OBSERVATIONS OF FREEZING NUCLEI OVER THE SOUTHWESTERN U.S.

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Abstract—Flight observations of numbers of freezing nuclei as a function of temperature were made at 5000 and 15,000 ft over southern Arizona every day from January 3 to January 31, 1955. The dates on which peaks in the nuclei counts occur do not support in any clearly recognizable way the hypothesis that they are associated with meteoritic dustfalls. The average temperature to which collected air samples had to be cooled to obtain given ice-crystal concentrations over Arizona are equal to within about two degrees centigrade, the corresponding values found at similar altitudes over Sydney, Australia, during the same period, thus suggesting a possibly worldwide similarity in nuclei populations. Observations of a remarkable nucleation activity noted on three occasions in air samples taken within clouds containing natural ice crystals are reported briefly but cannot be ascribed to any known physical processes.

DAILY NUCLEI-COUNT FLUCTUATIONS

Introduction—For four weeks during January, 1955, daily 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 [1953] 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 these 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 [Smith and Heffernan, 1954] 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 ft on Haleakala Crater and operated during the same January, 1955, period. Only the Arizona observations will be described in detail here.

Observational procedure—The Australian nuclei 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 4°C/min. A beam of light illuminates a ten-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, see the original report [SMITH and HEFFERNAN, 1954]. 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.

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 plane of the propeller. A 1·5-inch diameter rubber hose of total length about 15 ft 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 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 $-40°C$.

From January 3 to January 31, daily flights to 15,000 ft were made. Two observers went on each flight to permit cross-checks between independent observers. Each observer made one run at the 15,000-ft level and each made one run at the 5000-ft 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 touch-down of the aircraft. The samples taken at 15,000 ft are regarded as free from any contamination due to local dust which in the winter is not known to rise above a few thousand feet above terrain, particularly during the morning hours (10h 00m–12h 00m) during which these flights were almost invariably made. Westerly or northwesterly flow at 15,000 ft prevailed during most of the period, so the air at the 15,000-ft level cannot be said to have been uninfluenced by possible surface sources upwind (in the California area). The observations taken at 5000 ft 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.

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 1–4, the results for samples taken at 15,000 ft are presented in detail. As has been cited above, samples taken at this level are believed to be free from local contamination and hence should be fairly representative of the entire southwestern U.S. 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.

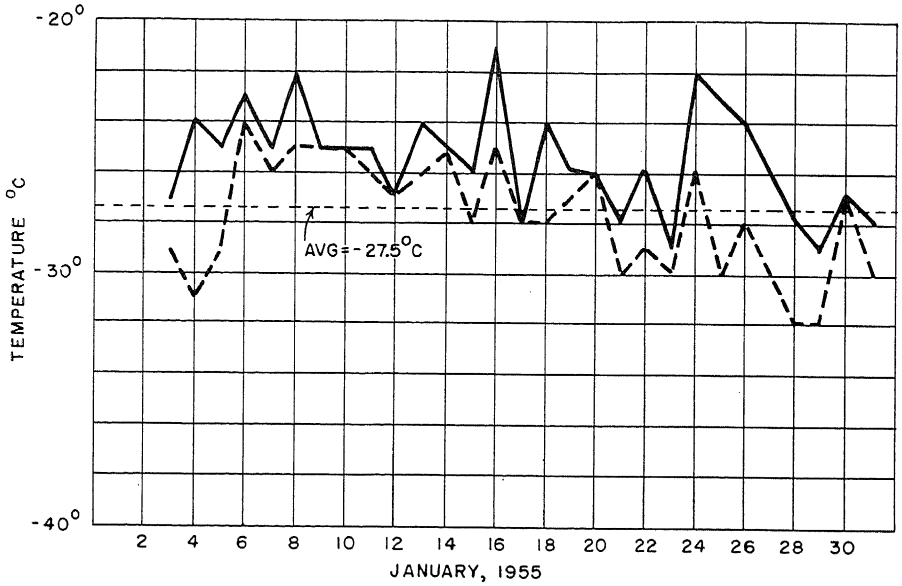

FIG. 1—Daily variation in temperatures at which ice-crystal concentrations reached 0·1 per liter in air samples taken at 15,000 ft over southeastern Arizona; solid curve refers to warmer one of each pair of daily values, dashed curve to colder values; average temperature difference between observers, 2.3° C

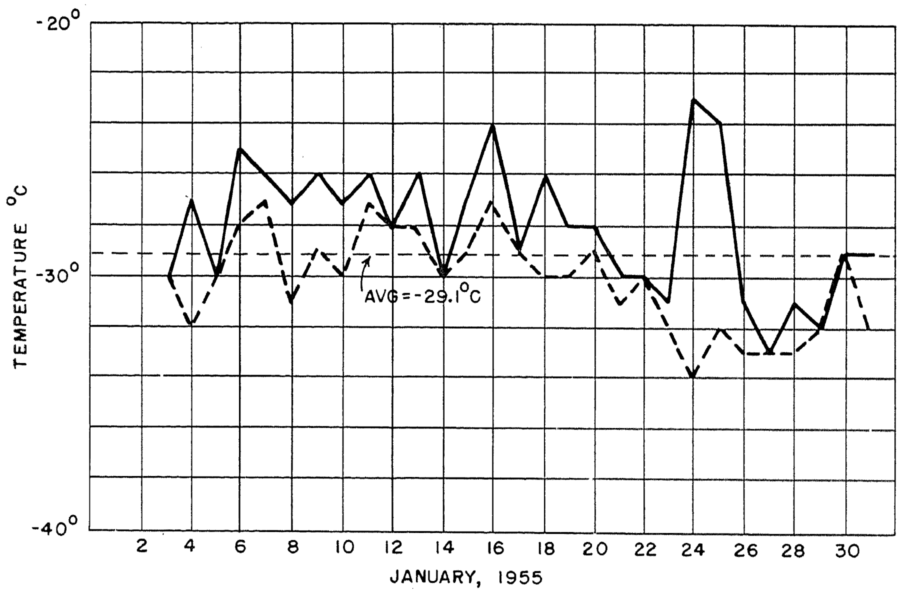

FIG. 2—Daily variation in temperatures at which ice-crystal concentration searched 1 per liter in air samples taken at 15,000 ft over southeastern Arizona; solid and dashed curves as in Fig. 1; average temperature difference between observers, 2·1°C

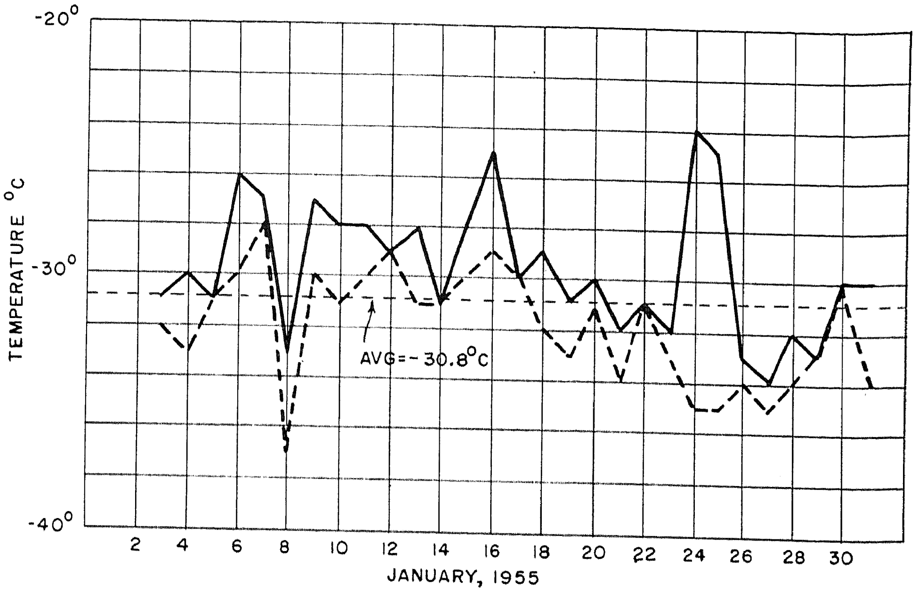

FIG. 3—Daily variation in temperatures at which ice-crystal concentrations reached 10 per liter in air samples taken at 15,000 ft over southeastern Arizona; solid and dashed curves as in Fig. 1; average temperature difference between observers, 2·3°C

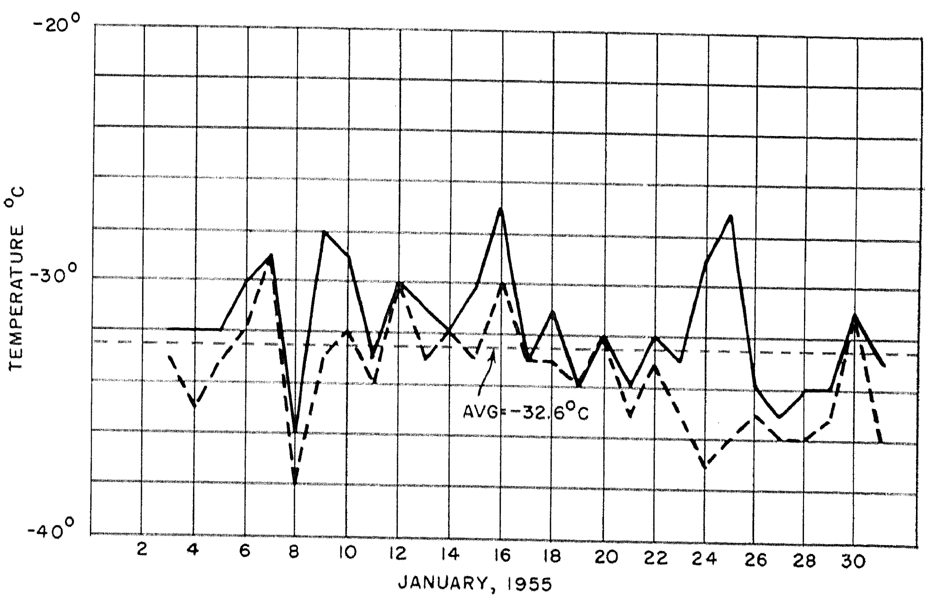

FIG. 4—Daily variation in temperatures at which ice-crystal concentrations reached 100 per liter in air samples taken at 15,000 ft over southeastern Arizona; solid and dashed curves as in Fig. 1; average temperature difference between observers, 2·0°C

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 degrees Centigrade, but on one occasion (January 24, concentration 10/L) it was as large as 11°C. It is entirely possible that even so large a difference between observers is a real one due to differences in nuclei populations at points several tens of miles apart on a given flight, though this possibility could only be attested by repeated observations at the air points in question.

These nuclei counts at 15,000 ft over Arizona do not reveal peaks on those dates which would agree with the worldwide meteor days as discussed previously by BOWEN [1953]. 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 briefly by BOWEN (talk to International Arid Lands Symposium, Albuquerque, N.M., 1955) 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 January 13, 23, and 29 in good agreement with the meteor days. Note that the Arizona curves running through points corresponding to the warmer temperature of each pair of observations 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 as 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 Table 1. In Fig. 5, comparative temperatures at surface, 5000 ft and 15,000 ft for a concentration of 1 per L are shown and in Fig. 6 the same types of curves are given for a crystal concentration of 100 per L.

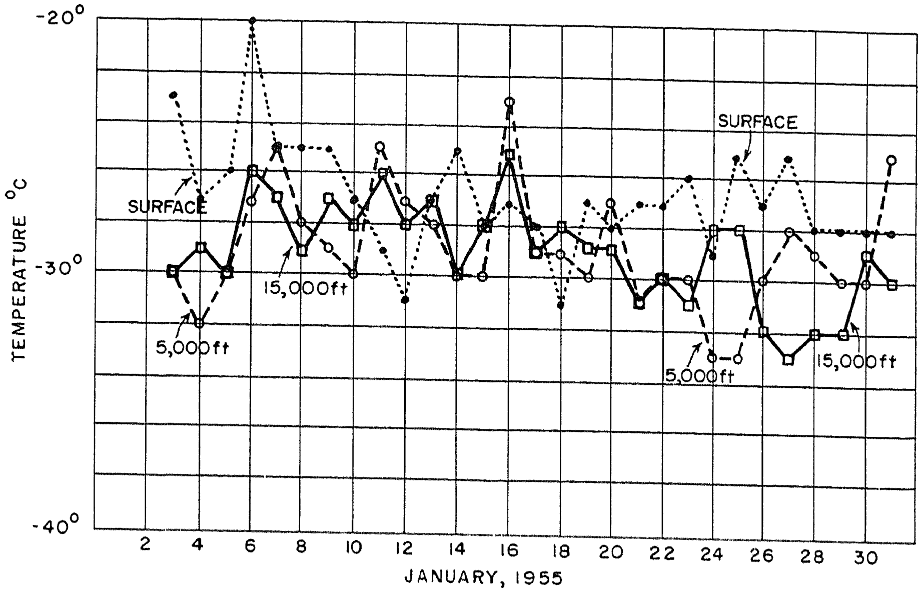

FIG. 5—Daily fluctuations in temperatures at which ice-crystal concentrations reached 1 per liter at surface, 5000 ft and 15,000 ft; curves for 5000 ft and 15,000 ft are for averages of daily pairs of observations

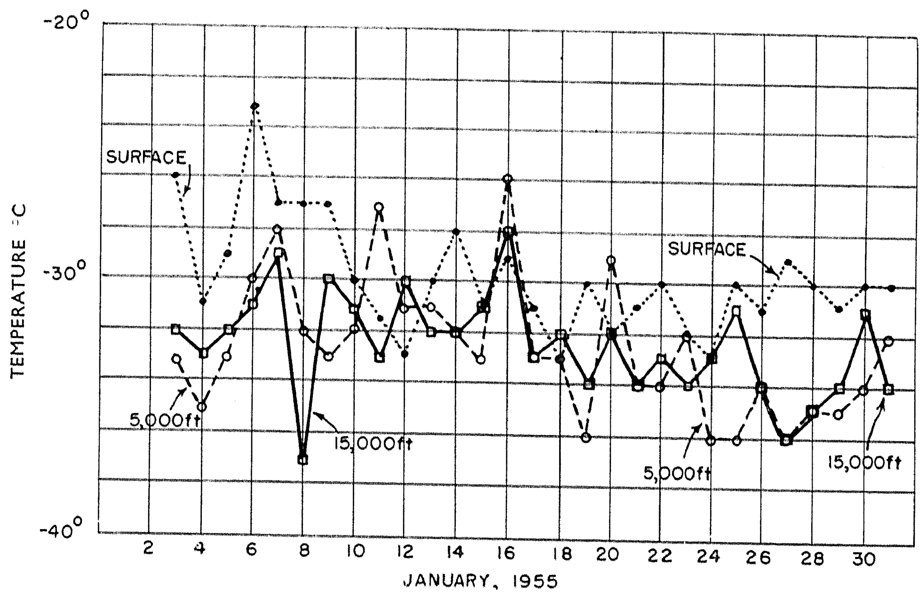

FIG. 6—Daily fluctuations in temperatures at which ice-crystal concentrations reached 100 per liter at surface, 5000 ft and 15,000 ft; curves for 5000 ft and 15,000 ft are for averages of daily pairs of observations

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 more 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-ft 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 -27°C while 15,000 ft air had to be cooled to -29°C. This difference is about equal to the average difference between observers, so it may not even be a real difference. In any event, these results provide some small 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.

Table 1. *Summary of the average temperatures at which the ice crystal concentrations rose to specified values at three altitudes over southern Arizona during January* 3–31, 1955

Altitude	Ice crystal concentration			
	0·1/L	1/L	10/L	100/L
ft	°C	°C	°C	°C
Surface	−25	−27	−28	−30
5,000	−26	−29	−31	−33
15,000	−27	−29	−31	−33

Finally, note that the Sydney and Tucson data are in remarkably good agreement as to the average temperatures at which nucleation develops a given ice-crystal concentration. Thus, for just the 15,000 ft data one finds that over Arizona cooling to $-29{\cdot}1$°C was required on the average during January, 1955, to develop an ice crystal concentration of 1/L, whereas the same figure for Sydney was $-27{\cdot}4$°C, or only 1·7°C 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.

It is to be noted that the Tucson and Sydney nuclei counts yielded results in substantial disagreement with the conclusions of AUFM KAMPE and WEICKMANN [1951], who suggested that in most air masses natural aerosols should produce ice crystal concentrations of the order of 1/L to 10/L by cooling only to about -15°C.

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 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 south-western 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 United States in midwinter. In general, it would seem that air must rise to an altitude of between 20,000 to 25,000 ft 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.

OBSERVATIONS ON EVAPORATION ICE NUCLEI

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 +10°C, 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 sec must have both melted and evaporated. Since air samples taken on the same day 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.

Observations in natural clouds—L. L. Sims, flying as one of the two observers on the January 8 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 5000 ft and tops at about 10,000 ft; the sample was taken at 5200 ft at a temperature of −3°C. Immediately after admitting the cloud-air sample into the cooling chamber which was at 13°C, 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 ten per cent as many) exhibited scintillation effects. No water droplet fog was in the chamber at this time. After noting the above peculiarities and waiting about two minutes to detect any changes in the appearance of the sample, fog was added in the normal fashion and the cooling process begun. Immediately after the chamber temperature fell below 0°C, large numbers of scintillating crystals appeared. At a temperature of −1°C their concentration was already up to about 100/L as estimated from intercrystal spacing. This high concentration was maintained to about $-10°C$, after which fallout reduced the value to only about 1/L. Not until the chamber reached $-25°C$ did the concentration of scintillating particles rise again to the 100/L value attained at $-1°C$. By way of comparison, a clear-air sample taken near this cloud showed no scintillating particles at all until $-11°C$ (one lone crystal) and gave a 100/L concentration only after cooling to $-32°C$, 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 January 8; but on January 9 two more cases of a similar nature were observed, but with less extreme departure from normal. In a cloud at 10,000 ft which contained ice crystals that were clearly discernible as they flashed into the chamber, a sample was taken and held at about 13°C for ten 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 $-5°C$ and by about $-11°C$ a concentration in excess of 100/L was observed. These fell out as before and not until about $-25°C$ 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 January 9 exhibited a high concentration of non-scintillating particles during the above-freezing stage and gave one scintillating crystal at $-4°C$ and 100/L at $-14°C$. 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 January 11 at 8000 ft 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 $-27°C$).

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 great potential importance of any natural process which might create highly active residues within ice crystals that would retain their nucleating effect even after melting and evaporation of the crystals. 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 obvious. In order to gain further information on this process some laboratory studies have been conducted by L. L. Sims. These will be briefly summarized next.

Laboratory observations—In an effort to duplicate the natural phenomenon just described, Sims attempted to form ice-crystal clouds within the cooling chamber of the nuclei counter by cooling to temperatures of the order of —40°C, then warming to above freezing and then cooling again. To maintain the cloud of ice crystals in suspension while carrying them through this cycle, a fan was introduced into the chamber and operated during the initial experiments. No successful laboratory reproductions of the phenomenon were obtained until the fan was removed. On carrying out the process without a fan in operation inside the chamber, residual nucleating effects were found at temperatures as warm as about —5°C. At the present time this work is suspended because of other Institute activities, and it is not possible to draw any firm conclusions as to the implications of the laboratory work. Just prior to suspension of the work it was discovered that the early nucleation effect was not then reproducible unless the initial cooling rate in the chamber was about double the rate employed in the flight work, so this point will have to be checked more completely in the near future.

It should be noted that a somewhat similar phenomenon has been found by Gourley and Crozier (unpublished) 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 —40°C 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.

Summary and conclusions—The point of greatest importance in these observations 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.

Acknowledgments—This paper reports work done jointly by a number of members of the staff of the Institute of Atmospheric Physics; thanks are due to all of them. The generous assistance of the Radiophysics Division of the Commonwealth Scientific and Industrial Research Organization is gratefully acknowledged. E. G. Bowen and E. J. Smith of that organization deserve particular thanks for their part in initiating the work described here.

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DISCUSSION

Dr. Everly J. Workman—How warm could you make the air sample and still have the ice reappear?

Dr. James E. McDonald—The first occasion was about +13°C, and the other +10°C.

Dr. Workman—We are interested in this problem through the work of Mary Gourley, in our laboratory. She finds a persistent nucleus from clean ice which has been desiccated to the point of invisibility at very high magnification.

Dr. McDonald—Is this at negative temperatures?

Dr. Workman—Yes, we wouldn't expect that it would occur after having been at warmer temperatures. We have not determined the limits of temperature yet.

Dr. McDonald—The Gourley results are as perplexing as ours. Some property apparently persists long after common sense indicates it should have gone. They differ in the respect that your group did the experiments at negative temperatures by dehumidifying the samples, so they would evaporate and not melt.

Dr. Helmut Weickmann—I'd like to offer a possible explanation of this phenomenon. Let's say your observation is correct. You didn't get any contamination from exhaust gas or from anything else. Once a nucleus has acted as a freezing nucleus it has an adsorption layer of H_20 molecules. This adsorption layer adheres to the crystal face and does not evaporate, not even at a low relative humidity. It also persists in temperatures above freezing and, this is important, with its molecules arranged in the right spacing. So, if you cool it down afterwards, you still have the oriented adsorption layer or, in other words, a very good freezing nucleus.

Dr. Edward M. Brooks—I'd like to make one point about these meteor showers. You mentioned that the Sydney and Haleakala Crater data supported the Bowen hypothesis. I think we should remember that meteor showers themselves may last many days, sometimes weeks, and it is very exceptional to find just a strong burst and then have nothing on the previous or following day.

Another thing you have to think of is the thirty-day period you mentioned. Of course, that could allow a little leeway, too. It seems to me you should not expect a sharp peak to verify the hypothesis but rather a gradual rise and fall. It seems that there should be some other explanation for that some day.

Dr. McDonald—The start of all this came from the rainfall peaks which are surprisingly sharp. This was Bowen's first point. One would ask for equally high peaks in the nuclei. But, I agree that from the astronomical and the meteorological aspects one would not expect it.

Dr. David Atlas—I wonder about the significance of these nuclei counts with regard to natural precipitation because of the great difference in the temperatures at which sufficient nuclei are presumably available to initiate the Bergeron process, and that at which actual clouds have been observed to release ice crystal precipitation. Nuclei counts of 10 per liter are not observed above $-28°C$, while observations of *Fallstreifen* by Peppler in Germany and our own radar observations of natural precipitation show a rather strong preference for the initiation of the Bergeron process in the neighborhood of $-15°C$. Can you explain this apparent inconsistency?

Dr. McDonald—If, in fact, there exists a residual property that would suffice to initiate crystal formation, then homogeneous nucleation at the cirrus clouds could be the source. They could fall out of cirrus, into lower clouds.

Dr. James P. Lodge, Jr.—I think all of us tend to underestimate the persistence of crystals. There was some work done in Germany by some of the medical researchers during the war. They were studying the nucleation of carbon dioxide bubbles in liquids. They discovered that a sodium chloride solution which had been made up, filtered, and allowed to stand 15 or 20 minutes still contained particles of something or other, presumably remaining polymolecular aggregates of sodium chloride capable of nucleating the formation of CO_2 bubbles; and only after something like standing for an hour could you pull a vacuum on them without getting bubble formation. Apparently, these crystalline entities, whatever they are, do stick around a lot longer than we think.

Dr. H. J. aufm Kampe—I have a few questions. First, do you have values of the temperature gradient above 15,000 ft, too? And what is the size of these particles if we assume it is meteoric dust that comes down?

Dr. McDonald—Several microns.

Dr. aufm Kampe—Still a particle of ten microns falls only about one centimeter per second if there is no convective action which brings it down more rapidly.

Dr. McDonald—Yes.

Dr. aufm Kampe—I remember our flights in Germany in connection with Professor

Regener's ozone measurements. Since Regener assumed that ozone is brought down from the ozone layer to lower layers by convection, it was important to know the gradient, that is, the instability or the stability of the layers above your sampling layer.

The next question is when you warmed the icebox to 13°C, was there water on the walls?

Dr. McDonald—No, the water all evaporated.

Dr. aufm Kampe—You mentioned two temperatures, at which you have ice crystals, and these are Rau's 'magic' temperatures. In his experiments he found peaks at −4 and −12°C. He said 'What I need as freezing nucleus is not necessarily an ice crystal but any dry dirt.' Therefore my question about the wet or dry state of your box.

I assumed you had cumulus clouds which formed from thermals. So you had air from the ground and with it you would have dust from the desert at least in Arizona. You mentioned, however that you also sampled other clouds that did not show the effect, and for those one would have to find another explanation.

Dr. McDonald—We took samples in water clouds on the same days and levels and they did not do it. It appears that the only differences was that some clouds had gone through the ice phase.