ICE NUCLEI IN THE ULTRAMICROSCOPE AND THE ATMOSPHERE

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Abstract. Various attempts have been made in recent years to overcome the difficulties inherent in the study of clouds by reproducing them on a small scale in the laboratory. The present article briefly reviews some of these experiments, particularly those relating to the formation of ice crystals, and discusses the extent to which the laboratory results accord with what is known of natural clouds. Recent experiments on the artificial stimulation of rainfall are also examined for the light they throw on the part played by ice nuclei in the formation of rain.

Résumé. Pendant ces dernières années on a essayé à plusieurs reprises de tourner les difficultés liées à l'étude des nuages naturels, en reproduisant ceux-ci à petite échelle au laboratoire. On passe en revue ci-dessous certaines de ces expériences, surtout celles qui traitent de la formation de cristaux de glace. On examine comment les résultats de ces expériences s'accordent avec ce que l'on sait des nuages naturels. De récents essais pour provoquer la pluie artificielle sont également étudiés dans la mesure où ils jettent quelque lumière sur le rôle que jouent les noyaux de sublimation ou de congélation dans la formation de la pluie.

An instrument has recently been constructed at Oxford, at the suggestion of Professor G. M. B. Dobson, in which the process of cloud formation on natural or artificial nuclei can be observed and studied ultramicroscopically. It consists, in effect, of a brass expansion chamber of about 1½ litres capacity, in the centre of which a volume of about 0·2 cm.^{3} is strongly illuminated in the field of view of a low-powered microscope by the light of a 250 watt high-pressure mercury arc lamp. Special precautions are taken to render the background particularly dark. The optics of the instrument are sufficiently good to enable small particles of a radius down to about 0·05 micron to be seen. Small but rapid expansions, whose volume ratio may be accurately controlled up to 1.10 by means of metal bellows, permit observation of the behaviour of the particles during small changes of temperature and relative humidity. For work at low temperatures provision is made for immersing the whole expansion chamber in a cooling bath of petrol and repeating the observations at any temperature down to −60° C. In this case special attention is paid to the nature of the condensation in order to determine whether it is in the form of supercooled droplets or of ice crystals. It is easy to distinguish between them, owing to the characteristic scintillation of the crystals. A detailed description of the apparatus and of the results obtained with it will be found in the *Quarterly Journal of the Royal Meteorological Society*,[1] but a brief account of the conditions under which ice crystals were observed may be of interest here.

The outdoor air at Oxford was examined on numerous occasions. Clouds formed in the chamber by expansions both above and below 0° C. were seen to consist entirely of water droplets as long as the temperature did not fall below a threshold which usually lay between −32° and −35° C. Below this threshold clouds were still formed mainly of water droplets but they also contained small numbers of ice crystals, perhaps 20 per cm.^{3} as compared with about 1000 water droplets per cm.^{3}. The proportion of ice crystals remained steady on further cooling until expansions brought temperatures below a second much sharper threshold, which appeared always to lie between −41° and −42° C. and which seemed to be the same as that discovered in clean air by Cwilong.[2] Below this second threshold clouds always consisted mainly if not entirely of ice crystals.

The most striking characteristic of all ice formation in outdoor air was that it occurred only at saturation with respect to water. Saturation with respect to ice was not sufficient. It seems therefore that direct sublimation from vapour to solid cannot be considered as a mode of ice formation in the atmosphere, unless on very rare nuclei. The normal process would appear to be condensation of water on to a nucleus, followed by freezing of the growing droplet.

Further support for this view was obtained from some experiments on artificial nuclei. A large

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variety of substances, including various sea salts, provided nuclei which, like the great majority of those in the atmosphere, gave water droplets at all temperatures above −41° C.; below that temperature they acted as freezing nuclei, giving ice crystals but only at saturation with respect to water. Silver iodide, first discovered to be active by Vonnegut,[3] promoted ice formation at temperatures as high as −6° C., but again only as a freezing nucleus. Cadmium iodide, on the other hand, provided what is as yet the only known example of true sublimation nuclei. After an ice cloud had been formed on them by the usual process of cooling to below −41° C., and had evaporated, it left behind nuclei which grew ice crystals as soon as saturation with respect to ice was passed, a phenomenon not observed with any other nuclei tested. This activity continued up to −10° C., when the nuclei apparently "melted," and reverted to their original rôle as water nuclei.

The ice-growing properties of the last two salts may be linked with the fact that they both exist as hexagonal crystals whose lattice constants are very close to those of ice. This does not explain, however, the difference in their behaviour, which may be connected with the solubility of cadmium iodide.

The ultramicroscope technique described above provides us with perhaps the only means of watching the behaviour of individual condensation nuclei without great interference in their natural surroundings. But for the study of outdoor air it suffers from the major drawback that the volume of air under observation is necessarily so small that the nuclei present in concentrations of less than 1 per cm.^{3} normally escape detection, whereas a concentration of 1 per litre is sufficient for nuclei to play an important part in atmospheric processes.

This difficulty was met by Findeisen, who, working in Germany during the war, constructed an expansion chamber of 2 cubic metres capacity, traversed by a beam of light from a 24 candle power lamp. Air from outside was admitted at a controlled humidity, and expansions were made at rates corresponding to the known velocities of vertical ascent in convection clouds. Although he was unable to distinguish individual cloud droplets, he could clearly identify ice crystals by their scintillation in the beam. In the outdoor air of central Europe he found crystals first appearing in his clouds between −8° and −15° C. according to the rate of expansion, but only in very small numbers, less than 1 per litre. The crystals remained rare until temperature fell below a second threshold between −30° and −35° C., when they increased very suddenly to at least 1 per cm.^{3}. The nuclei on which these crystals grew Findeisen termed sublimation nuclei Type 1 and Type 2 respectively, but from the details of his results there seems no doubt that they were in fact freezing nuclei.

Summing up the laboratory results, we may therefore say that of the large number of nuclei normally present in the atmosphere there do not appear to be any which permit true sublimation. There is a very small minority of nuclei, over land surfaces at least, which produce ice crystals in air saturated with respect to water below about −10° C.; a larger minority which become similarly active at about −32° C., while the great majority continue to condense water at all temperatures down to −40° C. It appears that activity in ice formation is linked with the chemical and crystallographic nature of the nucleus, and that certain artificial nuclei can be produced which grow ice at higher temperatures than any natural nuclei. By the use of these latter we may hope in some cases to influence the evolution of clouds and of precipitation.

Let us now turn to the atmosphere itself and examine the evidence as to whether in free air condensation follows the same course as in the laboratory. What may we expect to find?

In convection clouds, such as cumulus and cumulonimbus, we may expect ice crystals only when the top of the cloud has reached a temperature below −10° C., and then only in small numbers depending on the degree and quality of the pollution of the air. These crystals, though rare, will be large, having grown at the expense of the surrounding water droplets. They will fall rapidly and will therefore soon be found at all heights in the cloud above the freezing level. When the cloud develops further and reaches a temperature below −30° C. we may expect to find crystals in much greater numbers normally, sufficient to convert the upper region of the cloud entirely into ice, giving the well-known "anvil top." This will depend, however, on there being sufficient "Type 2" nuclei present, which may not always be the case in clean maritime air. In the absence of these nuclei the cloud will have to reach −40° C. before developing the anvil.

The observations which have been made in cumulonimbus are for obvious reasons very scanty, and in any case the measurement of temperatures in cloud presents difficulties which have not yet been entirely overcome. But the results which have been obtained do in general fit into the picture given above. Thus Dr. Frith,[4] of the Royal Aircraft Establishment, has found purely water clouds at temperatures down to −20° F. (−28·9° C.) and sometimes even lower. But it is more usual to find ice crystals appearing in small numbers at about −15° C. As for the development of the anvil top, the most striking sign of the presence of ice crystals, the author is unaware of any systematic measurements of the temperature at which it occurs. The general impression seems to be, however, that it appears at rather higher temperatures than one would expect from the laboratory results.

With regard to the second class of ice clouds, the cirrus type, one can do no better than refer to a recent paper by Ludlam.[5] He shows that an analysis of the forms of these clouds leads independently to the conclusion that ice crystals are formed only at the water saturation point or by the freezing of supercooled water droplets. The temperature at which cirrus appears shows wide variations. Frith has found it at all temperatures below 0° F. (−17·8° C.), but without any sign of a sudden jump in frequency at either −30° or −40° C.

From the absence of natural sublimation nuclei one might expect to find the atmosphere often supersaturated with respect to ice. There is very little evidence in support of this, but since even at −40° C. the difference between frost- and dew-point is only about 4° C., such slight supersaturation might well be difficult to detect.

The reason for the recent great interest in the occurrence of ice in the atmosphere arises of course from the important part which it is generally supposed to play in precipitation. The hypothesis that "every rain drop originates as an ice crystal," first advanced by Bergeron [6] in 1933, is based theoretically on the inability of the processes of selective condensation and of coalescence by collision to account for the growth of rain drops (radius of the order of millimetres) from cloud droplets (radius of the order of microns) at a rate corresponding to the observed intensities of rainfall; and empirically on the fact that, in temperate latitudes at least, moderate or heavy rain practically never falls except from clouds extending well above the freezing level. The "catalytic" action of ice crystals is to be attributed to the fact that the equilibrium vapour pressure over ice is always lower than that over water at the same temperature. Any crystals which form will therefore grow rapidly at the expense of the surrounding water drops, will fall through the cloud still growing by condensation and coalescence and eventually reach the ground as rain.

This theory has been the subject of some criticism. Many tropical meteorologists are convinced that heavy rain can and does fall from clouds that do not even reach the freezing level. Others have urged that the unexpected resistance to freezing of supercooled droplets militates against the acceptance of the idea that clouds must reach −30° C. before developing abundant rain. But whether or not we regard the presence of an ice phase as essential, there seems no reason to doubt that it will favour precipitation. We may therefore expect that if cloud droplets could be induced to freeze more readily rain might be stimulated in clouds from which otherwise it would never fall. The laboratory results indicate two possible ways of achieving this: either by cooling parts of the cloud to below −30° or −40° C. by dropping into it granules of solid carbon dioxide ("dry ice"); or by introducing into the cloud artificial nuclei which grow ice at higher temperatures than the natural nuclei. Both these methods have been tried.

The results have been by no means consistent or clear. Dry ice has been used on many occasions in Australia, South Africa, France and the United States. There is no doubt that on some occasions the dropping of dry ice has stimulated almost immediate rainfall and considerable growth of the cloud. But in other cases the cloud has merely dissolved into wisps of snow. It appears that for dry ice to have an immediate effect certain fairly stringent conditions of temperature and stability must be fulfilled, but it may be some time before the exact nature of these conditions is known. What does seem to be established beyond doubt is the ability of dry ice to promote the freezing of droplets in supercooled clouds. Perhaps the clearest example of this was the dissipation of supercooled stratus, described by Langmuir,[7] which points the way to the second important application of these techniques.

Silver iodide is the only substance so far to be tried in the form of artificial nuclei. While it has been claimed that a supercooled fog has been precipitated by silver iodide smoke, no reports are as yet to hand of its use on convection clouds. It should perhaps be noted in this connexion that a cloud formed purely of ice crystals is no more likely to give precipitation than one purely of water droplets. The most favourable conditions are only met by a mixture of the two phases, allowing selective condensation on to the ice. Success with artificial nuclei may depend upon the introduction into clouds of the right proportion of ice nuclei satisfying fairly narrow conditions of size and hygroscopicity.

But perhaps the most revealing experiment on artificial rainfall was that carried out in Hawaii in September 1947,[8] when dry ice stimulated heavy and prolonged rainfall from clouds which at the time of inoculation were entirely *above* 0° C., with their tops at least 5000 feet (1524 m.) below the freezing level. It is difficult in this case to attribute the rain to the action of ice crystals, since any crystals formed by the dry ice must have melted almost immediately. The most probable explanation has been put forward by Langmuir,[9] in the form of a new theory of rain formation. For the details one must refer to the original paper, but a brief summary may perhaps be attempted here.

According to the theory the necessary condition for rain to develop in a purely water cloud is the existence in the cloud of some droplets of radius greater than a certain critical value. This critical radius depends mainly on two factors, the liquid water content of the cloud and the velocity of the updraught. Once droplets of the critical size are formed, they will fall relative to the rest of the cloud and thus begin to grow by collision with other droplets. Eventually they will become unstable and break up, giving two or more fragments each greater than the critical size. This process repeats itself indefinitely, provided that the break-up of successive generations of droplets takes place each time at a higher level. The resultant large drops fall out as rain as soon as the updraught weakens or they escape from it. The action of the dry ice is attributed to the melting and breaking-off of the ice coating which forms round the granules as they fall through the moist air, each granule thus leaving behind it as it falls a trail of droplets each large enough to initiate the "chain reaction."

Natural rainfall from purely water clouds, as opposed to that produced by human interference, is to be explained in the same manner. Here an important part may be played by initial differences in droplet size, due to the difference in size and the hygroscopic properties of the nuclei. Collisions due to micro-turbulence will eventually produce droplets big enough to set off the chain reaction, provided that the conditions of temperature and stability are favourable. The chain reaction may also proceed in supercooled water clouds in the absence of ice, provided that the collisions do not cause the freezing of the droplets. In certain circumstances, with the base of the cloud above 0° C. and the top reaching −10° C., it may perhaps be initiated by the rare Findeisen nuclei. The snowflakes formed round these nuclei will melt when they fall below the freezing level and may give rise to droplets large enough to start the chain reaction if it has not already commenced. In clouds which lie wholly above the freezing level the chain reaction will not be started in this way, since the snowflakes will fall out of the cloud before melting. In this case the Bergeron process, aided perhaps by the multiplication of crystals through splitting in the upper parts of the cloud, will probably be responsible for any precipitation which falls.

Acceptance of Langmuir's ideas would thus entail a modification of the importance hitherto attached to the part which ice nuclei may play in precipitation, particularly in tropical and warm temperate zones. But they still possess a sufficient field of action to warrant a continued investigation of their character and activity.

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REFERENCES

1. Albe, E. M. Fournier d'. Some experiments on the condensation of water vapour at temperatures below 0° C. *Quarterly Journal Royal Meteorological Society*, Vol. 75, No. 323, 1949, p. 1–14.
2. Cwilong, B. M. Sublimation in a Wilson chamber. *Proceedings of the Royal Society*, Series A, Vol. 190, No. 1020, 1947, p. 137–43.
3. Vonnegut, B. Nucleation of ice formation by silver iodide. *Journal of Applied Physics* (Lancaster, Pa.), Vol. 18, 1947, p. 593–95.
4. Frith, R. Private communication.
5. Ludlam, F. H. The forms of ice-clouds. *Quarterly Journal Royal Meteorological Society*, Vol. 74, No. 319, 1948, p. 39–56.
6. Bergeron, T. On the physics of cloud and precipitation. *Union internationale géodésique et géophysique, Association de météorologie, Procés-verbaux* (Lisbon), 1933, II, 1935, p. 156–70.
7. Langmuir, I. *Final Report, Project Cirrus*, p. 121, General Electric Company, Schenectady, 1948.
8. Leopold, L. B. and Halstead, M. H. First trials of the Schaefer-Langmuir dry-ice cloud technique in Hawaii. *Bulletin of the American Meteorological Society*, Boston, Mass., Vol. 29, 1948, p. 525–34.
9. Langmuir, I. Production of rain by a chain reaction in cumulus clouds at temperatures above freezing. *Journal of Meteorology*, Vol. 5, 1948, p. 175.

AN ESKER IN PROCESS OF FORMATION: BÖVERBREEN, JOTUNHEIMEN, 1947

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Eskers have been variously defined but it is now generally agreed that they consist of winding ridges of cross-bedded or current-bedded sands, gravels and sometimes boulders. The best formed are flat topped and steep sided, and they may exhibit contortion, faulting and other disturbances which can well be attributed to the presence of ice during their formation. The nature of the bedding clearly indicates that the materials were laid down in water and their association with glacial deposits suggests that they were formed by melt water streams emerging from glaciers or ice sheets. They may extend any distance from a fraction of a mile to 150 miles if the gaps which usually occur in the longer eskers are included. They cross the country, continuing uphill and downhill over the minor features of the landscape. This latter characteristic favours Hummel's [1] explanation which regards them to be the casts of sub-glacial streams. This raised the question as to why sediments should be deposited in such places where one would have expected the streams to have flowed swiftly under hydrostatic pressure and so to have been fully capable of clearing out their channels. Also the frequently claimed perfection in the form of the cross section hardly suggested that they had been formed under moving ice. Furthermore the Moray Firth and other eskers subdivide and rejoin just like the distributary pattern of a glacial stream well laden with glacial outwash. This led me to follow Wright [2] in favouring Shaler's suggestion [3] that eskers originate when outwash streams end in standing water, as did the great eskers of Sweden in the waters of an expanded Baltic in Yoldia and earlier times. Where no such semi-permanent bodies existed the eskers were supposed to have been deposited in lakes formed by the ice blocking the normal drainage channels. It is easy to understand why an esker formed in such a manner would remain in an excellent state of preservation after the lake waters had disappeared.