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OBSERVATIONS OF FREEZING NUCLEI OVER THE SOUTHWESTERN U. S.

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ABSTRACT

The results of daily flights in January, 1955 in the vicinity of Tucson, Arizona for the purpose of detecting natural freezing nuclei are presented. Samples were taken each day at 15,000 feet, 5,000 feet and at the surface. No systematic correlation was noted with the long-term total rainfall record according to the Bowen meteoric dust hypothesis. However, the fact that average temperatures for given concentrations of nuclei were found to be within 2C° of temperatures for the same concentrations over Sydney, Australia is noted. Certain interesting observations on natural ice crystal clouds are also discussed.

I. DAILY NUCLEI-COUNT FLUCTUATIONS

1. Introduction. For four weeks during January, 1955, daily aircraft flights were made from Tucson, Arizona by staff members of the Institute of Atmospheric Physics of the University of Arizona for the purpose of observing day-to-day fluctuations in the population of natural freezing nuclei in the free atmosphere. These observations were made in cooperation with the Radiophysics Division of the Commonwealth Scientific and Industrial Research Organization, Sydney, Australia.

The work of Bowen [1] on the hypothesis that meteoritic dust may nucleate clouds and thereby influence rainfall amounts received at the ground has made it desirable to seek all possible means of studying the nature of daily variations in nuclei counts. A fairly direct test of this hypothesis should be afforded by flying suitable equipment well above the surface dust layer and counting the numbers of nuclei effective at successively lower temperatures on each of a series of days including several "meteor days", as we may briefly term those dates falling about 29 days after passage of the earth through a major meteor stream. To avoid possible bias and error due to any local peculiarities of atmospheric particulates Bowen has sought to carry out such measurements at more than a single place; so the Radiophysics Division built and generously donated to the Institute of Atmospheric Physics a replica of the nuclei counter reported by Smith and Heffernan [2] which was to be flown in Australia during January 1955. In addition to these Tucson and Sydney flight measurements, observations were made in Hawaii with a simpler device set up at an elevation of about 10,000 feet on Haleakala crater and operated during the same January, 1955 period. Only the Arizona observations will be described in detail here.

The results of the Arizona nuclei counts are reported here not only for the bearing they may have on the meteoritic dust hypothesis, but even more so for their value as systematic observations of natural freezing nuclei in a geographical area for which no previous measurements of this type have been made.

Throughout the following, the term "freezing nuclei" will be used to denote the unidentified particles which, when present in a suitably cold cloud of supercooled water droplets, leads to the appearance of ice crystals therein. The present study sheds no light on the still unsettled problem of whether natural nuclei are actually sublimation nuclei or freezing nuclei.

2. Observational procedure. The Australian nucleus counter consists essentially of a cold chamber having a volume of about 75 liters into which a sample of natural air is introduced and cooled at a rate of about 4C/min. A beam of light illuminates a 10-liter working volume which is continuously observed by the operator as the cooling proceeds. A source of water vapor continuously adds fresh vapor to the observation chamber to maintain a cloud of droplets throughout the chamber. Occurrence of freezing in a droplet is made evident by the scintillation of the resultant ice crystal. The operator logs the number of scintillating particles (or spacing between particles) present in the working volume as a function of the steadily decreasing temperature of the cloud. For other details of the procedure the original report [2] should be consulted. To insure comparability of results, E. J. Smith of the Radiophysics Division spent a month at the Institute of Atmospheric Physics in November, 1954 checking out all flight personnel in the use of the cold chamber.

Certain reservations were felt about the use of this cold chamber when it was found that rather strong temperature gradients existed in it, particularly in the vicinity of the vapor source. After a number of experiments were conducted, it was concluded that the thermometer was located in the coldest part of the chamber and that ice crystals were first formed here and rapidly moved into the light beam by convection so that they became visible with no time lag. The reproducibility of the results for several different operators supported this view and it must be concluded that from the standpoint of large sample volume and rapid cooling and reheating time, the chamber had many superior attributes.

For the Arizona flights, the nuclei counter was installed in a Lockheed Hudson aircraft with a collection tube projecting from the nose of the aircraft well ahead of the propeller plane. A 1.5-inch diameter rubber hose of total length about 15 feet carried the sample to the counter. Ram pressure was sufficient to flush out completely the previous sample and leave a fresh sample in the box in a time of about one minute. A run consisted of observing and counting the ice crystals appearing during the 10-12 minute period required to cool the sample to -40C.

From 3 January to 31 January, 1955, daily flights to 15,000 feet were made. Two observers went on each flight to permit cross-checks between independent observers. Each observer made one run at the 15,000 foot level and each made one run at the 5,000 foot level. One of the two made a single run immediately upon landing, employing an air sample whose collection was completed exactly at the instant of touchdown of the aircraft. The samples taken at 15,000 feet are regarded as free from any contamination due to local dust which in the winter does not rise above a few thousand feet above terrain, particularly during the morning hours (1000-1200) during which these flights were almost invariably made. Westerly or northwesterly flow at 15,000 feet prevailed during most of the period, so the air at the 15,000 foot level cannot be said to have been uninfluenced by possible surface sources upwind (in the California area). Knowledge of rates of turbulent elevation of surface dust is unfortunately too meager to permit any close estimates of the extent to which dust from the ground in California might affect the nuclei counts at 15,000 feet over Arizona.

The observations taken at 5,000 feet were made in order to detect, if present, any systematic height variations relating either to surface effects or, on Bowen's hypothesis, to descent of a stratum of nuclei from above. The surface observations were made in order to shed indirect light on the whole question of surface contamination and to extend the basic study of height variations of nuclei counts. During all but one flight, the aircraft flew within about 50 miles of Tucson. On one occasion the flight went to northern Arizona and back, but no significant differences characterized the results of this flight.

3. Results. For each of the approximately 150 samples taken in the course of the Arizona flights during January, 1955, the temperature at which the number of ice crystals reached a value equivalent to 0.1/L, (0.1 crystal per liter) 1/L, 10/L and 100/L was determined. This range of crystal concentrations spans the values customarily regarded as necessary to initiate the Bergeron mechanism of precipitation in clouds (10/L being a commonly suggested concentration).

In Figures 1 through 4, the results for samples taken at 15,000 feet are presented in detail. As has been cited above, samples taken at this level are believed to be free from any local contamination and hence should be fairly representative of the entire southwestern U. S. under normal conditions of westerly flow in winter. For each of these figures, the results of each of the two observers' determinations are plotted for each day in order to display the variability inherent in this observational technique. Since six persons rotated as observers during these flights, the slight differences resulting from individual observing techniques cannot easily be shown on figures, so the lines joining the points have been drawn simply to connect the points corresponding respectively to the warmer and the colder of the observations for each flight. The average difference, without regard to sign, between the temperatures at which the crystal concentrations rose to each given value are entered on the corresponding graph to provide a measure of the dispersion in results arising from personal differences of technique. It will be seen from Figures 1-4 that this difference averages about two Centigrade degrees, but on one occasion (24 January, concentration 10/L) it was as large as 11C. It is entirely possible that even this large a difference between observers is a real one resulting from differences in nuclei populations at points several tens of miles apart on a given flight, though this possibility could only be checked by numerous repeated observations at the air points in question, a check which could not be made in this study.

These nuclei counts at 15,000 feet over Arizona do not reveal peaks on those dates which would agree with the world-wide meteor days as discussed previously by Bowen [1]. The Arizona nuclei counts exhibit, in fact, no prominent peaks if we consider averages for both observers on each run. The results of the concurrent observations in Sydney have been described by Smith, Kassander, and Twomey [3] as offering some support for the meteoritic hypothesis and hence differ in this respect from the Arizona data. In particular the Sydney data for concentrations of 1/L exhibit peaks on 13, 23, and 29 January in good agreement with the meteor days. It is of some interest to note that the Arizona curves running through points corresponding to the <u>warmer</u> temperature of each pair of observations do display maxima near January 16 and 24-25 and that such maxima are displaced about two or three days from the Sydney maxima and from the meteor days of the Bowen hypothesis. Although so little is known of the vertical air motions in the stratosphere and ozonosphere in particular parts of the world that one must be willing to admit differences of a few days in times of descent of any strata of meteoritic particles incident upon the top of the atmosphere, we cannot find any strong reason for selecting the warmer points of the pairs of observations in question, so we cannot offer these points as indirect evidence for the meteoritic dust hypothesis. The conclusion, to repeat, is that the Arizona nuclei counts in January 1955 do not seem to support the hypothesis in the way that the Sydney data do.

The observations carried out by personnel of the Radiophysics Division in Hawaii during this same January period appear to support the Bowen hypothesis as well or better than the Sydney data. Furthermore, some concurrent nuclei counts made by Cwilong in Panama also tend to support the hypothesis, though in a way that is less clear-cut. Hence the Arizona data seem to stand alone as failing to substantiate the suggestion that there may be significant increases in the numbers of freezing nuclei on the days falling about 29 days after passage of the earth through meteor showers. It is clear that more work along these general lines is needed to make final decision possible in this interesting question.

Since the nuclei counts made in Arizona during the past January have considerable interest in themselves all aside from their implications for the Bowen hypothesis, additional results are presented in Figures 5 and 6 and in Table 1. In Figure 5, comparative temperatures at surface, 5,000 feet and 15,000 feet for a concentration of 1/L are shown and in Figure 6 the same types of curves are given for a crystal concentration of 100/L. Table 1 summarizes the average temperatures at which the nuclei counts rose to given concentrations at each of the altitudes at which samples were taken. Note that although there is an evident trend towards less active freezing nuclei at greater heights, there is only a difference of a few degrees between the surface nuclei activation temperatures and the 15,000-foot nuclei activation temperatures at corresponding crystal concentration. Thus, to develop a concentration of one ice crystal per liter, surface air had to be cooled on the average to about -27C while 15,000 foot air had to be cooled to -29C. This difference, it is to be noted, is about equal to the average difference between observers, so it may not even be a real difference. At any event, these results provide some small

TABLE 1. Summary of the average temperatures (°C) at which the ice crystal concentrations rose to specified values at three altitudes over southern Arizona during 3-31 January, 1955.

Altitude Feet	Ice Crystal Concentrations 0.1/L	1/L	10/L	100/L
Sfc.	-25	-27	-28	-30
5,000	-26	-29	-31	-33
15,000	-27	-29	-31	-33

basis for concluding that even surface nuclei counts should give a good first approximation to the upper air nuclei counts in the Arizona area. Daily fluctuations aloft are not, however, faithfully shown by corresponding surface fluctuations, as for example on January 6.

Finally, it is of interest to note that the Sydney and Tucson data are in rather good agreement as to the temperatures at which nucleation develops a given ice crystal concentration. Thus, for just the 15,000 foot data one finds that, over Arizona, cooling to -29.1C was required on the average during January, 1955, to develop an ice crystal concentration of 1/L, whereas the same figure for Sydney was -27.4C, or only 1.7C warmer despite the large geographical separation of these locales. An equally small temperature difference appears from other such comparisons in these two sets of data, so one is tempted to conclude that freezing nuclei populations may be much the same in all parts of the world. Such a conclusion cannot be drawn firmly, however, until very much more information is available than is presented here.

4. Summary and conclusions. The Arizona nuclei counts of January 1955 do not exhibit fluctuations that support in any clearly recognizable way the hypothesis that Bowen has formulated to account for certain irregularities in long-term rainfall records. Concurrent counts made in Sydney with the same apparatus and observing techniques do appear to support the meteoritic dust hypothesis, as do also concurrent counts taken in Hawaii and Panama. Whether some peculiarity in the high-level airflow over the southwestern Arizona could account for a failure to observe such peaks, or whether unrecognized differences

in observing techniques are basically responsible, or whether some phenomenon not related to meteoritic dust is in control here is not clear.

It is of considerable interest to learn that the nucleating efficiency of freezing nuclei in Arizona during January 1955 was nearly identical, on the average, to that of nuclei over eastern Australia, for this poses the possibility that some sort of large-scale uniformity may exist in this regard. The Hawaii and Panama data tend to support this speculation, though those observations were less detailed than the Tucson and Sydney observations.

The work reported here must be regarded as making its principal contribution by virtue of providing free-air observations of the activation temperatures corresponding to a series of different crystal concentrations in air that may be regarded as representative of the southwestern U. S. in midwinter. In general, it would seem that air must rise to an altitude of between 20,000 to 25,000 feet in winter in the southwestern U. S. in order to be cooled to such a low temperature as to yield crystal concentrations of the order of those currently felt to be necessary to initiate the Bergeron process in supercooled clouds.

II. OBSERVATIONS ON EVAPORATION ICE NUCLEI

1. Introduction. During two of the flights made during January 1955, a phenomenon of basic interest in the problem of natural freezing nucleation was observed. This phenomenon consisted of an unusually warm nucleation temperature for samples of air taken within ice-crystal clouds. The sampling procedure employed with the Australian nuclei counter consists in filling the chamber, warming to about 10C, and then starting the cooling process that leads to nucleation in the cloud of drops that is created in the chamber. Hence, the ice crystals drawn into the chamber from the cloud and held at above-freezing temperatures for a time of the order of 100-200 seconds must have both melted and evaporated. Since air samples taken on the same days (January 8, 9, see

below) and at the same altitude but outside the clouds exhibited no unusual nucleation properties, the observed phenomenon is suspected of being a property of some kind of residues left from the melting and evaporation of the natural ice crystals.

2. Observations in natural clouds. One of us (L. L. S.), flying as one of the two observers on the 8 January flight, directed the pilot to enter a small cumulus cloud from which snow virgae descended. His objective was to draw in a sample of air from within this cloud, which was one of a small fraction of such cumuli showing evidence of having been naturally nucleated. The clouds had bases at 5,000 feet and tops at about 10,000 and the sample was taken at 5,200 feet at a temperature of -3C. Immediately after admitting the cloud-air sample into the cooling chamber which was at 13C, two unusual phenomena were noted. First, there existed a large number of non-scintillating particles with average spacing of only about 0.5 cm. Second, a smaller number (roughly 10 per cent as many) exhibited scintillation effects. No water droplet fog was in the chamber at this time. After noting the above peculiarities, fog was added in the normal fashion and the cooling process begun. Immediately after the chamber temperature fell below OC, large numbers of scintillating crystals appeared. At a temperature of -1C their concentration was already up to about 100/L as estimated from intercrystal spacing. This high concentration was maintained until about -10C, after which fallout reduced the value to only about 1/L. Not until the chamber reached -25C did the concentration of scintillating particles rise again to the 100/L value attained at -1C. By way of comparison, a clear-air sample taken near this cloud showed no scintillating particles at all until -11C (one lone crystal) and gave a 100/L concentration only after cooling to -32C, which is close to the month-long average temperature required for 100/L (see Table 1).

It was not possible to secure a second ice-cloud sample on 8 January; but on 9 January two more cases of a similar nature were observed, but with less extreme departure from normal. In a cloud at 10,000 feet, which contained ice crystals that were clearly discernible as they flashed into the chamber, a sample was taken and held at about 13C for 10 minutes before being cooled. Again a high concentration of dull particles plus a few scintillating particles were found at above-freezing temperatures. On cooling, a few scintillations were observed at -5C and by about -11C a concentration in excess of 100/L was observed. These fell out as before and not until about -25C did the concentration reach 100/L. For comparison, samples were taken on this day in both clear air and in water-droplet clouds, yet neither type of sample displayed unusual characteristics.

The second ice-crystal cloud sampled on 9 January exhibited a high concentration of non-scintillating particles during the above-freezing stage and gave one scintillating crystal at -4C and 100/L at -14C. This second cloud was in the vicinity of a smelter at Douglas, Arizona and only slightly above the level of the smelter smoke plume so it was decided to take a sample well within the plume for comparison. Such a comparison sample taken on 11 January at 8,000 feet did in fact show a high concentration of non-scintillating particles at the start of the run (above-freezing temperatures) but did not show any unusual nucleation effects (to get 100/L required cooling to -27C).

The Australian flight personnel have reported observations of high concentration of non-scintillating particles similar to those described here as seen at above-freezing temperatures--they also describe seeing occasional scintillating particles at such times. They did not observe the peculiar nucleating effects that are reported here for three samples taken entirely inside ice-crystal clouds.

Despite continued search for ice-crystal clouds during the remainder of the period of flight work, no additional observations were possible on this peculiar effect. This is unfortunate in view of the obviously great potential importance of any natural process which might create highly active residues within ice crystals that could retain their nucleating effect even after melting and evaporation of the crystals themselves. The phenomenon becomes of principal interest in connection with its possible relation to natural seeding by cirrus clouds. If residual particles of some unidentified nature could survive descent from a cirrus deck through many thousands of feet of unsaturated air and still yield nucleation effects comparable only to those known to occur with silver iodide, the significance of the process would be very great. Laboratory studies attempting to duplicate this phenomenon by warming ice crystal clouds to temperatures above freezing and then recooling them have not been successful.

It should be noted that a somewhat similar phenomenon has been found by Gourley and Crozier (unpublished) in the laboratory at the New Mexico Institute of Mining and Technology. Their experiment consisted of evaporating (but not also melting) ice crystals formed by spontaneous nucleation at the -40C point. Residual nuclei of some unidentified type were left after evaporating the crystals and survived for times of the order of several hours. The physics of this phenomenon was not studied in detail by Gourley and Crozier and so, like the possibly related phenomenon described here, remains to be explained.

3. Summary and conclusions. The point of greatest importance in these observations made on January 8 and 9, 1956, seems to be that natural ice-crystal clouds forming in an air mass having no apparently abnormal nucleating properties yielded some type of residues capable of remarkably efficient nucleation. The chemical physics of the phenomenon is completely unknown. It is suggested that others working on nuclei-counting experiments should search for additional evidence of this potentially important phenomenon, both in the free atmosphere and in the laboratory.

4. Acknowledgments. This paper reports work done jointly by a number of members of the staff of the Institute of Atmospheric Physics; thanks are due all of them. The generous assistance of the Radiophysics Division of the Commonwealth Scientific and Industrial Research Organization is gratefully acknowledged. Dr. E. G. Bowen and Mr. E. J. Smith of that organization deserve particular thanks for their part in initiating the work described here. The flight program was supported as part of a grant from the Alfred P. Sloan Foundation.

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[2] Smith, E. J., and K. J. Heffernan, 1954: Airborne measurements of the concentration of natural and artificial freezing nuclei. Quart. J. r. meteor. Soc., 80, 182-197.

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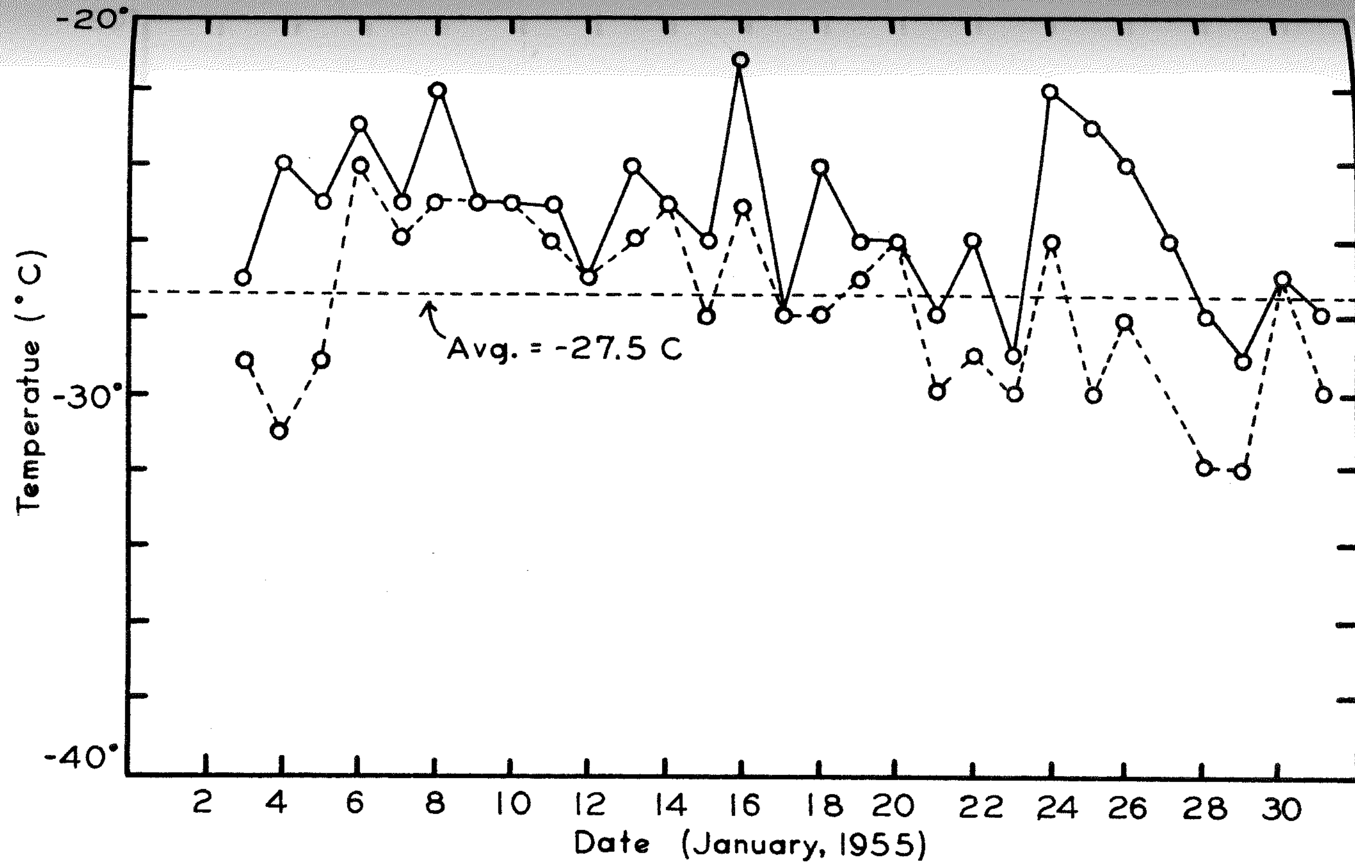

Figure 1. Daily variation in temperatures at which ice-crystal concentrations reached 0.1/liter in air samples taken at 15,000 ft over southeastern Arizona. Solid curve refers to warmer one of each pair of daily values, dashed curve to colder values. Average temperature difference between observers, 2.3C.

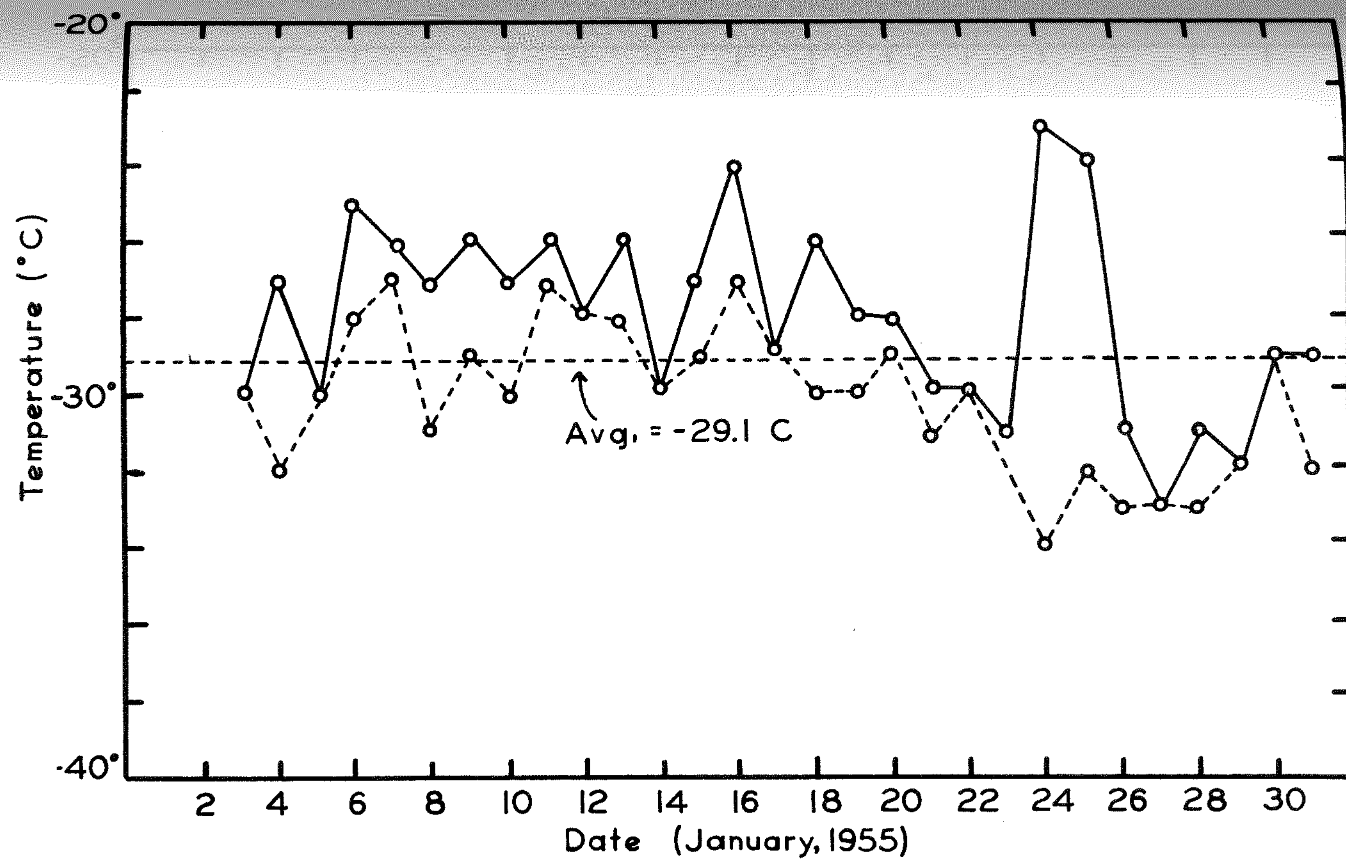

Figure 2. Daily variation in temperatures at which ice-crystal concentrations reached 1/liter in air samples taken at 15,000 ft over southeastern Arizona. Solid and dashed curves as in Figure 1. Average temperature difference between observers, 2.1C.

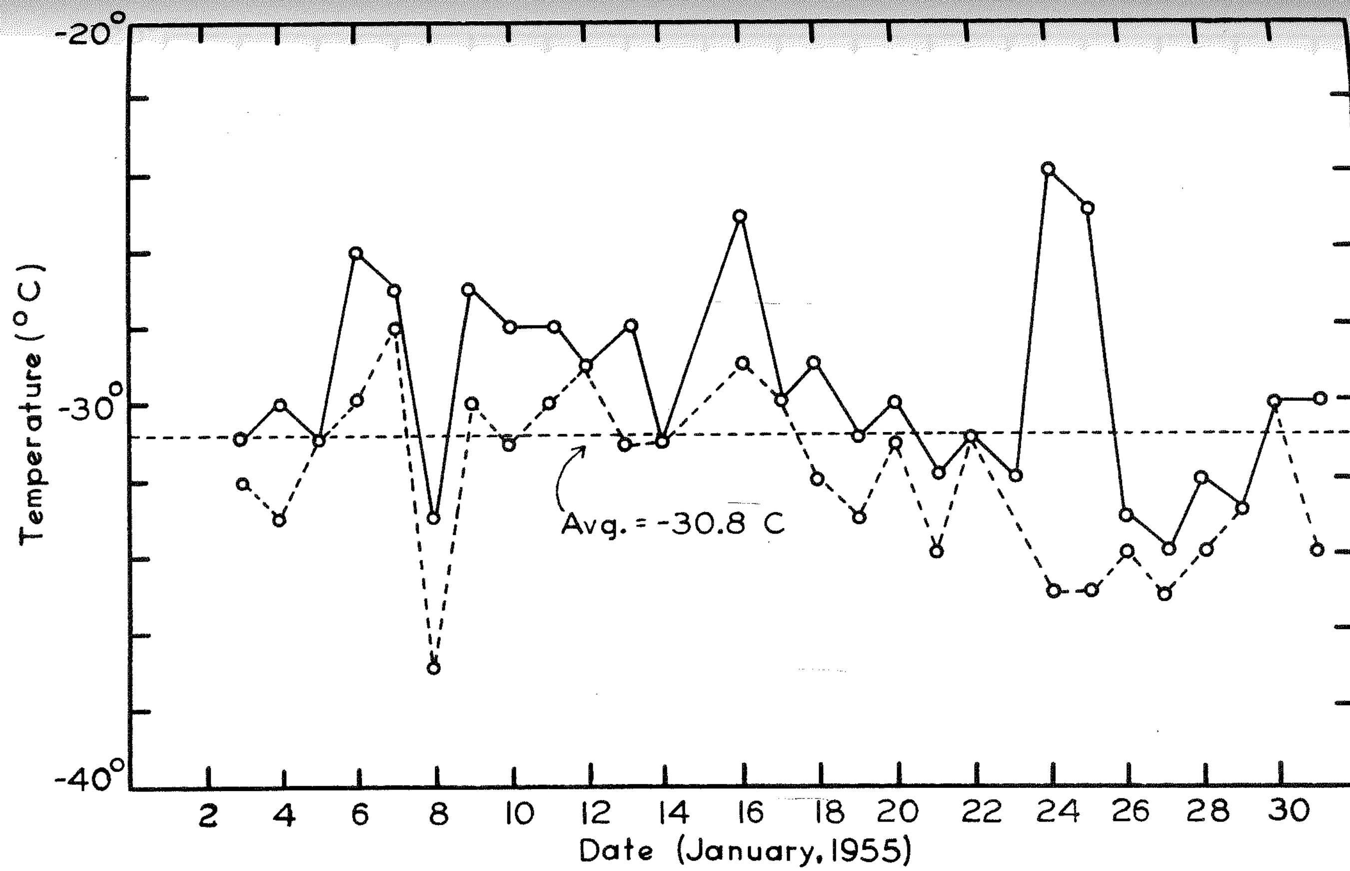

Figure 3. Daily variation in temperatures at which ice-crystal concentrations reached 10/liter in air samples taken at 15,000 ft over southeastern Arizona. Solid and dashed curves as in Figure 1. Average temperature difference between observers, 2.3C.

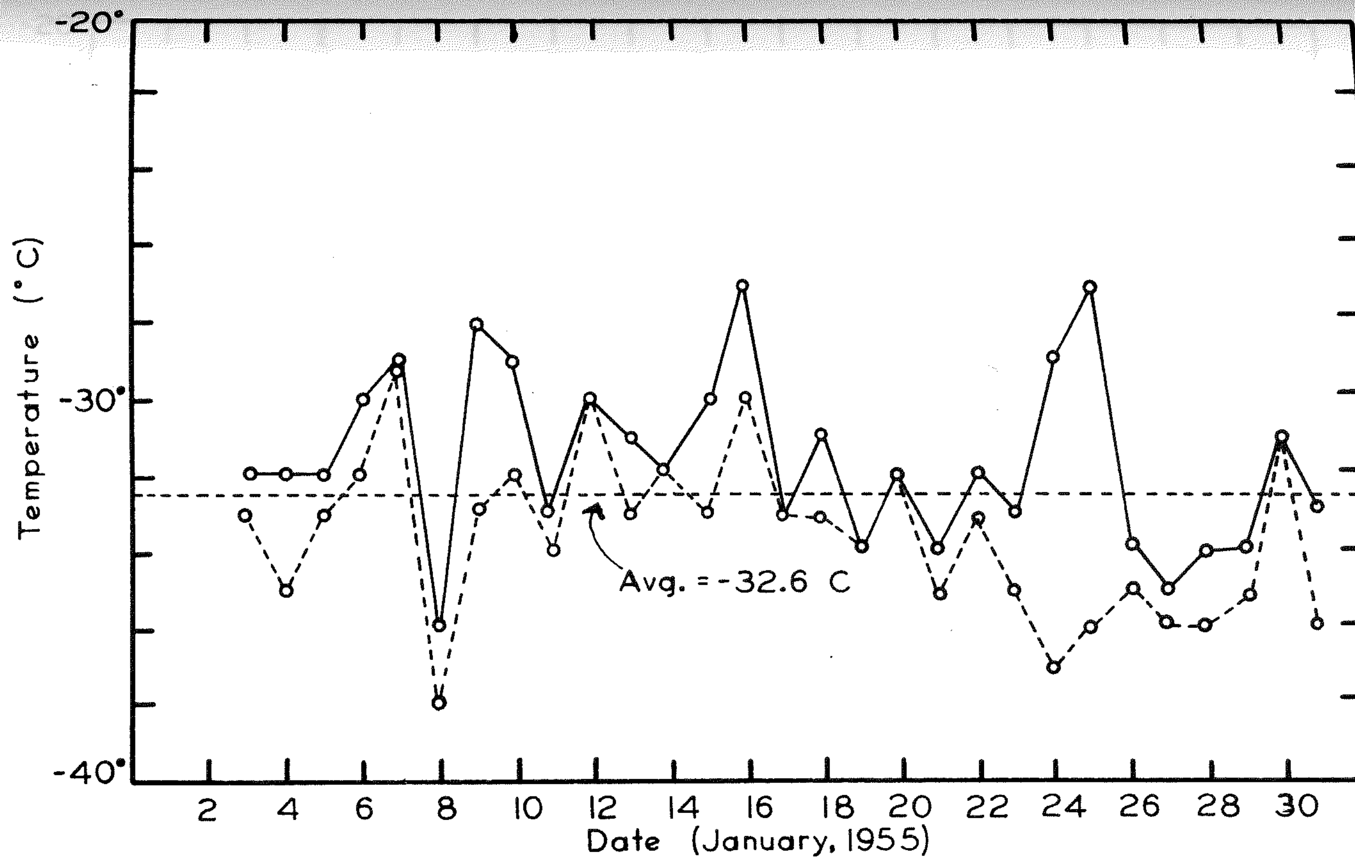

Figure 4. Daily variation in temperatures at which ice-crystal concentrations reached 100/liter in air samples taken at 15,000 ft over southeastern Arizona. Solid and dashed curves as in Figure 1. Average temperature difference between observers, 2.0C.

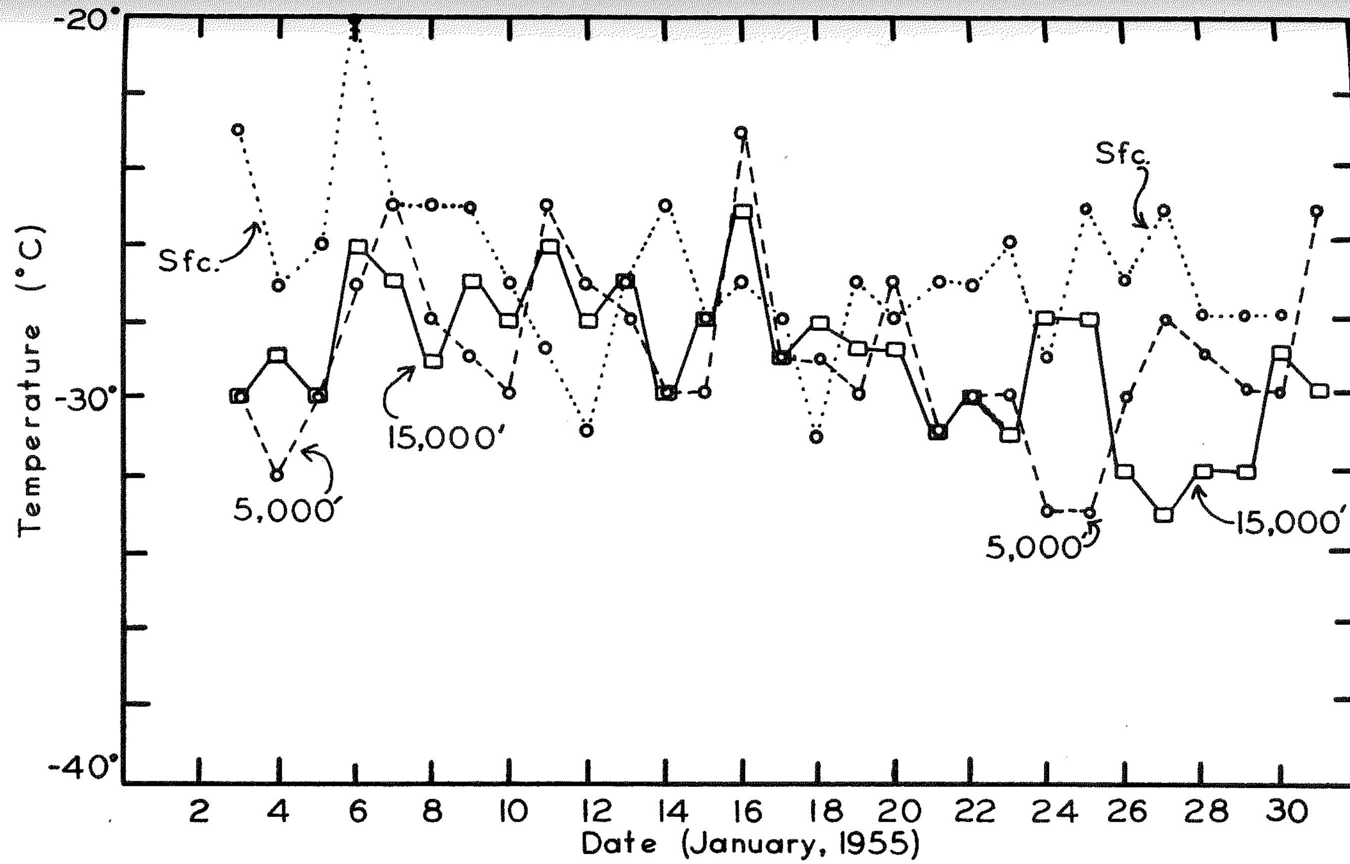

Figure 5. Daily fluctuations in temperatures at which ice-crystal concentrations reached 1/liter at surface, 5000 ft and 15,000 ft. Curves for 5000 ft and 15,000 ft are for averages of daily pairs of observations.

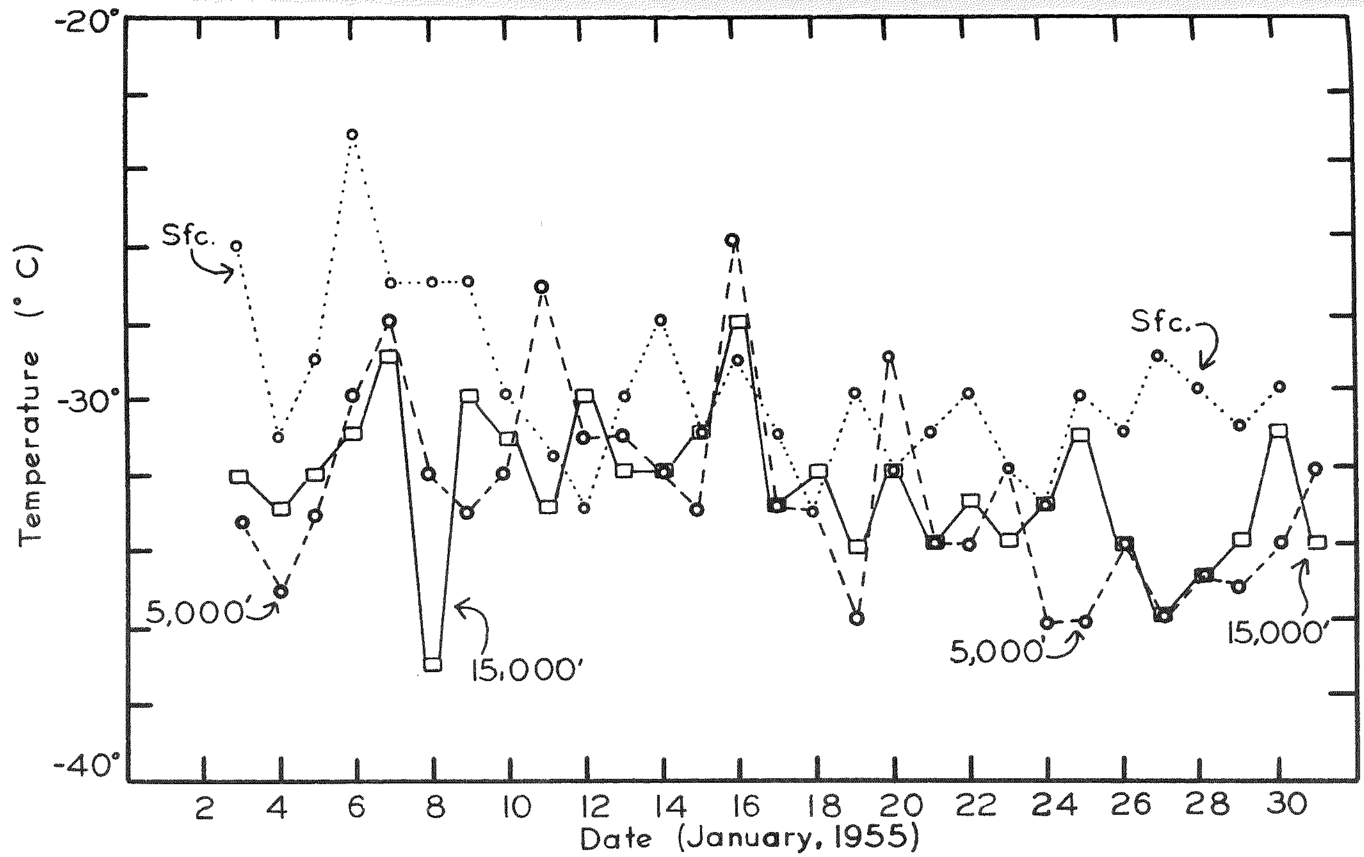

Figure 6. Daily fluctuations in temperatures at which ice-crystal concentrations reached 100/liter at surface, 5000 ft and 15,000 ft. Curves for 5000 ft and 15,000 ft are for averages of daily pairs of observations.