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SOME EXPERIMENTS ON THE MEASUREMENT OF NATURAL ICE NUCLEI¹

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ABSTRACT

A brief description is given of a new refrigerated expansion chamber apparatus based on an Australian C.S.I.R.O. design for measuring ice nuclei concentrations. The compatibility of results between five instruments of similar construction, a comparison of data obtained with a simple version of the mixing chamber method, and homogeneity of rapid expansion measurements at sites 8 and 100 miles apart are investigated. Except for uncertainties regarding the extrapolation of results to natural clouds, all indications are that, with care, the nucleation level in the atmosphere is capable of objective, compatible measurement with standardized equipment. However, a series of measurements verified the existence of significant differences between the rapid expansion and mixing chamber techniques. Both methods reflected similar trends during appreciable increases or decreases in nucleation activity. A surprisingly uniform geographical distribution of aerosols responsible for ice crystal nucleation is suggested in some of the results.

1. INTRODUCTION

The microphysical processes involved in the water-to-ice transformation in supercooled clouds has received increasing attention in recent years. It is generally accepted that one of the critical factors is the concentration of certain kinds of comparatively rare particles suspended in the atmosphere which serve as nucleating agents. This particular class of particles is variously referred to in the literature as "freezing," "sublimation," "deposition," or "ice" nuclei, depending on the physical mechanisms invoked. The more general nomenclature of "ice nuclei" is used here in view of the current uncertainties as to the exact physical nature of the process.

Since those particles under consideration normally constitute only an extremely minute fraction of the total atmospheric aerosol population, their detection on a routine basis at the present time requires the use of refrigerated devices and the production of supercooled droplets and ice crystals under what must be inferred as

highly artificial circumstances in the light of the scale of events in natural clouds. The a priori assumption is that the concentration of ice crystals produced in a given volume of air is a measure of the number of ice nuclei present which are active at temperatures between some threshold value and the particular cloud-chamber temperature at which the observations are obtained. The temperature dependency is extremely critical and somewhat variable; a 2° to 5° C. change results in roughly an order of magnitude difference in the number of particles activated.

While the basic assumption mentioned above appears valid, it is also an over-simplification in the sense that the ice nucleation process is known or suspected to involve subtle and delicate interactions of such additional parameters as instantaneous supersaturation levels, nuclei size, and droplet volume, along with crystallographic and surface properties of the nuclei. Nor is it yet entirely clear whether these nuclei participate directly in the condensation process, or are activated mainly by coagulation with the exterior surfaces of pre-existing cloud droplets, or possibly by deposition of moisture directly from the vapor state.

¹ These results were obtained in connection with an investigation sponsored jointly by the National Science Foundation (under NSF-GL29) and the U.S. Weather Bureau.

As a consequence of the complex nature of the problem and the present status of instrumentation in the field, it has seemed appropriate to adopt an inquiring attitude on the question of the measurability of ice nuclei concentrations in the free atmosphere. The main purpose of this paper is to summarize the results of a series of experiments designed to partially assess this problem, and incidentally, to describe an instrument which has now been in field service in multiple numbers over a sufficient period of time to be able to report an acceptable level of mechanical integrity and durability.

Although a modest number of ice nuclei measurements has been made in recent years, it has proved exceedingly difficult to evaluate or synthesize the data. Uncertainties exist regarding the compatibility of instruments and measuring techniques, sampling variations in the atmosphere as they reflect on the representativeness of such observations, as well as regarding the knowledgeable interpretation of cold box data as precise and meaningful cloud physics measurements. The absence of an absolute measuring technique requires that judgments of the reliability of such measurements must be, for the time being, based largely on evaluations of the internal consistency of such data.

The results presented here appear to contribute constructive and hitherto missing or sketchy information with respect to the confidence assignable to such measurements beyond a laboratory environment, along with indications of the compatibility of two methods most widely used to date. However, it must be emphasized that a large number of important questions on the interpretation of such measurements remains to be answered.

A discussion of the relationships of these and other daily measurements to meteorological events will be presented later. Some early and rather unexpected experimental results and synoptic clues pointing toward a potentially significant natural source of ice nuclei, utilizing the equipment described here, have been reported by Brier and Kline [1], Kline [2], and Battan and Riley [3]. Most of the measurements discussed in this paper were obtained in and around the Washington, D.C. area in conjunction with an observational program now in progress at a number of widely separated locations during selected periods with the assistance of a number of cooperating organizations. Although these data reflect the results of an evaluation of two particular techniques, the much broader objective of assessing the reliability of such measurements in the free atmosphere strongly motivated this study.

2. EQUIPMENT

An external view of one of the identically constructed instruments used in this investigation is shown in figure 1. The equipment is an adaptation of the basic design described by Warner [4]. The essential components enclosed in the console consist of: (1) a 10-liter insulated copper cylinder with the wall and bottom external surfaces

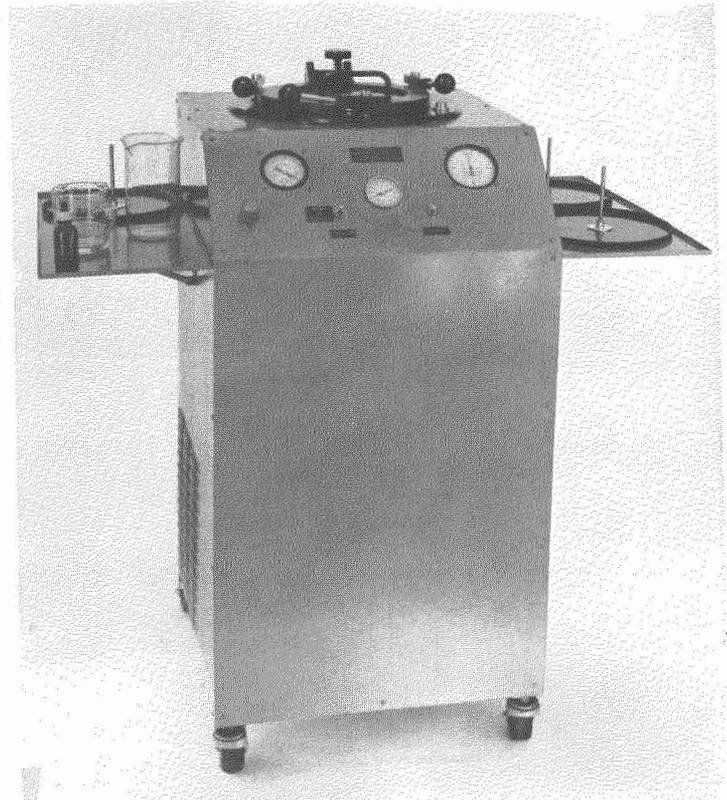


FIGURE 1.—Refrigerated expansion cloud chamber (Experimental Bigg-Warner Type) developed by U.S. Weather Bureau as a modified version of an original design by the Australian Commonwealth Scientific and Industrial Research Organization.

surrounded by copper tubing functioning as the evaporator in the refrigeration system, (2) a 1/3 h.p. hermetically sealed Freon 22 (b.p. -40.5° C.) compressor unit with a fan-cooled condenser, and (3) a 1/4 h.p. rotary air pump of 200 c.f.m. maximum capacity for pressurization of the chamber. Orifices flush with the inside chamber wall are vented to a sensitive differential pressure gage (modified air speed indicator) and the air pump. The latter is connected to a standard rubber air hose at the rear of the console. The dial of the air speed indicator is calibrated to indicate a temperature drop on expansion equivalent to the theoretical wet adiabatic value. The wall temperature indicator is a conventional dial-type bi-metallic thermometer in thermal contact with the exterior of the copper chamber about 1/3 of the distance from the bottom to the top of the chamber. The air seal is maintained between the Plexiglas lid and the chamber by adjustable clamps and a Neoprene O-ring. Decompression resulting in supercooled fog formation is accomplished by opening a spring-loaded valve in the lid after precooling of each air sample. Check and relief valves are installed in the air intake system to retain pressure between the pump and chamber and to prevent over-pressurization resulting in damage to the temperature calibrated pressure gage. The expansion

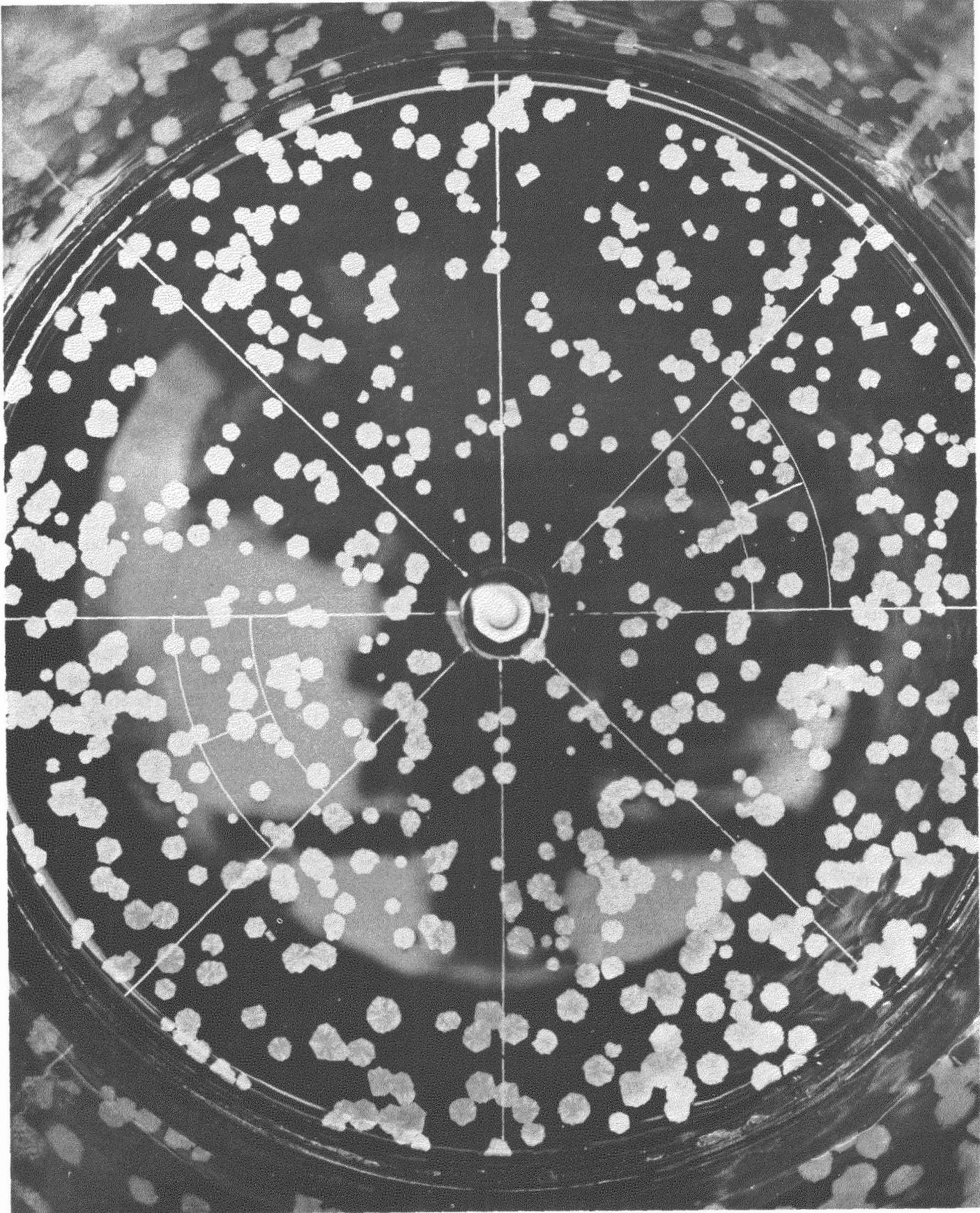


FIGURE 2.—Example of ice crystals produced in a removable tray containing a supercooled sugar solution and placed on the bottom of the cloud chamber. This photograph was obtained with a camera mounted at the top of the chamber.

ratios used in normal operation are approximately 1.08, 1.17, 1.26, and 1.37 for observations at -15°C ., -20°C ., -25°C ., and -30°C ., respectively, depending on the wall temperature.

Special attention was directed to the problem of achieving stable wall temperature control. The system adopted for primary use relies entirely on a manually adjustable automatic expansion valve. This valve serves in practice to meter only a sufficient amount of liquid Freon 22 to the cooling coils to maintain a constant evaporation pressure. Manual adjustment is required to compensate for the heat load imposed by ambient temperature conditions. The pressure differential across the expansion valve is monitored by reference to a vacuum pressure gage located on the front panel of the console. The compressor remains in continuous operation, with this control system thus eliminating the temperature instability problems inherent in the conventional thermostatically controlled on-and-off cycling methods. Provisions for the latter system are included, however, where the equipment must be used in ambient temperature conditions below which the refrigerant cannot be metered in sufficiently small amounts to prevent damage to the compressor due to back-pressure.

Thermocouple surveys on two units operated at the normal wall temperature of -10°C . indicated a uniform temperature distribution (within 0.3°C .) over the lower 2/3 of the chamber with an inversion of the order of 10°C . extending through the top 4 inches, depending on the temperature differential between the wall and outside environment. Although introducing some uncertainty on the "effective" sampling volume, a temperature inversion is an essential feature of the expansion technique to provide a reservoir of water vapor for reliable supercooled fog production at low expansion ratios. Measurements of temperature recovery times of the wall and bottom surfaces following purging with air at 23°C . were all within the range of 90 to 130 seconds, depending on the duration of the flushing cycle.

Three particularly attractive features characterize the expansion technique. One is the ease with which the sugar solution method developed by Bigg [5] may be used for objective detection and counting of the ice crystals produced in the entire 10-liter sampling volume, as shown in figure 2. Second is the rapidity of sampling over a spectrum of temperatures through controlled variations in the overpressure; and third, a normal operating wall temperature of -10°C . to -12°C . which considerably reduces the risks of contamination from splintering and fragmentation of frost particles on refrigerated surfaces over those encountered under humid conditions at colder wall temperatures. Glycerin is used as a freezing point depressant through careful coating of the walls of the copper chamber to prevent frost formation. The present version of the equipment was deliberately "over-designed" with respect to compressor capacity to provide both quick cooling capabilities (under 10 minutes from 30°C . to normal

operating levels) and optional use at colder wall temperatures for test with the mixing chamber method. Supercooled cloud temperatures as low as -30°C . can be obtained either by the expansion process or by selective changes in the wall temperature by means of the adjustable refrigeration expansion valve in the case of the mixing chamber method.²

3. PROCEDURES

The approach adopted for this investigation was dictated by considerations involving the intended use of the equipment. Most of the data were obtained during the course of a twice-daily observational program with the instruments used under conditions representative of field operation. In principle, greater precision in some of the results should be achievable in laboratory environment. However, possible influences on the results by different operating procedures, sampling variations, and uncertainties on the nature of the aerosols characterizing those in the free atmosphere were all considered potentially critical factors in evaluating the confidence which could be assigned to such measurements. The equipment was located out-of-doors or in a well ventilated enclosure at all times to assure purging with representative air samples by several rapid up-and-down movements of the tray within the chamber. Flushing with the air pump alone was found unreliable due to particle scavenging and problems associated with non-uniformity of airflow inside the chamber. Replication procedures were used throughout the measuring program. Efforts were made to obtain a minimum of five consecutive runs in the case of measurements confined to a single supercooled-cloud temperature, or a sufficient number of observations at varying temperatures to define a mean temperature-spectrum-concentration curve. A "reference temperature" of -20°C . was adopted for all observations confined to a single value.

The reproducibility of ice nuclei measurements between instruments (designated by manufacturer's serial number in the discussion to follow) was examined by simultaneous operation of two units located side-by-side. One of the instruments (No. 3) was retained as a "reference" unit throughout the test series, the choice being entirely arbitrary. Efforts were made to cover a representative range of ice nuclei concentrations to compare both the mean value and parallelism in the trends. Each unit was frequently operated by different observers with the ice crystal counts being made independently.

An examination of the areal representativeness of expansion measurements was made in connection with twice daily measurements at two fixed sites. One instrument was located on the roof of an office building approximately 130 feet above the street level in downtown Washington, D.C. A second unit was operated in a suburban resi-

² The term "mixing chamber method" is usually applied to techniques in which the wall temperature is maintained at a quasi-steady value. The air samples are allowed gradually to reach thermal equilibrium with the refrigerated surfaces. A supercooled cloud is usually produced by introducing moisture into the chamber from an external source.

dential section about 8 miles west of the metropolitan area. The downtown observations were normally made during the morning between 0900 and 1030 EST and during the afternoon between 1500 and 1630 EST. The normal observation times for the second series were 0530 to 0700 and 1830 to 2000 EST. A subsequent more limited comparative series was obtained between downtown Washington, D.C. and the U.S. Coast Guard Indian River Inlet Lifeboat Station located approximately 100 miles due east of Washington. These observations were performed at predetermined schedules during mid-forenoon and mid-afternoon periods and included measurements over a spectrum of temperatures in all instances.

In the case of the comparative runs between the expansion and mixing chamber techniques, the following procedure was adopted: A series of five expansion runs at -20°C . with different air samples was made with the wall temperature maintained at -10°C . ($\pm 0.5^{\circ}\text{C}$). Immediately following the expansion series, the wall temperature was cooled to -20°C . for a similar number of measurements with the mixing chamber method. Two cloud chambers were used for the latter series. One was operated at a temperature of -5°C . for precooling the sugar solution, thus permitting nearly instantaneous response to any ice crystals produced in the second chamber after purging and insertion of the cooled tray. Counts with the latter method were made at 1- and 2-minute intervals. Two minutes represented about the maximum upper limit practicable with this method due to parasitic wall frost problems, unreliable fog duration over longer periods, and merging of the growing crystals. Each complete set of sequential measurements in this test series spanned periods ranging from 60 to 120 minutes, depending on difficulties with contaminating frost. These experiments were conducted under summer temperature and humidity conditions such that a supercooled fog formed spontaneously in each fresh air sample drawn into the chamber when operated at -20°C . for the mixing chamber measurements, thus eliminating a need for the exhalation method or other more elaborate procedures for producing supercooled droplets. The sugar solution used at -10°C . consisted of equal parts of water and sugar by weight plus a wetting agent to reduce surface tension to provide uniform coverage of the trays. This mixture was modified to a slightly more concentrated strength to delay excessive supercooling resulting in spontaneous freezing of the solution and overlapping of the ice crystals which would otherwise grow too rapidly at the colder temperature used in the mixing chamber method. The severity of the frost problem at the operating temperature of -20°C . was such that it was usually necessary to reapply glycerin to the wall surfaces after each run with the mixing chamber method. Except for those instances where the counts were obviously contaminated by wall frost, all data were used. Because of the extraordinary tediousness of the mixing chamber method under humid

environmental conditions, most of these data were obtained by the same observer. The detection of ice crystals by the sugar solution technique in both methods was considered preferable to the "scintillation technique" usually used with mixing chamber method, in order to reduce sampling uncertainties.

4. RESULTS

(a) *Compatibility between instruments.*—A total of 89 sets of simultaneous runs using 5 instruments in pairs was performed on 61 days. Although a majority of the runs were confined to replicated observations at -20°C ., a few series included temperature-spectrum measurements. Four representative examples of the latter runs with two units are shown in figure 3. A graphical presentation of all series obtained at -20°C . is shown in figure 4 to indicate the relationship between the readings of the "reference" unit and each of the other units tested. Of these instruments, units No. 4 and 5 displayed the widest difference with a mean ratio with respect to the reference unit of 1.5, a statistically significant departure from unity. Unit No. 6 displayed values in somewhat closer agreement, the average ratios of all the series with this pair being 1.3. The comparative series with unit No. 1 gave a ratio of 0.8, a statistically significant discrepancy. Correlation coefficients between the paired series ranged from 0.95 to 0.99. The differences between units were therefore assumed to reflect a systematic bias, as is obvious from inspection of figure 4.

(b) *Comparison of rapid expansion and mixing chamber methods.*—This test series included 142 sets of observations on 74 days. The results corresponding to the 2-minute intervals are shown as a scatter diagram in figure 5 with each data point representing the average of the counts obtained in each sequential set of observations. The mathematically fitted regression has significant curvilinearity and indicates that both methods tend to give about the same average number of ice crystals only at higher concentration levels. However, the rare occurrence of extreme concentrations and counting difficulties introduces some degree of uncertainty in the location of the regression curve in this area. The correlation coefficient between the rapid expansion counts and the 2-minute mixing chamber values was 0.77. Overall, 2-minute mixing chamber counts at -20°C . were 6.8 times the rapid expansion values, but the ratios varied appreciably over the concentration range as shown by the regression curve in figure 5. An examination of the trends in mean values between consecutive sets of observations revealed that when the rapid expansion counts increased or decreased by a factor of 2 (70 cases), the trends in the mixing chamber measurements were in the same direction in 83 percent of the cases. Changes by a factor of 4 or greater (22 cases) resulted in both techniques reflecting parallel changes in all instances. In an analysis of variance, both techniques showed highly significant differences between the means of sets of obser-

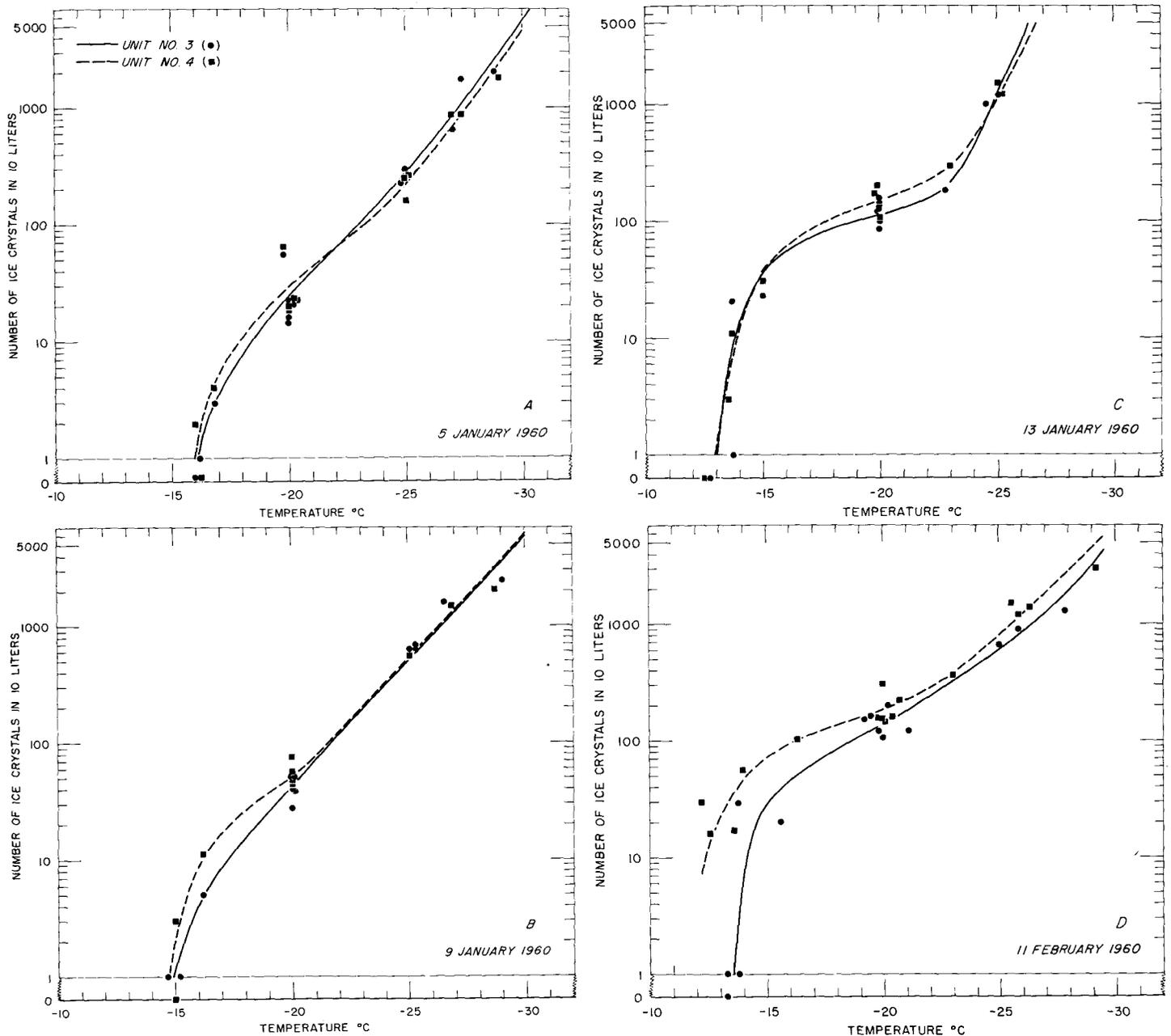


FIGURE 3.—Examples of nuclei temperature-spectrum concentrations obtained during simultaneous operation of two expansion cloud chambers. (a) January 5, 1960; (b) January 9, 1960; (c) January 13, 1960; (d) February 11, 1960.

variations, indicating that the variations of nuclei counts with time are real and much larger than the experimental "error."

If the counts obtained by the rapid expansion method are compared with the 1-minute mixing chamber counts (not given here) a correlation of 0.75 results, which is not much different than the correlation of 0.77 between the expansion chamber counts and the 2-minute interval counts. The correlation between the 1-minute interval counts and 2-minute interval counts was 0.84. This indicates that a large portion ($0.84^2=71$ percent) of the variation in the counts at 2-minute intervals can be ex-

plained in terms of values at 1 minute. When the counts were relatively high at the end of the 2-minute intervals there was a slight but statistically significant tendency ($r=0.23$) for a higher percentage of the crystals to appear during the first minute.

Another series of measurements data with a different instrument made over a spectrum of temperatures during low concentrations on six different days is shown in figure 6. These data were obtained by a continuous fog generation method using a 15-watt light bulb surrounded by a moist fabric and suspended in the center of the chamber, a method described by Soulage [6]. The sugar solution

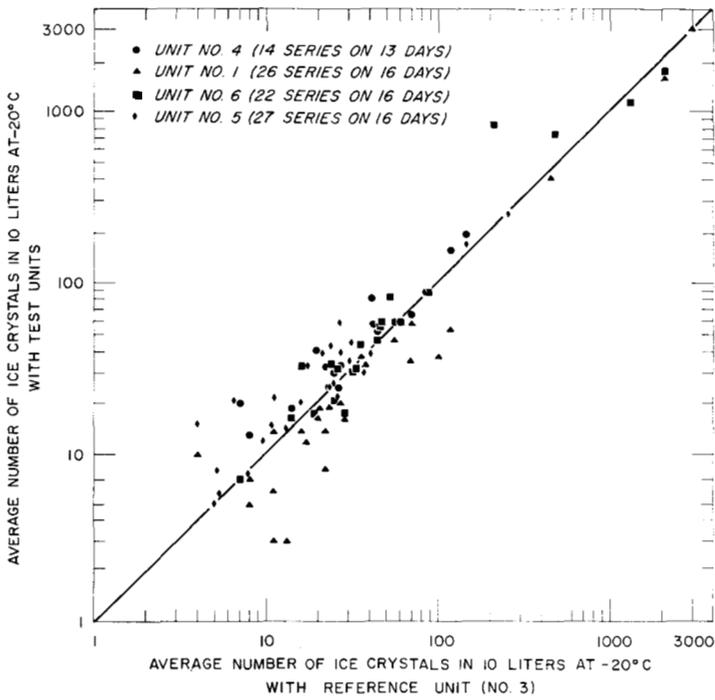


FIGURE 4.—Results of simultaneous expansion measurements at -20°C . obtained with five units, one of which was used throughout the tests as a “control” or “reference” instrument.

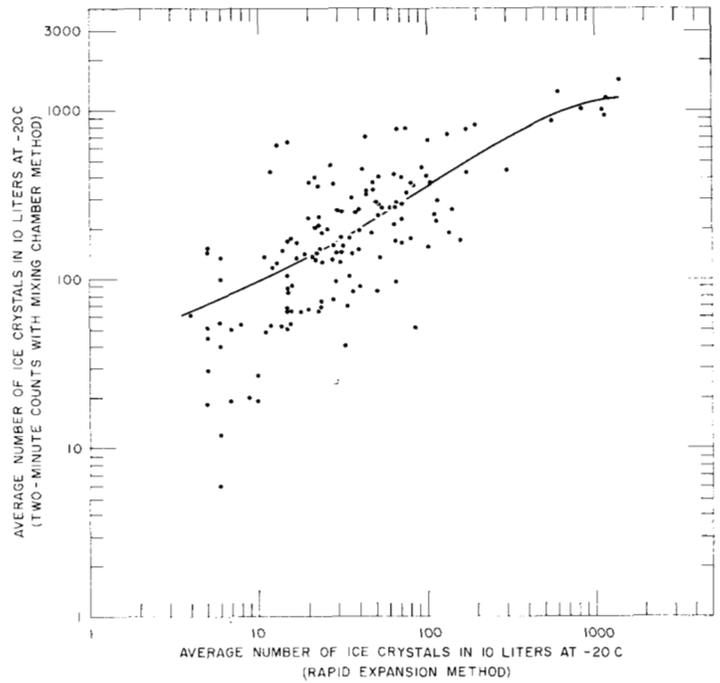


FIGURE 5.—Results of sequential runs using the rapid expansion and mixing chamber techniques at -20°C . Each data point is the average of at least five consecutive samples with each method.

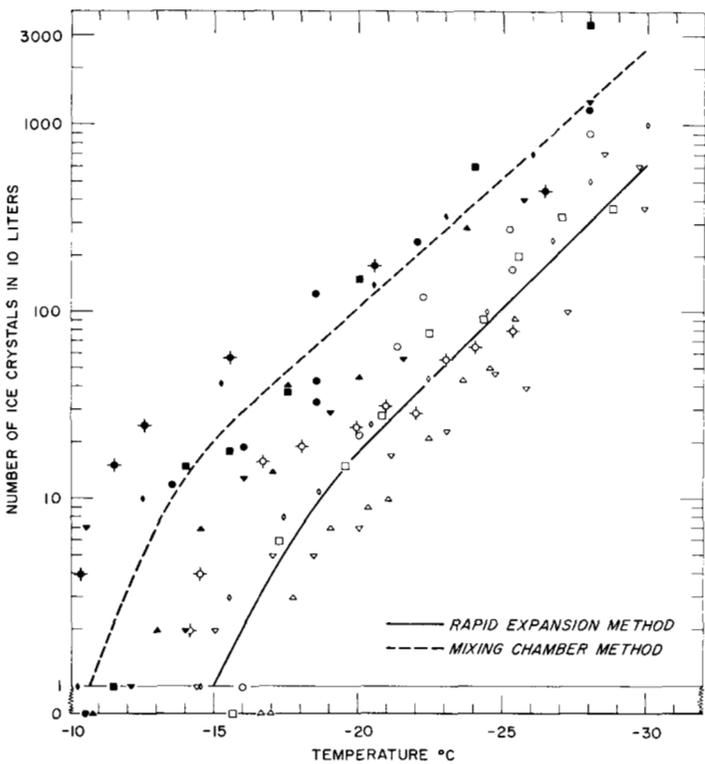


FIGURE 6.—Comparison of rapid expansion and mixing chamber measurements over a spectrum of temperatures on six different days. Data points applicable to the same series are indicated by similar geometric configurations.

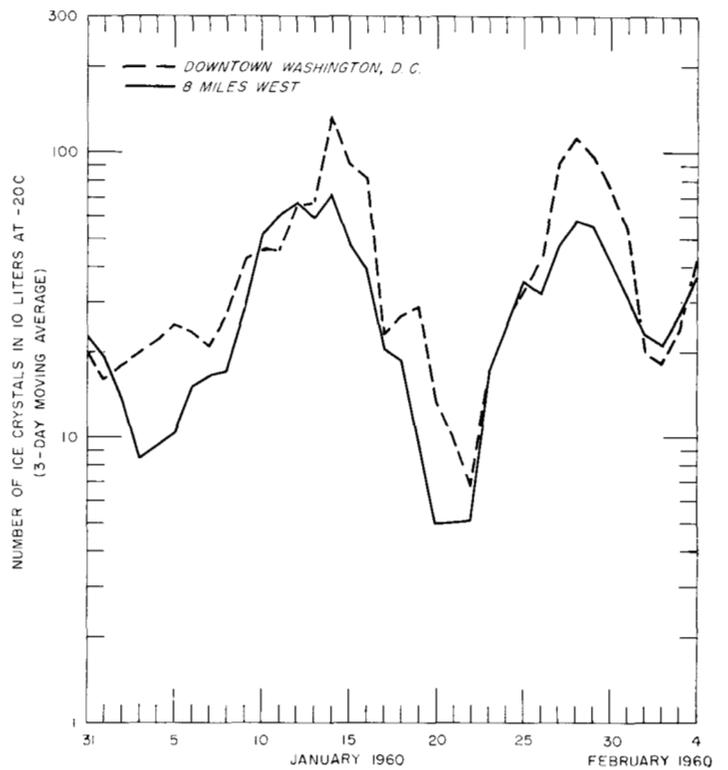


FIGURE 7.—Comparison of ice nuclei measurements at two sites separated by approximately 8 miles.

was diluted with increasing amounts of glycerin to forestall spontaneous freezing at colder temperatures. Counts were delayed until no additional crystals were observed to form. The results at -20° C. are considered in reasonable agreement with those in figure 5 (mean ratio of 5.7 compared with 6.5 for 2-minute values in the same concentration ranges), but indicate that the ratio also varies with the "reference" or fog temperature. An adjustment of the rapid expansion mean temperature-spectrum curve toward 4° to 5° C. warmer values would appear to bring the measurements into fair agreement. In the concentration range surveyed these results compare favorably with the conclusions reached from limited series of tests with several kinds of instruments as summarized by MacCready, Smith, Todd, and Beesmer [7].

(c) *Areal representativeness of ice nuclei measurements.*—The more extensive series of observations performed to evaluate the areal representativeness of ice nuclei measurements was conducted during January and early February 1960. Although considerable differences in absolute values occurred on a few days, the overall trends were similar, as indicated in figure 7, in which the data are presented as 3-day moving averages centered about the middle day. There was an apparent tendency for the highest counts to occur in the metropolitan area, the mean monthly values differing by a factor of 1.65. However, it can be shown that this is not a real but an instrumental effect. The units used for this series (Nos. 1 and 4) have not been operated side-by-side, but both were compared with unit No. 3, as shown in figure 4. When differences in the characteristics of the instruments are allowed for, the mean monthly values at the two sites separated by 8 miles differ by a factor of 1.26, with the lowest counts in the metropolitan area. The correlation coefficients in daily mean values and the 3-day moving averages shown in figure 7 were 0.71 and 0.85 respectively.

A further examination of the spatial compatibility of ice nuclei measurements was provided by a limited series of observations at the U.S. Coast Guard Indian River Inlet Lifeboat Station near the Atlantic shoreline 100 miles east of Washington, D.C. during March 1960. These data were obtained primarily for another purpose to be reported later. However, this series was coordinated with those obtained in downtown Washington, D.C., and included 18 simultaneous sets of data at both sites over the entire temperature spectrum during similar flow regimes on 13 days. These data are summarized in table 1.

TABLE 1.—Comparison of mean values of simultaneous ice nuclei observations during similar flow regimes

| | Threshold temp. ($^{\circ}$ C.) | No. of ice crystals per 10 liters | | |
|----------------------|----------------------------------|-----------------------------------|------------------|--------------------|
| | | -20° C. | -25° C. | -30° C. |
| Shoreline..... | -17.7 | 6 | 76 | 480 |
| Washington, D.C..... | -17.3 | 9 | 119 | ¹ 3,000 |

¹ Estimated.

The two units used in this series were Nos. 3 and 4. The latter was used at the Washington, D.C. site, and, as shown in figures 3 and 4, tended to indicate the higher values. When the "correction factor" of 1.5 is applied to the shoreline data it is apparent that the counts were similar in the warmer ranges of the temperature-spectrum concentrations. Unless these results are fortuitous, which seems unlikely from close inspection of the data, this indicates a surprising uniformity in the dispersion of the responsible particles. At the colder end of the spectrum, however, the discrepancy of a factor of more than 6 at -30° C. is believed considerably larger than the probable experimental error. This suggests that observations in the colder ranges may be particularly influenced by pollution from urbanized regions.

5. DISCUSSION OF RESULTS

In assessing the outcome of the experiments reported here, it is perhaps germane to point out that in some respects these tests were rather severe. The measurements were under environmental conditions representative of field usage. By design, several observers participated in the effort and many of the observations were made during meteorological conditions in which the reliable use of refrigerated devices is awkward and tedious to an extreme. Moreover, the fact that the tests were conducted in a densely populated area with the ever-present risks of contamination from domestic and other local sources conceivably introduced greater variations than might exist elsewhere.

Judgements on the quality of agreement between instruments and techniques depend, of course, on the objectives of any measuring program. Day-to-day fluctuations in ice nuclei concentrations may span up to several orders of magnitude with factors of 5 or greater not uncommon within individual sampling periods. Close inspection of the rapid expansion data indicates that fluctuations in time of the order of a factor of 2 or more in the *mean* values of replicated series were detected by all the instruments. The absolute values appear *capable* of measurement to within at least 25 percent on the average. Differences between instruments (and observers) of the order of 50 percent with respect to the "reference" unit and up to a factor of about 2 overall are indicated, however, without special attention to precise calibration of the equipment.

The results of the comparative runs between the rapid expansion and mixing chamber methods raise some potentially crucial questions on the measurability problem. Some, perhaps most, of the scatter in the results is attributable to uncontrolled factors, particularly with the latter technique. These include not only fluctuations in the aerosol population over the sampling period, but also non-uniformity in supercooled cloud density in the chamber from day to day, and small differences in the strength of the sugar solution influencing the growth rate of the ice crystals, along with the previously mentioned

problems with wall frost. Moreover, although the mixing chamber technique is sometimes referred to as a "constant temperature" method, this is far from the case in practice due to the instantaneous fog formation with the flushing of the chamber and the time required for the temperature of fresh air samples to reach that of the wall. This extends over an appreciable part of the 2-minute period in which ice crystal formation was kept under surveillance and varies depending on the temperature differential between ambient and operating levels. The mean discrepancies between the two methods, however, appear genuine. A satisfying physical interpretation of these differences is exceedingly difficult at this stage because of the elusive and contradictory nature of the evidence concerning the basic physical-chemical processes involved in nucleation phenomena.

Aside from subtle properties of the nuclei themselves, such factors as supercooled droplet size and the time dependency of the nucleation process are probable influences. The recent work reported by Hoffer [8] points to significant influences by soluble substances in depressing the freezing temperature of droplets. In general, each of these factors would operate in the direction of reducing the number of ice crystals observed in the rapid expansion method because of the smaller droplet sizes and extremely short time-intervals at which minimum temperatures are maintained compared with those characteristic of mixing chamber or similar techniques.

We are inclined to interpret our evidence as providing empirical confirmation that either of the techniques examined may be used for studying relative variations in concentration levels. The rapid expansion method was much simpler to use in practice under the prevailing experimental conditions. However, some rather elaborate variant of the mixing chamber method such as described by Bigg and Meade [9] may be necessary to place such measurements on a more physically realistic basis. The alternative would be to calibrate the expansion method against an agreed-upon standard. Our evidence on the suitability of this approach is not entirely clear. The data in figure 6 offer considerable promise in this regard. However, the convergence tendency in the higher concentration ranges noted in connection with the results shown in figure 5 hints that a lack of stability in any such calibration may be expected. The reason for this is not obvious, but the possibility exists that day-to-day differences with respect to the nature of the nucleating particles and their response to the cloud chamber characteristics may be critical factors. A similar trend has been reported by Soulage [10].

There is, of course, the basic question at this point of whether the highly artificial conditions (especially supersaturation levels) in any refrigerated devices of reasonable dimensions provide measurements directly relevant to natural cloud events. Practically no reliable information is available on this point because of the inadequacy of existing instrumentation for measuring either nuclei or ice

crystal concentrations in natural clouds. Moreover, qualitative observations indicate that natural cloud nucleation may often be facilitated by the sedimentation of pre-existing ice crystals from higher cloud formations into the supercooled portions of a cloud system and possibly also by the splintering and fragmentation of freezing particles. Natural nucleation under these circumstances would be more efficient than inferred from "cold box" measurements.

There remains the difficult problem of choosing a standard reference temperature in any compatible measuring system, since the mixing chamber and similar methods are not readily adaptable to both frequent sampling and temperature spectrum measurements. We arbitrarily adopted -20°C . when runs were confined to a single temperature. However, as shown in several examples by Kline [2] the mean temperature spectrum curves often undergo wide excursions in slope and shape so that measurements at any given temperature do not necessarily reflect conditions at other values. That is, high concentrations at -20°C . do not always indicate abnormal values at say -15°C . and warmer values. There is, however, a statistical tendency in this direction, and -20°C . appears to be an adequate compromise for some purposes.

6. CONCLUDING REMARKS

These results have been presented as a contribution toward placing some of the measuring problems in better, though still incomplete, perspective. Our evidence is that there are substantial grounds for believing that compatible and representative measurements can be achieved with carefully designed instruments and with attention to procedural quality control. At the same time, these results emphasize the existence of important instrumental and sampling problems requiring consideration in the design of future equipment and standardization of measuring techniques. That the natural aerosols responsible for ice crystal nucleation are other than of purely local origin and may be widely and fairly uniformly distributed at any given time, at least in the region and under the synoptic conditions surveyed, is strongly hinted. This would appear to have interesting implications in the cloud physics field meriting further investigation.

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