Ice Nucleating Properties of Meteoritic Material

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ABSTRACT

A description is given of an attempt to duplicate the small particles formed by evaporation and recondensation during the flight of meteors in the atmosphere by heating meteors at a low pressure.

A metallic meteorite produced entirely shiny spherules, almost all of which were in the size range 5 to 25 μ diameter, while a stony meteorite produced only irregular aggregates of tiny particles whose maximum dimensions were 0.1 to 0.2 μ. At water saturation and −10C, it is estimated that the iron meteorite creates about 10^5–10^6 ice nuclei per gram and the stony meteorite about 10^8–10^9. It is concluded that sufficient ice nuclei active at −15C are produced to explain observed concentrations in the troposphere.

1. Introduction

The origin of ice nuclei, the particles on which ice will form in a cloud of supercooled water drops, is a topic which is still debated. Specific local sources, such as steelworks, volcanoes and dust from desert areas, have been discovered, but it seems that the only important ice nuclei produced in such areas are those close to the source. The occurrence of ice nucleus "storms" at places without obvious specific sources also renders some of these identifications doubtful. Bowen's (1956) proposal that nuclei are meteoric in origin aroused a controversy of long standing. While it is now clear that the 10 μ diameter particles which he suggested formed the nuclei are usually too few to account for the commonly observed 100 ice nuclei per cubic meter active at −15C, it is quite possible that the smaller particles of extraterrestrial origin could supply these.

This seems a likely hypothesis when we consider the remarkable similarity between mean concentrations measured in places with very different concentrations of particles of terrestrial origin, such as:

1) Antarctica and Australia (Bigg and Hopwood, 1963);
2) Hawaii above and below the trade-wind inversion (Droessler and Heffernan 1965; Bigg, 1964);
3) above and below the tropopause (Bigg and Miles, 1963).

One difficulty in accepting Bowen's proposal has been the experiments by Schaefer (1957) which yielded no nuclei, even at −25C, from an iron meteorite. It is most unusual for any solid insoluble material when finely divided to give no ice nuclei active at this temperature.

Mason and Maybank (1958) and Mason (1960) have found both stony and iron meteorites to be relatively inactive when compared with the clay particles of terrestrial origin which are often found in the atmosphere. A more recent paper by Qureshi and Maybank (1966) confirms these results.

A little reflection shows that these experiments may be irrelevant. Meteors entering the atmosphere become hot enough to boil at the prevailing low pressures, and material will recondense from the supersaturated vapors in the meteor wakes. The surface properties of the recondensed material will depend critically on the mode of its formation, and surface properties are crucial in determining nucleating ability. It is therefore not a valid test to crush a meteorite and to test the nucleating properties of the powder, as Mason and Maybank or Qureshi and Maybank have done; and it is doubtful whether sparking a Tesla coil on it, as Schaefer attempted, or vaporizing it by heating it in a hydrogen flame, as Mason described, will produce realistic surface properties.

The experiments reported here represent an attempt to create particles which resemble in their surface properties those formed by meteors which boil during their fall through the atmosphere, and an attempt to assess the nucleating properties of these particles under properly controlled humidity.

2. Production of the particles

The meteorite to be tested was mounted on a zirconia rod at the focus of the solar furnace of the University of New South Wales, inside a 10-liter glass flask held by continuous pumping to an air pressure less than 2 mm of mercury. The equivalent altitudes were greater than

about 75 km, the lower limit of burn-out of the larger meteors. The arrangement is shown in Fig. 1a, and one of the collecting slides can be seen placed below the specimen and some distance to one side.

Before the experiment began the zirconia rod was cleaned by melting its end with the full heat of the furnace. The meteorite was then cleaned of surface contamination by heating (Fig. 1b). When the flask was evacuated the meteorite was heated until it was observed to boil vigorously for a length of time sufficient to catch a suitable amount of material on the slides.

Both types of meteorite boiled very readily at the low pressures with the application of only a fraction of the available heat.

3. Examination of the particles

The exposed collectors were examined with an optical microscope and with an electron microscope after detaching the small grids stuck to each slide. To detect the ice nuclei, a slide was placed in a miniature cold chamber with thermoelectric cooling. The walls of the chamber were lined with ice-impregnated blotting paper, which normally kept the slide at a humidity between ice and water saturation. A sudden cooling of the thermoelement allowed water saturation to be attained because of the time delay between cooling the base and the walls of the chamber. The ice crystals which formed grew slowly (Fig. 2) and after half an hour were easy to count.

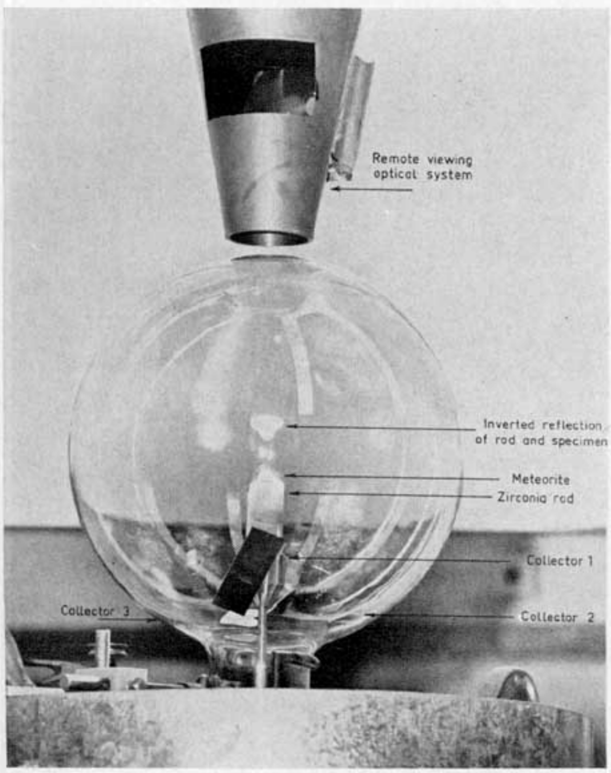

FIG. 1a. Arrangement for boiling meteorites at low pressure in a solar furnace.

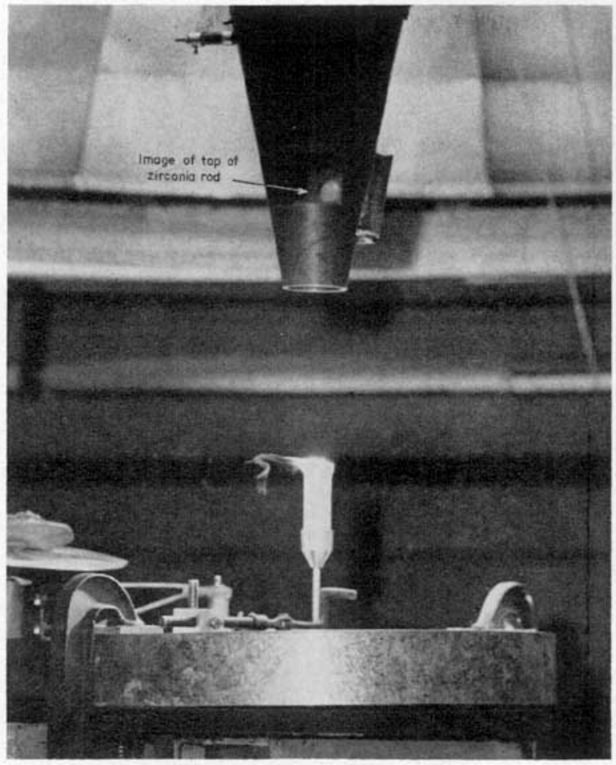

FIG. 1b. Removing the surface contamination from a meteorite specimen before heating it in a vacuum.

When crystals were numerous, as in this photograph, those nearest the vapor source (at the edges) reduced the humidity so that fewer formed in the center of the slide. This leads to underestimates of actual ice nucleus concentrations at the lower temperatures.

4. Results

a) *Particle size and appearance.* The meteorites used were a typical octahedrite, obtained from the Australian Museum and known as the "Mungindi No. 2," and a stony meteorite, with numerous chondrules and no visible metallic content, obtained from Dr. T. G. Vallance of the Geology Department of the University of Sydney.

Particles reaching the slide from the boiling iron meteorite were found to be entirely composed of shiny black completely spherical particles with diameters ranging from 5 to 25 μ. Their appearance was typical of those which have been found in the Antarctic, for example, those illustrated by Schmidt (1963).

On the other hand, all particles produced by the stony meteorite were too small to be seen with the optical microscope, and for counting and photographing an electron microscope had to be used. Fig. 3 shows the particles obtained on an electron microscope grid by boiling the meteorite for some minutes. It is curious that the particles, which were all much the same size, 0.1 to 0.2 μ in their largest dimension, had the appear-

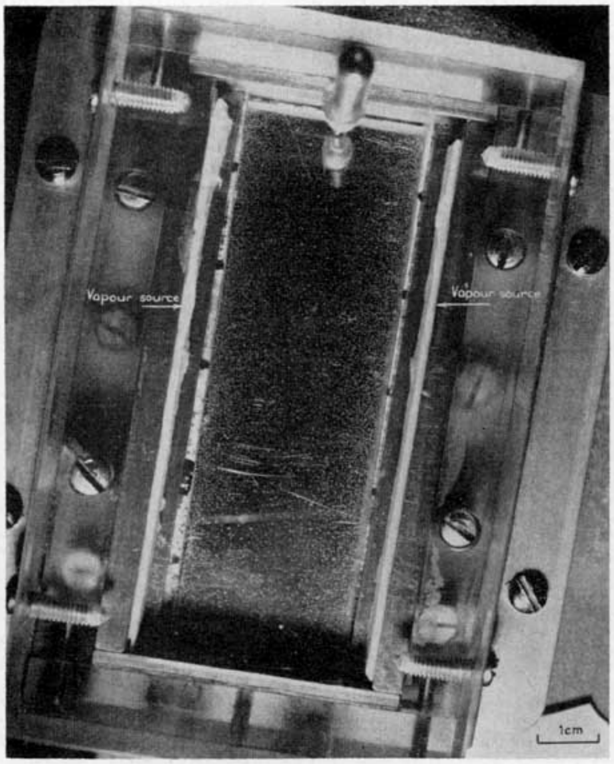

FIG. 2. Ice crystals growing at water saturation on a metal slide in a miniature cold chamber.

ance of being aggregates of large numbers of smaller particles. These are very like particles collected either by rockets from above 80 km, illustrated by Hemenway and Soberman (1962) and called by them "fluffy micrometeorites," or from U-2 flights in the stratosphere, as shown by Mossop (1965) in his Figs. 5 and 6.

The particles produced by low-pressure boiling when collected by sedimentation, therefore, appear to be consistent with types which have been identified as natural micrometeorites.

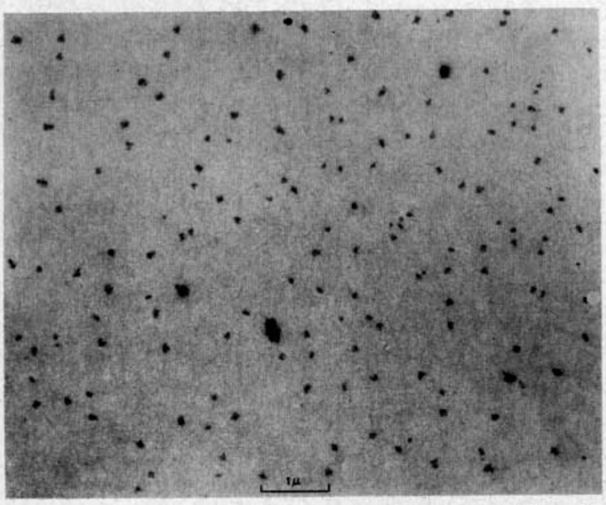

FIG. 3. Electron microscope photograph of particles collected after boiling a stony meteorite at low pressure.

b. Ice-nucleating properties. Considered as a ratio of active ice nuclei to total particles, the metallic spherules were the better nuclei, ranging from 3×10^{-4} at -7C through 10^{-3} at -10C to 5×10^{-3} at -14C. The stony meteorite ratios at the same temperature were about 10^{-5}, 5×10^{-5} and 5×10^{-4}. A test of the densely-covered slide corresponding to the grid sample shown in Fig. 3 yielded three ice crystals at -3.7C from about 10^9 particles. There are very few aerosols of a similar dispersion produced by more usual methods which yield a comparable proportion of ice crystals at this temperature.

Considered in terms of nuclei per gram of meteorite, the stony meteorites obviously win because of their small size and low effective density. Probably 10^{14} particles are formed per gram, so that more than 10^8 nuclei per gram active at -10C and water saturation are produced. The output for the iron meteorite is about three orders of magnitude less.

Thus, in contrast to the earlier experiments which have been mentioned, it can be claimed that evaporation of meteors in the high atmosphere may lead to appreciable concentrations of very active ice nuclei.

5. Discussion and conclusion

The discrepancy between these results, which show meteoritic recondensation products to be very active ice nuclei, and the earlier ones which showed meteorites dispersed in other ways to be completely inactive or only slightly active, is entirely reasonable. Theory and experiment have established the importance of surface structure and purity in all nucleation phenomena, and enhanced nucleating ability would be expected with the method of production that we have used. Since it more nearly resembles meteor disintegration in the atmosphere, we suggest that the results obtained are relevant to the problem of the origin of ice nuclei, while the earlier ones are not.

One problem which should be mentioned is that some meteoric material may have quite a different composition to the meteorites which reach the ground and may produce either better or worse nuclei. (If Bowen's theory is right, the shower meteors must produce better nuclei than the sporadics, in order to produce annual rainfall anomalies.) Since quite a high proportion of the mass intake is from sporadic meteors, of which our specimens are probably typical, this is not likely to alter our conclusions appreciably.

Another more serious problem is the destruction of ice nuclei during their journey to the ground. Junge *et al.* (1961) found, in the region from 15–25 km, a relatively high concentration of large hygroscopic particles, and subsequent investigations have confirmed that the layer is worldwide and consists mainly of ammonium sulfate and persulfate.

The fate of many small particles traversing this region has been clearly demonstrated by Mossop (1965), who

shows in his Figs. 4, 5, and 6, small particles, very similar to those of our Fig. 3, which have become coated with ammonium sulfate. Now if such a particle is subjected to increasing humidity, the soluble material forms a concentrated solution around the insoluble inclusions, preventing ice-formation. Mossop's (1964) discovery that volcanic dust in the stratosphere only a few weeks after its injection had a soluble coating suggests that the process is a rapid one.

Thus, the only extraterrestrial particles to reach the troposphere as ice nuclei are either those which traverse the sulfate layer relatively quickly, or those in air subsiding from higher levels which does not become completely mixed with its surroundings. For an extraterrestrial origin of ice nuclei to be important, it is therefore necessary that the input flux should be appreciably higher than is necessary to give the observed concentrations.

Newkirk and Eddy (1964) have discussed measurements of particle concentration above the sulfate layer, and a figure of one per cubic centimeter seems reasonable for the size of particle with which we are concerned. This is also consistent with Mossop's measured concentrations of insoluble inclusions within sulfate particles, although collection of dust by Hemenway and Soberman (1962) from much greater altitudes would suggest considerably higher concentrations. At -15C we have estimated about one stony meteorite particle in 1000 to be an ice nucleus, so that we are left with 1000 m^{-3}. If this air is transferred unchanged to the ground, without loss of nuclei, the concentration will increase to about 40,000 m^{-3}. This upper limit may be compared with a typically observed ice nucleus concentration of about 100 m^{-3} at the same temperature. If one particle in 400 survives as an ice nucleus during its residence in the sulfate layer and transfer to the ground, then meteor dust is likely to be the most important source of the more active ice nuclei. Only direct measurements of concentrations as a function of altitude at many places and times will show whether this is a reasonable figure.

If it is assumed instead that the stratosphere is a source of nuclei because of storage of particles of terrestrial origin injected by violent convection or air interchange, the same loss processes must occur. The absence of seasonal changes of ice nucleus concentrations, in the Southern Hemisphere at least, which could be related to convectional activity, and the absence of appreciable differences with latitude argue against this supposition.

The conclusion that most of the active ice nuclei are of extraterrestrial origin seems not only possible but probable. The most exciting possibility is that relatively large concentrations of ice nuclei above the sulfate layer would allow us to trace air movements with the relatively simple techniques required for detecting ice nuclei, while concentrations within the sulfate layer could yield information on destruction rates and residence times.

The most useful experiment that could be performed to test the results described here would be to collect dust from above 80 km by a recoverable rocket, as Hemenway and Soberman (1962) have done, and to test its nucleating properties.

REFERENCES

Bigg, E. K., 1964: Geographical differences in concentrations of ice nuclei. *Mon. Wea. Rev.*, **92**, 355–356.

——, and S. C. Hopwood, 1963: Ice nuclei in the Antarctic. *J. Atmos. Sci.*, **20**, 185–188.

——, and G. T. Miles, 1963: Stratospheric ice nucleus measurements from balloons. *Tellus*, **15**, 162–166.

Bowen, E. G., 1956: The relation between rainfall and meteor showers. *J. Meteor.*, **13**, 142–151.

Droessler, E. G., and K. J. Heffernan, 1965: Ice nucleus measurements in Hawaii. *J. Appl. Meteor.*, **4**, 442–445.

Hemenway, C. L., and R. K. Soberman, 1962: Studies of micrometeorites obtained from a recoverable sounding rocket. *Astron. J.*, **67**, 256–266.

Junge, C. E., C. W. Chagnon and J. E. Manson, 1961: Stratospheric aerosols. *J. Meteor.*, **18**, 81–108.

Mason, B. J., 1960: Ice-nucleating properties of clay minerals and stony meteorites. *Quart. J. R. Meteor. Soc.*, **86**, 552–556.

——, and J. Maybank, 1958: Ice-nucleating properties of some mineral dusts. *Quart. J. R. Meteor. Soc.*, **84**, 235–241.

Mossop, S. C., 1964: Volcanic dust collected at an altitude of 20 km. *Nature*, **203**, 824–827.

——, 1965: Stratospheric particles at 20 km altitude. *Geochim. Cosmochim. Acta*, **29**, 201–207.

Newkirk, G., and J. A. Eddy, 1964: Light scattering by particles in the upper atmosphere. *J. Atmos. Sci.*, **21**, 35–60.

Qureshi, M. M., and J. Maybank, 1966: Further tests on the ice nucleation potential of meteoritic material. *Nature*, **211**, 508–509.

Schaefer, V. J., 1957: The question of meteoritic dust in the atmosphere. *Artificial Stimulation of Rain*, London, Pergamon Press, 18–23.

Schmidt, R. A., 1963: Microscopic extra-terrestrial particles from the Antarctic peninsula. Res. Rep. Ser. 63-3, Univ. of Wisconsin Geophys. and Polar Research Center, Madison.