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CURRENT PROBLEMS IN RESEARCH

Freezing Nuclei, Meteors, and Rainfall

Do tiny particles from meteor streams influence rainfall over the earth's continents?

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New theories concerning mechanisms that exert a controlling influence upon the earth's weather are bound to be regarded with skepticism, particularly by meteorologists used to watching the orderly, though regrettably rather unpredictable, progression of cyclones and anticyclones, cold and warm fronts, over the earth's surface. Such indeed was the reception accorded the suggestion by E. G. Bowen, chief of the Radiophysics Division of the Australian Commonwealth Scientific and Industrial Research Organization, that there was a close connection between the earth's passage through meteor swarms and the whole terrestrial rainfall pattern some 30 days later.

In the eight years which have elapsed since the meteor hypothesis was first propounded it has excited sufficient interest and controversy that a great deal of experimental and statistical work has been undertaken in various parts of the world, designed to test the predictions of the theory and to extend our knowledge of the various links in the chain by which the mechanism is supposed to operate. Some of this work has already yielded interesting and important new information, while some experiments are still in the planning stages, and still others have revealed further puzzles for later investigation.

Condensation Nuclei and Clouds

To view the meteor hypothesis in proper perspective we need first to see what is at present known about the way in which clouds form and the way in which rain develops within them.

When warm moist air rises it expands and cools, and its relative humidity increases toward 100 percent; at this stage the air is said to be saturated with respect to water vapor. Any further cooling of the air would cause a further increase in humidity above 100 percent, and the air would become supersaturated if it were perfectly clean, but in the atmosphere this does not happen. Instead, water begins to condense on some of the numerous tiny particles of soluble material which are normally suspended in the atmosphere, and a cloud of tiny water droplets begins to form at a relative humidity just greater than 100 percent. In fact, even in very fast growing clouds, the supersaturation rarely exceeds 1 percent.

These soluble particles on which the water droplets form are called condensation nuclei and are produced by evaporation of tiny droplets of sea water, or, even more importantly, by the actions of dew and sunshine on soluble salts in the soil. The concentration of condensation nuclei in the atmosphere is normally quite large, and since their most important source is the soil, they are more numerous in air masses which have spent many days over a large continental land mass than in maritime air masses. Figure 1 shows the concentrations of condensation nuclei which become active at various supersaturations, as measured by Twomey in southeastern Australia.

When the condensation process is studied for different sets of possible conditions it is found that the maximum supersaturation reached is a few tenths of 1 percent, and from Fig. 1 this would lead us to expect that maritime clouds would have about 50 droplets per cubic centimeter, whereas continental clouds would typically have about 300 droplets per cubic centimeter. These values agree very closely with the results of droplet counts in clouds of maritime and continental origin reported by Squires.

Another thing which has been established is that in almost all reasonably large clouds, irrespective of size or origin, the liquid-water content averages about 0.5 gram per cubic meter. There appears to be some sort of mixing-in of dry air from outside the cloud which maintains this value. Taken with the droplet counts given above, this means that maritime clouds contain small numbers of rather large droplets, whereas continental clouds consist of large numbers of very small droplets.

Now, careful theoretical work by Hocking in England has shown that because of the complicated flow of air around small cloud droplets, only droplets with radii greater than about 18 microns are able to collide with each other; smaller drops deflect each other and can suffer only "near misses." Because of this effect, though the droplets of maritime clouds are big enough to form raindrops by repeated collisions and coalescences, most continental clouds are quite stable, in this respect,

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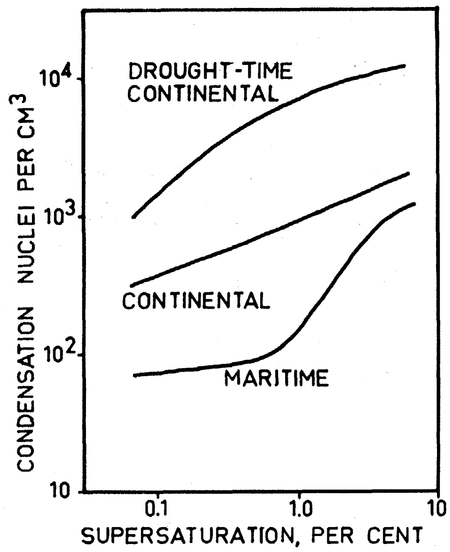

Fig. 1. Activity distributions of condensation nuclei in air masses of continental and maritime origin. [After Twomey]

have few such collisions, and can never yield rain by this mechanism. Since most important continental rainfall, with the exception of that falling on coastal regions, tends to originate in continental air masses, I shall now concentrate on the properties of these apparently stable clouds.

Freezing Nuclei and Ice Crystals

As a cloud rises above the condensation level its top continues to cool, and eventually its temperature falls below the freezing point. It might be expected that the cloud droplets would then freeze to form tiny ice crystals, but this is not the case, and in fact tiny droplets of pure water can be supercooled to temperatures near $-40°C$ before they will freeze spontaneously. The presence of small particles, usually called freezing nuclei (*1*), is required to initiate ice formation in clouds at temperatures higher than this.

These freezing nuclei also occur naturally in the atmosphere, but whereas condensation nuclei are present in hundreds per cubic centimeter, the concentration of natural freezing nuclei active above $-20°C$ is only a few per liter of air. The general form of the activity spectrum of these natural freezing nuclei is shown in Fig. 2.

We may expect, then, that under average conditions, when cloud-summit temperature falls to $-20°C$, the cloud top itself will contain about one ice crystal per liter. Though the concentration of these ice crystals is negligible as compared with that of water droplets, their physical importance is very great. At temperatures below 0°C the water-vapor pressure over ice is less than that over water, and at temperatures in the range $-10°$ to $-20°C$ this effect is so great that water droplets near one of these ice crystals evaporate quite quickly and the vapor is deposited upon the growing ice crystal. This process is illustrated in Fig. 3.

Here, then, is the process which leads to instability and precipitation in continental clouds. The ice crystals grow at the expense of neighboring water droplets until they are large enough to fall through the cloud, collecting further water droplets by collision as they do so. Whether the precipitation falls as rain, hail, or snow depends largely upon the total depth of the cloud and upon the temperature at its base. The concentration of ice crystals required to cause substantial precipitation from an average cloud is estimated to be between 1 and 10 per liter at the cloud top.

I might digress a moment to point out that this is the origin of our present rain-making techniques. Silver iodide smoke is a very efficient ice-nucleating agent at temperatures below about $-10°C$; hence, if this smoke is injected into a cloud it starts the precipitation mechanism operating much earlier than the natural freezing nuclei can, and in this way it should tend to increase the local precipitation.

The onset of glaciation in cloud tops can be quite easily recognized from the appearance of the cloud itself. Figure 4 shows a cumulus cloud that consists entirely of water droplets and has a sharp, cauliflower-like outline. Figure 5 shows the same cloud after the top has become glaciated, in this case after the introduction of silver iodide particles. The ice crystals are blowing away from the top of the cloud, giving it a characteristic anvil shape and soft outline, while heavy rain can be seen falling from the base. Exactly the same sort of behavior occurs with natural freezing nuclei.

This precipitation process has been investigated from many points of view, but here we are concerned with its very first stages—stages leading up to the production of ice crystals in the required concentration in the cloud top. The three major questions to be answered concern the origin of the natural freezing nuclei that are active at temperatures above about $-20°C$, their chemical composition, and the mechanism by which they cause the production of ice crystals. Each of these questions is still a source of discussion and controversy, but considerable progress appears to have been made toward their solution.

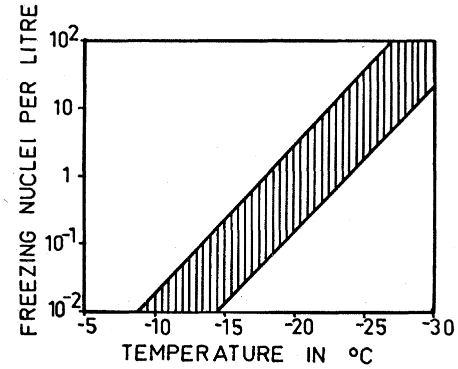

Fig. 2. Average activity distribution for ice-forming nuclei in the atmosphere. The activity curve varies from day to day but typically lies within the shaded region.

The Meteor Hypothesis

The meteor hypothesis, put forward by Bowen in 1953, states that at least a substantial fraction of the more active freezing nuclei in the atmosphere have their origin as micrometeoritic particles collected by the earth in its orbit through space. The evidence which leads to this conclusion is of an indirect and statistical nature, as I shall now show.

Rainfall on the earth's surface is influenced by many factors, some of which are periodic (like the seasons) and many of which, on a shorter time scale, are much more random, though there may be serial correlations due to such things as the movement of pressure systems. It is therefore to be expected that when a sufficient number of data from different places and different years are combined to give a single curve for rainfall as a function of day of the year, a smooth curve will result.

The great difficulty lies in estimating the degree of departure from smoothness which is statistically significant when a given number of records have been combined, and of course this also depends in detail on the exact method used to treat the data, since daily rainfall is far from normally distributed. Nevertheless, when Bowen combined the rainfall records for 300 stations widely distributed in both hemispheres (each station contributed records for a period of about 50 years), the deviations from smoothness were so large that it seemed reasonable to regard them as significant without any detailed statistical analysis.

Bowen's world rainfall curve for the months of November, December, and January is shown in Fig. 6. It can be seen that deviations from the mean are as large as $\pm$ 15 percent and that some peaks, particularly those of 12 and 22 January and of 1 February, are particularly prominent.

Since the publication of this curve, Brier of the U.S. Weather Bureau has carried out an independent analysis of rainfall data from 150 stations distributed over the United States. The choice of the stations and years involved was such that none of the data had been included in Bowen's curve. Using careful statistical techniques Brier showed that there was a high correlation between the dates of rainfall maxima and minima among the two sets of data which he used and the world rainfall curve of Bowen. The probability of the observed level of correlation occurring by chance was estimated to be less than 1 percent.

A less elaborate, but again completely independent, analysis of rainfall records, from stations in the U.S.S.R., by Dmitriev and Chili yielded 30 rainfall peaks over the whole year which were considered to be statistically significant. The agreement between the dates of these peaks and those in the appropriate part of Bowen's curve was again very good.

These studies appear to have established fairly firmly the existence of world-wide, annually recurring singularities in the terrestrial rainfall pattern.

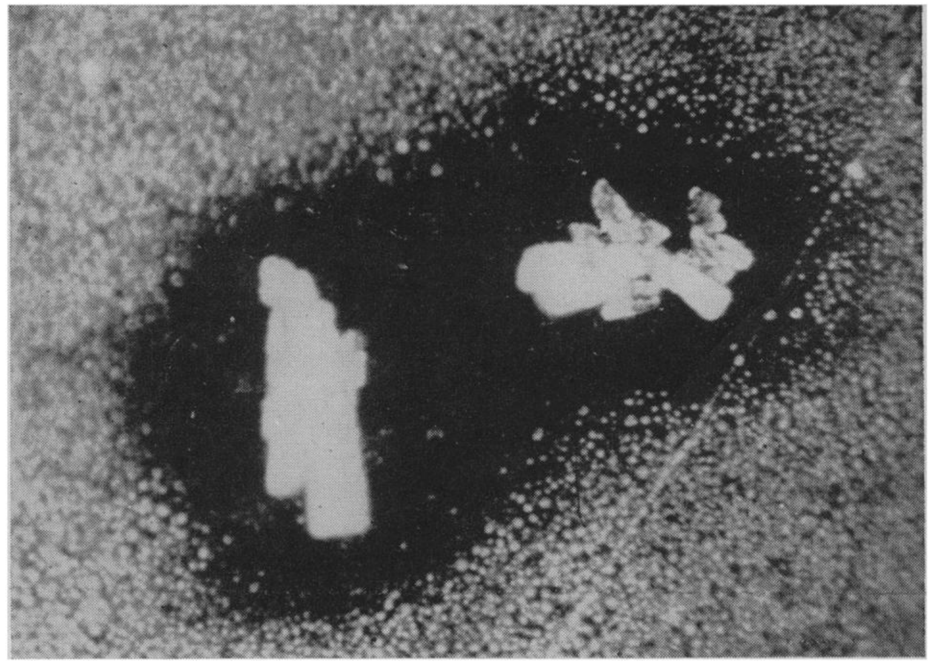

Fig. 3. Two ice crystals growing among a population of water droplets on a microscope slide. The ice crystals shown are about 100 microns in maximum dimension, while the water droplets are typically about 2 microns in diameter.

Fig. 4. A cumulus cloud consisting entirely of water droplets. Note the sharp outline and the uniform base level, which shows how definite is the onset of condensation. [Commonwealth Scientific and Industrial Research Organization]

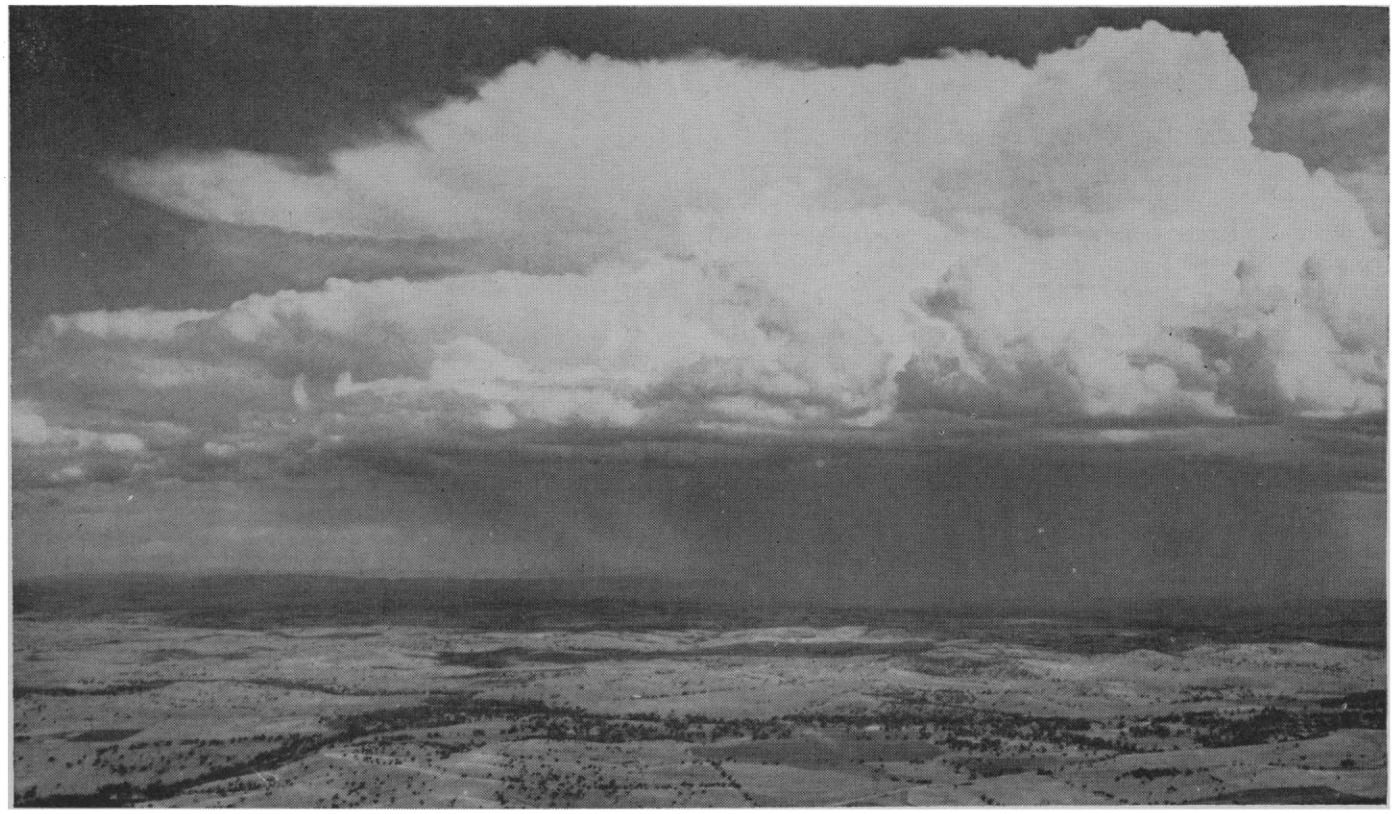

Fig. 5. The cloud shown in Fig. 4 about 30 minutes after seeding with silver iodide smoke. The top of the cloud now contains large ice crystals, which may be seen blowing downwind in a typical "anvil." Heavy rain can be seen falling from the cloud base. [Commonwealth Scientific and Industrial Research Organization]

If then, these rainfall peaks are real, they must be due to some physical phenomenon in the atmosphere, and both the global nature of the singularities and their annual recurrence appear to require some sort of extraterrestrial mechanism. It was for this reason that the meteor hypothesis was put forward.

When a correlation is sought between rainfall peaks and known meteor showers it is natural to expect that the meteor shower will precede the rainfall peak by an appreciable period, during which the meteoric dust settles through the stratosphere and reaches the level of convective mixing below the tropopause. With this in mind, Bowen found a correlation between his rainfall peaks and intense meteor showers occurring about 30 days previously, as shown in Table 1.

More extensive correlations have been found for other parts of the year, both by Bowen and by Dmitriev and Chili, and the correlation has been strengthened by the recognition of periodicities in rainfall peaks associated with periodic meteor showers such as the Perseids in September, the Giacobinids in October, and the Bielids in December. Most recent work has, however, concentrated upon the well-defined peaks and prominent meteor streams indicated with asterisks in Table 1.

A rather striking feature of the meteor hypothesis is the time lapse of 30 ± 2 days between the meteor shower and the rainfall peak with which it is associated. The most obvious conclusion is that this is the time required for the meteor particles, slowed to terminal velocity at a height of about 80 kilometers, to fall under gravity to the level of the tropopause. If this is so, then it can be estimated that the particles involved must have a diameter of about 10 microns, and the sharpness of the rainfall peak implies a corresponding sharpness in the size distribution. I discuss this aspect of the theory in more detail later.

Supporting Evidence

Evidence supporting the meteor hypothesis, or at least consistent with it, has come from a variety of sources. In the first place, if the rainfall peaks are real and due to a sudden increase in the freezing-nucleus content of the atmosphere, then it would be helpful to have a direct measurement of this increase. Now the concentration of freezing nuclei in the air can be measured directly by a fairly simple technique. An air sample of known volume is placed in a container, humidified, and cooled down in such a way that a supercooled cloud forms. If any freezing nuclei are present that are active at the temperature of the cloud, they form ice crystals, which grow and fall to the bottom of the container. If the bottom of the container is coated with a supercooled sugar solution, then, on falling into this solution, the tiny ice crystals grow rapidly to visible size and can easily be counted.

Freezing-nucleus counters of this type have, over the last few years, been operated daily during the month of January at a number of stations in Australia, New Zealand, South Africa, Sweden, England, France, Germany, and the United States. January was chosen for this study because the distinct peaks in the rainfall curve for this month led one to hope that the freezing-nucleus count might show a similar simple pattern.

The results of several years' operations have established that the freezing-nucleus count in most places does fluctuate greatly from day to day, and that concentration peaks of one or two days' duration frequently occur. The records often vary greatly from station to station; however, when a composite curve is drawn for all stations there appear to be fairly well defined peaks occurring within one or two days of the peaks in the rainfall curve.

More recently Bigg, at the Radiophysics Laboratory, has developed a counter which continuously monitors the freezing-nucleus content of the air around it. This device has established that freezing-nucleus peaks may be of very short duration, rising to a maximum in one or two hours and then decaying more slowly toward the background level. If purely local nucleus sources are ruled out by investigation, then it would appear that such sharp peaks occur after the breakdown of a temperature inversion separating nucleus-poor air in the lower layers from nucleus-rich air above. The occurrence of these rapid fluctuations requires that some sort of integrating technique be used if a representative daily nucleus count is to be taken.

A second study which also has given a measure of support to the general picture is a census of the occurrence of cirrus cloud over wide areas. Cirrus cloud consists of ice crystals formed high in the atmosphere, and a dense cirrus cover may reasonably be associated with either a high concentration of freezing nuclei or a high concentration of water vapor, or both, at that altitude. Bigg, who also carried out this work, examined available records of the occurrence of cirrus cloud over Australia and found maxima in the cirrus cover on or near peak days in the rainfall curve. In a similar study in the United States, however, Braham failed to find any significant correlation between the two curves.

A very important piece of supporting evidence has recently been obtained. It has been known for some time that, within the altitude limits conveniently available to ordinary aircraft, the freezing-nucleus concentration shows no over-all tendency to decrease with height. This finding is rather unexpected if it is supposed that the nuclei originate at the earth's surface, but it is completely in accord with the meteor hypothesis. Now, at an altitude of about 10 kilometers there is a more or less permanent inversion, the tropopause, which should act as a substantial barrier to material of terrestrial origin. In 1959, however, Telford of the Radiophysics Laboratory succeeded in making several measurements of freezing-nucleus concentration above the tropopause and established that the concentration at those altitudes was quite comparable to that below the tropopause.

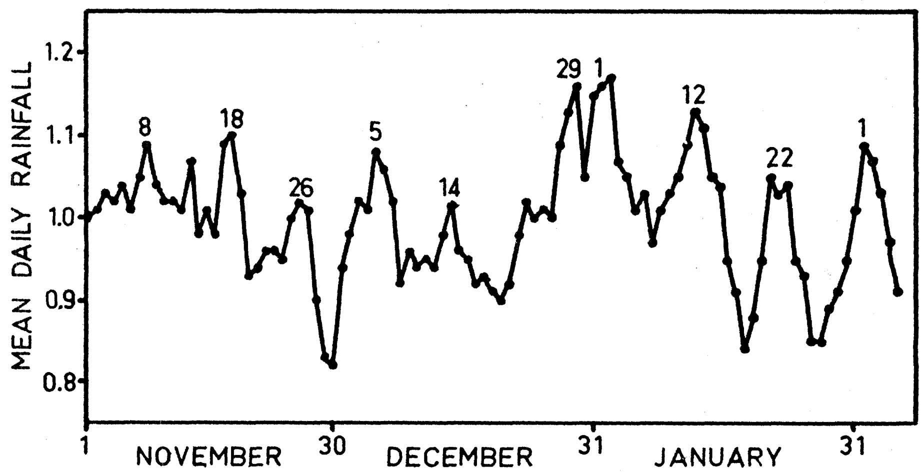

Fig. 6. Bowen's world rainfall curve.

Towards the end of 1960, using a new sampling technique and with the aid of U.S. Air Force U2 aircraft, Bigg was able to sample freezing nuclei to a height of 20 kilometers. These measurements confirmed those of earlier trials with balloon-borne equipment and again showed high nucleus counts above the tropopause, giving substantial support to the meteror hypothesis.

A related experiment at present being carried out promises to yield additional information about conditions in the high stratosphere. If small meteor particles enter the earth's atmosphere they either fall gently at the appropriate terminal velocity or else burn up during their deceleration. The dividing line between these two types of behavior occurs at a particle radius of about 10 microns. The smaller particles are slowed to a low velocity at a height of about 80 kilometers, and at this height their concentration becomes appreciable. The particle concentration at these heights can be studied by examining the light scattered from this layer in the twilight zone, when it is illuminated by the sun and when lower layers of the atmosphere are in shadow. This technique frequently shows pronounced concentrations of particles at this height and may provide valuable information about the number of particles captured from meteor showers, though of course it cannot be inferred that these particles are active as freezing nuclei.

Table 1. Correlation between rainfall peaks and meteor showers.

Rainfall peak (date)	Meteor shower		Time lapse (days)
	Meteor stream	Date	
8 Nov.	Giacobinids	9 Oct.	30
18 Nov.	Orionids	20–23 Oct.	29
26 Nov.	?		
5 Dec.	Taurids	3–10 Nov.	32
14 Dec.	Leonids	16 Nov.	28
29 Dec.	Bielids I	27 Nov.–15 Dec.	—
1 Jan.*	Bielids II	2 Dec.	30
12 Jan.*	Geminids	13 Dec.	30
22 Jan.*	Ursids	22 Dec.	31
1 Feb.*	Quadrantids	3 Jan.	29

* Well-defined peaks.

Criticism of the Theory

Many aspects of the meteor hypothesis and the evidence brought forward to support it have been strongly criticized, and agreement has by no means yet been reached.

Much of the initial criticism questioned the reality of the rainfall peaks from both a physical and a statistical point of view. At this stage the criticism was perhaps justified, since relatively few data had been combined to give the rainfall curve. The curve of Fig. 6, however, representing as it does some 15,000 station-years of measurements, seems adequately representative of the world rainfall pattern.

The associated statistical question relates to the significance of the "peaks" in Fig. 6 and is much more difficult to answer in view of the weighting and smoothing procedures used to obtain the rainfall curve, and of the non-normal distribution of rainfall records. The later work of Brier has, however, given very convincing support to the view that the peaks are real and significant. It would be of great value if similar independent analyses could be made of the almost countless alternate sets of rainfall records kept by the weather bureaus of the world. When such analyses are available, expert statisticians will be able to determine the significance of the correlations between them.

Perhaps the more interesting areas of disagreement concern physical aspects of the theory. For the meteor hypothesis to be regarded as vindicated, the following points must be established: (i) that meteor particles can act as efficient freezing nuclei at cloud-top level; (ii) that sufficient meteoric material is collected by the earth to account for a significant proportion of the observed freezing-nucleus population active above −20°C; (iii) that there is no terrestrial source of such freezing nuclei which greatly outweighs the contribution of the meteor particles; and (iv) that the arrival of the meteor particles at the earth's surface (or at the tropopause) as a sharp pulse after a delay of 30 ± 2 days is a necessary consequence of some reasonable mechanism.

None of these points has yet been conclusively proved or disproved, and it is in their investigation that most interest lies at the present time. Let us consider them in turn.

The theory of the action of solid particles as ice-forming nuclei is as yet in the early stages of its development. Enough is known, however, to suggest that an efficient ice-forming nucleus should be of the order of 0.1 micron in diameter, or larger, and that it should consist of some more or less polar compound whose atoms are so arranged that crystal faces, when exposed, can match fairly closely the molecular arrangement on some low-index face of ice. This is illustrated for the case of two of the most efficient nucleating substances known—silver iodide and lead iodide—in Fig. 7.

Now, meteoric particles may be either metallic or stony in nature. If metallic, they consist predominantly of an iron nickel alloy which seems most unlikely, on theoretical grounds and on the basis of laboratory tests, to have any appreciable activity as an ice nucleus. This conclusion, however, neglects an important possibility, and that is that small metallic particles may undergo chemical reactions as they fall through the atmosphere and may become coated with some ferrous salt which is itself a good nucleating agent. The situation is somewhat similar for stony meteoric particles. Powders or vapors made from stony meteorites have little activity in the laboratory, but it is not known to what extent such meteorites are typical in composition of the micrometeorites in which we are interested, or to what extent abrasion or vaporization duplicate the influences to which the micrometeorites are subjected. The question of the activity of meteoric particles thus still remains open.

Estimates of the amount of meteoritic material accreted by the earth are necessarily indirect and differ from each other by as much as six orders of magnitude, so to evaluate the sufficiency of meteors as a source of nuclei is almost impossible. Evidence from examining snowflakes and from using a particle-size discrimination counter on Bigg's continuous nucleus counter suggests that most active freezing nuclei in the atmosphere are of the order of 1 micron in diameter. If this is true, then most of the estimated meteor accretion rates are somewhat too low to account for the observed nucleus concentration, but the higher estimates of some workers and the general uncertainty leave this question open.

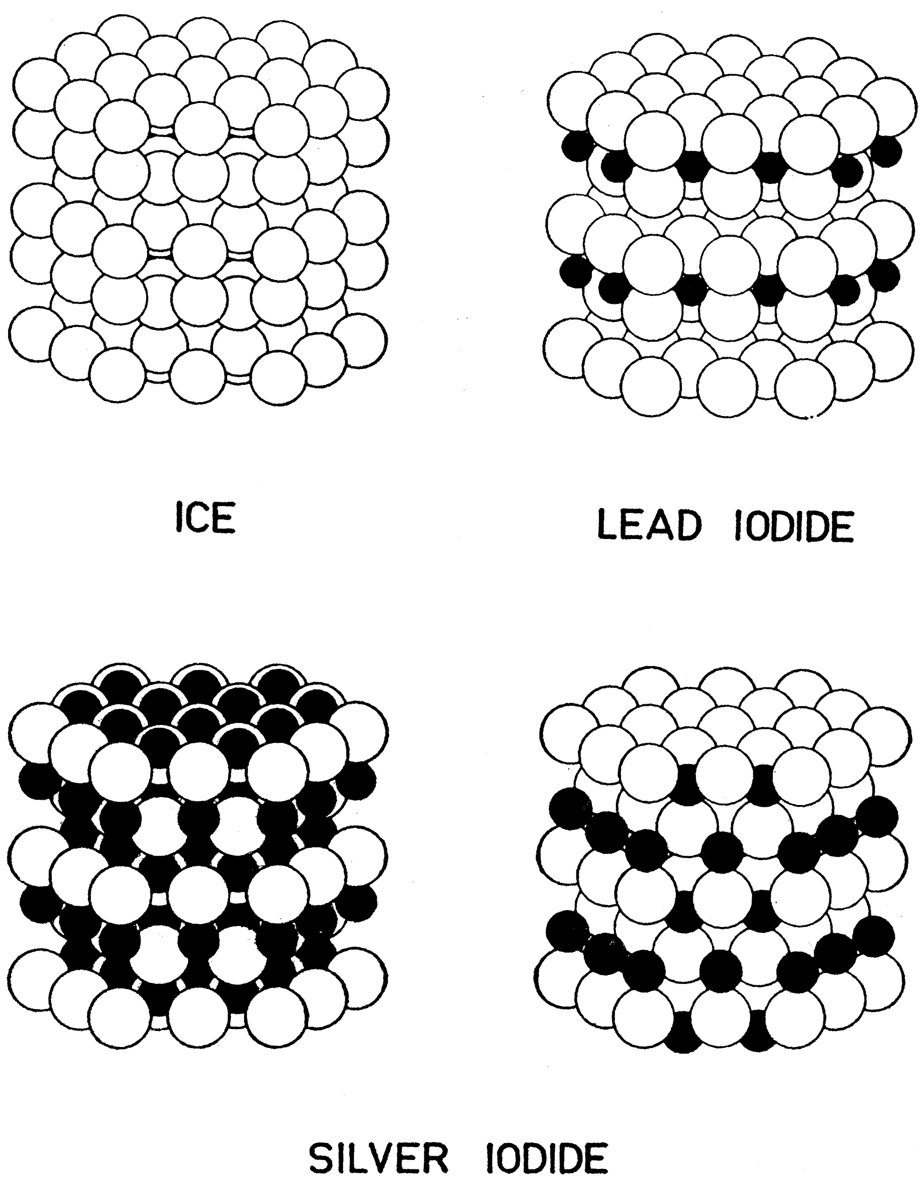

Fig. 7. Molecular models, all to the same scale, of small crystals of ice, lead iodide, and silver iodide. In ice, only the oxygen atoms are shown; in the other two crystals the white spheres represent iodide ions; the black spheres represent the metallic ions.

A related question concerns the relative accretion rates of shower and sporadic meteorites. If the fluctuations in daily freezing-nucleus counts are taken to be caused by fluctuations in the meteoritic accretion rate, then one would expect that the accretion rate during showers would be as much as 100 times the sporadic accretion rate. The weight of evidence at present available suggests that this is not so, and that despite the considerable increase in visible meteors during showers, the total accretion rate changes relatively little. Present meteor-counting techniques, however, are limited to particles rather larger than those we have been considering, and it is possible that meteor streams may contain rather large numbers of these submicron particles.

When the possibility of a terrestrial source of freezing nuclei is examined, data are much more easily found. Mason and his co-workers in England have examined the ice-nucleating properties of many naturally occurring minerals and have found at least one, kaolinite, which is apparently sufficiently common and active to account for all observed nuclei, and at the same time not so abundant as to cause a saturation of the air with its particles.

A criticism which may be made of these experiments is that we cannot be sure that the freshly ground mineral samples tested are typical in activity of the particles of this same mineral which might find their way into the atmosphere through processes of erosion. In fact it has been shown that the nucleation activity of many mica-like materials decays very rapidly on exposure to humid atmosphere conditions, and this may well occur with other minerals too.

The suggested activity of kaolinite is, however, supported by Japanese work in which the nuclei at the centers of snow crystals were examined by electron diffraction. The particles were, in most cases, several microns in diameter, and the diffraction pattern suggested a clay mineral of the general type of kaolinite.

Industrial processes, particularly those associated with ore smelting, which have been found to liberate vast quantities of active nuclei, are another possible source of freezing nuclei on the earth's surface.

Against this must be set, however, the fact that active nuclei are found in apparently undiminished concentration over the Southern Ocean, a thousand miles from any possible continental source, and high above the tropopause, where particles originating at the earth's surface would not be expected to penetrate. Our knowledge of the fate of tiny particles injected into the atmosphere is, however, not sufficiently exact at present for any firm conclusion to be drawn.

Finally, we consider the requirement that the meteoric nuclei arrive with a well-defined front at the tropopause after a delay of 30 ± 2 days. The only mechanism which has been suggested for this effect is the free fall of the particles at their terminal velocities through the stratosphere. Rough calculations of this process suggest that a 30-day fall time should be associated with particles about 10 microns in diameter. This accords well with the maximum size for a meteor particle that remains unmelted in its fall, and also with the general size of particles found in snowflakes and cirrus crystals. These particles do not, however, show any sharp size preference, and indeed the majority of nuclei measured at the earth's surface are less than 1 micron in diameter.

Again, it is hard to see why the "peak" should remain sharp after 30 days, since this would require an almost unbelievable uniformity of particle size, and no such uniformity is evident at the earth's surface. It seems that if the meteor hypothesis is valid there must be some as yet undiscovered mechanism which either limits the size range of efficient nuclei or else transports all nuclei through the stratosphere by some mechanism other than simple falling. Until such a mechanism is suggested, this will remain one of the most serious obstacles to any general acceptance of the meteor hypothesis.

Conclusion

During the past eight years the meteor hypothesis has provided a stimulus for much interesting and valuable work on the origin and properties of natural freezing nuclei in the atmosphere. In many ways the results of this work appear to support the hypothesis, but several steps of rather crucial importance to the argument are still unexplained, and other groups of experiments point towards a terrestrial origin for the nuclei.

It is still too early to decide which view is correct. If, however, as appears to be the case, the rainfall singularities are real, then the meteor hypothesis is the only one which has been advanced as yet which seems capable of explaining them.

Note

1. To call these particles "freezing nuclei" implies that they act by causing the freezing of a water droplet. Another possibility is that ice may form directly upon the foreign particles by sublimation from the vapor, in which case they should be termed "sublimation nuclei." Particles are known which act in both these ways, and what happens depends in detail upon temperature and humidity. For this reason the inclusive term *ice-forming nuclei* is perhaps to be preferred, but tradition has established the use of *freezing nuclei,* and I follow it here.

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