistic grows generally larger as one turns to more arid regions. The same figure for Yuma, Arizona, located in the driest section of the United States, is 0.62. Since cloud-seeding activities are perhaps most pressingly needed in the arid regions of the world, one recognizes an inherent bias in the direction of having the greatest background of meteorological "noise" in the precipitation record of those very stations most likely to be encountered in evaluation of seeding experiments.

TABLE IX. Natural variability of rainfall.* (From [104].)

Station	Years of record	Mean annual precipitation (inches)	Coefficient of variation
Iowa City, Ia.	70	35.2	0.14
Boston, Mass.	60	41.0	0.16
Cleveland, Ohio	60	34.0	0.16
Kansas City, Kans.	63	36.1	0.18
Portland, Ore.	60	42.2	0.19
Bismarck, N. Dak.	66	16.3	0.25
Cheyenne, Wyo.	70	14.6	0.25
Ogden, Utah	60	16.2	0.25
Wichita, Kans.	52	29.2	0.26
Sheridan, Wyo.	47	15.0	0.27
Tucson, Ariz.	87	11.2	0.30
Phoenix, Ariz.	75	7.6	0.40
San Diego, Calif.	60	9.9	0.44
Yuma, Ariz.	83	3.2	0.62

* The table gives length of record, annual mean precipitation, and coefficient of variation of annual precipitation for each station.

The fact that root-mean-square deviations of totals for 12-month periods can, as a consequence of purely natural fluctuations, range as high as 60% of the corresponding average values leads to evident difficulty in reliably identifying seeding increases of the order of only 10% in programs carried on for only a few seasons. In modification work, one always confronts the thorny problem of disentangling natural variability from true seeding effects. One central question is always lurking in the background: *Is it probable that the precipitation patterns that appeared immediately following seeding would have appeared even if no seeding had been carried on?*

5.1. Physical Evaluation

If meteorologists only knew so thoroughly all of the intricacies of cloud physics that a given cloud could be examined just prior to cloud-modification treatment to ascertain what its *natural* evolution was going to be, treatment could then be applied and the actual outcome contrasted with the predicted behavior. During the past dozen years, there has always been a certain segment of meteorological opinion which tacitly refused to admit the present impossibility of this approach. That is, many have insisted that one could "see" the effects of seeding, simply through observing individual cases. These and other workers, of course, may indeed have "seen" true seeding effects, but the difficulty of proving this to the satisfaction of anyone fully aware of present ignorance of cloud dynamics and cloud microphysics is not to be underestimated.

Several ways of attempting more nearly physical than purely statistical evaluations have been tried; and often physical and statistical techniques have been fused, as in the University of Chicago dry-ice and water-seeding experiments where appearance or nonappearance of radar echoes became the datum fed into the statistical machinery. There is so much room for improvement in physical evaluation methods that it seems almost irrelevant to describe existing attempts. However, every time radar, photographic, or aircraft observations of clouds can be obtained before, during, and after seeding, a step in the direction of physical evaluation is being taken. The in-cloud observations by research aircraft are potentially most informative, for in this way, drop-sizes, liquid water contents, updraft strengths, presence or absence of the ice phase and of hail or graupel particles, temperatures, and other very pertinent quantities can be obtained. In fact, however, not only is it difficult and expensive to operate suitable planes in this way, but also the whole area of flight instrumentation for cloud physics observations is sorely in need of far more attention than it has yet received.

It does not seem an overestimate of the present state of basic cloud physics theory to suggest that if truly complete physical observational data on a treated cloud could be turned over to the theoretician, he would, by dint of sufficient calculation based on basic physical principles, be able to determine conclusively whether seeding was or was not significant in its effects. This emphasizes the need for striving for all possible physical control in future seeding experiments for, even if the above-mentioned ideal of complete data-coverage is still far from attainable, exact physical measurements of even portions of the problem greatly reduce the statistical burden of evaluation.

5.2. Statistical Evaluation

In one way or another, every seeding experiment is subjected to some kind of statistical evaluation, even if only vaguely in the mind of the empiricist who has scattered dry ice into a cloud and notes what he feels to be an evolution not customary in other clouds he has previously observed. From this implicit and qualitative form of "statistical" evaluation (which was not infrequently the only kind of judgment made in some of the seeding trials of a decade ago), cloud-modification evaluation methods have progressed steadily toward more and more sophisticated schemes as one or another program has come under the scrutiny of meteorologists and statisticians who were distressed at early evaluation approaches. Because the subject is technically involved and basically lies outside the scope of this discussion, I shall not try to summarize more than the major outlines of the evolution of evaluation methodology. But to attempt to do at least this much here seems highly desirable, for one cannot understand the development of the field of cloud modification without appreciating the influence which statistical evaluation difficulties have exerted on the whole subject. Because silver iodide seeding has been carried on more extensively than any other type of seeding, the following is to be assumed to apply to that kind of operation unless otherwise specified.

One of the first evaluation refinements to be introduced, beyond that of noting whether or not rain fell soon after seeding in a given area, was to compare the total rainfall measured at the ground in what was called (often on doubtful grounds) the target area with the normal rainfall for that same area as deduced from climatological records. This so-called "per cent of normal method" took as its criterion of success the appearance of per cents of normal in excess of 100%. But often the seeded time periods were only of the order of a month and the data in Table IX, plus standard probability arguments, show that even with times of the order of a year, one could not incontrovertibly assert that anything but a very large per cent of normal could not have been due to chance meteorological fluctuations. After a time, the per cent of normal method was abandoned by scientifically minded evaluators.

An obvious improvement lay in examining the per cent of normal in the "target area" with the same quantity in one or more "control areas" which were selected so that they lay as near as possible to the target area without, however, lying in the prevailing direction of flow of seeding agent from the target. At this stage of evaluation evolution, appearance of a larger per cent of normal in the target area than in one or more control areas was offered as evidence of seeding efficacy. This was becoming more convincing, but operators who used this method had then to answer the following question. How often, in the historic past, might even larger differences in per cent of normal between these two geographic areas (the seeder's target and control areas) have occurred as a consequence of natural processes? Self-evident as such a question should seem it is still not always asked, or if asked, it is not always satisfactorily answered.

The question stated near the end of the preceding paragraph led next to resort to regression methods. The historic (preseeding) rainfall records of all available stations in the target area were correlated against contemporaneous records for stations in the several control areas, and the standard error of estimate (or other equivalent statistic) was then computed for the regression of target on control area amounts. The time period involved might be units of a year, a season, a month, or even individual storm periods. By plotting on a regression diagram the point representing simultaneous values of target and control for the current (seeded) period, and computing the residual deviation of the target ordinate above (if an apparent increase had occurred) the regression-line ordinate corresponding to the given period's control-area precipitation, a probability statement could be formulated expressing the likelihood that the observed target excess over regression prediction could have arisen solely as a result of random fluctuations measured by the yardstick of the historic standard error of estimate. This regression method was a very real improvement over earlier methods, and, despite weaknesses, it would be capable of discerning real effects if seeding could be conducted for a long enough period of time and if enough historic data were available for use in calculating the basic regression relations.

However, statisticians soon pointed out that this regression method may contain pitfalls, too. One difficulty fairly easily suppressed hinges upon the concept of heteroscedasticity, that is the property of a bivariate distribution by virtue of which the variance of the dependent variate is not the same for all intervals of the independent variate. In some geographical areas, the precipitation heteroscedasticity occurs in the sense that the dispersion is greatest for high values of the control-area precipitation. If, then, a seeded year is one in which generally heavy precipitation occurs, use of the ordinary standard error of estimate will lead to bias in the direction of regarding as significant a positive deviation from regression that is not really significant. To correct this error, various coordinate transformations such as the square-root, cube-root, incomplete-gamma [105], and logarithmic transformations may be used, though all must be recognized to be only empirical adjustments at the present time.

But still more fundamental objection is raised on the ground that any kind of historic regression method may, when applied to a seeding project carried on for only a few years, give quite misleading evaluation results because of the varying proportions of differing "storm types" that may influence the experimental area. Imagine that two distinctly different types of rain-bringing storms tend to affect the vicinity of a given cloud-seeding-project area during the season in question. Suppose, further, that some years tend to be characterized predominantly by Type A, other chiefly by Type B, for reasons of oscillations in the general circulation. Finally, let Type A storms have such characteristics that they tend to deposit relatively large amounts of rain on the target area but little on the seeder's control area, while Type B storms have circulation characteristics such that they tend to distribute rain in just the opposite manner. Then, if the seeder unwittingly prepares his regression relation from a limited amount of available data that came from a period of years when it happened that Type A predominated, and if a secular trend brings, at about the time seeding begins, a shift to relatively more of Type B storms, the seeder will be misled by his regression analyses into concluding that his efforts are *less* effective than they may actually be. Conversely, if he should happen to base his historic regression relation on data from a period of years when Type B predominated and the seeding period begins after a swing to relatively more storms of Type A, his regression methods will incorrectly lead him to believe that his seeding is producing significant results when it really may be doing nothing at all, or might even be yielding decreases over what would have occurred in the absence of seeding. In short, the hypothesis of storm-types confronts regression methods with a very basic uncertainty. The first discussion of storm types in this particular context of seeding evaluation was present by Jeeves *et al.* [106] in 1953. The reality of storm types is not in doubt; but the importance of their quantitative effects on evaluation statistics has not been settled [105]. (Note that here is a problem lying basically in the field of synoptic climatology which, like so many in cloud physics proper, could not be decisively answered on the basis of existing information when it was encountered in the course of work in cloud modification. Many others might be cited in this same category.)

The statisticians, disturbed with the above shortcomings of even the regression methods, are satisfied only with seeding designs wherein one or another acceptable form of *randomization* is incorporated into the experiment from the start of operations. Details are not pertinent here, but the basic point is that the seeder must do something equivalent to *first* selecting the cloud or time interval as "seedable," and *after* announcing his decision, he must then effectively flip a coin (randomize) to decide whether he will or will not actually seed that "seedable" situation. After many repetitions of this process, two sets of results will have been obtained, those for the seedable-and-seeded and those for the seedable-but-not-seeded cases. These data, processed in any of a number of ways, fulfill the philosophic requirements posed by sampling theory and enable the statistician to define confidence limits or significance limits for the observed effects of treatment (whatever the measure of seeding efficacy might be in a given case).

Whereas several aircraft seeding investigations had been started as much as five years ago with randomized designs, it was not until 1957 that any ground-generator silver iodide seeding project was set up in this fashion in the United States. The latter is now underway near Santa Barbara, California, and will require at least three winter seasons of operation before meaningful results will be forthcoming. The detailed design of this experiment is somewhat too involved to be summarized here. A brief account of some aspects of this undertaking has been published [107].

Randomization can assume many possible forms in seeding trials, but in all cases its function is simple: It greatly reduces (and in theory eliminates) bias entering into the selection of those cases that go, respectively, into the seeded and the unseeded populations between which an evaluative comparison is ultimately to be made. In the absence of randomization, hidden bias of many kinds may enter, sometimes being forced into the design unwittingly by imposition of other criteria. To give a single example, decision to count only treated clouds which last for, say, 30 min following treatment may be made in what seems the sensible effort to avoid diluting the results with clouds which at time of seeding were so near dissolution that they are not fair tests of seeding efficacy. But the lifetimes of all clouds are so short, and the extent to which wholly natural drop-growth processes may succeed in approaching a state of precipitation is so dependent upon cloud lifetime, that such a criterion may inherently bias the selection of ultimately analyzed data quite strongly in the direction of counting only clouds that were going to precipitate *regardless* of seeding. This single example suffices to document the fact that the real problem throughout evaluation is ignorance of cloud physical details.

Why did randomization not appear in evaluation work a dozen years ago? Partly, I feel sure, because there was nothing in the background of earlier meteorological practice comparable to this problem of detecting effects of treatments of complex natural phenomena. And, of course, even in fields where such problems are highly typical, as for example in agricultural research, it has not been more than about two or three decades since these and related ideas have infused standard practice. But, in addition, an obstacle to randomization in those numerous projects where some water using group was retaining the services of a commercial seeder lay in the reluctance of the client to pay for a program in which roughly half of all seedable storms must be allowed to pass by unseeded. Perhaps, viewed in this light, a decade of postponement of randomized projects in the hope that an answer would be otherwise forthcoming was only about what one could expect from groups desperate for more water. The consequence has been, however, that much effort has gone into contract seeding without past or present knowledge of whether the clients did or did not receive any extra precipitation. It is to be hoped that increasing numbers of contract-seeding projects in this country will be set up on a randomized basis, now that one such (Santa Barbara County, California) project has been established. Quick and easy answers cannot be expected; in adopting this resort, we pay with time for information which we cannot now obtain immediately for the reason that fundamental physical research on clouds and precipitation has progressed too short a distance into its complex subject.

6. Concluding Remarks

It would, of course, be unreasonable to expect that a mere dozen years of research should have led to final answers in a problem with as many ramifications as have come to be recognized in the physics of cloud modification. In retrospect, one sees that this complexity was not fully appreciated when present seeding techniques were introduced in 1946–1947. From the viewpoint of pure research, one of the great benefits of the recent interest in cloud seeding has been its stimulation of effort to unravel this web of interacting factors that enter into the precipitation problem.

I believe that most persons in close contact with current research in cloud physics feel that the surface of this field has barely been scratched. It will have been noted by the reader that in the discussions of each of the four main seeding techniques, those involving dry ice, silver iodide, water spray, and giant salt nuclei, it has been necessary to admit that key aspects of the problem which might hold very real promise have simply not been adequately examined up to the present time. Also, even the most sceptical of those who reject as valid evidence the occasional reports of apparently very marked changes in cloud behavior after seeding has been performed must admit that those statistically insignificant cases may have been instances where, though in ignorance of the detailed reasons, the seeder, quite by chance, carried out his operations in what was exactly the optimal procedure. It is this aspect of cloud modification, the prospects of advances that may await studies not yet begun, rather than the often indecisive results of work already completed, that I believe deserves principal emphasis in summarizing the present position of the field. In short, the great need is for much more intensive research effort on all fronts, for the importance of water in our economy is mounting so rapidly that no prospects for augmenting our limited supplies can be overlooked.

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References

1. Hutchinson, G. E. (1954). The biochemistry of the terrestrial atmosphere. *In* "The Earth as a Planet" (G. P. Kuiper, ed.), 751 pp. Univ. Chicago Press, Chicago, Illinois.
2. Fox, C. S. (1952). "Water," 148 pp. Philosophical Library, New York.
3. Sutcliffe, R. C. (1956). Water balance and the general circulation of the atmosphere. *Quart. J. Roy. Meteorol. Soc.* **82,** 385–395.
4. Braham, R. R., Jr. (1952). The water and energy budgets of the thunderstorm and their relation to thunderstorm development. *J. Meteorol.* **9,** 227–242.
5. Huff, F. A. and Stout, G. E. (1951). A preliminary study of atmospheric moisture-precipitation relationships over Illinois. *Bull. Am. Meteorol. Soc.* **32,** 295–297.
6. Reitan, C. H. (1957). The role of precipitable water vapor in Arizona's summer rains. *Tech. Repts. on Meteorol. and Climatol. Arid Regions, No.* **2,** 18 pp. Inst. Atm. Physics, Univ. Arizona, Tucson, Arizona.
7. Los Alamos Scientific Laboratory (1950). "The Effects of Atomic Weapons," 456 pp. U. S. Govt. Printing Office, Washington, D. C.

7a. Mason, B. J. (1957). "Cloud Physics," 481 pp. Oxford Univ. Press, London and New York.

8. Houghton, H. G. (1950). A preliminary quantitative analysis of precipitation mechanisms. *J. Meteorol.* **7,** 363–369.
9. Junge, C. E. (1958). Atmospheric chemistry. *Advances in Geophys.* **4,** 1–108.
10. Howell, W. D. (1949). The growth of cloud drops in uniformly cooled air. *J. Meteorol.* **6,** 134–149.
11. Houghton, H. G. (1938). Problems connected with the condensation and precipitation processes in the atmosphere. *Bull. Am. Meteorol. Soc.* **19,** 152–159.
12. Kraus, E. B., and Smith, B. (1949). Theoretical aspects of cloud drop distributions. *Australian J. Sci. Res.* **A2,** 376–388.
13. Squires, P. (1952). The growth of cloud drops by condensation, I, II. *Australian J. Sci. Res.* **A5,** 59–86, 473–499.
14. Junge, C. E. (1956). Recent investigations in air chemistry. *Tellus* **8,** 127–139.
15. Woodcock, A. H. (1952). Atmospheric salt particles and raindrops. *J. Meteorol.* **9,** 200–212.
16. Woodcock, A. H., and Blanchard, D. C. (1955). Tests of the salt-nuclei hypothesis of rain formation. *Tellus* **7,** 437–448.
17. Krastanow, L. (1941). Beitrag zur Theorie der Tropfen- und Kristallbildung in der Atmosphaere. *Meteorol. Z.* **58,** 37–45.

18. Turnbull, D., and Fisher, J. C. (1949). Rate of nucleation in condensed systems. *J. Chem. Phys.* **17,** 71–73.
19. Mason, B. J. (1952). The spontaneous nucleation of supercooled water. *Quart. J. Roy. Meteorol. Soc.* **78,** 22–27.
20. McDonald, J. E. (1953). Homogeneous nucleation of supercooled water drops. *J. Meteorol.* **10,** 416, 433.
21. Schaefer, V. J. (1946). The production of ice crystals in a cloud of supercooled water droplets. *Science* **104,** 457–459.
22. Bigg, E. K. (1953). The formation of atmospheric ice crystals by the freezing of droplets. *Quart. J. Roy. Meteorol. Soc.* **79,** 510–519.
23. Jacobi, W. (1955). Homogeneous nucleation in supercooled water. *J. Meteorol.* **12,** 408–409.
24. Pound, G. M., Madonna, L. A., and Peake, S. L. (1953). Critical supercooling of pure water droplets by a new microscopic technique. *J. Colloid Sci.* **8,** 187–193.
25. Fournier d'Albe, E. M. (1949). Some experiments on the condensation of water at temperatures below 0°C. *Quart. J. Roy. Meteorol. Soc.* **75,** 1–14.
26. Schaefer, V. J. (1951). Snow and its relationship to experimental meteorology. *In* "Compendium of Meteorology," (T. F. Malone, ed.) 1334 pp. Am. Meteorol. Soc., Boston, Massachusetts.
27. Isono, K. (1955). On ice-crystal nuclei and the other substances found in snow crystals. *J. Meteorol.* **12,** 456–462.
28. Kassander, A. R., Jr., Sims, L. L., and McDonald, J. E. (1956). Observations of freezing nuclei over the southwestern United States. *Sci. Rept. No.* **3,** 18 pp. Inst. Atm. Physics, Univ. Arizona, Tucson, Arizona.
29. Smith, E. J., and Heffernan, K. J. (1954). Airborne measurements of the concentration of natural and artificial freezing nuclei. *Quart. J. Roy. Meteorol. Soc.* **80,** 182–197.
30. Murgatroyd, R. J., and Garrod, M. P. (1957). Some recent airborne measurements of freezing nuclei over southern England. *Quart. J. Roy. Meteorol. Soc.* **83,** 528–533.
31. Findeisen, W. (1938). Die kolloidmeteorologischen Vorgänge bei der Niederschlagsbildung. *Meteorol. Z.* **55,** 121–133.
32. Simpson, G. C. (1941). On the formation of cloud and rain. *Quart. J. Roy. Meteorol. Soc.* **67,** 99–133.
33. Schmauss, A., and Wiegand, A. (1929). "Die Atmosphäre als Kolloid," 74 pp. Vieweg, Braunschweig, Germany.
34. Humphreys, W. J. (1922). How fast can a droplet grow by falling in a cloud? *Bull. Am. Meteorol. Soc.* **3,** 59–60.
35. Milham, W. I. (1912). "Meteorology," 549 pp. Macmillan, New York.
36. Langmuir, I. (1948). The production of rain by a chain reaction in cumulus clouds at temperatures above freezing. *J. Meteorol.* **5,** 175–192.
37. Swinbank, W. C. (1947). Collision of cloud droplets. *Nature* **159,** 849–850.
38. Blanchard, D. C. (1950). The behavior of water drops at terminal velocity in air. *Trans. Am. Geophys. Un.* **31,** 836–842.
39. Gunn, K. L. S., and Hitschfeld, W. (1951). A laboratory investigation of the coalescence between large and small water drops. *J. Meteorol.* **8,** 7–16.
40. Bowen, E. G. (1950). The formation of rain by coalescence. *Australian J. Sci. Res.* **A3,** 193–213.
41. Ludlam, F. H. (1951). The production of showers by coalescence of cloud droplets. *Quart. J. Roy. Meteorol. Soc.* **77,** 402–417.

42. Weickmann, H. K., and aufm Kampe, H. J. (1953). Physical properties of cumulus clouds. *J. Meteorol.* **10,** 204–211.
43. Marshall, J. S., Hitschfeld, W., and Gunn, K. L. S. (1955). Advances in radar weather. *Advances in Geophys.* 1–56.
44. Reynolds, S. E., and Braham, R. R., Jr. (1952). Significance of the initial radar echo. *Bull. Am. Meteorol. Soc.* **33,** 123.
45. Battan, L. J. (1953). Observations on the formation and spread of precipitation in convective clouds. *J. Meteorol.* **10,** 311–324.
46. Adderley, E. E. (1953). The growth of raindrops in cloud. *Quart. J. Roy. Meteorol. Soc.* **79,** 380–388.
47. Telford, J. W., Thorndike, N. S., and Bowen, E. G. (1953). The coalescence between small water drops. *Quart. J. Roy. Meteorol. Soc.* **81,** 241–250.
48. Telford, J. W. (1955). A new aspect of coalescence theory. *J. Meteorol.* **12,** 436–444.
49. Pearcey, T., and Hill, G. W. (1957). A theoretical estimate of the collection efficiencies of small droplets. *Quart. J. Meteorol. Soc.* **83,** 77–92.
50. East, T. W. R., and Marshall, J. S. (1954). Turbulence in clouds as a factor in precipitation. *Quart. J. Roy. Meteorol. Soc.* **80,** 26–47.
51. Arenberg, D. (1939). Turbulence as a major factor in the growth of cloud drops. *Bull. Am. Meteorol. Soc.* **20,** 444–448.
52. East, T. W. R. (1957). An inherent precipitation mechanism in cumulus clouds. *Quart. J. Roy. Meteorol. Soc.* **83,** 61–76.
53. Houghton, H. G. (1951). On the physics of clouds and precipitation. *In* "Compendium of Meteorology" (T. F. Malone, ed.), 1334 pp. Am. Meteorol. Soc., Boston, Massachusetts.
54. Ludlam, F. H., (1948). The forms of ice-clouds. *Quart. J. Roy. Meteorol. Soc.* **74,** 39–56.
55. Smith, E. J. (1949). Experiments in seeding cumuliform cloud layers with dry ice. *Australian J. Sci. Res.* **A2,** 78–91.
56. Squires, P., and Smith, E. J. (1949). The artificial stimulation of precipitation by means of dry ice. *Australian J. Sci. Res.* **A2,** 232–245.
57. Bowen, E. G. (1952). Australian experiments on artificial stimulation of rainfall. *Weather* **7,** 204–209.
58. Braham, R. R., Jr., Battan, L. J., and Byers, H. R. (1957). Artificial nucleation of cumulus clouds. *In* "Cloud and Weather Modification: A Group of Experiments," Meteorol. Monographs, **11,** 47–85.
group of field experiments. *Meteorol. Monogr.* **2** (11).
59. Braham, R. R., Jr., and Sievers, J. R. (1957). Overseeding of cumulus clouds. *In* "Artificial Stimulation of Rain." Proc. Conf. Phys. of Cloud and Precipitation Particles, Woods Hole Oceanog. Inst., Pergamon, London, in press.
60. Vonnegut, B. (1947). The nucleation of ice formation by silver iodide. *J. Appl. Phys.* **18,** 593–595.
61. Vonnegut, B. (1949). Nucleation of supercooled water clouds by silver iodide smokes. *Chem. Revs.* **44,** 277–289.
62. Smith, R. B., in ref. [62] and Boucher, R. J. (1956). Operation overseed. *Final Rept.* 116 pp. Advisory Comm. on Weather Control, December 1955–January 1956. Mt. Washington Obs., Milton, Massachusetts.
63. Warner, J., and Twomey, S. (1956). The use of silver iodide for seeding individual clouds. *Tellus* **8,** 453–459.
64. Reynolds, S. E., Hume, W., Vonnegut, B., and Schaefer, V. J. (1951). The effect

of sunlight on the action of silver iodide particles as sublimation nuclei. *Bull. Am. Meteorol. Soc.* **32,** 47.

65. Inn, E. C. Y. (1951). Photolytic inactivation of ice-forming silver iodide nuclei. *Bull. Am. Meteorol. Soc.* **32,** 132–135.
66. Vonnegut, B., and Neubauer, R. (1951). Recent experiments on the effect of ultraviolet light on silver iodide nuclei. *Bull. Am. Meteorol. Soc.* **32,** 356.
67. Reynolds, S. E., Hume, W., and McWhirter, M. (1952). Effects of sunlight and ammonia on the action of silver iodide particles as sublimation nuclei. *Bull. Am. Meteorol. Soc.* **33,** 26–31.
68. Birstein, S. J. (1952). The effect of relative humidity on the nucleating properties of photolysed silver iodide. *Bull. Am. Meteorol. Soc.* **33,** 431–437.
69. Smith, E. J., Heffernan, K. J., and Seely, B. K. (1955). The decay of ice-nucleating properties of silver iodide in the atmosphere. *J. Meteorol.* **12,** 379–385.
70. Smith, E. J., and Heffernan, K. J. (1956). The decay of ice-nucleating properties of silver iodide released from a mountain top. *Quart. J. Roy. Meteorol. Soc.* **82,** 301–309.
71. Wyckoff, R. W. G. (1948). "Crystal Structures," Vol. 1. Interscience, New York. (Looseleaf.)
72. Manson, J. E., (1955). X-ray diffraction study of silver iodide aerosols. *J. Appl. Phys.* **26,** 423–425.
73. Mason, B. J., and Hallett, J. (1956). Artificial ice-forming nuclei. *Nature* **177,** 681–683.
74. Manson, J. E. (1957). Ice nucleation and the structure of nuclei crystal faces. *In* "Artificial Stimulation of Rain," (H. K. Weickmann and W. E. Smith, eds.). 427 pp. Pergamon Press, London.
75. Pruppacher, H. R., and Sänger, R. (1955). Mechanismus der Vereisung unterkühlter Wassertropfen durch disperse Keimsubstanzen. I, II. *Z. angew. Math. u. Phys.* **6,** 407–416, 486–493.
76. Schaefer, V. J. (1954). Silver iodide and lead iodide as ice-crystal nuclei. *J. Meteorol.* **11,** 417–419.
77. Mason, B. J., and Hallett, J. (1957). Ice-forming nuclei. *Nature* **179,** 357–359.
78. Birstein, S. J. (1955). The role of adsorption in heterogeneous nucleation. I. Adsorption of water vapor on silver iodide and lead iodide. *J. Meteorol.* **12,** 324–331.
79. Birstein, S. J. (1956). The role of adsorption in heterogeneous nucleation. II. The adsorption of water vapor on photolysed silver iodide. *J. Meteorol.* **13,** 395–398.
80. Braham, R. R., Jr., Seely, B. K., and Crozier, W. D. (1952). A technique for tagging and tracing air parcels. *Trans. Am. Geophys. Un.* **33,** 825–833.
81. Crozier, W. D., and Seely, B. K. (1954). Concentration distribution in aerosol plumes 3 to 22 miles from a point source. *Trans. Am. Geophys. Un.* **36,** 46–52.
82. Kassander, A. R., Jr. (1957). A study of the trajectories and diffusion patterns of ground-generated airborne particles under orographic wind flow conditions. *Sci. Rept. No.* **5,** 16 pp. Inst. Atm. Physics, Univ. Arizona, Tucson, Arizona.
83. "Proceedings of the Conference on the Scientific Basis of Weather Modification," April 10–12, 1956. Inst. Atm. Physics, Univ. Arizona, Tucson, Arizona. (Bound mimeo abstracts.)
84. McDonald, J. E. (1956). Scientific basis of weather modification. *Science* **124,** 86–87.
85. Thom, H. (1957). An evaluation of a series of orographic cloud seeding opera-

tions. *Tech. Rept. No.* **2,** 78 pp. Advisory Comm. on Weather Control, Washington, D. C.
86. Bergeron, T. (1949). The problem of artificial control of rainfall on the globe. I, II. *Tellus* **5** (1), 32–33; **5** (3), 14–32.
87. Ludlam, F. (1955). Artificial snowfall from mountain clouds. *Tellus* **7,** 277–290.
88. Kramer, H. P., and Rigby, M. (1950). Selective annotated bibliography on cloud physics and "rain making". *Met. Abstr. Bibl.* **1,** 174–205.
89. Thuronyi, G. (1955). Annotated bibliography on weather modification. *Met. Abstr. Bibl.* **6,** 1433–1513.
90. Woodcock, A. H. (1950). Condensation nuclei and precipitation. *J. Meteorol.* **7,** 161–162.
91. Lodge, J. P. (1955). A study of sea-salt particles over Puerto Rico. *J. Meteorol.* **12,** 493–499.
92. Twomey, S. (1955). The distribution of sea-salt nuclei in air over land. *J. Meteorol.* **12,** 81–86.
93. Reitan, C. H., and Braham, R. R., Jr. (1954). Observations of salt nuclei over the midwestern United States. *J. Meteorol.* **11,** 503–506.
94. Fournier d'Albe, E. M., Lateef, A. M. A., Rasool, S. I., and Zaidi, I. H. (1955). The cloud-seeding trials in the central Punjab, July–September 1954. *Quart. J. Roy. Meteorol. Soc.* **81,** 574–581.
95. Langmuir, I. (1950). Studies of tropical clouds. *Occasional Rept. No.* **25,** 17 pp. Project Cirrus, Contract No. W-36-039-SC-38141. General Electric Res. Lab., Schenectady, New York.
96. Coons, R. D., Jones, E. L., and Gunn, R. (1949). Second partial report on the artificial production of precipitation, cumuliform clouds, Ohio, 1948. *U. S. Weather Bur. Res. Pap. No.* **31,** 58 pp. Washington, D. C.
97. Bowen, E. G. (1952). A new method of stimulating convective clouds to produce rain and hail. *Quart. J. Roy. Meteorol. Soc.* **78,** 37–45.
98. Braham, R. R. Jr., Battan, L. J., and Byers, H. R. (1955). Artificial nucleation of cumulus clouds. *Rept. No.* **24,** 106 pp. Cloud Physics Project, Contract No. AF19(604)618. Dept. Meteorol., Univ. Chicago, Chicago, Illinois.
99. Schaefer, V. J. (1953). Laboratory, field, and flight experiments. *Final Rept.* Part I. Project Cirrus, Contract No. DA-36-039-SC-15345. General Electric Res. Lab., Schenectady, New York.
100. Davies, D. A. (1954). Experiments on artificial stimulation of rain in East Africa. *Nature* **174,** 256–258.
101. aufm Kampe, H. J., Kelly, J. J., and Weickmann, H. K. (1956). Seeding experiments in supercooled stratus-type clouds. Paper presented at the Conference on the Scientific Basis of Weather Modification, Univ. Arizona, Tucson, Arizona, April 10–12, 1956.
101a. aufm Kampe, H. J., Kelly, J. J., and Weickmann, H. K. (1957). Seeding experiments in subcoded stratus clouds. *In* "Cloud and Weather Modification: A Group of Experiments," Meteorol. Monographs, **11,** 86–111.
102. Weickmann, H. (1957). Current understanding of the physical processes associated with cloud nucleation. *Beit. Phys. Atm.* **30,** 97–118.
103. Thom, H. (1957). A method for the evaluation of hail suppression. Tech. Rept. No. 4, Adv. Comm. for Weather Control, Washington, D. C., 42 pp.
104. McDonald, J. E. (1956). Variability of precipitation in an arid region: A survey of characteristics for Arizona. *Tech. Repts. on Meteorol. Climatol. of Arid Regions, No.* **1,** 88 pp. Inst. Atm. Physics, Univ. Arizona, Tucson, Arizona.

105. Thom, H. (1957). A statistical method of evaluating augmentation of precipitation by cloud seeding. *Tech. Rept. No.* **1,** 62 pp. Advisory Comm. on Weather Control, Washington D. C.

106. Jeeves, T. A., LeCam, L., Neyman, J., and Scott, E. L. (1955). On the methodology of evaluating cloud seeding operations. *Weather Modification Operations in California, Bull.* **16,** 271 pp. California State Water Resources Board, Sacramento, California.

107. Neyman, J., McDonald, J. E., Elliott, R. D., Reynolds, R. R., and Scott, E. L. (1957). Unusual opportunity for studies of atmospheric physics. *Bull. Am. Meteorol. Soc.* **38,** 175–177.