

## EVIDENCE OF GEOGRAPHICAL DIFFERENCES IN ICE NUCLEI CONCENTRATIONS<sup>1</sup>

DWIGHT B. KLINE

U.S. Weather Bureau, Washington, D.C.

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### ABSTRACT

Concentrations of ice nuclei detected with refrigerated devices vary with observational techniques and may be influenced by small differences in the thermal compatibility of otherwise similar instruments. To provide improved homogeneity in the observations obtained for this investigation, all mechanically identical equipment was calibrated against a "reference" instrument. The median concentration levels of the standardized measurements at 15 observational sites spanned approximately 2 orders of magnitude at  $-20^{\circ}\text{C}$ . Data obtained at the 11,150-ft. elevation of Mauna Loa Observatory, Hawaii, clearly revealed low values compared with all other locations. It may be inferred from this evidence that terrestrial aerosols contribute dominantly to the ice nuclei budget of the lower atmosphere, at least as detected by the expansion method under conditions surveyed in the United States.

### 1. INTRODUCTION

The hypothesis that there might be sources of cloud nucleating agents and modulating influences on rainfall from extraterrestrial factors such as meteoritic dust, as proposed by Bowen [4], has been neither generally accepted nor clearly disproved, although the recent paper by Volz and Goody [19] contributes important information in this regard. Other recent papers by Bigg [3] and Rosinski and Pierrard [16] suggest modifications to the original hypothesis which continue to raise provocative issues in need of clarification. Investigations such as those reported by Brier [7] and O'Mahoney [14] tend to lend support to the reality of a singularity component in rainfall as proposed by Bowen. A similar phenomenon has been reported with respect to daily fluctuations in ice nuclei measurements during the month of January at widely separated locations based, for instance, on evidence presented by Bowen [5], Kline and Brier [10], and Carte and Mossop [8]. Before one can conclude that any parallelism between the two phenomena can be interpreted as a reliable clue of a direct physical linkage, it would appear desirable to demonstrate that ice nuclei data used for the purpose of examining the singularity hypothesis in the context of extraterrestrial considerations are relatively independent of regional or local sources of nucleating agents. Otherwise any association may be a reflection of more subtle and indirect consequences of such factors as air mass influences and atmospheric circulation or air trajectory considerations of the type concluded by Georgii [9], Rau [15], Kline [11], and Battan and Riley [1].

The literature indicates considerable diversity of evidence regarding this vexing problem. The practical dilemma posed in assessing the interpretation of ice nuclei measurements (in addition to the fact that they are an indirect and possibly somewhat synthetic type of data relative to natural cloud physics processes) is that anomalies may in effect be "generated" by storm situations. That is, the geometry of changing flow regimes in migratory atmospheric disturbances may bring about a redistribution of terrestrial particles in such a manner as to show a correlation with large-scale precipitation. Intuitively, one might expect this, particularly at sites where the primary sources of the nuclei may be in other than the prevailing wind direction. There is synoptic-type evidence that this in fact is the most proximate explanation of the apparent validation of the January singularities in ice nuclei anomalies in the Washington, D.C. area as described by Kline and Brier [10] and Kline [11]. This implies that there could be a confounding of results or a possible misinterpretation of connecting mechanisms caused by a singularity phenomenon in large-scale circulation of the type noted by Brier [6].

This paper summarizes evidence on a poorly defined aspect of the problem derived from a cooperative ice nuclei observational program at 15 locations in the United States during the period from 1959 to 1962. The results bear on the specific question of whether gross spatial or regional differences in such data are in fact identifiable using mechanically similar and "calibrated" instruments. No attempt is made to offer evidence on either the physical nature of ice nuclei, or the relevancy of these kinds of measurements to natural cloud nucleating

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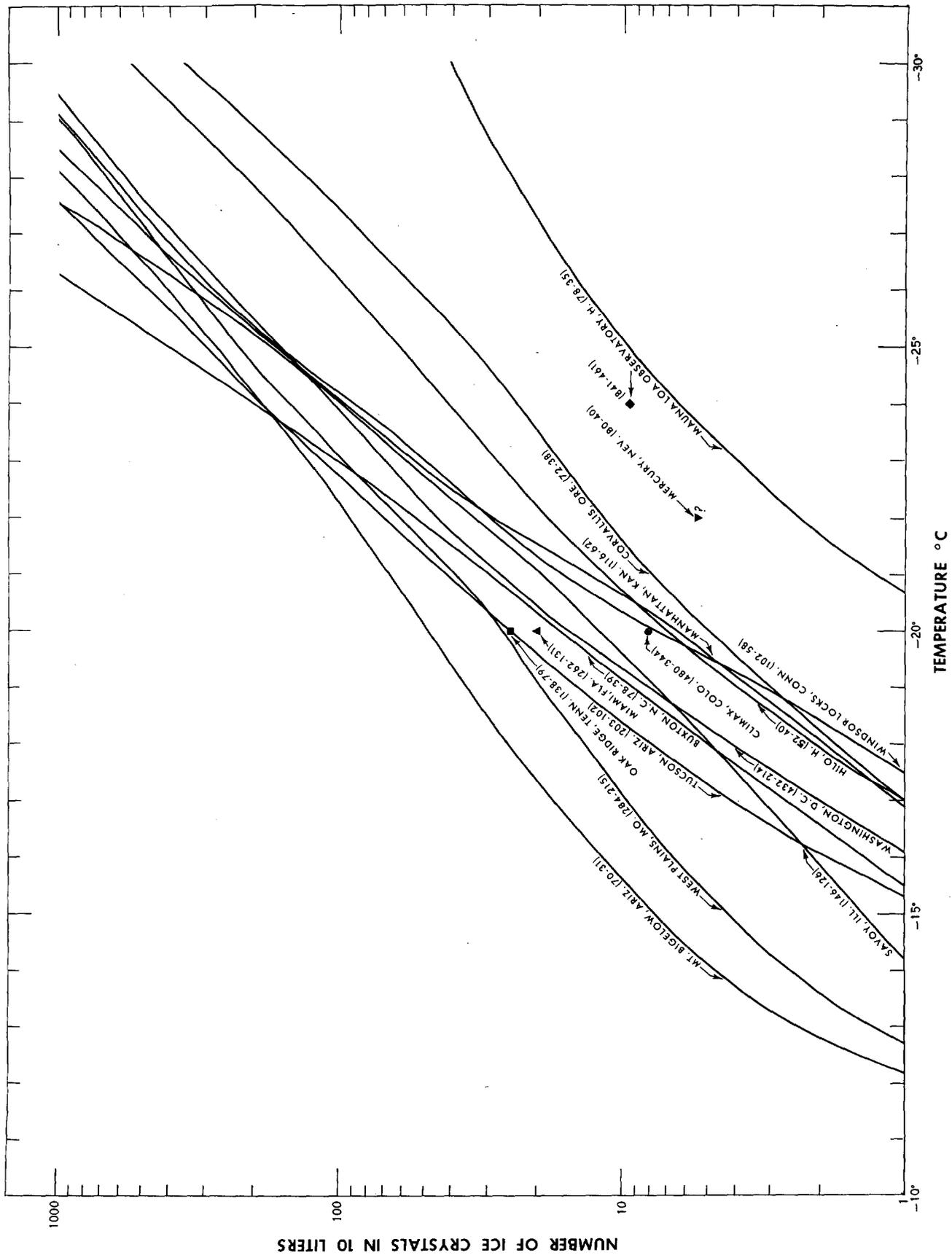


FIGURE 1.—Median standardized ice nuclei concentrations at 15 sites based on the number of observational series on the number of days shown respectively in the parenthesis following the location designator. Those sites indicated by individual points conducted routine measurements only at the specified temperatures. Both types of data are shown for Mauna Loa Observatory. The abscissa values are the temperature after expansion in the sampling chamber.

TABLE 1.—Summary of all ice nuclei measurements corrected to reference instrument at  $-20^{\circ}$  C. except as indicated

Station	No. of observational series	No. of days	Number of ice crystals per 10 liters			Primary periods of measurement
			Average	Median	Range	
Buxton (Cape Hatteras), N. C.	78	39	29	17	3-160	12/29/60- 2/ 5/61
Climax (High Altitude Obs.), Colo.	480	344	17	8	0-450	11/23/59- 2/18/60 7/10/60-12/18/60 5/ 1 61-12/ 9/61
Corvallis, Oreg.	72	38	6	5	0-32	12/30/60- 2/ 5/61
Hilo, Hawaii	52	40	15	7	0-180	1/ 2/61- 2/10/61 12/ 1/59- 4/ 4/60 7/25/60- 9/ 9/60 12/27/60- 2/ 5/61
Manhattan, Kans.	362	199	10	7	0-130	
Mauna Loa Obs., Hawaii (data at $-24^{\circ}$ C.)	841	461	21	10	0-715	10/15/59- 2/ 4/61
Mercury, Nev. (data at $-22^{\circ}$ C.)	80	40	6	6	0-22	12/28/60- 2/ 5/61
Miami, Fla.	262	131	34	20	1-840	12/ 1/59- 2/29/60 12/28/60- 2/ 5/61
Mt. Bigelow, Ariz.	70	31	72	49	11-335	7/25/60- 9/ 9/60 12/28/60- 2/ 5/61 4/10/61- 5/19/61 9/11/61- 9/29/61
Oak Ridge, Tenn.	138	79	34	25	2-145	12/ 3/59- 2/29/60 7/ 1/60- 8/31/60 12/29/60- 2/ 3/61
Savoy, Ill.	195	162	36	14	0-140	12/ 1/59- 3/14/60 12/27/60- 2/ 5/61 1/11/59- 9/ 4/59 11/ 2/59- 4/ 1/60
Tucson, Ariz.	274	141	56	47	0-490	7/25/60-10/23/60 12/30/61- 2/ 5/62 6/ 2/60- 8/28/60 6/12/61- 8/31/61 6/11/62- 8/31/62
Washington, D.C.	1,144	492	51	22	0->5000	6/ 2/59- 9/ 4/59 12/ 3/59- 2/29/60 7/25/60- 9/ 9/60
West Plains, Mo.	284	215	47	25	3->670	
Windsor Locks, Conn.	343	194	8	5	0-140	

mechanisms, or to assess their detailed linkages with synoptic meteorological events at individual observational sites. Rather, the particular contribution claimed for these results is that an attempt has been made to minimize internal heterogeneity in the data caused by instrumental variations, thus improving the degree of confidence with which any genuine differences could be discriminated. Moreover, and perhaps of most acute interest for eventual clarification of the subject, the data include a series of measurements at an isolated, aerosol-limited site which permit a comparison with those of less remote locations believed more typical of the environments of a majority of such measurements heretofore reported, especially in the Northern Hemisphere.

2. THE DATA

The basic measurements were obtained for this investigation by use of a 10-liter sampling volume and the expansion method described by Warner [20]. The observational program tended to emphasize selected periods of the year. However, in several instances, measurements spanned considerably longer or differing periods to provide information of interest to cooperating research groups who in turn kindly supplied their data for inclusion in this study. A few series of data coincided with experimental cloud seeding projects. Periods or days subject to local contamination by artificial nuclei, as reported by the principal investigators, were excluded from this summary. Where the time limitations pre-

vented measurements over a spectrum of cloud chamber temperatures, routine observations were confined to  $-20^{\circ}$  C. ( $-24^{\circ}$  C. and  $-22^{\circ}$  C. in the case of Mauna Loa Observatory and Mercury, Nev. respectively), and therefore should be assigned the greatest overall reliability in terms of the number of daily observational series as well as the quantity of replicated samples within each series. Likewise, the calibration procedure stressed measurements at  $-20^{\circ}$  C. and the resulting correction factors for each of the instruments relative to the reference instrument can with greatest confidence be applied to counts at this temperature. The means of the ratios of the replicated counts with the "reference" unit to those of the individual instruments ranged from about 0.5 to 2.0, or a variation between the 13 instruments used for this investigation of up to a factor of 4. Each calibration series included from 50 to in excess of 100 paired samples to cover a representative range of natural concentration levels.

It should be noted that standardization of the data to a common instrument does not imply that the measurements are thereby corrected to an absolute reference inasmuch as none exists in this field. The reader is referred to the paper by Kline and Brier [12] for a more complete description of both the measurability problem and the instruments used in the observational program.

3. RESULTS

Since day-to-day fluctuations in ice nuclei concentrations may span several orders of magnitude at any given temperature, medians<sup>2</sup> of the adjusted measurements were selected as most representative of the central tendencies for comparative purposes since some of the observational series were of limited duration and therefore subject to distortion by extreme values. Composite results of the standardized measurements from the 15 observational sites are presented in graphical form in figure 1. Where both spectrum runs and measurements confined to a single temperature were available from a particular station, the curve was derived entirely from the spectrum-type measurements. Single data points on the other hand identify those sites where most of the routine measurements were confined to the indicated abscissa value. The single exception shown is the Mauna Loa Observatory data which included both types of measurements, presented for visual comparison in view of its unique geographical location. Since several stations obtained extensive series with both methods, all usable measurements applicable to the single temperature (including those incorporated in fig. 1) are summarized in table 1 showing

<sup>2</sup> In practice, the concentrations in any individual series of measurements were obtained either from mean temperature spectrum distribution curves or, in the case of data confined to a single temperature, were the averages of the replicated ice crystal counts in the air samples that constituted the individual observational series. These mean values were then adjusted (standardized) to the reference instrument and the medians of all the data obtained at each observational site determined. The observational procedures at Climax, Colo. (High Altitude Observatory), Mt. Bigelow, Ariz., and Mauna Loa Observatory included a temperature correction factor equivalent to moist adiabatic expansion to mean sea level ambient pressure.

average and extreme values in addition to the median concentrations to permit comparison with other published data.

Overall, the results indicate differences in median values of about an order of magnitude for data obtained within the continental United States, and at least a factor of 50 with respect to the Mauna Loa Observatory data. The latter, if extrapolated to  $-20^{\circ}\text{C}$ . would indicate a range of differences approaching a factor of 100 at this temperature.

The data for Mercury, Nev., are indicated as questionable. Although these measurements were obtained at an indicated temperature of  $-20^{\circ}\text{C}$ ., there is evidence that the thermal contact between the thermometer gage and the cloud chamber wall was disrupted during shipment to the site. Thermocouple surveys indicated that a probable correction of  $-2^{\circ}\text{C}$ . should be applied to the data.<sup>3</sup>

#### 4. DISCUSSION AND CONCLUSIONS

While not crucial for the overall conclusions from this study, it should be noted that the observational program from which these results are derived included sampling periods ranging from about 5 weeks to in excess of 15 months. The obvious question arises regarding the representativeness of those data limited to the shorter periods. The Mauna Loa spectrum concentrations obtained during a 35-day period compare reasonably well with the long-period median values found at  $-24^{\circ}\text{C}$ . as shown in figure 1. This would indicate that measurements typical of conditions at a given site might be achieved in a relatively short period of time. However, this conclusion is suspect without further scrutiny. Examination of the three most extensive series of data, for Washington, D.C., the High Altitude Observatory at Climax, Colo., and the complete Mauna Loa results, on a monthly basis for those periods with at least one series of replicated measurements on each day revealed an extreme range in the monthly median values from a factor of 7 at Washington, D.C., to a factor of 8 at the High Altitude Observatory, and a factor of 10 at Mauna Loa. The individual monthly median and average values were within a factor of 2 of the longer-period values in about three-fourths of these monthly series. While not conclusive, the longest-period measurements suggest that approximately 3 months of data are required for a high degree of confidence that the resulting central tendencies will be within a factor of 2 of the "climatological" values. The data are, of course, too limited for an assessment of whether there may be seasonal or annual trends of possible interest.

A comparison of these results with those obtained elsewhere is difficult in view of the problems of reconciling the data from various techniques and instruments. Never-

theless, the summary of world-wide data compiled by Bigg [2] resulted in evidence that there may be appreciable differences between mean concentration levels observed in the Southern Hemisphere and those measured in the Northern Hemisphere, the latter being higher overall by a factor of 5 with the expansion technique. Variations between sites ranged in the extreme from a factor of 20 in the Southern Hemisphere to nearly 70 in the Northern Hemisphere at  $-20^{\circ}\text{C}$ . The measurements summarized here support his evidence with respect to Northern Hemisphere data, about half of which were United States measurements. Bigg also proposed that those sites with mean concentrations in the neighborhood of 4 ice crystals per 10 liters at  $-20^{\circ}\text{C}$ . with the expansion method offered the greatest likelihood of obtaining uncontaminated results. It would appear that, of all the United States sites sampled in this study, only Mauna Loa Observatory unequivocally meets this specification.

This leads to perhaps the most fundamental issue raised by our data. Aircraft surveys of the vertical distribution of ice nuclei as summarized by Smith and Heffernan [17], Smith, Kassander and Twomey [18], and Murgatroyd and Garrod [13] have in general indicated no substantial evidence of decreasing concentrations with elevation as might be expected if terrestrial sources were the major contributors to the ice nuclei budget of the atmosphere. The unmistakably low values found at the isolated high-altitude site of Mauna Loa Observatory, when compared with those at all other stations, may be interpreted as evidence that the concentrations are appreciably less under circumstances where a semi-permanent inversion extends over a broad geographical region and might be expected to serve as an extensive and reliable barrier to the vertical transport of particulate matter over sustained periods of time. The validity of this interpretation is strengthened by comparing the spectrum curves for Hilo, Hawaii and Mauna Loa Observatory as shown in figure 1. Both series of measurements were obtained during January and early February 1961 and show that median concentrations measured at the 11,150-ft. elevation of the Observatory and thus well above the mean altitude of the trade inversion, were approximately an order of magnitude lower than those at sea level. These results would appear to offer substantial evidence that low-level influences contribute dominantly to the ice nuclei budget of the lower atmosphere under the conditions sampled, unless an environmental conditioning or "preactivation" process, such as exposure to humid conditions in the sub-inversion layer, is a factor or the low humidity typical of the Mauna Loa Observatory environment influenced the reliability of the measurements. More detailed information on these and related results obtained in connection with these two coordinated series of measurements will be offered in a subsequent paper in view of their implications to a further assessment of terrestrial versus extraterrestrial sources of ice nuclei.

Finally, it should be noted that the Hilo results obtained

<sup>3</sup> The author is indebted to Mr. A. Don Bourquard of the University of Chicago for performing the laboratory tests and directing attention to evidence of a change in calibration for this instrument which was also used in the West Plains, Mo. measurements. Those portions of the latter data obtained after the change in calibration occurred were similarly adjusted.

near the Hawaiian shoreline show evidence that the association between abnormal ice nuclei concentrations and the influx of air with a maritime trajectory as discussed by Kline [11] and Battan and Riley [1] does not appear to represent an obvious influence in the Pacific environment. These contradictory results suggest appreciable regional differences in need of further investigation.

#### APPENDIX

Because of its implications for improving future measurements in this field, attention is directed to a feature of the instrument compatibility problem which appears to have been overlooked in both laboratory and field observations heretofore reported in the literature and which escaped diagnostic attention in this investigation until the time-consuming calibration phase was well advanced. In the course of operating the various instruments in paired series of replicated runs, for the most part confined to  $-20^{\circ}\text{C}$ ., occasional periods were noted in which seemingly atypical (compared with most series with the same instruments) but nevertheless consistent discrepancies between instruments were indicated. These discrepancies did not appear to be attributable to either operating technique, instrumental variations, or non-uniformity of particle concentrations. While the acute sensitivity of ice nucleation to temperature is universally recognized, it has become apparent that variations between instruments cannot be construed as a monotonic function of the temperature parameter as normally used. Small deviations in the thermal characteristics and temperature gaging systems of otherwise similar devices may at times compromise the compatibility of the data beyond normal expectations due to what amounts to a magnification of the thermal effects by fluctuations in the nuclei temperature spectrum distribution. If the number of crystals increases steeply with decreasing temperature, a small uncertainty in temperature will give a greater uncertainty in number. This is illustrated in figure 2 using two examples from Washington D.C. observations. Whereas a  $1^{\circ}\text{C}$ . variation between instruments in the January 30, 1959 case would have yielded counts differing by a factor of 1.3 at  $-20^{\circ}\text{C}$ ., a discrepancy of 1.8 is indicated in the April 9, 1959 example at the same temperature and concentration.

In retrospect, it therefore appears that a potentially more precise calibration procedure would be to perform the comparative runs for a range of spectrum values to establish a calibration factor in terms of temperature rather than on the basis of the ice crystal counts at a single temperature as used in this investigation. Where routine measurements are to be conducted at a given temperature with multiple instruments, the required correction could then be incorporated in advance into the indicated values for each instrument to provide data compatibility. Alternatively, complete surveys of the thermal characteristics

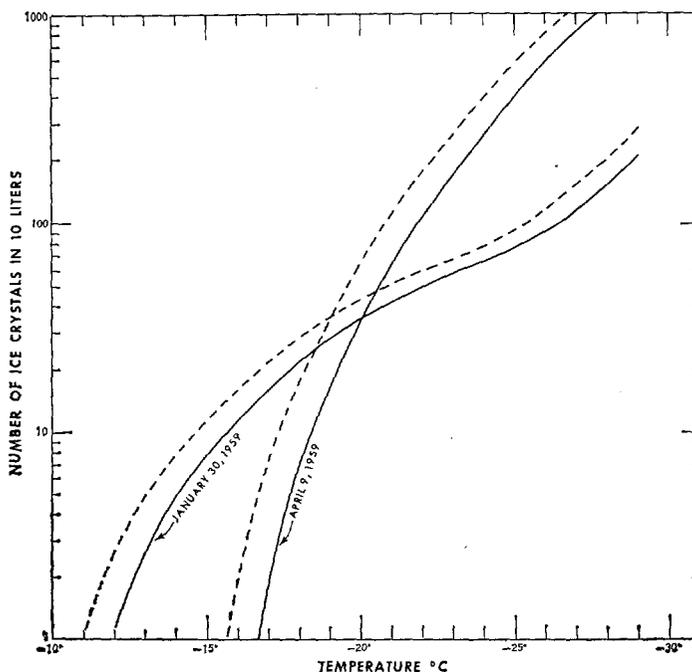


FIGURE 2.—An illustration of the variable consequences of a  $1^{\circ}\text{C}$ . discrepancy in the thermal characteristics between instruments as influenced by differing nuclei spectrum concentrations. The examples are based on two observational series in the Washington, D.C. area selected on the basis of identical indicated concentrations at  $-20^{\circ}\text{C}$ . in the original measurements (solid curves) but differing values at other temperatures. Simulated measurements (dashed curves) for an instrument with a thermal discrepancy were obtained by displacing the original measurements  $1^{\circ}\text{C}$ .

and gage calibrations could be performed on each instrument. However, this approach was abandoned early in our case because of the complexity of identifying and combining all potential sources of differences between instruments, particularly those involving subtleties in the temperature gradients over the refrigerated surfaces. If our experience is representative, precise thermal compatibility is difficult to achieve in instruments of this type requiring rugged design for routine field use.

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#### REFERENCES

1. Louis J. Battan and James J. Riley, "Ice-Crystal Nuclei and Maritime Air," *Journal of Meteorology*, vol. 17, No. 6, Dec. 1960, pp. 675-676.
2. E. K. Bigg, "Summary of Measurements of Ice Nucleus Concentrations," *Bulletin de l'Observatoire du Puy de Dôme*, No. 3, July/Sept. 1960, pp. 89-98.
3. E. K. Bigg, "A Lunar Influence on Ice Nucleus Concentrations," *Nature*, vol. 197, No. 4863, Jan. 12, 1963, pp. 172-173.
4. E. G. Bowen, "The Influence of Meteoritic Dust on Rainfall," *Australian Journal of Physics*, vol. 6, No. 4, Sept. 1953, pp. 490-497.
5. E. G. Bowen, "January Freezing Nucleus Measurements," *Australian Journal of Physics*, vol. 9, No. 4, Sept. 1956, pp. 552-555.
6. G. W. Brier, "A Note on Singularities," *Bulletin of the American Meteorological Society*, vol. 35, No. 8, Oct. 1954, pp. 378-379.
7. G. W. Brier, "A Test of the Reality of Rainfall Singularities," *Journal of Meteorology*, vol. 18, No. 2, Apr. 1961, pp. 242-246.
8. A. E. Carte and S. C. Mossop, "Measurements of the Concentration of Atmospheric Ice Nuclei in Southern Africa, 1956 to 1960," *Bulletin de l'Observatoire du Puy de Dôme*, No. 4, Oct./Dec. 1960, pp. 137-149.
9. H.-W. Georgii, "Bemerkungen zu den 'Januar-Singularitäten' der Konzentration Atmosphärischer Gefrierkerne," *Geofisica Pura e Applicata*, vol. 44, 1959/III, pp. 249-256.
10. Dwight B. Kline and Glenn W. Brier, "A Note on Freezing Nuclei Anomalies," *Monthly Weather Review*, vol. 86, No. 9, Sept. 1958, pp. 329-333.
11. Dwight B. Kline, "Recent Observations of Freezing Nuclei Variations at Ground Level," "Physics of Precipitation," *Geophysical Monograph No. 5*, American Geophysical Union, 1960, pp. 240-246.
12. Dwight B. Kline and Glenn W. Brier, "Some Experiments on the Measurement of Natural Ice Nuclei," *Monthly Weather Review*, vol. 89, No. 7, Aug. 1961, pp. 263-272.
13. R. J. Murgatroyd and M. P. Garrod, "Some Recent Airborne Measurements of Freezing Nuclei over Southern England," *Quarterly Journal of the Royal Meteorological Society*, vol. 83, No. 358, Oct. 1957, pp. 528-533.
14. G. O. O'Mahoney, "Singularities in Daily Rainfall," *Australian Journal of Physics*, vol. 15, No. 3, Sept. 1962, pp. 301-326.
15. von W. Rau, "Die Zeitlichen Änderungen des Atmosphärischen Gefrierkerengehalts und Ihre Beziehungen zu den Meteorströmen und zur Atmosphärischen Zirkulation," *Geofisica Pura e Applicata*, vol. 46, 1960/II, pp. 110-124.
16. J. Rosinski and J. M. Pierrard, "On Bowen's Hypothesis," *Journal of Atmospheric and Terrestrial Physics*, vol. 24, Dec. 1962, pp. 1017-1030.
17. E. J. Smith and K. J. Heffernan, "Airborne Measurements of the Concentration of Natural and Artificial Freezing Nuclei," *Quarterly Journal of the Royal Meteorological Society*, vol. 80, No. 344, Apr. 1954, pp. 182-197.
18. E. J. Smith, A. R. Kassander, and S. Twomey, "Measurements of Natural Freezing Nuclei at High Altitudes," *Nature*, vol. 177, No. 4498, Jan. 14, 1956, pp. 82-83.
19. F. E. Volz and R. M. Goody, "The Intensity of the Twilight and Upper Atmospheric Dust," *Journal of the Atmospheric Sciences*, vol. 19, No. 5, Sept. 1962, pp. 385-406.
20. J. Warner, "An Instrument for the Measurement of Freezing Nucleus Concentration," *Bulletin de l'Observatoire du Puy de Dôme*, No. 2, Apr./June 1957, pp. 33-46.