A new Technique for Counting Ice-Forming Nuclei in Aerosols

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(Manuscript received September 9, 1956)

Abstract

An objective method for measuring freezing nucleus concentration with improved accuracy is described and the results of a series of measurements made on the coastline in an on-shore wind near Carnarvon, Western Australia are given. Fluctuations in ice crystal counts existed which were well beyond the probable experimental error and appeared to be independent of weather conditions, except for the occurrence of cirrus cloud. A remote terrestrial source or an extraterrestrial source of nuclei is indicated.

1. Introduction

The use of silver iodide to stimulate rainfall has increased interest in the measurement of naturally-occurring freezing nuclei. The widely varying temperatures at which clouds in the free atmosphere have been observed to contain the ice phase suggest that the nucleus concentration is far from homogeneous. Yet few measurements have been made on a continuous basis over a long period in order to observe these variations in concentration. Some recent exceptions are the experiments described by SMITH, KASSANDER and TWOMEY (1956) in which daily observations were made for about one month. In common with many earlier experiments, the counting technique was not entirely objective so that the real extent of the fluctuations was uncertain.

The present paper describes first a simple and objective method of measuring ice crystal concentrations in an artificially formed supercooled cloud, and then presents the results of a 3 months' series of observations using this method.

SMITH and KASSANDER avoided possible effects of local pollution by making their measurements from an aircraft, while TWOMEY conducted his experiments on the top of 10,000-ft mountain in the Hawaiian Islands. The present measurements were made on the coast at Carnarvon in Western Australia where reliable on-shore winds were known to exist. This site had the further advantages for these experiments of uniform weather and little cloud or rainfall.

2. Measurements of freezing nucleus concentrations

Most freezing nucleus measurements so far reported have been based on visual estimates of the concentration of ice crystals in a strong beam of light. The method relies on the ability of the operator to estimate concentrations consistently, to reproduce conditions such as the density of the cloud and to decide correctly what is an ice crystal and what is not. The latter is made difficult if there are particles in the atmosphere which scintillate, but are not ice, and there may be ice crystals, irregular or partly formed which do not scintillate. CWILONG's (1947) apparatus overcame these difficulties at the expense of reducing the information obtained. In the small expansion chamber which he used was a pool of supercooled water, and its freezing signified that one or more ice crystals had fallen into it.

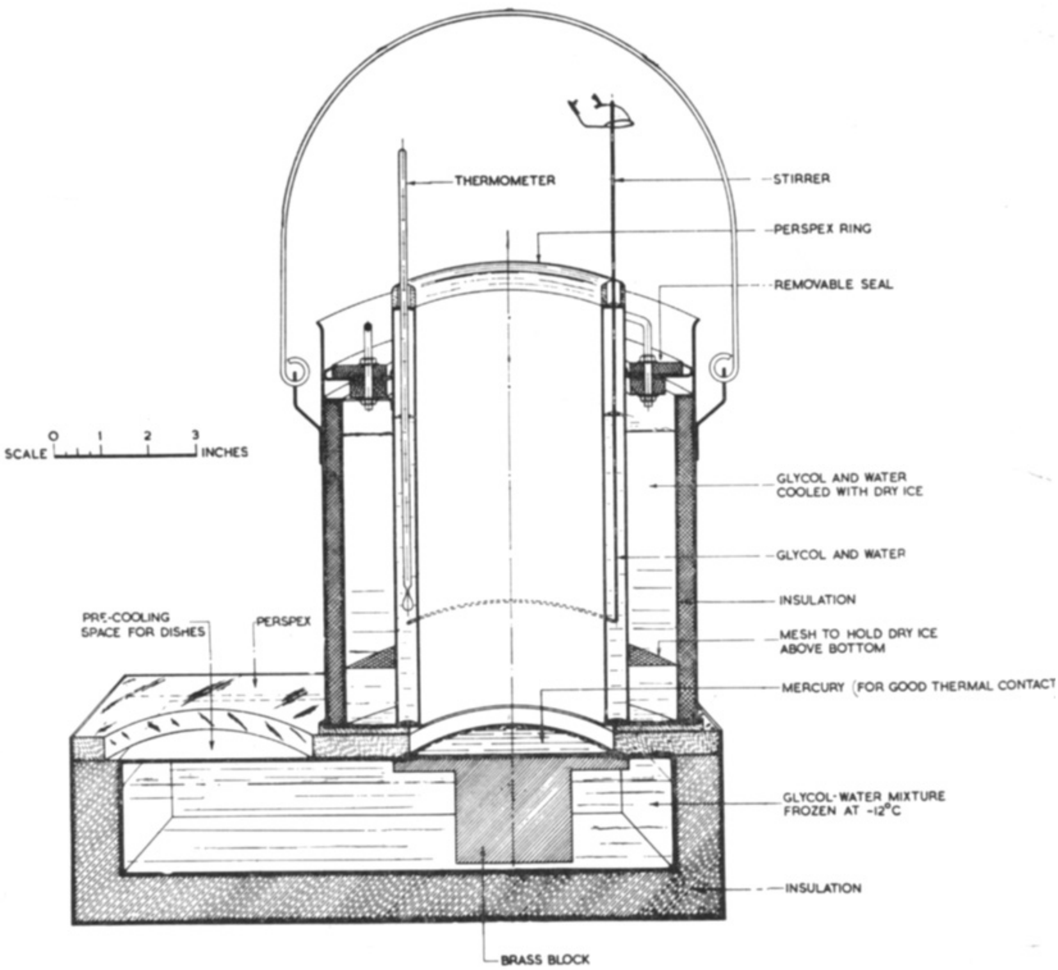

Fig. 1. Cloud chamber and cooling tank.

The information was therefore limited to a threshold temperature for the appearance of ice crystals. The small sampling volume and very short time at minimum temperature were further disadvantages.

The procedure adopted in the present case was a modification of CWILONG's method in which the supercooled water on the bottom of the cold chamber was replaced by a strong aqueous solution. Ice crystals falling into it from the cloud did not freeze the whole of the solution, but grew as individual crystals at a rate controllable over a wide range by varying the concentration of the solution, or the degree of supercooling. In practice one of the simplest and least critical solutions to use is cane sugar in water, as it is readily supercooled to low temperatures. As a typical example, 1.2 kg of sugar added to one litre of water will allow crystals to grow to about 2 mm diameter in 2 minutes when held at $-12°$ C, the melting range being -10 to $-7°$ C. At temperatures warmer than $-15°$ C risk of nucleation in the sugar itself by foreign substances is negligible.

A sketch of the cold chamber and the device for supercooling the sugar solutions is given in Fig. 1. The cold chamber consisted of a hollow cylinder (in which the cloud formed) surrounded by two concentric cylinders partly filled with ethylene glycol-water mixtures. The outer of these was cooled with dry ice, the inner being designed to prevent cold spots. The temperature in the inner jacket, measured after adequate stirring, is the quantity referred to throughout as the "temperature of the cloud". In fact, the average temperature of the cloud is higher, by about 5° C at −20° C but its minimum temperature closely approximated the recorded value.

The cold chamber itself stood on a rectangular thermally insulating base plate in which were two holes to accommodate petrie dishes (not shown in the figure). The petrie dishes contained sugar solution and made good thermal contact with the heavy metal tank below the base plate containing a melting mixture of glycol and water at −12° C.

In making a measurement of freezing nucleus concentration the sequence of events was as follows: the cold chamber was lifted from an insulated box in which it was kept when not in use and placed on the base plate above a petrie dish. The act of lifting the chamber replaced the air within it, and on being lowered to the base plate, a cloud formed together with a small number of ice crystals, which fell into the supercooled solution. After a fixed interval—in this case 2 minutes—the crystals were counted. Although the cooled volume of air was only 1 litre, larger volumes could be sampled by repeating the experiments many times at the one temperature.

Some precautions were essential. Perhaps the most important was the removal of frost from all surfaces in contact with the cloud. This was ensured by careful wiping with glycerol after each measurement. Another possible source of trouble was spontaneous nucleation in the sugar solution. This could occur if the cold sugar solutions absorbed water from the air, or if the temperature fell too low. It was avoided by frequent replacement of the sugar solution on humid days and by careful regulation of the temperature.

As a typical example of the counts obtained, Fig. 2 shows the measurements made at Carnarvon, Western Australia on 19 January 1956. In the diagram, the points marked by crosses are arithmetic averages over the temperature ranges −20.0° to −21.2° C and −21.7° to −22.5° C. It then becomes possible

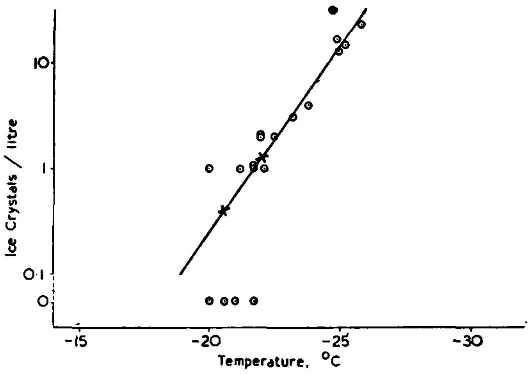

Fig. 2. A typical measurement of freezing nucleus concentration.

to draw a straight line providing a satisfactory representation of the points, the 0.1/l level being reached by a straightforward extrapolation. Occasionally, however, a straight line plot was not acceptable and the number of experiments then had to be increased to provide a reasonable estimate of the temperature for 0.1/l. The number of points necessary for a good determination varied from 15 to 50 depending on the slope of the line (which varied from day to day) and the scatter of the points.

3. Sources of error

The true numbers of nuclei present could differ from the number counted for three main reasons.

(1) Ice crystals may not have grown large enough to fall out of the cloud. It was found that few fell out after 90 seconds although wisps of fog were then still visible and it is considered that the time allowed of 2 minutes was sufficient.

(2) The high probability of obtaining a count other than the mean when counting low numbers, if the nuclei are distributed at random in space. This was diminished by taking many observations and on occasions when the curve was a straight line, representing slightly more than 80 % of the total, the error was probably less than ± 2°, ● 1°, ± 1° C for 0.1/l, 1/l and 10/l respectively. On the remaining occasions, including 4 out fo the 12 measurements for which more than 0.1/l was estimated at —10° C, the error may have been twice as great. Three 0.1/l estimates

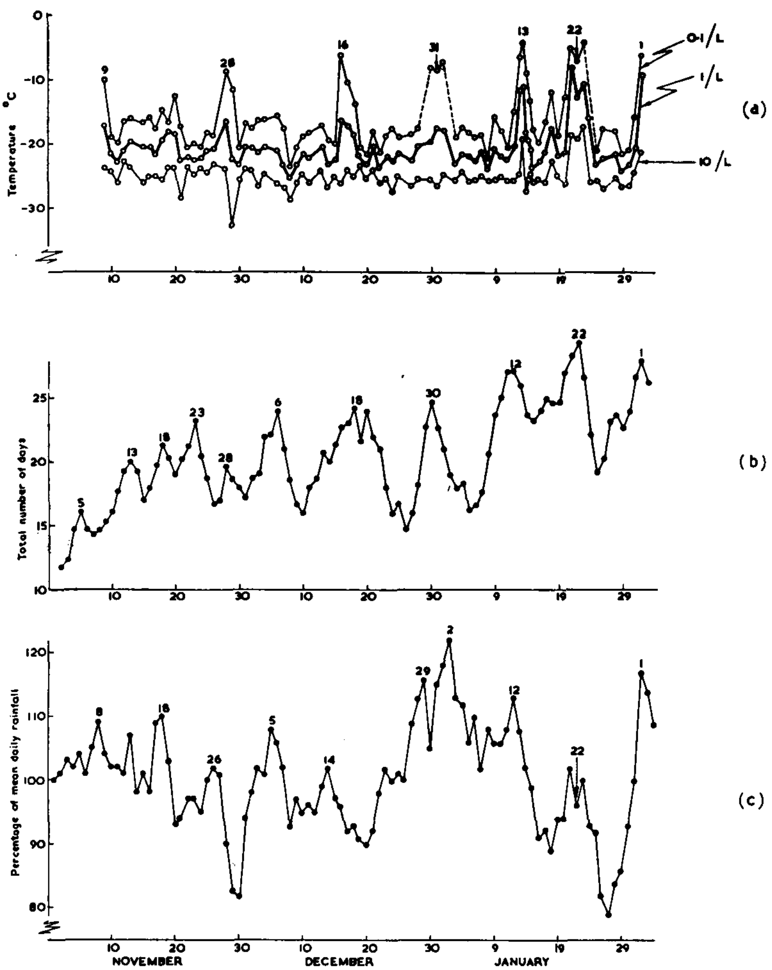

Fig. 3. (a) Temperatures of given freezing nucleus concentrations, Carnarvon 1955—56. (b) The frequency of occurrence of cirrus clouds over Western Australia, 6 stations for about 10 years. (c) Percentage of mean daily "world rainfall", after BOWEN.

were omitted from the graph because of high scatter, thought to be due to a patchiness in the distribution of the nuclei in the atmosphere. These were 27 November, 14 January (3rd reading) and 24 January.

(3) The possibility of spurious ice crystals from frost or nucleation in the detecting solution. Because standard solutions were used at fixed temperatures and a rigid procedure of glycerining the walls of the cold chamber was followed, it is not thought that errors from this source were serious.

Temperature was measured with an alcohol-in-glass thermometer accurate to ± 0.5° C over the range used. With its long time constant, and the possibility of incomplete equilibrium in the liquid, errors in measurement of the minimum cloud temperature may have been ± 1° C. As pointed out earlier, the mean temperature of the cloud was higher.

4. Results

Fig. 3a shows a graph of the temperature for three concentrations of ice crystals from 9 November 1955 to 1 February 1956 inclusive. Days missed, through lack of dry ice, were 14 November, 5, 12, 26, 29 December, 2 and 27 January. Two measurements were made

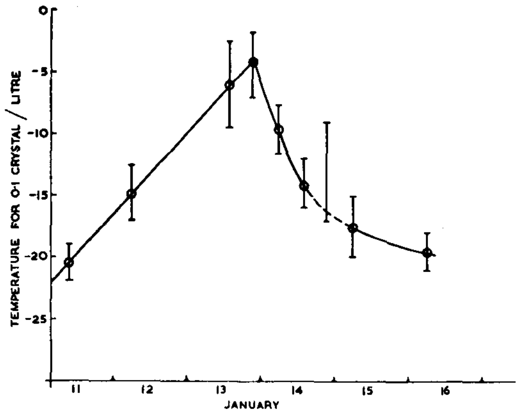

Fig. 4. Temperatures for 0.1 crystal/litre and estimated errors 11—16 January.

on 13 January and three on 14 January, but otherwise only one daily. It can be seen that there were small fluctuations from day to day interspersed with a few major changes, labelled in the diagram. The changes, which were most conspicuous at the 0.1/l level, were always seen in the 1/l count but only in January at concentrations of 10/l.

The major fluctuations were considerably greater than the estimated probable errors and represented systematic changes over several days. This is well illustrated by the measurements of 11—16 January when extra experiments were performed which are shown in Fig. 4. Here the limits of error shown were estimated only from the scatter of the points, thus ignoring systematic errors. The concentrations of nuclei active at −10° C present on 13 and 21—23 January and 1 February were as high as those commonly supposed to produce continuous rainfall, while on most other days this concentration was attained only below −20° C. Since their activity is comparable with that of silver iodide their influence on weather situations favourable to rain may have been considerable.

The average temperatures for concentrations of 0.1, 1 and 10/l were −16° C, −20.5° C and —25° C respectively. The temperature for 1/l was thus about 8° C higher than those found in Sydney and Arizona in January 1954 and 1955 by Smith and Kassander. This may be partially due to their measurements being made from aircraft and partially to the different equipment. The equipment described here, when operated in Sydney on the ground in 26 observations during September 1955, yielded a mean temperature of −18.7° C for 1 crystal/l.

5. Source of the nuclei

It has been reported by several observers that combustion residues, soil, or even microorganisms may act as freezing nuclei (see, for example, Soulage 1955) so that it is important to determine whether the observed fluctuations in ice crystal counts were due to purely local sources of nuclei. At the site chosen, air passed over varying amounts of land according to the wind direction, with a minimum of less than 1 km (consisting of a tidal swamp) for directions between SW—the prevailing wind—and NW.

Graphs of mean wind speed and direction during the period of observation were prepared and compared with the corresponding measurements of temperature for 0.1 crystal/l. There was clearly no consistent relationship between the graphs and the inference is that the nuclei were not of local origin. The lack of correlation with wind speed does not favour the possibility of an oceanic source.

Combustion nuclei could have been only rarely present in the air sampled, the country being virtually uninhabited, except for the township to the east of the site. Notice that the mean temperature for 0.1/l in a smoky industrial suburb of Sydney in September was only 1.8° C higher than for Carnarvon in the period November to February. Evidently only a minute proportion of combustion nuclei has the property of being freezing nuclei, and their effect may be neglected at most sites.

No correlation with any other meteorological phenomenon was found during this period, with the exception of the occurrence of cirrus cloud. This type of cloud appeared, on the average, only on one day in three, yet it occurred on 11 out of the 15 days on which the temperature for 0.1 crystal/l was unusually high. Now, cirrus cloud will only form if a region simultaneously contains sufficient water vapour and nuclei capable of growing ice crystals at its temperature. If the relation observed is real and not fortuitous, it means that nuclei at the ground appear at about the same time as at cirrus levels—probably higher than 6 km in this latitude. If the terrestrial source of nuclei is so remote that considerable vertical mixing has had time to occur, it is strange that large fluctuations should be observed on some occasions. An alternative explanation of the frequent simultaneity is that the nuclei are transported rapidly to low levels in ice crystals falling from the cirrus. The question of how they reached the cirrus in the first place then arises.

BOWEN (1953) has suggested that active freezing nuclei may be formed by dust accompanying (or resulting from) meteor showers and has summarized measurements (BOWEN 1956) which appear to support this contention. Cirrus may be regarded as an indication of the presence of active nuclei, for the more active the nuclei present, the more frequently will the conditions for formation of the cloud be satisfied.

Thus, according to BOWEN's theory, the frequency of occurrence of cirriform cloud should depend on date, and should be related to measured freezing nucleus concentrations. Of course, in any one year, the presence or absence of water vapour will also determine whether or not the cirrus forms and may influence the time of arrival of the nuclei at the ground. Consequently a similarity, rather than a complete correspondence between cirrus and freezing nucleus concentrations would be expected.

Records of the frequency of cirrus cloud occurrence over Western Australia have been compiled from the available records (about ten years') of six widely separated stations. These are shown in Fig. 3b as three-day means in order to compare them with BOWEN's (1956) published curve of rainfall, based on about 50 years' records of 300 places distributed round the world. This is shown in Fig. 3c.

Quantitative comparison of Figs. 3a, 3b, 3c through correlation coefficients may be rendered inexact by the fact that departures from the mean are not randomly distributed and each series has some serial correlation. McDONALD (1957) has summarized arguments from the literature on statistics which show that errors arising from such causes are more likely to be of academic than practical interest when dealing with geophysical data. If the correlation coefficient, assuming normally distributed variables and no serial correlation were significant at the 99 % level, it would be immaterial that the rigorously derived significance figure should be 98 %. The point of making a quantitative comparison at all is to test whether one's opinion is due to bias, and as such is a useful procedure. In the author's opinion, Figs. 3a, 3b and 3a, 3c show a degree of association which would not be expected in unrelated series. The opinion is confirmed by correlation coefficients of 0.38 and 0.22 respectively, significant at about the 99.9 % and 95 % levels. Moreover, the Southern Hemisphere countries' contribution only to Fig. 3c (i.e. Argentine, Australia, New Zealand and South Africa) produces a curve which has the much higher correlation coefficient of 0.42 with Fig. 3a, significant at the 99.99 % level.

Thus, even allowing errors in these estimates of significance, there is undoubtedly far more agreement between the curves than is likely to arise from chance.

The implication is that the nuclei, the cirrus and the rainfall have some common control, and this is consistent with BOWEN's theory of a meteoric influence on rainfall.

A terrestrial source of nuclei is not ruled out. If the quoted results are typical the source must be distant, in order to explain the observed relation between phenomena at high and low levels and the lack of correlation with weather. The dependence of variations in the supply of nuclei on calendar date implied by the relation between curves including 1, 10 and 50 years' data is difficult to explain in terms of a terrestrial source. Note, however, that as there is not a one-to-one correspondence, the pattern is probably only partially repeated from year to year.

It will require many more such series of measurements at different sites before the source can be fixed with certainty. Desirable information which could be obtained by the method described is: the correlation between simultaneous measurements at sites separated by varying distances, the importance of local sources of nuclei in other parts of the world, and the vertical distribution of nuclei. A knowledge of the nature and size distribution of effective nuclei is clearly essential but will have to be obtained by other techniques.

6. Conclusion

The equipment described represents an advance on former techniques in giving an objective measurement of ice crystal concentration in a supercooled cloud. This should be as valuable in the search for useful cloud-seeding substances as in the measurements of natural nuclei. In the form used in these experiments it is extremely simple, reliable and easy to use, but for many applications larger working volumes, expansion techniques or other means of refrigeration have proved desirable.

The series of readings taken with it has shown that there occur large changes in the ice crystal concentration, often of short duration. The fluctuations are large enough to be a factor worth considering in weather modification programmes, and must have some bearing on whether or not natural clouds will rain.

The need for a world-wide series of observations of this sort is obvious, for conditions at this one site may not be typical. The problem of the source of the nuclei requires a similar approach if it is to be resolved.

7. Acknowledgements

I should like to thank the Director of Meteorological Services for providing the information on occurrence of high cloud, and the meteorological staff at Carnarvon airport for local data.

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