## The Second International Workshop on Condensation and Ice Nuclei

Organized Under the Auspices of
Subcommittee on Nucleation
International Commission on Cloud Physics
International Association of Meteorology and Atmospheric Physics
International Union of Geodesy and Geophysics
Sponsored by
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Host
Colorado State University
Fort Collins, Colorado


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## DEDICATION

The Proceedings of this Second Workshop on Nucleation are dedicated to the memory of Professor Henri Dessens. The concept of such international workshops was his idea and he was also host to the first workshop in 1967 in his Institut de Recherches Atmospheriques at Lannemezan, France. Professor Dessens' interest ranged from the microphysics of fogs and clouds to the mesophysics of the artificial stimulation of convection and finally to the large scale suppression of hail damage. His ideas initiated and enriched research projects not only in his home country but in many regions of the globe. His personality, colorful and modest at the same time, will be missed but his spirit will live.

Helmut K. Weickmann
President, International Commission on Cloud Physics, IAMAP


HENRI DESSENS

# Report on Workshop Goals, Program, Instruments, Observations, and Preliminary Analyses 

5-19 August 1970

Compiled by:
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This report covers the activities during the second International Workshop on Condensation and Ice Nuclei (IWCIN) held at Colorado State University in Fort Collins, Colorado during August, 1970. A description of Workshop activities, including background, planning, and organization, is included as part of the addresses presented during the opening session by Drs. H. Weickmann, J. Podzimek, and V. Schaefer. Keynote addresses during opening session by Dr. Patrick Squires, on condensation nuclei, and Dr. John Hallett, on ice nuclei, served as a technical starting point for the workshop and are included in this report.

Detailed descriptions of the laboratory, the sampling facilities and the nuclei counters being compared are also included. These descriptions and a listing of nearly all data collected are followed by a section which presents brief, preliminary summaries of analyses of both the condensation and ice nuclei observations. Sections of the report dealing with the laboratory facilities, procedures, equipment, etc. have been prepared by the individuals most concerned. Preliminary analyses of the CCN and IN data have been prepared by selected representatives from participants who made the CCN and IN measurements.

## PREFACE

This description of the program, facilities, equipment, and data collected constitutes the nitial, detailed report of the second International Workshop on Condensation and Ice Nuclei (IWCIN).

Most participants and members of the organizing committee agreed at the end of the workshop that a report of workshop activities and the data should be prepared as soon as possible following its completion. The consensus of the organizing committee was that this report should not be delayed until such time as detailed analyses were completed by individual participants, but instead should serve to provide all workshop data to all participants and other interested researchers. The receipt date of materials from the various contributors, delays in the final tabulation of all data, and the checking by each participant of his own data have made May 1971 the "earliest possible" date for the completion of the report.

It is expected that, when completed, certain analyses of the workshop data will be submitted to technical journals. A compilation of these and other analyses, completed during approximately the next year, is being considered for a final report of the Workshop. Consequently, a second volume workshop report, composed of an abridgement of individual analyses, will be prepared if sufficient papers are available.

Several comments seem warranted, based on the experience gained in compiling this report, since they may be of value in planning future IWCIN workshops. First, the cooperation of all participants, in supplying material for this report has been outstanding. This is noteworthy since contributions from 60 individuals are included. This shows that, from an organizational standpoint, large cooperative efforts to concentrate on specific problems during a workshop are practical. The technical value of the workshop must wait for future evaluation. Two other comments deal with deficiencies in workshop data output from the standpoint of report preparation. The data collection format used was a substantial improvement over the many different forms used by the various participants at the first workshop in Lannemezan in 1967. Further development should be a prerequisite in planning for any subsequent workshops. A format should be developed and reviewed by the various participants well before the data collection procedure gets under way. The one that evolved toward the end of this second IWCIN proved reasonably satisfactory but changes made during the course of the workshop presented very substantial data handling problems. These data handling problems have been accentuated by non-uniformity of reporting, by the various participants, of the formats that were available. Greater care must be exercised by the participants in making data entries more legible and in checking their data before submission.

An additional comment is directed primarily to those who will use data from this report but who did not personally participate in the Workshop. The various participants brought their own equipment. This involved packing, shipping, and recalibration after arrival in Fort Collins. Even in a three week workshop it is difficult with some of the equipment used to be certain of its operation after such a short recalibration. The three weeks allocated to the workshop included calibration of the workshop facilities and the individual instruments, and the data collection phases for both CCN and IN aerosols. Only some three days were available for intercomparisons of ice nuclei counters using natural, NaCl , teflon, and AgI aerosols (produced thermally, from pyrotechnics, and from AgI-NaI and $\mathrm{AgI}-\mathrm{NH}_{4} \mathrm{I}$ solutions). The task for such a short period was compounded since tests with each aerosol were undertaken at various super-saturations for the CCN and at various temperatures for the IN. During certain tests when any participant's equipment was not working to his satisfaction, time generally was not available for him to make adjustments or determine the causes. Consequently, while the quality of the data collection with the respective units is believed to be generally good, in at least some cases, the various counters were probably not operating in an optimum mode.

A last comment relates to the magnetic tapes upon which all workshop data is stored. Copies of these tapes can be made and charged to the recipient. Computer analyses can also be made on the Colorado State University CDC 6400 computer, again the requesting scientist would have to assume computer and programming charges. The analyses required could be described by the requesting scientist and subsequently programmed and processed at Colorado State University. Alternately, a computer ready program could be supplied and processed from the data tape.

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## SECTION I

## SECTION II

## INTRODUCTION

The role of atmospheric nuclei in the formation of both cloud droplets and ice crystals has been considerably clarified during the past decade. Interest and support for research in atmospheric nuclei have been accentuated by (1) expanding evidence that cloud droplet and ice crystal characteristics can have broad influences on the development and precipitating characteristics of clouds and (2) the developing technology to produce artificial nuclei with varying characteristics.

Continued refinement of basic nucleation theory as applied to clouds, and further development of appropriate microphysical models to describe cloud processes, are obviously dependent on both laboratory and field measurements of atmospheric nuclei. Experience has shown that there are many pitfalls in making such observations. Very precise control of environmental conditions in the nuclei counter is required and this must be accomplished in a manner in which the device being used does not itself influence the results. Further, it is now becoming apparent that different nuclei can become effective by different processes, e.g., deposition, freezing, contact. Perhaps, the most stringent limitation in the development and use of ice nuclei counters is the lack of a readily usable "standard" for cal ibrating the instruments. This problem of having a "standard" is emphasized when we consider that in the atmosphere, nucleation takes place under a very wide range of conditions and by various processes. In lieu of a "standard", an intercomparison of a wide variety of nuclei measuring devices can provide a critical base for interpretation of observations and their subsequent use for various analyses. It is within this general framework that the 2nd International Conference on Atmospheric Nuclei was ogranized and conducted.

## OBJECTIVE

The basic objective of the 2nd International Conference Workshop was to study and compare various techniques for observing atmospheric nuclei. Investigation of certain nucleation processes is virtually inseparable from this objective.

The specific goals of the 2nd IWCIN were formulated as:

1) Survey of the state of the art in the field of the measurement of cloud condensation nuclei (CCN) and of ice nuclei (IN).
2) Compare the operating characteristics of various types of CCN and IN counters by measuring the activation characteristics of well defined ("in situ" aerosol size distributions measured by the Minnesota Aerosol Analyzing System) distributions of CCN and IN to determine:
a) Reproducibility of the measurements using the same apparatus under the same conditions.
b) Humidity activation characteristics in various types of CCN counters (at $S=0.3,0.75$, 1.0).
c) Temperature activation characteristic in various types in IN counters (at $T=-8$ to $-26^{\circ} \mathrm{C}$ ).
d) Effect of particle size on IN counters (and possibly on CCN counters) using well-defined size spectra of nuclei such as AgI with mean radii of 0.01 , 0.03 , and 0.10 microns.
3) Familiarize the participants with the methodology and the work of their colleagues.
4) Provide for discussion, in an informal way, of some important problems related to the function of IN and CCN in the clouds and of the principles of measuring their physical-chemical characteristics.

## SECTION III

Wel come
by
Dr. Charles 0. Neidt

The workshop participants were cordially welcomed to Colorado State University by Dr. Charles 0. Neidt, Academic Vice-President for Colorado State University.

## Address

by
Helmut K. Weickmann
Following the initiative of Professor H. Dessens, the distinguished member of the International Commission on Cloud Physics, to convene a Workshop on condensation and ice nuclei in 1967 in Lannemezan, France, the second Workshop was planned and held on the campus of the Colorado State University, Fort Collins, Colorado, U.S.A., August 3 to 22, 1970. The Workshop was hosted by the Department of Atmospheric Sciences of Colorado State University. The Workshop would not have been possible without the support of the National Science Foundation, whose interest is gratefully acknowledged.

The Workshop was planned and organized under the enthusiastic and able leadership of Professor R. Steele, now with Desert Research Institute, jointly with his organizing committee. Credit for the numerous details during the final arrangements, construction of laboratory facilities, housing and entertainment of the participants is due to the unceasing energy and devotion of Professor L. Grant, Dr. U. Katz, Dr. J. Podzimek, Chairman Subcommittee, and their team of able colleagues.

The experiences gained during the first Workshop were a most welcome asset for planning the second Workshop as will be apparent when the reader studies the subsequent report. The most important improvement was the cooperation of Dr. K. Whitby, University of Minnesota, and his co-workers for the determination of the nuclei spectra.

The report makes clear the great complexity of the nuclei measurements, which indeed is not the straightforward measurement of a particle but the measurement of a particle which has initiated a process of phase transition, or in the words of $A$. Wegener we measure the "ausloesende Wirkung" (catalytic effectiveness) of a particle. While the Workshop dealt with the measurement of Aitken, cloud condensation and ice nuclei, the overriding concern was the measurement of ice nuclei. Ice formation requires either the detour via a process of condensation or the introduction of the nucleus into a subcooled water droplet. Considering that at a temperature of $-10^{\circ} \mathrm{C}$ one freezing nucleus is outnumbered by factors of $10^{7}$ to $10^{9}$ by Aitken nuclei and $10^{6}$ to $10^{7}$ by cloud
condensation nuclei, then it is obvious that we are dealing here with some of the most difficult measurements of trace constituents. Conventionally one measures therefore at a temperature of $-20^{\circ} \mathrm{C}$ because there the misrelationship is about 3 orders of magnitude smaller. In addition one either takes vessels with a great volume of 10 to 20 liters or one collects the nuclei with millipore filters through which great air volumes can be sampled. An additional difficulty for the measurements of freezing nuclei is that we do not know exactly how nature inside clouds forms ice crystals. Essentially there are four ice forming processes:

## vapol 1. Homogeneous ice formation directly from the water phase.

2. Heteorogeneous ice formation on a surface from the vapor phase (sublimation nucleus).

## 3. Homogeneous formation directly from the liquid

 phase.4. Heteorogeneous formation on a surface from the liquid phase (freezing nucleus).

It appears nowadays that the sublimation nucleus process does not occur in nature. Homogeneous nucleation from the gas phase can also be excluded but homogeneous nucleation from the liquid is possible for instance in cumulonimbus anvils, when the cloud is being carried through the $-40^{\circ} \mathrm{C}$ level. Ice will form primarily through processes involving freezing nuclei. To accomplish this the freezing nucleus a) may already be contained in the droplet and starts crystallization when its threshold temperature is reached; b) may act through contact with a supercooled droplet, or c) may act at first as a condensation nucleus and then immediately as a freezing nucleus. Here it is possible that through capillary action condensation already begins before water saturation is reached, or that the nucleus is so small that supersaturation would be necessary for condensation to occur and that therefore the only chance for that nucleus to act is to come into contact with a supercooled water droplet.

It is conceivable that various methods for measuring freezing nuclei concentrations respond differently to this spectrum of freezing nuclei and that we therefore cannot expect all methods to give the same result.

With this reservation in mind we would like to assess the main methods currently used for ice nuclei measurements:

## 1. Bigg-Warner Expansion Chamber Method

The basic principle is that of the Aitken counter; a moist air volume is adiabatically expanded which causes a temperature drop to $-20^{\circ} \mathrm{C}$. The ice crystals which have formed on the activated nuclei fall into a supercooled sugar solution where they grow into visible macroscopic crystals. For an expansion ratio of about $1: 1.25$ all wettable particles larger than $60 \AA$ act as condensation nuclei; according to Fletcher such a particle being an ideal freezing nucleus forms ice for all temperatures colder than $-5^{\circ} \mathrm{C}$. Smaller particles are lost. All particles within the Aitken nuclei range could theoretically be counted, but it appears that considerable losses occur due to the short time interval which is
available for activation. This method can be considered to give an integral number of ice nuclei which would be active from -5 to $-20^{\circ} \mathrm{C}$.

## 2. Diffusion Chamber Method

The essential difference of this method to the one described above is the smaller supersaturation during the formation of the drops. Supercooled fog is formed either through a moist warm surface or through a droplet generator. In one counter cloud formation is accomplished through a semi-permeable membrane filled with warm $\left(35^{\circ} \mathrm{C}\right)$ water; it is possible that this method causes considerable supersaturations. In a different counter droplets are generated using a droplet generator and the walls are continuously flushed with glycol to prevent formation of rime ice; here at best 100 percent to 103 percent relative humidity may be achieved. In this counter we can therefore measure only those freezing nuclei which are also well wettable condensation nuclei, or which make contact within the residence time of the dir sample in the counter of about one minute with a droplet, or those which were present in the water. There may easily be an order of magnitude or more droplet, or those which were present in the water. There may easily be an order of magnitude or more difference between the two methods without any method being wrong. This method therefore could be called a differential method which counts essentially large freezing nuclei.

## 3. Millipore Filter Method.

The nuclei contained in a known air volume are being deposited on a millipore filter and then "developed" on a cold chamber. The method is attractive but the nuclei spectrum measured is poorly defined, since the great number of Aitken nuclei sampled simultaneously may easily poison some of the freezing nuclei.

It is indeed my great pleasure to thank all contributors and participants who have helped to make this Workshop a success. It is hoped that future workshops will be held and will help to clear up unsolved problems in the measurement of nuclei concentration.

> Address
> of
> Joseph Podzimek
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> Prague, Czechoslovakia

Three years ago we met in Lannemezan in order to test the apparatus measuring the content of condensation and ice nuciei in the air and to discuss some general problems related to this subject. In this connection allow me, fellow colleagues, to mention that, thanks to the enthusiasm and extreme diligence of Professor Dessens and all of his fellow workers, we were able for the first time in the history of cloud physics to organize an international workshop in such a large scale trying to compare the apparatus and to deduce some practical conclusions for the world-
wide use of some recommended methods. Now, opening the second workshop, we have to look back and, first, thank Professor Dessens, who unfortunately is not able to attend this meeting, for his successful management of the workshop and for the beautiful atmosphere created by the well-known French hospitality; and second, to look at our own work during the Lannemezan meeting and compare it with our intentions here in Fort Collins. The second workshop is organized at the foot of the beautiful Rocky Mountains, thanks to the understanding and appreciation of our task shown by the National Science Foundation and by the Colorado State University in Fort Collins, represented by it's President, Dr. A. R. Chamberlain, and by the Department Head of the Atmospheric Science Department, Dr. Elmar R. Reiter. Dr. H. Weickmann, President of the International Commission on Cloud Physics, assisted during the entire preparation of this workshop and helped very much in elaborating the report of the last meeting and preparing with us the program for this meeting.

Many thanks to Professor R. Steele, Chairman of the Organizing Committee, to Professor L. Grant, representing Colorado State University in the Organizing Committee, to Dr. U. Katz, also of CSU, and to the other members of the Organizing Committee for their successful work in preparing this workshop, which is certainly also due to the effort of many other employees of the Cloud Simulation and Aerosol Laboratory and of the Department of Atmospheric Sciences of Colorado State University.

Now let us look at the experiences made organizing such a workshop and then review some of its most important results. We learned that the most efficient work can be done when the majority of workshop participants are actually dealing with the testing of the apparatus. Therefore, I recommend to those who did not bring their own apparatus, that they join their colleagues and thus learn their apparatus and their method of work. It seems appropriate to me not to disperse our attention during the workshop organizing many plenary sessions with scientific communications which are not immediately connected with the methodology of our measurements. Bearing this in mind, the Organizing Committee decided to dedicate the opening and concluding plenary sessions to the communications of general importance. In addition to the sessions there will certainly be plenty of time during the coffee breaks and in the evenings for the exchange of opinions between the conference participants on an informal basis. If some of these discussions lead to conclusions having importance for other participants of the workshop, I would recommend summarizing the main conclusions and submitting them to the Steering Group.

After the Lannemezan meeting, Professor Dessens generously published all the papers presented during the plenary sessions, including some preliminary comparisons of the apparatus tested. We did not succeed in publishing all the data submitted by the individual operators. The reasons are as follows; not all of the participants submitted the data in a uniform way immediately after the measurements. Some of the participants made additional corrections in the measurements two or three months after the end of the workshop. You can look at the original records submitted by the individual operators in my office and you will probably come to the conclusion that there was sometimes a lack of discipline among the operators. On the other hand, there was not suffi-
cient initiative and not enough pressure on the part of the Organizing Committee to improve it. These reasons led the Organizing Committee of the Fort Collins meeting to create the Steering Group for supervising the work during the workshop and for making recommendations to the Organizing Committee in order to reach the goals of this meeting. Therefore, for your convenience, please keep all the instructions and rules posted on the bulletin board. When we succeed in our task, there will be the possibility of printing all the data and all the main remarks related to the methodology of our work in the series of publications of Colorado State University, Department of Atmospheric Science.

Evaluating the results of the Lannemezan meeting, I came to the following conclusion. It was not possible to compare all apparatus present at the workshop. Some of them were not in good condition throughout the meeting and the operators did not succeed in repairing them. Some of the participants measured in different times comparing it with the originally recommended ones and there is a principal difficulty in comparing different types of apparatus having different sampling time. The main conclusion drawn from the comparisons of the measurements using natural ice nuclei made approximately at the same time and at the same temperature is as follows. The apparatus based on the same principle having similar dimensions gave comparable results. It can be seen from the ice nuclei measurements with the aid of the mixing chambers of the Soulage-Admirat type presented in Table 1.

TABLE 1

|  | Correlating <br> factor at <br> $-21^{\circ} \mathrm{C}$ | No. of <br> Measure- <br> ments |
| :--- | :---: | :---: |
| Admirat-Benesch | 0.60 | 5 |
| Admirat-Podzimek | 0.80 | 4 |
| Admirat (basement)- | 0.85 | 7 |
| Admirat (first floor) |  |  |

On the other hand, there is a big difference between the apparatus based on different principles like the Soulage-Admirat chamber and the LangerRossinski acoustic counter where the correlating factor was only 0.3 ( 11 cases) showing a constant lower concentration measured by the Langer counter (in the range from $5 \%$ to $50 \%$ ). In general, the highest concentrations were measured in the 300 Liter expansion chamber (Mossop) and in all mixing chambers of the Soulage-Admirat type. Not many large differences between the apparatus brought along by Admirat, Grant, Steele, and ESSA were found in the domain of temperatures above $-20^{\circ} \mathrm{C}$. The measurements of ice nuclei above $-10^{\circ} \mathrm{C}$ can hardly be compared. Unfortunately, the number of measurements made simultaneously at the same time and temperature by the operators of the apparatus mentioned is too low and we cannot arrive at any general conclusions.

The measurements of the condensation nuclei show a similar picture as the measurements of ice nuclei. The most difficult problem was to select the apparatus working with the same supersaturation or with the same supersaturation levels for a comparison. For example, good response was shown by the apparatus presented by Jiusto, Gagin, and Auer. The corresponding corre-
lation factor based on 16 measurements (at the supersaturations $0.1 ; 0.3 ; 1.0$; and $3.0 \%$ ) in the case of Jiusto's and Auer's apparatus was 0.91 and in the case of Jiusto's and Gagin's device was 0.86 (for 8 comparable measurements). There is very weak correlation between Gardners and the General Electric counter and Jiusto's chamber working at the supersaturation of $1 \%$, but there is a strong correlation between two Gardners counters, one of which was made like a portable counter. Some measurements support the idea that in the experimental laboratory and dealing with the storing bags we did not succeed in storing well-controlled aerosol particles for any extensive time. This can be clearly seen from the counting of NaCl particles in the apparatus with variable low supersaturation.

From these facts we want to discuss how to improve our work during the second workshop. In my opinion we need more information about the apparatus to be tested including all correction constants. We need more information about aerosol particles used for testing and we have to decide the evaluation system of data permitting us to evaluate the basic data right here. In general it seems to me that we need more discipline in our work.

Following the wishes of the participants, the Organizing Committee decided to present all main kinds of apparatus to the participants before starting the workshop. It should be done this afternoon. In order to save you time, we selected a main speaker for each group of the apparatus, who should give you a short description of the apparatus or of the method used. All the other operators of the apparatus being presented are kindly asked to complete the explanation of the main speaker adding only those remarks characterizing the pecularities of their own apparatus. The form of these presentations should be adapted to what degree the description of the apparatus was given or published in detail before. Please give short instructions on how to evaluate the measured data in such a way that everybody knowing the correction constants could replace the operator.

We shall try to improve our knowledge about aerosol particles generated in the laboratory. We can, of course, tell little about their activity in cloud generating or cloud transforming processes, but we hope to know to some degree their physicalchemical nature including some information about size spectrum distribution curve or the geometry of individual particles using electronomicrophotographs. We assume that everyone received the information of the laboratory facilities carefully prepared by Dr. Katz. From it and from the working program set by the Organizing Committee, it can be seen that we will try to use many kinds of artificial nuclei, like NaCl , teflon, AgI, phloroglucinol, enabling us to know more about their nucleating activity relating to different sizes of particles and different temperatures used in the apparatus.

We know that very few of the apparatus are actually simulating the processes occurring in the nature, speaking about the duration of supersaturations actually occurring in the clouds. We have not sufficient knowledge of the turbulent transfer of heat, momentum and concentration of water vapor in different kinds of clouds presenting the non-steady case of the embryo generation. Besides, most of the active nuclei occurring in the atmosphere have the nature of mixed nuclei and the state of theoretical description of heterogeneous nucleation or of epi-
taxial growth is not sufficient in order to permit us with an acceptable occurrence to calculate the migrational movement of water vapor molecules on a non-active substrate toward a nucleus of active substance. One is certain that the nature of both and the geometry of both play a great role. A more complicated situation is related to the explanation of the freezing of supercooled water droplets through migration of active nuclei or to the mechanism of freezing of very thin layers of water on a substrate having very complicated geometry and nonhomogeneous composition. Bearing all these facts in mind, we can hardly describe the processes taking place on the surface of the active nuclei inside individual apparatus in which we usually do not know sufficient information about the micro-variations of the temperature and humidity field. That these factors play an important role we discovered during the Lannemezan meeting. For example, I was able to observe that in all chambers of the Soulage-Admirat type, there was in the proximity of the source of water vapor much higher concentration of ice crystals formed on the nuclei than on the more remote places. The same was observed in the proximity of the borders of the trays used for developing ice crystals in a sugar solution. During the "nuclei week" in the laboratories of Professor Whitby preceding this workshop, some preliminary results strongly suggested the idea that we have first of all to check the size characteristics of different types of counters. The general method enabling us to measure the number of active ice nuclei in the population of supercooled droplets should be carefully examined.

On the other hand, we know that the majority of the apparatus used show a close response with the water phase transition observed in the nature. Some of the apparatus show a similar trend of the records made in the same environment. On these statements we can base our working program during this workshop. We assume that we do not know which of the apparatus tested gives, in a certain domain of temperatures, humidities or sizes of particles, the right counts; but we can compare the efficiencies in counting the nuclei under well defined conditions. The words, "well defined conditions" mean to use the natural nuclei or to generate the aerosol particles in such a way that we know their size spectrum distribution, their concentration, and in the case of an artificial aerosol also their physical-chemical nature. Naturally, we have to check the temperature, humidity and the purity of the samples of aerosol stored for the testing. The use of an aerosol analyzing device brought with it some complications concerning the methodology of our work. We have to check the size spectrum of particles at higher concentration, which means that we have to work on two levels taking samples with the Aerosol Analyzing System from a small balloon ( 125 cfm ) and then to dilute this sample in a balloon ( 2000 cfm ) of a low level diluting system. This implies some complications speaking about possible changes of size spectrum curves of aerosol particles in both systems, which we tried to eliminate to a minimum rate. We hope that it will be possible to keep aerosol in the main storing bag for 1 hour in a well controlled state. Thus, during the first days of the workshop we will try to get all necessary information about the behavior of aerosol particles in the storing bag, including the use of electron microscopy. The shakedown period will be character-
ized without doubt by finding the most suitable methods for comparing the apparatus based on different physical principles. In my opinion, we should be able at the end of the workshop to deduce very important aerosol size characteristics of the apparatus tested for at least some of the used artificially generated particles like AgI. Thus, success of this trial is dependent upon the success in preparing monodisperse, well defined aerosol. The aerosol size characteristics are closely connected with the time lag characteristics of the apparatus or the catching efficiencies of the filters.

The second aim of our work should be to try to deduce the temperature characteristics of the ice nuclei counters for selected kinds of ice nuclei. The basic temperatures, $-8^{\circ},-16^{\circ}$, and $-20^{\circ} \mathrm{C}$, are recommended for doing it, and in addition we hope that some operators will be able to measure also at the temperature of $-12^{\circ}$ and $-26^{\circ} \mathrm{C}$. It is now difficult to foresee exactly how much time it will take before reaching the temperatures wanted in different types of apparatus in order to announce the final time schedule. It will be settled during the first days of the workshop.

In a similar sense we can speak about vapor pressure characteristics of the cloud condensation nuclei counters using some well defined aerosol particles like NaCl . The recommended supersaturations will probably be $0.1 \%, 0.3 \%, 1.0 \%$, and $3.0 \%$. The very important characteristics could be obtained according to my opinion when we would be able to measure the number of active nuclei also in nonsaturated domain like at $90 \%$ or $95 \%$.

A very interesting experiment could be done using the hydrophobic particles of well defined size, like teflon, during the workshop.

In concluding the workshop, I hope that we shall be able to deduce some correlation factors permitting us to compare the different apparatus or to check the reproducibility of the measurements made using the same type of instrument.

For these aims we prepared simple statistical methods and we will keep you informed during the IWCIN. In my eyes, these are the goals of this workshop, and I would like to know your opinion on this subject. However, you know how complicated these questions sometimes are when viewed from the relatively narrow perspective of a special kind of apparatus or a method of working. Therefore, I would like to encourage you to think about it and sometime during the first three days kindly acknowledge your suggestions to the Steering Group who will prepare the final proposal of our working program for the Organizing Committee. After this deadline, please do not ask substantial changes to be made concerning the program generally accepted.

In closing this introduction, I would like to thank all the participants who brought along apparatus and who prepared themselves for this meeting. Expecting cooperation from all to build a friendly atmosphere--as it was always so during our meetings-I am personally convinced that we shall reach our goals.

## Remarks <br> by <br> Vincent J, Schaefer <br> State University of New York

I have been asked to say something about the procedures and operations which might be followed at this Workshop based on the experience we gained during the first gathering of this kind at Lannemezon, France in 1967, which many of those present attended. It is hardly necessary to do this since all of the essential recommendations which I summarized at the conclusion of the Lannemezon Workshop have been incorporated in the facilities here at Fort Collins, thanks to the hard work of Roger Steel, Josef Podzimek, Lewis Grant, Ulrich Katz, Helmut Weickmann, and others who have spent so much time preparing for this present meeting.

The pioneering effort of Dr. Henri Dessen, his son Jean, and his devoted staff, which made the first workshop three years ago such a stimulating experience, should be recognized at this time for helping us to prepare for the present activity. I should also mention the inspiration which Dr. Leo Pollak gave to some of us who are here when he initiated the ICCN conferences a generation ago. He and some of the other pioneers like Findeisen, Kohler and Nolan would be very pleased to see the many young scientists, such as are in this audience, who are tackling the fascinating, complex and often bewildering problems which confront us in our attempt to properly understand the intertwined problems of gas to particle conversion, the aerosol climatology of regions and the challenging and sometimes baffling aspects involved in our attempts to devise instruments which are compatible and which we hope will permit us to properly measure the gases and particulates of the atmosphere so we can better understand the role they play in atmospheric dynamics, air pollution and weather modification.

I hope I will be forgiven if I am somewhat critical of a trend I see in the design of particle measuring devices. As these instruments get larger and larger in an engineering attempt to more closely simulate the atmosphere, there is a very real danger that these efforts will lead the researcher away from the real atmosphere. I am very much afraid that in seeking physical precision, we may lose contact with reality.

The real atmosphere and its properties are far from being an ideal or uniform system. One only needs to monitor any one of the myriad of properties to become aware of this factor. While it is nice to have precision, and some individuals are never happier than when seeking it, at this stage of the "art" I believe we need lightweight, portable, simple and practical devices which can give us data with an accuracy of ten or twenty percent. The concentrations of atmospheric particulates often vary by orders of magnitude and their effect must be better understood before we can afford the luxury of high precision in our measurements:

Some of you may remember my presentation at the Prague meeting last September, when I departed from my scheduled subject to briefly announce a strange phenomenon which I had recently encountered while measuring submicroscopic lead particles. At that
time I suggested that the effect I had observed may be a universal feature possessed by clean country air. Since then I have found this to be true. I have measured the effect on the edge of the Japanese sea and in the snowy mountains of Hokkaido, at 30,000 feet above the northwest coast of the United States, in the California deserts, near the top of Mt. San Jacinto and Mt. Baldy in that same area, in various parts of New York State, and within the past few months, in northern Arizona, Utah, Wyoming and just outside our meeting place here at Fort Collins.

Since my initial disclosure, I have discovered that vapors other than from iodine produce similar reactions.

This effect clearly illustrates the degree of ignorance which still challenges our ingenuity. By catching a sample of "country air" in a container such as a plastic bag, and exposing this air for a few seconds to the gaseous vapor of substances such as iodine, turpentine, limonene or beta terpene, an air sample which ordinarily has a concentration of 400 cloud nuclei (CCN) and 2000 Aitken Nuclei (CN) will immediately rise to values of $30-50,000$ cloud nuclei (CCN) and over a million Aitken Nuclei (CN) per milliliter (ml) or (cc).

I have also found that the cause of this amazing reaction is not a true gas to particle reaction such as is believed to occur when unburned hydrocarbons are catalyzed by ozone and oxides of nitrogen. I find that if I pass air which contains the potential for causing this reaction through an absolute filter, the effect completely disappears. Thus, I believe that all relatively clean air contains clusters of molecules, perhaps like large ions, but which are not large enough to be activated with a supersaturation of 300 percent (such as is imposed by a small particle detector), but that the saturated vapor of iodine, turpentine, limonene or the pinenes is sufficient to cause an instantaneous polymerization of these clusters to form both a very high concentration of Aitken Nuclei. What is even more striking is the extremely high and quite unnatural concentration of cloud nuclei which also form.

I first observed this effect in the early summer of 1969 when I put an iodized sample of relatively clean Arizona air in my cold chamber. A similar spectacular effect can be seen when similar air samples are compared in two simple Wilson Cloud Chambers, one of which contains a drop of turpentine or tiny crystal of iodine. An even simpler demonstration of the effect is to permit outside air to ventilate a shower room. When the hot water is turned on, a cloud soon forms. Again ventilating the room until the cloud disappears, a few drops of turpentine put on a piece of paper towelling and waved a few times in the air will produce a noticeable smell of turpentine. Again turning on the shower, the cloud that then forms will be one of the most dense "pea soup" fogs you have ever seen!

Interestingly - this effect will not be found in badly polluted air. Apparently the clusters, or pseudo embryos as I have called them, become adsorbed on the pollution particles and thus lose their mobility to react.

I have digressed in telling you about this intriguing phenomenon to suggest the degree of our
present ignorance about atmospheric particulates. Thus, as you embark upon the effort to compare instruments, I hope you will subsequencly make a real effort to devise instruments which are as simple, small and inexpensive as your ability can devise. While it is convenient and sometimes necessary to have things automatic, data without understanding and perception is worse than useless. Nothing along this line is of more value than to be present when the measurement is being made. Nothing can replace the inquiring mind, intelligent eyes, perseverance and serendipity! The latter never occurs to an automatic instrument:

## Keynote Address

by
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Laboratory of Atmospheric Physics
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DIFFUSION CHAMBERS FOR THE MEASUREMENT OF THE SPECTRUM OF CRITICAL SUPERSATURATIONS

OF CLOUD NUCLEI

## ABSTRACT

Some limitations of diffusion chamber design are discussed. The chamber depth should not exceed about 1 cm ; the measurements may not be reliable below about $0.2 \%$; excessive nucleus concentrations can cause serious errors, the presence of which can be detected by dilution experiments; if the top plate is heated to a temperature above ambient, the entering sample should be warmed to at least that temperature in contact with dry surfaces. Presented at the International Workshop on Condensation Nuclei and Ice Nuclei, Fort Collins, August, 1970.

## IUTRODUCTION

The reasons for interest in cloud nuclei are well known. The theories of cloud formation of Howell (1949), Mordy (1959) and Neiburger and Chien (1960) based on Köhler's theory for nuclei consisting of a pure soluble salt, indicate that the most important property of a nucleus is its critical supersaturation, which determines whether or not it will grow unstably. The effect of the properties of nuclei on rates of growth is of secondary importance. Although it is known that aerosol particles do not in general consist of pure soluble species, the general conclusions appear to remain valid: that every particle behaves more or less according to a Köhler type law, and has a critical supersaturation; and that the process of cloud formation could be fairly well simulated if it were assumed that each droplet began to grow on a nucleus at the moment when its critical supersaturation is exceeded, and then grew according to a simple law, independent of the properties of the nucleus. Thus, it is preferable that measurements be made of the spectrum of critical supersaturations rather than, for example, of sizes. Particle mass would correspond
one-to-one with the critical supersaturation only if the particles all consisted of a pure species (an articical aerosol of NaCl , for example).

The majority of investigators have used a diffusion chamber more-or-less similar to that described by Twomey, (1963) in which the cloud of drorlets is photographed. The following remarks concern certain experimental difficulties which arise. The discussion is based on the treatment given by Wieland (1956) who showed that the maximum supersaturation, occurs nearly midway between the horizontal plates, and varies as $(\Delta T)^{2}$, where $\Delta T$ is the temperature difference. The discussion given here ignores second order effects which result in the gradients of temperature and water vapor density being slightly non-linear, such as the temperature dependence of conductivity and diffusivity, thermal diffusion effects, etc. The slight displacement of the level of maximum supersaturation from the midpoint is also ignored.

## TRANSIENT SUPERSATURATIONS

Certain situations can give rise to transient supersaturations during the passage of the sample air into a diffusion chamber. If, for example, moist air moves close to a relatively cold, dry surface which is sufficiently hydrophobic to resist the nucleation of water droplets, the sample may be cooled below its dew point. Since one degree depression of the temperature below the dew point corresponds to several percent supersaturation, this could result in nucleating droplets on particles with critical supersaturations larger than that at which it was intended to make the measurement. These droplets once formed may or may not reevaporate to haze droplets, depending on the subsequent course of events.

If the cold surface is wet, it presents a sink of water vapor as well as of heat. Twomey (1963) has pointed out that since the diffusivity of water vapor exceeds that of heat, proximity to a cold wet surface cannot result in a transient supersaturation. The same questions have been discussed by Saxena et al. (1970), Fitzgerald (1970).

For example, if a semi-infinite volume of air at temperature $T_{0}$ and vapor density $\rho_{0}$ is placed close to a wet surface at temperature $T$, the temperature at distance $x$ from the wall ${ }^{1}$ 's given by

$$
T(x t)-T_{1}=\left(T_{1}-T_{0}\right) \operatorname{erfc}\left(\frac{x}{2 \sqrt{k t}}\right)
$$

where $k$ is the diffusivity of heat. Hence

$$
\frac{\partial T(x, t)}{\partial t}=\frac{\left(T_{1}-T_{0}\right) x}{2 \pi^{\frac{1}{2}} \kappa^{\frac{1}{2}} t^{3 / 2}} \exp \frac{-x^{2}}{4 \kappa t}
$$

If the saturated vapor density at temperature $T_{1}$ is $\rho_{1}$, and $k^{\prime}$ is the diffusivity of water valpor, a similar relation holds for $\rho$, and at a constant $x$ it follows that:

$$
\frac{d \rho}{d T}=\left(\frac{k}{k^{\prime}}\right)^{\frac{1}{2}}\left(\frac{\rho_{1}-\rho_{0}}{T_{1}-T_{0}}\right) \exp \left[\frac{x^{2}}{4 t}\left(\frac{1}{k}-\frac{1}{k^{\prime}}\right)\right]
$$

Consider a $T$, $p$ diagram on which the points $T_{0}, \rho_{0}$ and $T_{1}, \rho_{1}$ are plotted, the latter lying on the curve of saturated vapor density versus temperature and the former below it. Since $k^{\prime}>k$, the curve along which the point $T, p$ moves with increasing time from $T_{0}, \rho_{0}$ to $T_{1}, \rho_{1}$ must begin by being steeper than the line joining these two points (for $t$ small), but finally becomes less steep for large $t$. Obviously, the slope can never vanish, or reverse sign. Thus, if $T_{0}>T_{1}$, it is graphically obvious that the air can never become supersaturated, but if $T_{0}<T_{1}$, it will do so if the asymptotic value of $\mathrm{do} / \mathrm{d} T^{1}$, that is $\left(k / k^{\prime}\right)^{\frac{1}{2}}$ ( $\rho_{1}-\rho_{\rho} / T_{1}-T_{0}$ ) is less than the slope of the $T$, $\rho_{S}(T)$ curve at the point $T_{1}, \rho_{1}$.

These considerations show that there is some risk in heating the upper plate of a chamber above ambient temperature unless, before injection, the sample air is heated to at least the top plate temperature, in contact with dry surfaces. If the lower plate is cooled and the top plate held close to ambient temperature as recommended by Twomey, this problem is avoided.

## DIAMETER TO HEIGHT RATIO

The simple theory applies to diffusion between infinite plates. In practice the ratio of diameter to height is limited by the need to illuminate and observe small droplets. With a given source, the illumination which falls on a droplet is proportional to the solid angle over which it is illuminated, and the flux of scattered light which can be used to observe it is similarly proportional to the solid angle over which this light is collected. Thus, with a given light source and a given method of detecting droplets, the need to observe droplets while they are still small in practice limits the "aspect ratio". Twomey (1963) has computed the disturbing effect of the walls for a chamber with a diameter four times its height, and with the walls and top at ambient temperature, the bottom plate being cooled. Since the walls are warmer than the central air at all levels, the heat conducted inwards raises the temperature and reduces the available supersaturation. In contrast to the quadratic curvature effect, this disturbance is linear in $\Delta T$ and so becomes more important at low supersaturations. Twomey found that the overall results could be approximately reproduced if the temperature difference between the plates were reduced by about one degree before calculating $S$ in the usual way, that is, as if for infinite plates. In general, if the diameter is $m$ times the height, the temperature increase at the midpoint which is due to the walls ( $\delta \mathrm{T}$ ) is given by:

$$
\frac{\delta T}{\Delta T}+\frac{1}{2}=\sum_{n=1}^{\infty}\left[\frac{1}{\alpha_{n} J_{1}\left(\alpha_{n}\right) \cosh \frac{\alpha_{n}}{m}}\right]
$$

where $\Delta T$ is the difference in the plate temperatures, and $\alpha_{n}$ is the $n$th positive zero of $J_{0}$. The resulting decrease in $S$ is about $(7 \delta T) \%$, and it is easily shown that the effect of the walls can be allowed for by reducing the measured value of $\Delta T$ by about $(90 \delta \mathrm{~T} / \Delta \mathrm{T})$ deg. C. before calculating S This correction decreases rapidly with increasing $m$, as would be expected; at $m=8$, it is only about
$-3 \times 10^{-30}$ C. and hence quite negligible. Elliott (1970) has calculated $\delta T$ for several values of $m$. At $m=5$, he finds a value of $2 \times 10^{-3}$ for $\delta T / \Delta T$, corresponding to a correction to $\Delta T$ of about $-0.2^{\circ} \mathrm{C}$, and considers that wall effects are negligible provided $m \geq 6$.

Other disturbing effects such as convection could interfere with the operation of a chamber. Gagin and Terliuo (1968) have overcome these difficulties by providing metallic walls of high thermal conductivity, so that the temperature gradient in them matches that of the air.

## SAMPLE ENTRY

As the sample enters and flushes the chamber, some of the air must pass along the vertical walls which are usually dry. If a gradient of temperature exists in the wall, some parts of it will be cooler than the top plate, and if air which has been moistened there later passes down the walls and is cooled, the possibility exists that large supersaturation will result. For this reason it may be desirable that the sample enter through the top plate at the center of the chamber, and exhaust through ports located near the vertical wall.

A reasonably conservative assumption concerning the flushing process is that the contents at any moment are perfectly mixed, so that aerosal concentration approaches the desired value exponentially:

$$
\left(n-n_{2}\right)=\left(n_{1}-n_{2}\right) \exp (-a t)
$$

where $n_{2}$ is the concentration in the entering sample, $\alpha$ is the rate of flushing in chamber volumes per second, and $n_{1}$ is the initial concentration in the chamber. For $n_{1}<10^{4}, n_{2}>10$, as would normally be true for cloud nuclei, the chamber concentration lies within $1 \%$ of the desired value provided at $>12$, that is provided the chamber is flushed with twelve times its volume. If flushing is accomplished at high flow rates, it may be desirable to slow down the flow rate towards the end, since otherwise droplets will appear before the air has come to rest, the diffusivity of momentum due to molecular transfer being rather less than that of heat or water vapor.

## APPROACH TO EQUILIBRIUM

Once the air has essentially come to rest and equilibrium conditions are being established as a result of conduction of heat and diffusion of water vapor, it is clear that the designed maximum supersaturation ( $\mathrm{S}_{\mathrm{m}}$ ) will be approached asymptotically. If the chamber is too deep - irrespective of its "aspect ratio" this approach can be so slow that tens of seconds will elapse between the moment when the air becomes saturated $(S=0)$ and that when $S$ reaches, say, $90 \%$ of the intended level, $\mathrm{S}_{\mathrm{m}}$.

The difficulties which can arise in a deep chamber are simply illustrated by considering the following example. Suppose that the sample air is initially warmer than either plate but has a mixing ratio such that, on cooling to the mean temperature $\left(T_{1}+T_{2}\right) / 2$, the vapor density will be $\left(\rho_{1}+\rho_{2}\right) / 2$.
Then, that part of the sample which comes to rest in the middle of the chamber will neither lose nor gain
water vapor. The variation with time of $T$ at midpoint between the plates is given by:

$$
T=\frac{\left(T_{1}+T_{2}\right)}{2}\left[1-\frac{4}{\pi}\left(y-y^{9} / 3+y^{25} / 5 \ldots \cdot\right)\right]
$$

where $y=e p\left(-\pi^{2} k t / \ell^{2}\right), \ell$ is the depth of the chamber, and the origin of temperature is taken at the initial sample temperature.

An equilibrium supersaturation of $1 \%$ corresponds to a dew point excess of about $1 / 7^{\circ} \mathrm{C}$. If the initial temperature exceeds $\left(T_{1}+T_{2}\right) / 2$ by a few degrees, the sum of the infinite series is less than 0.1 by the time the air reaches saturation. The sum of the first two terms is, a fortiori, less than 0.1, and it is obvious that only the first term is significant, so that the approach to equilibrium thereafter is exponential, with a time constant of $\ell^{2} / \pi^{2} k$, that is, about $\ell^{2} / 2$ seconds, where $\ell$ is in centimeters. From the moment when $\mathrm{S}=0$ to that when $\mathrm{S}=0.9 \mathrm{~S}_{\mathrm{m}}$, the time elapsed will be about two and a half time constants, that is about $\ell^{2}$.

It is observed that once a cloud forms, only a few seconds elapse before it begins to fall out of the illuminated region. It would therefore seem necessary that the depth of the chamber should not be greater than about 1 cm , so that the time which elapses between moments when $\mathrm{S}=0$ and $\mathrm{S}=0.9 \mathrm{~S}_{\mathrm{m}}$ is only about a second. If, for example, $\ell=5 \mathrm{~cm}$, this time will be about 25 seconds; during the slow rise of $S$ from zero to $0.9 S_{m}$, many nuclei with critical supersaturations of less than $0.9 \mathrm{~S}_{\mathrm{m}}$ may have formed droplets and fallen out of the observing region well before $S$ has approached within $10 \%$ of its design value. Disregarding the short time required for a droplet to become observable once it is nucleated, the rate of appearance of drops is given by $(\mathrm{dN}(\mathrm{S}) / \mathrm{dS}$ $d S / d t)$, where $N(S)$ is the usual cumulative distribution function. This function is almost always convex upwards, so that $\mathrm{dN}(\mathrm{S}) / \mathrm{dS}$ is a decreasing function of $S$ : As $S$ rises towards its asymptotic value, $d S / d t$ is also a decreasing function of $t$. Hence, their produce must steadily decrease. In a chamber with $\ell$ of order 5 cm , the observed "maximum cloud" will probably occur a few seconds after the central air becomes saturated, when $S$ is still only a small fraction of $\mathrm{S}_{\mathrm{m}}$. The observed maximum droplet concentration in such a chamber may bear little relation to the true value of $N\left(S_{m}\right)$.

Some mitigation of this limitation would follow from the consideration that droplets forming initially above the central observing region would fall into it. However, in view of the observed short duration of the approximately constant maximum cloud, this would probably be a minor aspect.

## THE LOWER LIMITS OF $S_{m}$

The objective sought with diffusion cloud chambers is to make an observable dichotomy between nuclei which, on being exposed to a given supersaturation, grow in an unstable manner, and those which do not. At supersaturations much below $0.1 \%$, this becomes difficult, since the haze droplets formed on nuclei with low critical supersaturations are quite large, and these may be recorded along with unstably growing droplets.

In order that a droplet may be detected reliably by photography, or otherwise, its radius must exceed
some minimum value $r_{f}$; commonly, droplets are recorded when $r$ is about $2 \mu$. The equilibrium radius of a haze droplet (formed on a soluble nucleus) ranges from $6 \times 10^{3} \mathrm{M}^{\frac{1}{2}}$ at saturation to $10^{4} \mathrm{M}^{\frac{1}{2}}$ at the critical supersaturation, where $M$ is the effective molar mass of the nucleus. The corresponding critical supersaturation is about $7 \times 10^{-10} \mathrm{M}-\frac{1}{2} \%$. Thus, haze droplet radii can range up to $7 \times 10^{-6} / \mathrm{S}_{\mathrm{C}}$, or about $2 \mu$ for $S_{C}=0.03 \%$.

A more stringent limitation follows from a consideration of the growth and fall of droplets at low supersaturations. Droplet growth may be approximately represented by:

$$
\frac{d(r+\sigma)^{2}}{d t}=A S
$$

where, following the ideas of Rooth (1954), $\sigma$ is a length of order a few microns, which depends on the condensation coefficient for water vapor and the accommodation coefficient for air, and $A$ is a constant about equal to $10^{8}$ if S is in percent. Combining this with Stokes' law, $\mathrm{v}=\mathrm{Br}^{2}$, and treating $S$ as constant, the distance fallen below the midpoint of the chamber is given by:

$$
z=\frac{2 B}{A S}\left(\frac{r^{4}}{4}+\frac{\sigma r^{3}}{3}\right)
$$

The value of $\sigma$ is still in some doubt, but even if it is as small as $1 \mu$, the droplet falls $(0.15 / \mathrm{S})$ by the time it grows to have a radius of $2 \mu$; that is, at $1 \%, 1.5 \mathrm{~mm}$, and at $0.2 \%, 8 \mathrm{~mm}$. When account is taken of the limitation on total chamber depth discussed above, these considerations place a limit on the minimum supersaturation at which diffusion chambers can be used, in the vicinity of $0.2 \%$.

The major inaccuracy in the simple growth law used above arises from the neglect of the Kelvin effect, which at $r=1 \mu$ reduces the supersaturation available to the droplet by about $0.1 \%$. The growthretarding effect of capillarity at low ambient supersaturations would act to increase still further the difficulty illustrated above.

The value of $S$ is approximately a parabolic function of $z$, but since only the central region of the chamber (where $S$ is nearly constant) should be illuminated, treating $S$ as constant does not affect the conclusion: that at sufficiently low supersaturations, the droplets tend to have fallen out of the beam before they have reached the size at which they can be reliably recorded.

## MAXIMUM CONCENTRATION

The supersaturation computed on the basis of linear gradients of temperature and vapor density is, of course, strictly correct only when no growing droplets are present. A growing droplet represents a sink of water vapor and a source of heat, and both effects tend to reduce the supersaturation. The magnitude of the disturbance increases with the concentration of growing droplets, so that obviously there is some upper limit to the concentration which a chamber can measure reliably. Even an approximate calculation of these effects in the presence of a parabolic distribution of supersaturation is complex;

Twomey (personal communication) has carried out numerical computations of the effect, and concluded that, for a chamber 1 cm deep, an appropriate maximum concentration at $S=1 \%$ is about $10^{3} \mathrm{~cm}^{-3}$.

This was confirmed experimentally when polluted air was being sampled near Denver, Colorado, using a chamber about 1 cm deep (Squires, 1966). If after diluting 1 part of sample air with ( $n-1$ ) parts of filtered, nucleus-free air, the concentration measured was $C_{n}$, this would indicate an original sample concentration of $n C_{n}$. It was found that as $n$ was increased, from 2 to about 10, the resulting value of $\mathrm{nC}_{n}$ at first increased and then leveled off, indicating that the disturbing effects of excessive concentrations were no longer affecting the result. This indeed occurred at a value of $n$ which resulted in a value of $C_{n}$ of about $10^{3} \mathrm{~cm}^{-3}$.

## CONCLUSION

Despite the limitations of diffusion chambers, they have been used with success by Squires and Twomey (1961) and Twomey and Warner (1967) to predict the concentrations of droplets in convective clouds, with a degree of success which is entirely satisfactory when the problems of securing representative and comparable measurements are considered. In these comparisons, samples of air from below cloud base were taken and the spectrum of critical saturations measured. An estimated or measured updraft speed was used in a calculation of the cloud forming process, using the approximate analytical method of Twomey (1959), and the resulting concentration of activated nuclei compared with the observed droplet concentration in the clouds above. It is clear that fluctuations occurring in space and time in all three measurements would result in a considerable scatter when the predicted droplet concentrations are plotted against those observed.

The satisfactory degree of success achieved in these comparisons seems to indicate that both our present understanding of the cloud-forming process, and the methods used for measuring the spectrum of critical supersaturations are reasonably satisfactory.

## ACKNOWLEDGMENT

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Keynote Address<br>by<br>John Hallett<br>Laboratory of Atmospheric Physics Desert Research Institute University of Nevada System

## ICE CRYSTAL NUCLEATION

## ABSTRACT

A review is given of methods of measurement of ice nuclei. It is pointed out that results depend on the test method, and moreover, that ice nucleus spectrum in the atmosphere depends on the particular cloud process considered. A one to one relation between ice nucleus counter and ice crystal in cloud is only to be expected when the change of temperature and saturation ratio to which the particles are subjected in the counter and cloud are comparable. Presented at the International Workshop on Condensation Nuclei and Ice Nuclei, Fort Collins, August, 1970.

## INTRODUCTION

Much of the precipitation which occurs in mid and high latitudes originates as ice in the upper
levels of the troposphere. Interest in the initiation of the ice phase was first expressed as early as 1911 by Wegener who postulated the existence of small foreign particles which assisted the change from water vapor to ice. Since that time a number of devices designed to predict the concentration of ice crystals produced by these foreign particles - ice nuclei - under differing conditions in the laboratory and the atmosphere have been developed. These all rely on the growth to detectable size of an ice crystal which has been nucleated by the foreign particle under specified physical conditions. Before looking closely at the detail of these devices, it is, however, essential to examine mechanisms whereby ice crystals could be produced in the absence of these particles and also whether one ice crystal, once produced, may have means of self propagation. Lack of appreciation of the role of these processes could easily lead to false conclusions in any attempt to compare ice crystal concentrations measured in the atmosphere and those observed in the test chamber.

## ICE CRYSTALS IN CLOUDS

Systematic measurements of ice crystal concentrations in clouds has received, until recently, somewhat sparse attention in relation to their importance to precipitation processes. From the viewpoint of the physics of the situation, two extreme situations are of importance; the highest temperature at which ice crystals occur, and the lowest temperature at which ice crystals are entirely lacking.

Koenig (1963), Braham (1964), and Mossop et al. (1968) have reported ice crysfal concentrations in cumulus clouds of up to $100 \ell^{-1}$ with cloud top temperatures of $-4^{\circ} \mathrm{C}$. On the other hand, measurements of ice crystals in lenticular clouds and observations of lack of downwind ice in similar situations, lead to a conclusion that no ice crystals ( $<0.1 \ell^{-1}$ say) are present under these situations at temperatures higher than $-35^{\circ} \mathrm{C}$. Measurements of ice crystals at intermediate temperatures show an appreciable variation in concentrations, with a maximum value near $800 \ell^{-1}$. With present ideas, we would interpret ice crystais at low temperatures, below $-35^{\circ} \mathrm{C}$, as being caused by homogeneous nucleation of water drops and at higher temperature by nuclei.

The cloud processes themselves, however, cannot be ignored in examining nucleus activity; a well defined laboratory study may be irrelevant if we cannot specify the cloud or drop temperature to sufficient accuracy. Koenig (1965) pointed out that since the temperature of a cloud drop is equal to the wet bulb temperature ( $T_{W}$ ), under some circumstances freezing may occur at apparently high dry bulb temperature. For example, when the drop is near the edge of a cloud the values of $T_{W}$ may change drastically. Should a cloud drop be involved in mixing at a cloud edge, the difference of $T_{W}$ between cloud and environment might be $-5^{\circ} \mathrm{C}$ at $-4^{\circ} \mathrm{C}$, with smaller differences at lower temperature. The thermal relaxation time of a $50 \mu$ radius drop is $-10^{-4}$ seconds which may be neglected; the evaporation time -50 seconds with a fall distance several meters before evaporation. Hence, in the case of a sharp gradient of $\Delta T_{W}$ at the cloud edge or at the cloud top in case of Kelvin-Helmholtz instability waves at an inversion, the drop has adequate time to fall into drier air, reach its wet bulb temperature, freeze, and be reincorporated into the cloud. The larger the drop, the longer its evaporation time and the more this process
is likely to occur; at low temperatures the possible $\Delta T_{W}$ is small and the effect becomes unimportant. Here, then, is a distinct influence of the detail of the mixing process and the sharpness of gradients of $T_{W}$ on the cloud micro physics in the form of ice nucleation.

Additional complications arise if we consider the micro physical processes themselves. Evidence exists, Pruppacher (1963), Abbas and Latham (1969) and Gitlin and Lin (1969), for the initiation of the ice phase at quite small supercoolings by cavitation phenomena. This may be caused by rapid mechanical deformation associated with drop break up, induced by hydrodynamical or electrical process. The precise role of this process in the atmosphere, however, is still debatable, as the presence of a solid surface may be necessary to nucleate the vapor bubble. Drops may shatter and produce small ice splinters on freezing by internal pressure buildup, a process enhanced when they depart by only a small amount from solution and thermal equilibrium with their environment (Johnson and Hallett, 1968). This is a process which may well be enhanced during freezing at the cloud edge, particularly for larger drops. There is also evidence that shattering is more likely when cloud drops accrete such that freezing is roughly symmetrical from front and back, a situation which is most likely during the riming of small graupel particles (Brownscombe and Hallett, 1967). These different processes are unlikely to occur uniformly as a cloud; ice would appear only while physical conditions were appropriate - at cloud edge or in the region of small graupel growth.

## ICE NUCLEI

Lacking further evidence, then, there is some case for linking both the initiation and the multiplication of the ice phase in clouds with the microphysics - the presence of large drops which can either fall out of the moist cloud air, freeze and perhaps shatter, or be preferentially accreted on falling ice crystals leading to multiplication.

Unfortunately, these are not the only problems. Nucleation processes are statistical, by virtue of the probability of a sufficiently large molecular arrangement forming, either in the bulk of the liquid, or on a solid surface. This randomness has been clearly demonstrated in the former case (Langham and Mason, 1958) amounting to a spread of about $1^{\circ} \mathrm{C}$ in freezing temperature. For solid nuclei, the effect has proved difficult to demonstrate explicitly because of the difficulty of reproducing a given spectrum of particles for identical freezing conditions. Even consecutive measurements on one drop cannot be uniquely interpreted in this way, as the freezing process may lead to changes in the particle size distribution (Hoffer and Braham, 1962). Experimental work has led to the concept of an activation temperature for a particular nucleus material and an exponential increase of activity with temperature below $0^{\circ} \mathrm{C}$, reflecting the increase of numbers of sites on the surface of the particles capable of nucleation.

Laboratory studies of the activity of natural and artificial ice nucleating substances by different workers have been characterized by lack of reproducibility of results. This is not surprising when we consider how many complications can exist during a test. Materials prepared in different ways - by
grinding, quenching from the vapor by chemical reaction, are subjected to different thermal and mechanical stress histories which lead to different surface and bulk structure - such as distribution of trace impurities and distribution of crystalline defects, to different size distribution and more surface area.

## NUCLEI DETECTION

However, subtle than these problems are those involving the exact method of test. These are best examined in light of the historical development of devices for counting ice nuclei which rely on growth of ice crystals to a size which can be detected either visually or by some more sophisticated technique. Concentrations of ice, crystals in clouds have been measured from $0.1 \ell^{-1}$ up to $800 \ell^{-T}$. The lower limit has coincided with the lower limit of detection by devices available and has approximated that which would lead to detectable fallout from cloud base. It is of interest to look at possible reasons for an upper limit in concentration. Cloud drops have a maximum concentration of $-1000 \mathrm{~cm}^{-3}$ and a mean separation of -1 mm . This can be interpreted in terms of the number of cloud nuclei activated in a specified updraft and mean supersaturation. Cloud drops grow quickly by condensation, a radius of $5 \mu$ being reached a few hundred seconds in a moderate updraft. For this situation the mean drop separations is -100 radii, so that the mean heat and vapor diffusion fields around each drop is influenced to about $1 \%$ by neighboring drops. Drops separated by less than this distance interact significantly with each other. In the case of ice crystals the situation is different. If we consider growth at water saturation, the effective supersaturation is up to 50 times greater, the crystals are linear hollow prisms or platelike dendrites and of low effective density so that their dimensions increase more rapidly than in the case of water drops. This leads to radii of order $50 \mu$ in the same time period and a mutual interaction distance of $\sim 1 \mathrm{~cm}$, equivalent to a concentration of $100 \ell^{-1}$. As this situation is reached, the supersaturation falls well below water saturation, leading to evaporation of all drops and a value of ice supersaturation inadequate for further ice nucleation, from the vapor (about $12 \%$ is required as a minimum). We may, therefore, regard 1000 crystals $e^{-1}$ as approaching on upperlimit. It follows then, that devices for counting nuclei must be designed to examine volumes not less than about ten litres and to measure crystal concentrations up to $1000 \ell^{-1}$.

The earliest attempt to investigate the presence of ice nuclei experimentally was made by Findeisen. He attempted to reproduce in the laboratory not only conditions which existed in the clouds, but also the rate of change of those conditions and to have an appropriate supersaturation. He used a chamber containing moist atmospheric air which was expanded and cooled at different rates to simulate natural updrafts of different magnitude. As it was used, this apparatus maintained supersaturation just above water saturation.

During the mid 1940's work at General Electric of Langmuir and Schaefer used the much simpler technique of a 'mixing chamber' - a cold box with a supercooled droplet cloud formed by adding warm moist air, the test air or material being added as the temperature became steady. This apparatus produced a supersaturation over ice and a slight subsaturation over water. Particles suddenly introduced were subjected
to a variable supersaturation depending on the humidity of the air in the sample and the details of the injection and mixing process.

Several simplified versions of the expansion system were subsequently developed. These were somewhat smaller than the original Findeisen chamber and in order to eliminate wall effects, were designed to have considerably more rapid expansion rates. This led to the disadvantage of short duration times for nucleation events. These devices, however, had the advantage of independently controlled temperature and saturation ratio.

While the thermal diffusion chamber has been used extensively in measurement of condensation nuclei, its use for ice nuclei has been somewhat limited. Whilst having the advantage of an independently controlled temperature and supersaturation, (including operation between water and ice saturation) the principles of its operation have precluded its use for measuring the lower range of concentration of ice nuclei - since only small volumes of aerosol may be injected without changing its diffusion equilibrium. However, this device, somewhat modified, has recently been in considerable use for studying the ice nucleation of particles caught on a millipore filter, (Stevenson, 1968). The general filter technique has the great advantage that nuclei from a large volume of air, up to 2000 litres - may be collected on 1" diameter filter paper. These may then be subjected to independently controlled temperature and supersaturation changes. Problems arise in two ways; the more numerous condensation nuceli may be caught along with the ice nuclei leading to a significant change in the cahracteristics of their surfaces. Secondly, working near water saturation may activate condensation nuclei and prevent a reliable attainment of slight water supersaturation. Table 1 gives a summary of these different methods.

## CONDITIONS FOR ICE NUCLEATION

This discussion of techniques reveals some interesting problems. The first question to arise is the extent to which the physics of the test nucleation process is related to any process which takes place in the atmosphere, with respect to temperature supersaturation history. The following situations may be distinguished. If the nucleus does not contain a hygroscopic component, it may pass through ice to water saturation, and then, at a rate depending on . its size, diffuse to the surface of a cloud drop and ultimately be incorporated within the drop, the diffusion process leading to an exponential fall in concentration with time constant typically $10^{3}$ seconds for atmospheric concentrations. If the nucleus contains both hygroscopic and insoluble component, there will be a critical water subsaturation ( $78 \%$ for NaCl , somewhat higher for other atmospheric salts, e.g., $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{SO}_{4}, 86 \%$ ) beyond which dissolution occurs. Depending on the relative mass and distribution of soluble and insoluble components, the liquid may or may not enclose the particle. If it should not, then there arises the question of whether the soluble part may migrate over the surface to form an effective surface coat, a process which could take a time comparable with the test processes. Eventually ice saturation will be reached, then for example, a NaCl solution drop at $-15^{\circ} \mathrm{C}$ would have the effective melting and nucleation temperature depressed by about $17^{\circ} \mathrm{C}$ (about 4 molar solution). As water saturation is reached, the drop dilutes and if the nucleus is

TABLE 1 CHARACTERISTICS OF ICE NUCLEI DETECTION SYSTEMS

| Instrument | Sensitive Time | Saturation Ratio | Limitations |
| :---: | :---: | :---: | :---: |
| Mixing | $10^{2} \mathrm{sec}$ | Just less than water saturation | Mixing leads to local supersaturation fluctuations; supersaturation not controlled. |
| Expansion (slow) | $\begin{aligned} & \text { Variable, to } \\ & 10^{3} \mathrm{sec} \end{aligned}$ | Water saturation | Wall effects - loss of water vapor, nuclei |
| Expansion (rapid) | 0.1 sec | Variable | Soluble particles fail to dissolve. Insufficient time for surface processes to equilibrate. Insufficient time for Brownian capture of small particles. |
| Diffusion | $10^{2} \mathrm{sec}$ | Variable | Size - large air volumes cannot be examined continuously to monitor low concentrations. Particles lost by fallout and diffusion. |
| Diffusion with millipore | $10^{4} \mathrm{sec}$ | Ice to slight water supersaturation | Sample contamination with Aitken nuclei, surface removal of vapor at water supersaturation. |

sufficiently large to be activated, grows to become effectively pure water. If it should not be activated, the drop still dilutes, and has a depression of its nucleation temperature between 0.4 and $4^{\circ} \mathrm{C}$ depending on the supersaturation present in the cloud. These drops are typically a few tenths of micron in diameter and are too large for significant Brownian capture by cloud drops and too small for coalescence to be of importance. The time scale of the cloud process may vary from -100 seconds, with a smooth increase in saturation ratio as a nucleus moves along a stream line in the smooth flow of a lenticular cloud to fractions of a second in the mixing of a cloud edge as ascending air penetrates a dry inversion or in the breaking of a wave with KelvinHelmholtz instability. These processes are summarized in Figure 1.

Laboratory experiments which aim to reproduce the above process have given a somewhat disconcerting divergence of nucleation temperatures depending on conditions. For illustration, the following examples are quoted:

Silver iodide requires water saturation to nucleate ice above $-12^{\circ} \mathrm{C}$; below this temperature $12 \%$ ice supersaturation is sufficient.

Particles embedded within drops have a lower nucleation temperature than those liable to Brownian capture at a drop surface.

The median freezing temperature of distribution of particles of AgI and kaolinite is lowered by $8^{\circ} \mathrm{C}$ when they are immersed in water.

The particle dissolves - freezing by homogeneous nucleation. AgI particles radius $0.01 \mu$ dissolve completely in a $10 \mu$ radius drop in a time -100 seconds.

Preactivation - montmorillonite becomes effective at $-4^{\circ} \mathrm{C}$ as opposed to -25 C at the first time of nucleation.

A few direct comparisons of ice nuclei counts and ice crystals concentrations in clouds have been carried out. Mossop (1968) measured nuclei in a $10 \ell$ mixing chamber mounted in an aircraft. His results showed a greater ice crystal concentration by a factor of almost $10^{3}$ at $-18^{\circ} \mathrm{C}$, decreasing to about 10 by $-28^{\circ} \mathrm{C}$. Although the air was heated to $+2^{\circ} \mathrm{C}$ before test, destroying any possible preactivation, his measurements appear to have been made under conditions where preactivation would have not persisted naturally. Hobbs (1969) and Auer et a1. (1969) measured nuclei in a mountaintop cloud with an NCAR counter. This produces a high supersaturation, with uncontrolled mixing in the pretest stage. Ice crystals concentrations were larger than nuclei count (extrapolated from values at -10 C ) by more than $10^{3}$ at $-5^{\circ} \mathrm{C}$, this ratio falling to about 10 by $-20^{\circ} \mathrm{C}$. In view of the above discussion, these results are open to criticism because the conditions of test - at any temperature differ significantly from the conditions of ice nucleation in the cloud itself.

## CONCLUSION

The direct application of any ice nucleus detector lies in its prediction of the number of ice crystals which will be produced in the atmosphere. From the practical standpoint, the cloud physicist is faced with a number of quite formidable problems. Having obained a nucleus count at a point in space and time, he must be reasonably sure that air from the trajectory that he has sampled is the one leading to the regions of atmosphere of interest - where the cloud is growing. He must be alert not only to the possibility that ice may be produced by homogeneous nucleation - a well predictable process - but also to the possibility of ice forming by a multiplication process, the details of which currently represents a serious gap in our knowledge.

The core question, however, is still whether, in the absence of the above problems, an ice nucleus counted gives one crystal in the cloud. The foregoing discussion has shown that we cannot assign a


Figure 1 Ice nuclei life history.


Figure 2 Cloud processes.
given nucleation temperature for a given nucleus there is not a unique nucleus spectrum of a given sample of air. Different physical condition (Fig. 2) - saturation ratio, temperature history, incorporation of the particles in or at the surface of drops give different temperatures of nucleation. Certain tests may yield a higher count than a typical atmospheric situation, for example, when a soluble ice nucleus fails to dissolve quickly in a rapid expansion. Conversely, other test procedures, for example, when all nuclei are centered within drops at high temperatures and subsequently cooled, may yield a lower count than the atmosphere.

The problem is therefore removed one step to our ability to simulate in our apparatus sufficiently closely those conditions of the cloud which we have found by experiment to influence ice nucleating behavior. This was the basic concept of Findeisen; the wheel has turned full circle with the current concept of chambers for atmospheric simulation. As far as comparing different ice nuclei counters, we only hope to compare those in which particles undergo similar saturation - temperature histories, only those where the particulates have comparable histories with respect to Brownian capture at the surface and incorporation within drops.

With respect to the carryover to the atmosphere, the counter must reproduce to a sufficient approximation the life history of the particles during their approach and incorporation into the cloud. Final justification for any technique will be a comparison of ice nuclei counts in air prior to entry into a particular cloud and the ice crystal concentration in the same air inside the cloud, carried out over a considerable range of cloud types and atmospheric conditions. It may ultimately turn out that the nucleation of ice is less sensitive to the conditions imposed by the trajectory and cloud dynamics than the evidence discussed here suggests; this is a hope however, which has to be shown rather than assumed.

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## SECTION IV

## WORKSHOP PROGRAM, PROCEDURES AND FACILITIES

Program Description<br>by<br>Joseph Podizmek

The program of the 2nd IWCIN in Fort Collins was prepared by the Organizing Committee on the basis of general recommendations of the Subcommittee on Nucleation ICCP IAMAP and on the basis of the experiences made during the "Nuclei week at the University of Minnesota" (May 18 to May 21). This pre-workshop session was held to explore sampling and testing procedures which could be employed during the workshop.

In order to reach the main goals the Organizing Committee, having responsibility for the success of the 2nd IWCIN, created the Steering Group, headed by J. Podzimek and having the members: U. Katz, K. Bigg, A. Gagin, R. Ruskin, G. Soulage. These men had the responsibility to:
a. Steer the work in the testing laboratory, including the preparation of aerosols to be used for testing of the apparatus. The following artificial nuclei were intended to be used:

AgI - thermal (3 s1zes, 0.01; 0.03; 0.10) Whitby
pyrotechnic (LW 83) - Katz, Whitby
$\mathrm{AgI}+\mathrm{NaI}$ - Acetone - Grant
$\mathrm{AgI}+\mathrm{NH}_{4} \mathrm{I}$ - Acetone - U.S. Navy
NaCl - Whitby
Phloroglucinol - Hoffer
Teflon - hydrophobic CN - Kocmond
b. Collect the descriptions of the apparatus presented during IWCIN
c. Collect and evaluate all data measured
d. Prepare the results of the 2nd IWCIN for publication (J. Podzimek).
e. Submit the proposals to the Organizing Committee related to the main goals of the IWCIN and their personal and financial support.

During the workshop the forms below were used for submitting the reports.

Before starting the program of the 2nd IWCIN the Organizing Committee discussed the following remarks which were submitted in a written form:

Prof. Kenji Isono, Nagoya: "On the scope of the 2nd IWCIN," 2 pp .

Dr. K. Spurny, Praha: "The use of pore filters in measurements of ice nucleus concentration," 18 pp .
Dr. G. Vali, Laramie: "Nucleation terminology," 3 pp.
Some of the recommendations and proposals were used in preparing the laboratory work, or communicated to those participants of the IWCIN who were mostly interested in similar kinds of measurements. The "Nucleation terminology" presented a subject of one evenings discussion.

Starting the shakedown period of the 2nd IWCIN on August 4, Dr. U. Katz presented to all the participants a description of the laboratory activities available in Fort Collins. On August 5 Dr . J. Podzimek gave a short description of the system of the collection and evaluation of data including the computation of the main characteristics of the apparatus to be tested.
a) Today's Program. A daily announcement including the following data:

b) The form for the reports on nuclei measurements including:
EXP. TYPE __ MACHINE \#__ DATE ___ OUTLET \# ON DUCT__ TIME OF BAG FILLING___ NUCLEI TYPE___NUCLEI SIZE____

ROOM TEMP. $\qquad$ ROOM REL. HUM. $\qquad$ ROOM PRESSURE $\qquad$


1) Room or Duct
2) Walls of mixing or expansion chambers. Base of filter chambers, Air Temp. NCAR counter
3) Cloud temp. for sample

## AEROSOL SAMPLE PREPARATION AND DISTRIBUTION SYSTEM USED DURING THE INTERNATIONAL WORKSHOP ON CONDENSATION AND ICE NUCLEI (IWCIN)

(This description was also presented in the opening session of the AMS Cloud Physics Conference, Fort Collins, Colorado, 24-27 August, 1970).

Ulrich Katz

Colorado State University, Fort Collins, Colorado 1)
Major prerequisites for a meaningful comparison of ice or cloud nuclei detectors are the proper choice of test aerosols, and, even more important, a system to distr bute this aerosol in such a manner that at a given t'me all instruments receive samples of the same characteristics. In addition, it is very desirable to provide samples of constant characteristics over a prolonged time in order to allow instruments to change experimental parameters during that period.

Final decisions on above points were made by the Organizing Committee after evaluating the experiences of a trial workshop at the Particle Technology Laboratory of the University of Minnesota. The following test materials were chosen:
a) Natural aerosol
b) NaCl , by evaporation
c) AgI, by evaporation
d) AgI from LW-83 pyrotechnics
e) AgI - $\mathrm{NH}_{L} \mathrm{I}$ - Acetone burned in Davis generator
f) AgI - NaI - Acetone burned in Skyfire generator
g) Phloroglucinol
h) Teflon

The atmospheric aerosol was an obvious choice since most instruments are designed to detect natural nuclei. However, clues for what causes differences in the reaction of the various counters can better be found by using test aerosols whose characteristics can be controlled to a certain extent. Prime representatives of that category are NaCl and AgI aerosolized by evaporation, and subsequently treated to result in the various desired size distributions. These well defined aerosols were vary valuable in assessing the characteristics of the various instruments, the hydrophyllic NaCl mainly for cloud condensation nuclei (CCN) counters, and AgI chiefly for the ice nuclei (IN) detectors.

Quite well defined with respect to material, but hardly controllable size wise are phloroglucinol and Teflon; the latter strongly hydrophobic substance was in powderous form and pneumatically dispersed. Phloroglucinol on the other hand is the representative of the organic ice nuclei; it was prepared by spraying and drying its alcohol ic solution according to Hoffer and Perthel (1).

1) Present affiliation: Cornell aeronautical Laboratory, Inc., Buffalo, V. Y.

A last group consists of three AgI aerosols as prepared in field projects, namely smoke from LW-83 flares (courtesy NWC, China Lake). AgI-NH 4 I - Acetone solution burned in a small smoke generator by Bryant Davis, and finally AgI-NaI-acetone $(2 \%-1 \%)$ solution burned in a Skyfire generator. With respect to material as well as size, these three preparations are not too well known; fuel and additives lead to very mixed particles, and the natural aerosol is present, too; burning fuel alone results in very similar size spectra.

Whereas above three AgI smokes were prepared in the vertical dilution tunnel, and then transferred in a 100 liter syringe into Bag A (see below), all other nuclei were produced in a closed system connected to Bag A.

For the following description of the sampling system one is referred to the schematics of Figs. 1 \& 2 .

The initially mentioned condition that every instrument must get a sample of the same kind could best be fulfilled by placing all counters along a 10 cm diameter sampling pipe through which a constant flow of sample air (several liters per second) is maintained. The length of the experiment room ( 14 m ) and the number of devices expected to participate (29) called for 15 pairs of 19 mm diameter outlets at intervals of about 90 cm . Wherever possible instruments were placed less than 50 cm from the sampling pipe in order to reduce aerosol losses. For this latter purpose the short length of the sampling pipe ( 13 m ) and sample velocities within the pipe ( 1.5 to $.5 \mathrm{~m} / \mathrm{s}$ ) were beneficial, and outweighed the general crowdedness. A preliminary analysis of results does not show any indication that instruments downstream would get samples of lower concentration.

In order to provide a sample over a period of about one hour a polyethylene bag of $54 \mathrm{~m}^{3}$ capacity (Bag B) served as storage container. This also solved two other problems:
a) Some instruments require ambient or positive pressure at their sampling inlet; although an atmospheric air sample could be brought to the counters by an upstream blower, this could cause considerable aerosol losses. Therefore, by evacuating a rigid tank built around the bag, the sample flows without obstruction into storage, and then, by pressurizing the tank, the sample is forced through the sample pipe.
b) In case of artificial ice nuclei, the concentration should be considerably below $1 \mathrm{~cm}-3$ for most instruments, whereas for the Minnesota Aerosol Analysing System (MAAS) concentrations considerably above $10^{3} \mathrm{~cm}^{-3}$ are necessary; therefore, a sample is prepared at a concentration suitable for MAAS, and put into a small ( $5 \mathrm{~m}^{3}$ ) nylon bag (BAG A). By adding a small known volume from Bag A to the clean air stream from fan-filler while filling Bag $B$, the dilution ratio can be determined much easier and better than by a dynamic dilution system.

Various measures were taken to exclude possibilities of inadvertent contamination of samples or instruments:

The fan-absolute filter combination No. 4 adds $20 \mathrm{~m}^{3} / \mathrm{min}$ of clean air at approximately 1 mm W.G over pressure to the experiment room.


Figure 1


Figure 2

It was requested that, whenever possible, instruments should be connected to the exhaust pipe which was held at about 30 mm W.G. below ambient pressure, and which would carry all spent samples into the absolute filter No. 3.

The blower No. 2 serves to pressurize ( 15 mm W.G.) or evacuate the tank around Bag B. To prevent contamination in the case of leaks in Bag B, another absolute filter was added (on Aug. 7) on the pressure side of this fan. The air cooler shown on the diagram chills the air pressurizing the tank. This seemed to help prevent chemical reactions in atmospheric air samples leading to high Aitken nuclei concentration.

During a few days, when evaporative AgI samples were used, a number of instruments, chiefly CCN counters were on a trial basis, fed with a high concentration sample directly from Bag A. A number of disadvantages, however, made this a very limited success. Bag A could not be pressurized enough to produce the necessary flow rate through the 20 mm copper pipe; also, losses along this narrow pipe were not negligible any more, and the volume of Bag A was used up too soon. The desirability of a complete dual system - high concentration and low concentration - for many experiments had been foreseen; however, lack of space and finances prohibited the realization at the large scale which has been proven necessary by the improvised version.

The choice of materials for the aerosol distribution system was mainly governed by findings at the Minnesota Particle Technology Lab. There were no indications that aluminum, rubber, and polyethylene would produce nuclei at room temperature. This was largely confirmed by the actual use of aluminum pipe, butyl rubber sleeves to connect them, rubber balloons as valves, and polyethylene for Bag B. The only difficulty arose from the use of adhesive tape as tanksealing material. Some component of the adhesive seemed to penetrate the polyethylene and to combine inside 3ag B with some constituent of atmospheric air to form extremely high concentration of Aitken nuclei. The problem of permanent contamination of the system by AgI samples turned out to be practically non-existent. However, as precaution all the atmospheric air samples experiments were done prior to any AgI test. Blank runs after the latter experiments showed that mere flushing of pipes and bag with filtered air cleaned the system sufficiently. The only reason why, as a precaution, Bag B was replaced after several days of use was a slow increase in Aitken counts which probably would not have been detrimental.

Since condensation inside the sampling system would lead to unacceptable losses of nuclei, the dew point of the samples had to be closely watched. Instead of monitoring the sample humidity and temperature directly in the sampling duct (as originally planned), the experiment room temperature and the temperature and humidity of the sample "source" (outdoors or air conditioned particle preparation room) were continuously measured. From the recorded values, it can be concluded that the sample dew point temperature was, in the case of natural nuclei, at least $5^{\circ} \mathrm{C}$ below the experiment room temperature (equals minimum temperature of samples prior to entering instruments), and much lower for artificial samples.

## ACKNOWLEDGEMENT

Many thanks go to Messrs. D. Dah1, C. Swain, and C. Carlson whose untiring assistance made it possible to have the above described system ready on time.

## REFERENCE:

Hoffer, T. E. and R. Perthel, 1968, J. Res. Atmosph. 1968, 203-206.

THE MINNESOTA AEROSOL ANALYZING SYSTEM
Particle Technology Laboratory Publication No. 148
(This description was also presented in the opening session of the AMS Cloud Physics Conference, Fort Collins, Colorado, 24-27 August, 1970).

## by

K.T. Whitby and R.B. Husar

## INTRODUCTION

The Minnesota Aerosol Analyzing System (MAAS) is a system of instruments designed to make rapid insitu measurements of aerosol particle size distribution and number concentration. As used during the International


Workshop on Cloud and Ice Nuclei it consisted of the following instruments operating over the given size ranges.

1. Single particle optical counter operating over the range from 0.6 to $12 \mu \mathrm{~m}$.
2. Whitby Aerosol Analyzer (WAA), an electrical aerosol analyzer, operating over the range from 0.0075 to $0.6 \mu \mathrm{~m}$.
3. Standard General Electric Condensation Nucleus Counter (CNC).

In Figure 1, the operating ranges of the three automatic particle counters are compared with selected properties of average atmospheric aerosols at the earths surface. It is seen that the MAAS covers the size range containing most of the aerosol number and mass. The MAAS is shown in Figure 2.

Following is a brief description of the system as used during the IWCIN, its calibration and some of the characteristics of the instruments that should be kept in mind when interpreting the data. Further details on the various instruments making up the system may be obtained from the following references:

Whitby and Clark (1) described the prototype instrument and its principle of operation. Whitby et al. (2) have described the present system and further developments in the WAA. Reference 3 is a detailed report describing the MAAS system used during an atmospheric research project in Pasadena, California during the summer of 1969. The system used there is essentially the same as that used during the IWCIN. This report was distributed to the workshop participants in a preliminary form and should be available for larger distribution by the fall of 1970.

## AEROSOL PLUMBING ARRANGEMENT

The MAAS and the Minnesota aerosol generating facilities were located in a first floor laboratory because of the large amount of space required and also to minimize contamination of the workshop laboratory by aerosol generation activities.


Fig. 1. Operating ranges of the components of the MAAS compared to some properties of average urban aerosol.


Fig. 2. MAAS in use at the Aerosol and Cloud Simulation laboratory. Left - Univ. of Minn. Condensation Nuclei Counter, Second from left - GE-CNC top and Optical Particle Counter bottom, Third from left - Whitby Aerosol Analyzer, and right - HewlettPackard data acquisition system.

The connection of the MAAS to the various aerosol sources is shown in Figure 3.

Direct sampling of outside atmospheric aerosol was through a 7.5 cm ID aluminum pipe projecting 3.6 m above the roof. Aerosol for the three instruments was distributed by short lengths of 1 cm ID rubber tubing. Two hundred twenty $1 / \mathrm{m}$ of aerosol was also drawn through the clean air circuit of the WAA and the humidity sensor to provide sufficient flow down the stack to transport the aerosol.

Aerosol from Bag B was sampled by connecting the line from the instruments to a 1.9 cm ID rubber tube that was connected to the aerosol distribution manifold in the workshop laboratory.

Aerosol from Bag A was obtained by connecting instruments to a 1.2 cm ID rubber tube connected to a copper tube inserted into the bag.

When the Minnesota CNC or the electrostatic sampler used for obtaining electron microscope samples was used, these instruments were connected into the system using the shortest possible amount of tubing between the source and the samplers.

## OPERATION OF THE MAAS

The three measuring instruments have been integrated into an automatic system under the control of a data acquisition system so that measurements can be made automatically and all data recorded in printed and punched tape form. For most of the measurements the system was allowed to run automatically on a 10 min cycle as shown in Figure 4 . During the first 7 sec the DAS measured channels $1,2,3$,


Fig. 3. Schematic of the Aerosol distribution system. Figures in parentheses are the approximate aerosol flows in the lines in liters/min.

10,11 , and 24, which were connected respectively to 1-solar pyroheliometer on the roof, 2-WAA temperature, 3 -sampling line relative humidity, 10 -wind velocity, 11 -wind direction and 24-GE-Condensation Nucleus Counter. The DAS then dwelt on channel 25 in the monitor mode and transferred control to the internal programmer in the WAA. The WAA then stepped through its program in approximately 4 min , recording each current reading on the DAS as it was completed. At the beginning of the cycle the DAS also initiated the 5 min count cycle of the OPC which was then recorded separately on the teletype and tape punch at the end of the five mincte count.

Although the MAAS was usually allowed to run automatically or the ten minute cycle, the aerosol sampling inlet for the entire system was switched from Bag A, to Bag B or to the outside atmospheric aerosol line as desired. This procedure permitted convenient time coordination of the records from the various instruments with the other data of the workshop.

Because it was essential to provide size distribution data to the workshop participants immediately after an artificial aerosol was generated and


Fig. 4. Sampling and recording intervals and their relation to the 10 min cycle times used.
delivered, a Hewlett-Packard 9100B electronic calculator was used to calculate some of the data immediately. The remaining results were calculated after our return to Minnesota.

The data sheet, with an example calculation, used for data reduction is shown as Table I. The upper part for the WAA gives the particle sizes, the corresponding voltages, and space for calculating $\Delta N / \Delta D_{p}$. The lower part provides space for calculating the results from the OPC data. The 123 channels of data are grouped into 14 groups for convenience and to fit the calculator capacity.

The MAAS consists of three measuring instruments and a data acquisition (DAS) which provides automatic control of the instruments and automatic digital data recording. The three instruments and how they have been calibrated are described below.

## GENERAL ELECTRIC NUCLEI COUNTER

The University of Minnesota GE-CNC was calibrated against an automated expansion type counter having the same optical path length as the Pollak counter. This U. of M. CNC was calibrated in 1963 against a standard Pollak and found to agree. More recently it has been compared with the WAA on a Nichrome wire aerosol which had an optimum size distribution for accurate counting by the WAA (e.g., $0.02<D_{\mathrm{D}}<0.3 \mu \mathrm{~m}$ ). The results of this comparison are shown in ${ }^{\text {Pigure }} 5$. It is seen that the theoretical calibration of the $U$. of M. CNC agrees with the numbers from the WAA within better than $\pm 50 \%$ over the concentration range from 10 K to 250 K per cc .

During the period when NaCl aerosol was being generated during the workshop, it was also found that the GE-CNC operated by Austin Hogan agreed within about 15\% or better with ours. Thus we believe that our GE-CNC agrees as well as can be expected with the


Fig. 5. Comparison of the number concentrations measured by the WAA with the Pollak calibration of the Univ. of Minnesota condensation nuclei counter. The size distribution of the aerosol produced by the heating of the Nichrome wire was in the optimum sizing range of the WAA. The theory assumes that all of the available water vapor is condensed onto the droplets.

TABLE I MINNESOTA AEROSOL ANALYZING SYSTEM (MAAS)
WHITBY AEROSOL ANALYZER
Aerosol Flow Rate: 0.32 cfm

| $D_{p}$ | $\Delta D_{p}$ | $\mathrm{D}_{\mathrm{pi}}$ | $\frac{\Delta N}{\Delta I}$ | $\frac{\Delta N}{\Delta I \Delta D_{p}}$ | Volts |  | I | $\Delta \mathrm{I}$ | $\Delta N$ | $\frac{\Delta N}{\Delta D_{p}}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| . 0075 | . 0025 | . 00875 | 487,000 | 195,000,000 | 210 | 0 | . 1248 | 0.0106 | 13250 | 5,300,000 |
|  |  |  |  |  |  |  |  |  |  |  |
| . 01 |  |  |  |  | 390 | 1 | . 1142 |  |  |  |
| . 015 | . 005 | . 0125 | 274,000 | 54,800,000 | 920 | 2 |  | . 0097 | 6178.9 | 1,235,000 |
|  | . 005 | . 0175 | 181,000 | 36,200,000 |  |  | . 1045 | . 0062 | 2126.6 | 425,000 |
| . 02 | . 01 | . 025 |  |  | 1,400 | 3 | . 0983 |  |  |  |
|  |  |  | 110,000 | 11,000,000 |  |  |  | . 0094 | 1532.2 | 153,000 |
| . 03 | . 01 |  |  |  | 2,410 | 4 | . 0889 |  |  |  |
| . 04 |  | . 035 | 75,700 | 7,500,000 | $3,240$ | 5 | . 0824 | . 0065 | 579.8 | 58,000 |
|  | . 02 | . 05 | 50,000 | 2,500,000 | 4,460 |  |  | . 0077 | 367.3 | 18,400 |
| . 06 |  |  |  |  |  | 6 | . 0747 |  |  |  |
|  | . 02 | . 07 | 30,000 | 1,500,000 |  |  |  | . 0088 | 235.8 | 11,800 |
| . 08 | . 02 | . 09 | 21,200 | 1,060,000 | 5,800 | 7 | . 0659 | . 0091 | 166.5 | 8,330 |
|  |  |  |  |  |  |  |  |  |  |  |
| . 1 | . 025 | . 1125 |  | 628,000 | 8,300 | 8 | . 0568 | . 0096 | 131.5 |  |
| . 125 |  |  | 15,700 |  |  | 9 | . 0472 |  |  | 5,260 |
|  | . 025 | . 1375 | 11,900 | 476,000 |  |  |  | . 0077 | 82.4 | 3,300 |
| . 15 | . 05 | . 175 | 9,200 | 184,000 | 9,250 | a | . 0395 | . 0148 | 123.4 | 2,470 |
|  |  |  |  |  |  |  |  |  |  |  |
| . 2 | . 1 |  |  |  | 12,300 | b | . 0247 |  |  |  |
|  |  | . 25 | 6,210 | 62,100 |  | c | 0058 | . 0189 | 109.4 | 1,090 |
| . 4 | . 1 | . 35 | 4,460 | 44,600 |  |  |  | . 0107 | 43.9 | 438.7 |
|  |  |  |  |  | 13,400 | d | -. 0049 |  |  | 438.7 |
|  | . 2 | . 5 | 3,140 | 15,700 |  |  |  | . 0000 | 0.0 | 0000 |
| . 6 |  |  |  |  | 14,000 | e | -. 0049 |  |  |  |

ROYCO PC-220/240 Sampling Time: $5 \mathrm{~min} .470 \mathrm{ccm} \quad N_{\text {tot }}=$

| Channe1 | $\mathrm{D}_{\text {pi }}$ | $\Delta \mathrm{D}_{\mathrm{p}}$ | $\overline{N_{\text {count }}{ }^{\Delta D_{p}}}$ |  | $N_{\text {count }}$ | $\Delta N=\frac{N_{\text {count }}}{2350}$ | $\frac{\Delta N}{\Delta D_{p}}$ |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 2 | .66 | .12 | .00355 | 0 | 4371 | 1.86 | 15.52 |
| 3 | .80 | .16 | .00266 | 1 | 1695 | .721 | 4.51 |
| 4 | .96 | .17 | .00250 | 2 | 893 | .380 | 2.23 |
| $5-6$ | 1.10 | .30 | .00142 | 3 | 982 | .418 | 1.39 |
| $7-8$ | 1.5 | .30 | .00142 | 4 | 483 | .206 | .686 |
| $9-11$ | 1.82 | .35 | .00121 | 5 | 386 | .164 | .467 |
| $12-15$ | 2.21 | .42 | .00101 | 6 | 303 | .129 | .306 |
| $16-19$ | 2.71 | .58 | .00073 | 7 | 186 | .079 | .136 |
| $20-29$ | 3.55 | 1.1 | .00039 | 8 | 199 | .085 | .078 |
| $30-39$ | 4.70 | 1.1 | .00039 | 9 | 126 | .054 | .049 |
| $40-49$ | 5.80 | 1.1 | .00039 | a | 79 | .034 | .031 |
| $50-69$ | 7.44 | 2.18 | .00020 | b | 90 | .038 | .018 |
| $70-89$ | 9.66 | 2.27 | .00019 | c | 52 | .022 | .010 |
| $90-126$ | 11.9 | 2.20 | .00019 | d | 40 | .017 | .008 |

G.E. COND. NUCLEI: $\qquad$
U. of M. COND. NUCLEI : $\qquad$
$\mathrm{I} / \mathrm{I}_{0}=$
other CNC on atmospheric and NaCl aerosols. However, there is evidence both from this workshop and from our previous work with the GE-CNC that there are sometimes large disagreements between the GE-CNC and other types of CNC on certain kinds of aerosols (and at high concentrations).

## WHITBY AEROSOL ANALYZER (WAA)

The aerosol size distribution in the diameter range between $0.0075 \mu \mathrm{~m}$ and $0.4 \mu \mathrm{~m}$ was measured with a slightly modified commercial model 3000 Whitby Aerosol Analyzer (WAA)*. In the present section, the principle of its operation is reviewed and the main components of the instrument are described, including the modifications made on the standard commercial instrument. Particular emphasis is placed on the calibration procedure and results since the latest calibration was more elaborate and detailed than the earlier ones. Finally, estimates on the accuracy of the WAA are given and in an example a sample calculation illustrates the determination of several aerosol parameters such as the number spectra, total number, etc.

## PRINCIPLE OF OPERATION

The electrical mobility of a charged particle, i.e., the ratio of the particle velocity to the force acting on it, is a function of the particle size. In a proper electrical and flow field, therefore, particles of different sizes may be classified according to their electrical mobility. For diffusion charging and for particles less than $1.0 \mu \mathrm{~m}$ in size (diameter) the electrical mobility is a monotonically decreasing function of the particle size and it varies only slightly at sizes larger than $1.0 \mu \mathrm{~m}$. Accordingly, for particles less than $1.0 \mu \mathrm{~m}$ in diameter, the electrical mobility is uniquely related to the particle size. If diffusion charged particles are then introduced into an electrostatic precipitator through a point or line source, the precipitation distance increases with increasing particle size.

The Whitby Aerosol Analyzer as described by Whitby and Clark (1) was developed starting with the ideas and findings as stated above. The objective of their work was to extend these findings and to combine them in such a way that the number of particles in different size ranges could be measured by properly varying some of the parameters of the system. The five years of work on the development of the instrument resulted in the following arrangement and measuring procedure.

The aerosol first enters a jet-charger, specially developed for this purpose, where the charging results from the fast mixing of uncharged particles with high concentration of negative ions (generated on a corona needle). The p pysical mechanism of charging is governed by molecular and turbulent eddy diffusion and in a small part by the existing electrical field in the charger vessel. A schematic of the WAA is shown in Figure 6.

As a result of the prevailing diffusion charging, the particles leaving the charger vessel are charged such that the number of elementary charges on a particle is approximately proportional to its diameter for $D_{p}>0.04 \mathrm{\mu m}$.


Fig. 6. Schematic of the WAA.

The aerosol leaving the charger is then introduced into the precipitating tube, as an annular cylinder surrounding a core of clean air. The aerosol collector, a metal rod, passes axially through the center of the tube. Depending on the voltage on the collecting rod, all particles with an electrical mobility exceeding a certain value will be collected on the precipitating rod, while those with smaller mobility (larger size) pass through the precipitation section and are then collected on an absolute filter.

The absolute filter itself is electrically connected to a sensitive electrometer, which measures the total current due to the charge given up by the particles to the collecting filter. A step increase of the rod voltage will cause particles of a larger size range to be precipitated with the resulting decrease of the electrometer current.

The number of particles $\Delta N$, in a given size range $\Delta D_{p}$, corresponding to the current charge $\Delta I$ may be calculated from the known values for the mean number of elementary charges per particle, the flow rate and the fraction of particles lost by space charge and diffusion.

[^0]The aerosol size distribution in the size range between $0.075 \mu \mathrm{~m}$ and $0.6 \mu \mathrm{~m}$ is obtained by scanning the precipitation voltage from 210 volts (for 0.075 $\mu \mathrm{m}$ ) to $15,000 \mathrm{~V}$ (for $0.6 \mu \mathrm{~m}$ ) in 15 steps, measuring the corresponding currents on the electrometer and then, using the calibration curve, converting the voltage vs. current curve to a discrete (14 point) size distribution curve.

To allow the new particle trajectories in the precipitation tube to stabilize, the times between two consecutive voltage steps must be kept sufficiently long. These times range from 30 seconds at the lowest voltage to 12 seconds at the highest voltage, the time required for the entire cycle being about four minutes.

The scanning cycle for a size distribution measurement is controlled by an internal timing system that also commands a digital readout for the current before each sten to a higher voltage. With a digital voltmeter connected to the electrometer of the WAA, the size distribution measurement can be fully automated and the system is then suitable for monitoring time-dependent aerosol size distributions.

MODIFICATIONS MADE ON THE STANDARD COMMERCIAL INSTRUMENT

The main components of the WAA were subjected to extensive experimental and numerical investigations as well as to changes in design and operating conditions, before the instrument was used in the Los Angeles Smog project in the summer of 1969.

First, in a numerical investigation, Whitby et al. (2) (1969) Section IV, the flow field and particle trajectories in the precipitator section of the WAA were studied with the ultimate objective of reducing the geometrical and hydrodynamical distortion effects on the quality of size classification. As a result of this numerical study, the ratio of the aerosol and clean air flow rates was reduced from $1 / 3.1$ to $1 / 14$. This reduced the standard deviation of the mobility classification from $\sigma_{\mathrm{g}}=1.4$ to $\sigma_{\mathrm{g}}=1.18$.

In Section $V$ of the same report, Whitby et al. (2) (1969), the results of experimental studies on the performance of the WAA are discussed, with particular emphasis on the effect of aerosol flow rate and the aerosol concentration on the resolution of the instrument. The optimal ratio of aerosol/clean air flow was found to be $1 / 14$ at a total flow rate of $2.12 \mathrm{1} / \mathrm{sec}(4.5 \mathrm{cfm})$. The aerosol concentration was found to affect the level and quality of the charging only slightly, if the total current due fo the deposited charged aerosol was between $2 \times 10^{-12}$ and $50 \times 10^{-12} \mathrm{amp}$.

In a further series of experiments (as yet unpublished) aerosol losses due to diffusion and space charge in the charger were investigated. The results indicated that the losses in the charger depend strongly on the particle size and on the aerosol flow rate, decreasing in magnitude with increasing particle size and increasing flow rate. For particle size of $0.08 \mu \mathrm{~m}$ for instance, the aerosol penetration through the original 221 charger were found to be $20 \%$ at an aerosol flow rate of $0.15 \mathrm{1} / \mathrm{sec}$ (optimal flow rate for the classification) and increased to $40 \%$ at a flow rate of $0.5 \mathrm{l} / \mathrm{sec}$.

A significant reduction of the aerosol losses in the system was achieved by replacing the charger vessel of the standard instrument which had a volume of 22 liter, with a much smaller, 1.7 liter charger vessel. The corresponding penetrations for the same particle size of $0.08 \mu \mathrm{~m}$ but with the smaller charger vessel were $58 \%$ at an aerosol flow rate of $0.15 \mathrm{l} / \mathrm{sec}$ and increased to $70 \%$ at $0.5 \mathrm{l} / \mathrm{sec}$. The increase in the aerosol penetration is attributed to the reduction of the aerosol residence time in the small vessel. In a highly recirculating flow field, such as the jet-driven recirculation in the larger charging vessel, the aerosol is forced several times to the vicinity of the walls before it leaves the vessel. In the small vessel, this contact time is reduced, by that reducing both the diffusion and space charge losses. The jet entrance and the primary aerosol mixing section of the charger remained unchanged. The new cylindrical charger vessel is 7.5 cm in diameter and 34.0 cm long with its axis coinciding with the jet axis. The aerosol exit is placed radially on the cylindrical vessel, 3.0 cm from the jet entrance.

There were also several minor changes and adjustments made on the electrical circuit of the standard model 3000 TSI instrument.

First, it was found that the stepping of the lower to a higher voltage on the collecting rod occurred simultaneously with the read command to the digital voltmeter. This caused a distorted current reading. This interaction was eliminated by inserting a time delay of about 0.2 seconds between the read command and the subsequent voltage stepping.

In the intermittent cycling mode, the instrument automatically scans through the 15 voltages, and then returns to zero voltage and then awaits the pulse from the digital clock to start a new cycle. In a further modification, a switch was installed which permitted the selection of either a zero or 15 K voltage, for the time period when the instrument was standing by awaiting the pulse for a new cycle. With the added choice of 15 Kv in the stand-by position, the instrument could be used for size distribution measurement, let's say every hour (for four minutes that is needed for the scanning) while for the rest of the time it would be classifying the aerosol on the rod. This mode permits then the correlation of size distribution data with those obtained by classification.

The scanning times between the steps 1 through 6 were found to be too short, so that the correspond ing currents were not stabilized before the next voltage step occurred. Accordingly, the time between first and second step was prolonged from 23 seconds to 30 seconds, then the step between 2 and 3 from 20 to 25 and those between 3 and 6 from 15 to 22 seconds. The stepping times for the remaining steps were unchanged at 12 seconds.

The stepping voltages were adjusted according to the newly calculated calibration curve discussed in the following paragraphs.

## CALIBRATION OF THE WAA

With the main components of the WAA restudied and the necessary modifications made, new operating conditions were chosen and the instrument was recalibrated.

The calibration required the determination of three independent quantities as a function of particle size, namely, the electrical mobility $Z_{p}$, the fraction of particles lost in the system, and the fraction of particles carrying a charge (for small particles only). It was attempted to obtain as much of these data as possible experimentally. For particle sizes less than $0.05 \mu \mathrm{~m}$ in diameter this was a rather difficult task since several of the experimental procedures had to be develoned for this particular work.

THE ELECTRICAL MOBILITY $Z_{p}$ AS A FUNCTION OF THE PARTICLE DIAMETER $D_{p}$

The electrical mobility $Z_{p}$ may be determined in at least two different ways: 1) using monodisperse aerosols of known size, the electrical mobility may be calculated from the precipitating voltage required for that particular size, or 2) running the WAA in classification mode with polydispersed aerosols and then calculating $Z_{p}$ from the precipitation distance on the rod. Because of the lack of a reliable aerosol generator for $D_{p}<1.0 \mu \mathrm{~m}$, the earliest mobility determinations for the WAA were nerformed using the second, more tedious approach. A recently developed condensation aerosol generator, Liu, Whitby, Yu, (1966) (5), provides monodisperse DOP particles in the size range between $0.06 \mu \mathrm{~m}$ and $1.3 \mu \mathrm{~m}$ (depending on the DOP concentration) and therefore for the present work the first approach was chosen.

The aerosol leaving the condensation aerosol generator was diluted to sufficiently low concentration so that the total current on the electrometer at zero voltage or the rod would not exceed about 10-11 Amp. A current vs. voltage curve was then obtained. The median voltage was chosen as the characteristic voltage, i.e., at which the current dropped to $50 \%$ of the value at zero voltage. This procedure was repeated for several different aerosol sizes, and the electrical mobility $Z_{p}$ was calculated using the formula (1), Whitby, Clark (1) (1966).

$$
\begin{equation*}
z_{p}=\frac{q_{t}\left(r_{2} / r_{1}\right)}{2 \pi L V} \tag{1}
\end{equation*}
$$

with $q_{t}$ the total flow rate (clean air and aerosol), $r_{1}$ and $r_{2}$ the analyzer tube radius and rod radius, respectively, $L$ the rod length and $V$ the median voltage.

The resulting mobilities are presented in Figure 7. Because, as is indicated in Figure 7, the electrical mobility at $D_{p}>0.6 \mu \mathrm{~m}$ decreases only slightly with particle size, the useful upper size limit was chosen to be 0.6 m . At small sizes, $\mathrm{D}_{\mathrm{p}}<0.06 \mu \mathrm{~m}$ the electrical mobility approaches the single charged particle mobility. In between these regions, the particles are multiple charged, carrying elementary charges roughly proportional to their diameter. For the present calibration, no experiments were performed with particles with $D_{p}<0.08 \mu m$, because no smaller monodisperse particles could be produced for which the size was known accurately enough.

The mobility of single charged particles is well established since it assumes only a value for the Cunningham slip correction.


Fig. 7. Electrical mobility versus particle diameter calibration for the WAA for two different calibrations and for the operating conditions and configuration described in the text.

## FRACTION OF PARTICLE CHARGED

The electrical mobility curve describes the mobility of the particles as a function of size, including those small particles which carry only a single elementary charge. When such small particles are charged in the diffusion charger, however, a fraction of those particles will escape the charger vessel without acquiring a single charge, i.e., without colliding with a negative ion.

Since for the size distribution measurements, the concentration of particles in a given size range is calculated from the current due to the particles giving up their charge, it is of basic importance to know what fraction of particles are carrying an elementary charge.

The fraction of particles charged was determined by passing monodisperse aerosol through the analyzer and then measuring the concentration of particles penetrating to the collecting filter, just ahead of the filter. The concentration was then measured twice, first with no voltage on the rod so that the particles, charged or uncharged, would penetrate to the collecting filter, and the second time with high voltage on the rod so that all the charged particles would be collected and only the uncharged would penetrate to the collecting filter. The ratio of the number of particles penetrating the analyzer section with high voltage on the rod to the total number at zero voltage is the fraction of particles that are not charged.

The most difficult part of the calibration procedure was to find proper monodisperse aerosols in the size range around $0.01 \mu \mathrm{~m}$. Fast diluted combustion generated aerosols were found to serve surprisingly well for this purpose. A home type propane torch, for instance, produces 0.0055 to $0.006 \mu \mathrm{~m}$.
aerosol reproducibly. Another generator used for the calibration of the WAA was a heated nichrome wire spiral, immersed previously in salted water ( NaCl ). Varying the concentration of NaCl in the water, the generated particle size could be varied between 0.01 and $0.06 \mu \mathrm{~m}$.

The aerosol prior to entering the analyzer, was diluted and well mixed in a $4.5 \mathrm{~m}^{3}$ plastic bag. All of the concentrations were measured with a General Electric condensation nuclei counter. Since for the calibration only relative concentration measurements were required, only the linearity of the instrument's reading was important rather than the absolute concentration it indicated.

For larger size particles ( $\mathrm{D}_{\mathrm{p}}>0.06 \mu \mathrm{~m}$ ) the Condensation Aerosol Generator (Liu, Whitby, Yu (5)) (1966) was used with DOP aerosol. Pure DOP, being hydrophobic, could not be detected by the GE nuclei counter, but minute amounts of the strong wetting agent Triton X-100 ( $1 \%$ of the amount of DOP) added to DOP increased the hygroscopicity of the DOP particles so that practically all the particles were detected by the GE nuclei counter. Since the condensation aerosol generator is provided with internal diluters, the aerosol generator was connected directly to the WAA.

The monodispersity of the aerosol was tested by running a current I vs. voltage $V$ curve before every test. The steepness of the I(V) curve was taken as a measure for the monodispersity of the aerosol.. If the I(V) curve for a run was not steep enough, the run was rejected and a new aerosol was generated. The particle size was determined using the median voltage to calculate the mobility according to formula (1) and with the known mobility from Figure 7 the particle size was obtained.

## diffusion and space charge losses in the system

Simultaneousiy with the experiments for the evaluation of the fraction of particle charged, the aerosol losses were also determined. In a similar manner as described in the previous paragraph, the concentration of monodisperse particles was measured at the aerosol entrance to the WAA, position 1, Figure 5, after the charger, position 2, and then at the collecting filter, position 3 . The ratio of the concentrations 1 and 3 , corrected by the dilution ratio for the clean air core, gave the fraction of particles penetrating through the system, P. This is plotted in Figure 8. The ratio of the concentrations 1 and 2, also corrected by the fraction of clean air introduced by the jet, indicated the losses in the charger only.

The aerosol losses on the walls of the system were caused mainly by Brownian and turbulent eddy diffusion as well as by the space charge of the unipolarly charged mixture of aerosol particles and ions. The two mechanisms for the aerosol losses were detected separately by first measuring the concentrations at positions 1, 2 and 3 , without charging (diffusion losses) and then with the charger operating, which gave the results for the total losses, diffusion and space charge together.

In the following, the steps are listed which were taken in order to obtain an experimental point for the penetration and for the fraction of particles charged:


Fig. 8. Penetration and fraction charged measurements for the WAA. $P$ gives the fraction not lost by diffusion in the analyzer. The fraction charged, $f$, gives the fraction that will receive a charge in the charger and $K$ is the factor by which the observed number at a given size must be divided to obtain the true number.

1. The plastic bag was filled with well mixed monodisperse aerosol.
2. The monodispersity of the aerosol and its size was checked by running an I vs. V curve.
3. Concentration $C_{1}$ of position 1 was measured by the GE nuclei counter.
4. With charger off, concentrations $C_{2}$ and $C_{3}$ were measured. $C_{1}$ and $C_{2}$ provided the diffusion losses in the charger whife $C_{1}$ and $C_{3}$ gave the diffusion losses in the entire system (collector rod voltage zero).
5. With the charger on, concentrations $C_{2}$ and $C_{3}$ were again measured, and the total losses (diffusion and space charge) were obtained at both positions after the charger and at the current collecting filter.
6. Concentration $C_{3}$ was again measured with high voltage $(20 \mathrm{Kv})$ on the rod. The ratio of this concentration to $C_{3}$ at no voltage on the rod gave the fraction of particles not charged.

The separation of the diffusion and space charge losses makes the assumption that the two mechanisms are mutually independent. This assumption is not strictly correct, but for the determination of K itself the separate contributions are not important as long as the total loss is known and it can be measured accurately. As a matter of interest, most of the losses occurred in the charger section (high turbulence level and recirculation) and diffusion losses were always larger than the space charge los ses.

## EVALUATION OF THE CALIBRATION CONSTANTS

With a knowledge of the previously discussed parameter, namely the electrical mobility, fraction of particles charged, and the fraction of particles
lost in the system, one can calculate the constants which are used to transform the current vs. voltage curve into a particle size distribution curve.

The fraction of particles penetrating the system, $P$, and the fraction of particles charged, $f$, may be combined into a single correction factor K , where $K=f \cdot P$. The true differential current $\Delta I$ between two voltage steps is therefore $\Delta I_{m} / K$, where $\Delta I_{m}$ is the measured current. The number of particles $\Delta N$, in a discrete size range (corresponding to a voltage range) is then calculated from the relationship:

$$
\begin{equation*}
\Delta N=\frac{\Delta I_{m}}{K q_{a} n_{p} e} \tag{2}
\end{equation*}
$$

where $q_{a}$ is the aerosol flow rate, $n_{p}$ is the number of elementary charges in the size range under consideration, and $e$ is the unit electron charge. The experimental calibration data for the penetration p , the fraction charged and the calculated K , are shown in Figure 8.

## WAA ACCURACY AND RESOLUTION

Knowledge of the sizing accuracy and the size distribution resolution capabilities of the instrument are necessary to the interpretation of the data. However, it is not easy to specify either quantity for the WAA for reasons which will be explained here.

From Figure 7 it is seen that the slope of the $Z_{p}$ vs. $D_{p}$ curve goes from about zero at a $D_{p}$ of about $1.3 \mu \mathrm{~m}$ to -2 at $\mathrm{D}_{\mathrm{p}}$ 's below about $0.01 \mathrm{\mu m}$. Thus the sizing resolution is essentially zero at $D_{p}$ 's greater than $1 \mu \mathrm{~m}$. As was explained earlier, the measured $\sigma_{g}$ of the I vs. V curve for a $1.3 \mu \mathrm{~m}$ monodisperse aerosol was found to be 1.18. Since the size dispersion of the aerosol would not affect the $\sigma \mathrm{g}$ of the I vs. V curve at this size significantly, this means that the inherent resolution of the instrument at this size plus the dispersion of the particle charge must be less than this figure. As is explained by Whitby et al. (2) (1969), the $\sigma_{g}$ of the residual size distribution is estimated to be about 1.04 so that the inherent mobility resolution of the instrument is 1.15 . If the dispersion of the particle charge were constant with particle size it would be a relatively straightforward matter to calculate the resolution as a function of particle size using the $\sigma_{\mathrm{g}}$ of 1.18 and the curve of Figure 7. However, the charge dispersion increases with decreasing particle size until below about 0.01 H all particles carry only a unit charge at which size the charge dispersion is zero. The problem is to determine how it varies in between, e.g., from about 0.01 to $1 \mu \mathrm{~m}$. Theoretical and experimental work now in progress is being carried out to provide answers to this question. Indications are that the maximum charge $\sigma \mathrm{g}$ is 1.4 at a particle size of about 0.03 $\mu \mathrm{m}$. However, from Figure 7 it will be noted that the slope is approaching -2 at this size. Putting all of this together we have estimated that the poorest resolution corresponds to a $\sigma_{\mathrm{g}}$ of about 1.25 up to a particle size of about 0.3 above which the $\sigma_{g}$ of resolution increases to infinity at about $1.3 \mu \mathrm{~m}$. At particle sizes around 0.01 and 0.1 the resolution will be better than 1.25, perhaps as small as 1.1.

It may be concluded that over the size range in which the WAA was used in this study ( 0.0075 to 0.4 $\mu \mathrm{m})$, the resolution is quite adequate to resolve the aerosols measured.

There are similar problems in estimating the accuracy of the calculations of $D_{p}$ and $N$. The measurements of air flow, collector voltage and electrometer current are quite accurate and are not the limiting factors in estimating the accuracy. Rather the limiting factor seems to be the accuracy of the determination of $K$ for particle sizes below about $0.02 \mu \mathrm{~m}$ and the distribution weighting effects of the decreasing size resolution above about $0.3 \mu \mathrm{~m}$.

From Figure 8 it may be seen that $K$ varies from a maximum of 0.75 at $0.4 \mu \mathrm{~m}$ to 0.06 at $0.0075 \mu \mathrm{~m}$, the smallest size measured in this project. Since the absolute accuracy of the determination of $K$ is on the order of $\pm 10 \%$ for the larger sizes and $\pm 20 \%$ for the smaller sizes it is seen that the accuracy of the calculation of $\Delta N$ is about $10 \%$ for $D_{p}>0.1 \mu \mathrm{~m}$, $20 \%$ at $0.04 \mu \mathrm{~m}$, and $100 \%$ at $0.01 \mu \mathrm{~m}$ and $200 \%$ at $0.0075 \mathrm{\mu m}$. While this means that probable error at the smallest sizes measured is perhaps a factor of two in concentration, it must be remembered that the relative concentrations over the four orders of magnitude of particle size measured in these experiments vary by 6 to 8 orders of magnitude. Thus the instrument still provides useful information at the smallest sizes.

## OPTICAL PARTICLE COUNTER (OPC)

The OPC used a modified Royco 200 right angle sensor. The standard sensor has been modified by incorporating a passive sheath air inlet arranged so that although the total flow is $2.81 / \mathrm{m}$ the actual aerosol flow is $.471 / \mathrm{m}$. The pulse amplitudes have also been increased by more careful alignment of the optical system. In the modified form an ideally monodisperse aerosol would be indicated as having a standard deviation of about 12\%. This is adequate for the measurement of atmospheric aerosols and the artificial aerosols being generated during the workshop.

Pulses from the sensor are analyzed by a HewlettPackard 5400 A pulse height analyzer into 123 channels covering the size range from 0.6 to $12 \mu \mathrm{~m}$. These are recorded by a standard teletype printer and paper punch.

The sensor was calibrated over the size range from 0.5 to $2.9 \mu \mathrm{~m}$ using polystyrene latex. The calibration for larger sizes was based partially on a calibration with 6-14 $\mu \mathrm{m}$ latex and on the theoretical curves. The calibration will only be strictly correct for perfect spheres of 1.6 refractive index. For irregular particles, Whitby and Vomela (4) have shown that the effect is to spread out the indicated distribution without changing the indicated size much. This effect would not be of any great significance for the size distributions measured during the workshop.

The effect of refractive index differences between the aerosols measured and P.S.L. is more difficult to predict because it depends very much on size. However, as is explained in (3), if the refractive index was actually 1.5 instead of 1.6 the effect would at worst be to cause the particle size to be indicated about $1 / 3$ too small. Perhaps the best indication of the fact that the WAA and the OPC are not off more than a factor of two where they overlap is the fact that in all cases where there are sufficient counts to expect agreement the two curves pair together smoothly.

The OPC can count at most about 20 per cc without serious error. This limit is more due to the limitations of the pulse converter and to the spurious production of pulses by the statistical adding of subcountable particles than it is to coincidence. However, the concentrations of natural and artificial aerosols in the optical range measured during the workshop have been low enough so that there have been few problems.

## DATA PRESENTATION

The MAAS measures the number size distribution of an aerosol. This may be presented in a number of ways depending on the way in which the data is to be used. Since cloud and ice nuclei counters basically measure the number of particles larger than some particle size we have chosen to present the data in the form of cumulative number greater than size calculated to the concentration in the particular system from which the nuclei counters are drawing their samples. Whenever the concentration in Bag B was high enough to measure with the WAA, the measurement was made directly from the system. However, for the AgI runs the concentration in Bag B was so low that it could not be measured directly by the WAA. In that case the size distribution was measured in Bag A before dilution and it was assumed that the size distribution was not changed significantly by dilution or by storage in Bag B. This assumption is reasonable for all except the very small aerosols with mean sizes below $0.01 \mu \mathrm{~m}$ where diffusion would perhaps remove more of the smallest particles than the largest.

During the various experiments with artificial and atmospheric aerosols, all or portions of the size distributions in Bag B were monitored to determine the changes in the size distribution and concentrations of the aerosols with time. Figure 9 is a comparison of size distribution of an atmospheric aerosol measured from the Bag B distribution manifold at several different times with the size distribution measured during the filling of Bag B but sampled directly from the roof by the 3 inch line shown in Figure 3. It is seen that in the size range from 0.15 to $1 \mu \mathrm{~m}$ there was very little change. Above $1 \mu \mathrm{~m}$ there was a factor of 2 or 3 decrease in concentration during the run. It is impossible to determine the losses for sizes below $0.15 \mu \mathrm{~m}$ because of the formation of chemical aerosol in Bag B.

Figures in the section on aerosol generation show the number concentration in the 0.6 to $0.72 \mathrm{\mu m}$ range for several artificial aerosols over 1 hr periods.

From all of these data it may be concluded that the changes in the concentration in Bag B over a one hour period are about a factor of $2.5 \mathrm{at} 1 \mu \mathrm{~m}$, a factor of $1.5 \mathrm{at} .1 \mathrm{\mu m}$ and a factor of 3 for sizes smaller than $0.01 \mu \mathrm{~m}$.

For some of the runs artificial aerosols were also delivered directly from Bag A via a 2.5 cm ID copper tube to the cloud condensation nuclei counters in the upstairs lab. Because of the smaller size of the plumbing involved and the small volume of Bag A, the losses with respect to time were several times those for Bag $B$. The magnitudes of these losses can be estimated from the CCN and CiNC data measured in the upper lab.


Fig. 9. 'Illustrative particle size distributions of atmospheric aerosol measured at different times in Bag B for run 11. The large difference between the curves for the measurements in Bag B compared to the direct measurement on outside aerosol is due to chemically formed aerosol in Bag B.

For all of the AgI aerosols delivered to Bag B , the size distribution was measured in Bag $A$ just before a 2 or 3 liter sample was withdrawn for transfer to Bag B. The cumulative number distribution was then calculated from the Bag A cumulative number distribution using the appropriate dilution factors.

## THE CUMULATIVE NUMBER DISTRIBUTION

There are many ways of presenting particle size distributions. The cumulative number distribution was chosen because whenever a cloud or ice nucleus counter is sensitive to particle size it will be sensitive to the number of particles that are larger than some critical size or to a function which contains the integral above some given size.

The cumulative number distribution is calculated from the data by summing the $\Delta N$ 's in Table I and plotting the result as shown in Figure 10. For the artificial aerosols the data was submitted both in table form and as graphs similar to Figure 10.


Fig. 10. Atmospheric aerosol presented in cumulative number form.

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## MEASUREMENT OF SIZE DISTRIBUTION OF AEROSOLS BY ELECTRONMICROSCOPY

Aerosols were sampled from Bag A on meshes coated with collodion, using an electrostatic precipitator. The electronmicroscopic pictures were taken at NCAR. The electronmicroscope was a Carl Zeiss Model EM9A (Accelerating voltage, 60 Kv ; Beam Current, 6 and 0.5 $\mu \mathrm{A})$. The pictures for the measurement of size distribution were taken at a magnification of 5800 and were then enlarged to a final magnification of 20000. The diameter of a particle was defined by the average of the maximum and minimum dimensions.

## GENERATION OF ARTIFICIAL AEROSOLS

> Particle Technology Laboratory Publication No. 149

## by

## K.T. Whitby and R.B. Husar

## INTRODUCTION

A number of artificial aerosols were generated by Whitby and Husar during the IWCIN. The purpose of this report is to describe the technique and to make a few comments on characteristics of the different aerosols.

The thermal generation technique that was used during the workshop was chosen as the best compromise between reasonable monodispersity and the necessity to generate a wide range of concentrations and sizes on a very tight time schedule. Although more sophisticated methods that can produce more monodisperse size distributions are available, these were deemed to be impractical under the conditions of the workshop.

The requirements for monodispersity are not as critical if the cumulative number distribution is
known accurately, since whenever a cloud or ice nucleus counter is sensitive to particle size it will be sensitive to the number of particles that are larger than some critical size, or to a function which contains the integral above some given size. Thus, because the MAAS was capable of quite accurate measurement of the cumulative number distributions, the somewhat heterodisperse size distributions produced by the aerosol generators were reasonably satisfactory.

## GENERAL EXPERIMENTAL ARRANGEMENT

The general experimental arrangement is shown in Figure 1. A small amount of the substance to be aerosolized (AgI or NaCl ) is placed in a glass tube that has been preflamed to drive off absorbed contaminants. The tube is then heated while filtered air flows over the salt. The volatilized salt condenses into particles in the air stream, coagulates in the coagulating tube and then either passes directly into the clean air stream going into bag $A$ or else passes through the glass bead diffusion tube before entering the balloon. The diffusion tube is used to remove very small particles when monodisperse aerosols larger than a few hundredths of a micron are desired. The particle size and concentration in bag A can be adjusted by varying the amount of salt used, the air flow rate, heating rate and duration, the length of the coagulating tube and the diffusion tube.


Fig. 1. Schematic of the thermal generation apparatus used to generate most of the artificial aerosols.

Because the Whitby Aerosol Analyzer (WAA) requires that the number concentration of the aerosol be in the range from 5 K to $100 \mathrm{~K} / \mathrm{cc}$, the concentration in bag A is adjusted to this range. After measurement with the WAA the aerosol is allowed to flow through the transfer flask until it is filled with aerosol at the concentration in bag A. The flask is then quickly transported to the entrance tube of bag B and flushed into the entering clean air stream with clean compressed air. For these experiments this resulted in a concentration in bag $B$ of $1.2 \times 10^{-5}$ times that in bag $A$. In some experiments with NaCl the whole contents of $A$ were flushed into B.

Before introducing the aerosol into bag $B$, it was ordinarily inflated to about $3 / 4$ of its capacity. The remaining $1 / 4$ was introduced to convey and mix the aerosol.

After the aerosol was mixed in bag B, it was allowed to flow through the distribution manifold to the various instruments.

Following are some comments on the generation of particular aerosols.

## SODIUM CHLORIDE

Sodium chloride aerosol was prepared by heating approximately $0.5 \mu \mathrm{~m}$ of reagent grade NaCl in the tube. The salt is first fused without airflow through the tube and then the aerosol generated by reheating the salt for the appropriate time with the appropriate air flow. Exact air flows used and heating times and intensities were not recorded but rather the whole process was controlled by watching the rise of the current on the WAA and the count on the GE CNC. Small particle sizes were generated by using high airflows, brief but intense heating, a short coagulating tube and no diffusion bed. Large sizes were generated using very low airflow rates, prolonged and intense heating, long coagulating tube and the diffusion bed to remove the small particles. A flow rate of approximately $11 / \mathrm{min}$ for 0.05 and $40 \mathrm{l} / \mathrm{min}$ for $0.01 \mu \mathrm{~m} \mathrm{nmd}$ aerosol was used.

Since the NaCl was generated primarily to test the Cloud Condensation Counters, the whole contents of bag $A$ were flushed into bag B in order to obtain high enough concentrations in bag $B$.

For some of the runs, samples for electron microscopy were taken directly from bag $A$ using the ThermoSystems Model 3100 electrostatic aerosol sampler.

## SILVER IODIDE

Silver Iodide Aerosols were prepared from powder supplied to us by Uhlrich Katz by the same technique described above for NaCl . Since the concentrations desired in bag B were several orders of magnitude lower, only two to four liters of aerosol were transferred by flask from bag $A$ to the filling line of bag $B$.

Because the concentration of AgI in bag B was less than the residual concentration of non-active ice nuclei particles, and less than the reliable threshold of all but the INC, the only independent method for arriving at the concentration and size distribution of the ice nuclei in bag B was to calculate it from the concentration and size distribution in bag A multiplied by the appropriate dilution factors. It is difficult to assess the accuracy of this procedure by any direct method. However, there are several indirect indicators which may be used to arrive at an estimate.

First, all of the volumes involved in the dilution factor are known well enough so that the maximum error in the dilution factor is certainly less than a factor of two. Second, measurement of the particle concentration in bag B with the optical counter during several runs when the concentration of particles in the optical range was high enough to give a valid count (including run 41 on AgI), gave a maximum change in relative count over a one hour period of about 3 for the 0.6 to $0.72 \mu \mathrm{~m}$ size range. Since the decay rate for particles smaller than $0.6 \mu \mathrm{~m}$ but larger than $0.01 \mu \mathrm{~m}$, would be smaller,
it is estimated that the maximum error in the calculated concentration of AgI in bag B is less than a factor of three and probably a factor of two if a correction for the aerosol decay is made.

Decay curves of the concentration in the 0.6 to 0.72 m range for different aerosols are shown in Figures 2 and 3 .


Fig. 2. Decay of aerosol concentration in bag B as measured by the OPC, in the 0.6 to $0.72 \mu \mathrm{~m}$ size range. Data for the 1.33 to $1.66 \mu \mathrm{~m}$ size range for teflon is also shown.


Fig. 3. Decay of aerosol concentration in bag B as measured by the OPC, in the 0.6 to $0.72 \mu \mathrm{~m}$ size range.

NaCl SIZE DISTRIBUTION VARIATIONS WITH TIME IN BAG B
Of interest is the variation in the size distributions and concentration in Bag B of the artificial aerosols that were generated. These were not easy to measure for most runs because the concentrations were too low for the Whitby Aerosol Analyzer to measure accurately. For all of the AgI runs and for most of the NaCl runs the concentration was below that for which accurate size distributions could be measured.

However, NaCl runs 29 and 30 were sufficiently high in concentration so that the size spectra could be measured with the WAA every 10 minutes for the entire period of the run.

The cumulative number greater than size and the Condensation Nuclei Counter count for these runs are tabulated in the attached tables and the first and last size distributions are plotted in the Figures for the runs.

From tables 1 and 2 and figures 2 and 3 it will be seen that there was little change in the aerosol during the 50 minutes duration of these runs. Further, it will be noted that the number concentration calculated from the WAA is in satisfactory agreement with the G.E. CNC counter data. Although the optical counter data is included, the counts were so low that the results are not significant.

THE CHANGES OF CUMULATIVE (GREATER THAN) DISTRIBUTIONS FOR NaC1 AEROSOL IN BAG B

August 13, 1970


THE CHANGES OF CUMULATIVE (GREATER THAN) DISTRIBUTIONS FOR NaCl AEROSOLS IN BAG B
Run 30
August 13, 1970

| $D_{p}-\mu \mathrm{m}$ | 1420 | 1430 | $11 / 40$ | 1450 | 1500 | 1510 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $\begin{aligned} & .0075 \\ & .01 \\ & .015 \\ & .02 \\ & .03 \\ & \hline \end{aligned}$ | $\begin{aligned} & 4310 \\ & 4310 \\ & 4310 \\ & 3930 \\ & 2840 \end{aligned}$ |  | $\begin{aligned} & 4570 \\ & 4570 \\ & 4570 \\ & 4160 \\ & 2840 \\ & \hline \end{aligned}$ | $\begin{aligned} & 4720 \\ & 4720 \\ & 4720 \\ & 4720 \\ & 3040 \\ & \hline \end{aligned}$ | $\begin{aligned} & 4.10 \\ & 4.410 \\ & 4.10 \\ & 3 / 10 \\ & 2580 \\ & \hline \end{aligned}$ | $\begin{aligned} & 4670 \\ & 4670 \\ & 4350 \\ & 3740 \\ & 2530 \end{aligned}$ |
| $\begin{aligned} & .04 \\ & .06 \\ & .08 \\ & .1 \\ & .125 \\ & \hline \end{aligned}$ | $\begin{array}{r} 1990 \\ 1050 \\ 547 \\ 282 \\ 147 \\ \hline \end{array}$ | O | $\begin{array}{r} 1960 \\ 797 \\ 456 \\ 259 \\ 127 \\ \hline \end{array}$ | $\begin{array}{r} 1450 \\ 608 \\ 289 \\ 14.6 \\ 94.0 \\ \hline \end{array}$ | $\begin{array}{r} 1710 \\ 84,1 \\ 404 \\ 181 \\ 102 \\ \hline \end{array}$ | $\begin{array}{r} 1640 \\ 778 \\ 373 \\ 183 \\ 99 \\ \hline \end{array}$ |
| .15 .2 .3 .4 .6 | $\begin{gathered} 89.6 \\ 35.4 \\ .04 \\ .04 \\ .04 \end{gathered}$ | 알 | $\begin{gathered} 75.8 \\ 32.4 \\ .02 \\ .02 \\ .02 \end{gathered}$ | 45.1 <br> . 02 <br> . 02 <br> .02 <br> .02 | $\begin{aligned} & 66.4 \\ & 27.2 \\ & .01 \\ & .01 \\ & .01 \end{aligned}$ | 57 $\begin{aligned} & .00 \\ & .00 \\ & .00 \\ & .00 \end{aligned}$ |
| $\begin{aligned} & .6 \\ & .72 \\ & \hline \end{aligned}$ | $\begin{aligned} & .02 \\ & .01 \\ & \hline \end{aligned}$ |  | $\begin{aligned} & .012 \\ & .008 \\ & \hline \end{aligned}$ | $\begin{array}{r} .01 \\ .01 \end{array}$ | $\begin{aligned} & .01 \\ & .01 \end{aligned}$ | $\begin{aligned} & . \infty \\ & . \infty \\ & \hline \end{aligned}$ |
| $\begin{aligned} & .88 \\ & 1.02 \\ & 1.3 \\ & 1.66 \\ & 2.02 \\ & \hline \end{aligned}$ | . 01 |  | $\begin{aligned} & .007 \\ & .006 \\ & .005 \\ & .003 \\ & .0009 \end{aligned}$ | $.01$ | $\begin{aligned} & .01 \\ & .007 \\ & .005 \end{aligned}$ |  |
| $\begin{gathered} 2.46 \\ 3.13 \\ 4.12 \\ 5.55 . \\ 6.6 \\ 8.55 \\ 10.78 \end{gathered}$ |  |  | $\begin{aligned} & .0009 \\ & .00 a_{4} \\ & .0004 \\ & .0004 \\ & .0004 \\ & .0004 \\ & 0 . \end{aligned}$ |  |  |  |
| G.E. Cond. Nuclei Counter | 4700 | -- | 4500 | 4400 | 4200 | 4100 |

## SECTION V

## DESCRIPTION OF INSTRUMENTS OPERATED BY PARTICIPANTS

A. Aitken Nuclei
$\frac{\text { THE POLLAK "PHOTO-ELECTRIC CONDENSATION }}{\frac{\text { NUCLEUS COUNTER MODEL } 1957 \text { WITH }}{\text { CONVERGENT LIGHT BEAM" }}}$
by


The instrument operated at the workshop is a copy of the classic Pollak Aitken nuclei counter. The fog tube is positioned vertically, 58.74 cm long, 2.54 cm diameter, with electrically surface tinted glass plates at top and bottom. The walls are lined with a wetted porous ceramic lining that provides the moisture supply to the air sample. A light beam from the top of the chamber traverses the tube to a photo cell at the sottom. The chamber is ventilated with a $2 \ell \mathrm{~min}^{-1}$ air sample flow for 60 sec , than an overpressure ratio of 1.21 of clean air is applied at the longitudinal center of the tube. After a 60second conditioning period, the air is allowed to expand to ambient air pressure, from the center of the tube. Attenuation of the photocell current has been calibrated in terms of the concentration of Aitken nuclei in the air sample. The Pollak counter is capable of measuring the Aitken nuclei concentration from less than 10 to 250,000 nuclei $\mathrm{m}^{-1}$.

This particular instrument, ESSA \#920, was calibrated in Dublin, Ireland, and is now used only in direct comparison and calibration procedure with other Aitken nuclei counters.


Fig. 1. The Pollak photo-electric condensation nucleus counter Model 1957.

## THREE COMMERCIAL AITKEN NUCLEI COUNTERS

Description of Instruments
by
A. W. Hogan ASRC SUNY, Albany, N. Y.

## INTRODUCTION

Three Aitken nucleus counters, all of current U.S. Commercial manufacture, are used to measure the total aerosol concentration in the diluting and sample bags. These instruments are all of the photo electric type, base their calibrations on the POLLAK 1957 counter and its calibration and operate at the Aitken expansion of 1.2 to 1 , in an attempt to activate all aerosol particles as condensation nuclei.

## DESCRIPTION OF INSTRUMENTS

1) The Gardner Associates Small Particle Detector, Type CN (1) - This instrument is a portable battery powered instrument based on the instrument described by RICH (1955). It utilizes a fog tube 39 cm long by 2.5 cm diameter with a lamp and photocell located concentrically at opposite ends by 0 ring seals, the inside of the fog tube lined with blotting paper to provide a moisture supply. It is manually aspirated in the field, or by an accessory pump in the laboratory.

The instrument can be obtained with an engraved logarithmic scale face, or with a uniform 100 division scale, accompanied by an individual calibration curve, based on a comparison with a Pollak counter. The calibrated range of the instrument is from $200 \mathrm{n} / \mathrm{cm}^{3}$ to around $1 \mathrm{million} \mathrm{n} / \mathrm{cm}^{3}$. Because of the compressed scale and the finite time required to perform an observation ( 30 to 60 sec ) the instrument is most accurate over the range of $1000 \mathrm{n} / \mathrm{cm}^{3}$ to around $150,000 \mathrm{n} / \mathrm{cm}^{3}$, which covers the general range of continental aerosol.

This instrument has proven reliable in field and laboratory use, and has been successfully employed in Oceanic surveys over the Atlantic Ocean.
2) The General Electric Condensation Nucleus Counter ( $\overline{C N C}$ ) (2) - This instrument is a large ( 60 x $60 \times 60 \mathrm{~cm}, 50 \mathrm{~kg}$ ) instrument designed for laboratory use described by Scala (1963). It operates at five (5) expansions per second, giving a pseudo "continuous" record of nucleus concentrations.

The instrument compares the ratio of light scattered by the chamber before the expansion to the light scattered by the fog in the chamber after the expansion, utilizing a dark field patch in the light path, and a photo multiplier. Because the reference signal is the light scattered by the fog free chamber, this chamber, and the lenses at the light source and photocell end must be kept clean for optimum performance. The fog chamber, purging valve, and humidifier are monobloc, insuring a uniform temperature throughout the system.

This instrument is calibrated over the range of zero to ten million nuclei per $\mathrm{cm}^{3}$, with linear scales to $100,000 \mathrm{n}^{\mathrm{cm}}$, and two logarlinic scales above. When the fog chamber and its associated optics are clean, the instrument has a threshold of 50 to $100 \mathrm{n} / \mathrm{cm}^{3}$. The upper logarithmic scales are quite cramped, but because of the fast flow ( $100 \mathrm{~cm}^{3} / \mathrm{sec}$ ) of this instrument, are quite useful in determining peak values in laboratory experiments.

While this instrument was originally designed for laboratory use, it has proven quite useful in observatory studies (Reiter) of Natural aerosol concentrations.
3) The Environment One Corp. Condensation Nucleus Monitor, Model Rich 100 (3) - The Environment One counter is a relatively new instrument, not as yet described in scientific or engineering literature. The instrument used at the workshop is a prototype portable unit, ( $40 \times 40 \times 15 \mathrm{~cm}, 15 \mathrm{Kg}$ ), operating on 12 VDC power. Similar units are available for 115 V 60 Hz operation.

The instrument uses a small fog tube, about 1 cm in diameter by 15 cm long, in tight contact with a humidifier housing. The fog is sensed by a photo resistor, measuring the attenuation of light transmitted from a lamp at the opposite end of the tube. The lamp and photo resistor can be removed quickly to allow cleaning of the tube. An expansion is performed once per second in the fog tube and the signal stored electronically to provide a pseudo continuous record.

The instrument has linear scales to $300,000 \mathrm{n} / \mathrm{cm}^{3}$ and a single nonlinear scale above $300,000 \mathrm{n} / \mathrm{cm}^{3}$.

The prototype unit has shown some difficulties with low concentrations, the effective threshold being somewhere around $600 \mathrm{n} / \mathrm{cm}^{3}$. At higher concentrations, the instrument tracks with the other nucleus counters.

Two features of this instrument are continuous flow, which can be varied about the base level of $50 \mathrm{~cm}^{3} / \mathrm{sec}$ with a built in flow meter, and a full scale adjust which facilitates laboratory diffusion loss experiments.

## CONCLUSIONS

The three commercial Aitken nucleus counters tracked well, and were invaluable in monitoring the total aerosol concentration in the test chambers during the workshop program. Specific information on these instruments can be obtained from the manufacturers listed in the Appendix.

## APPENDIX

1. Gardner Associates, 3435 Carman Road, Schenectady, New York 12302.
2. General Electric Company Electronic Systems Division, Ordnance Systems, 100 Plastics Avenue, Pittsfield, Massachusetts 01201.
3. Environment One Corporation, Balltown Road, Schenectady, New York 12302.

UMR ABSOLUTE AITKEN NUCLEI COUNTER: DEVELOPMENTS SINCE 1967 AND OPERATING PROCEDURE

## by

J.L. Kassner, Jr., V.K. Saxena, A.H. Biermann

Graduate Center for Cloud Physics Research
University of Missouri - Rolla

## INTRODUCTION

The UMR Absolute Aitken Nuclei Counter consists basically of a Wilson cloud chamber which has been used in this laboratory for various studies (1-12). Since 1967 two refinements have taken place which are briefly described here.

## NEW FEATURES OF UMR ABSOLUTE NUCLEI COUNTER

The diagram of the cloud chamber operated as condensation nuclei counter is displayed in Fig. 1. Figure 2 exhibits a photograph of the counter in operation at our laboratory. It is felt that this counter constitutes a precision standard against which other field instruments can be evaluated and calibrated. Moreover, it provides a means of studying the aerosols in more detail than can be accomplished with simpler field instruments inasmuch as a wide range of operating supersaturations can be precisely obtained. In Table 1, the specifications for the counter are given. It may be mentioned that this counter is capable of measuring concentration once in each 5 minute interval.


Fig. 1. Drawing of cloud chamber.


Fig. 2. UMR Absolute Aitken Nuclei Counter General View.

The counter is now equipped with a new vent valve which has proved to be perfectly leak tight and a new programmer-timer which has been found to be more reliable for controlling the functions of the counter.

## VENT VALVE

The design of the new valve is shown in Fig. 3 which differs (4) from the old one (11) inasmuch as
a new section is added in order to insert a rubber diaphragm. The latter provides a positive seal for the valve stem and prevents the passage of the gas along it without hindering the motion of the stem. The diaphragm flexes slightly as the stem moves through its paces. In this design, it is also possible to vary the spring constant which controls the pressure on the 0 -ring sealing the orifice to the expansion chamber. This orifice lies at the bottom of an inverted conical frustrum at the base of the valve.


Fig. 3. Vent in valve.

The new design also improves the geometry of the flush line which purges the volume in the vicinity of the valve stem. A slant inlet directs the sample flow towards the orifice. All adjustments can now be made on the valve without ever exposing the orifice to atmospheric air. Plumbing connections to the valve are $0-$ ring sealed.

## ANALOG PROGRAMMER TIMER UNIT

This unit is responsible for the precision with which the automation of the cloud chamber cycle is carried out. It controls every function (11) of the counter. Automation of the counter is largely dependent on the reliability of this unit. This is what led us to build a new programmer timer.

Design parameters for the new timer were determined by experience gained during the past three years. Some of the ideas employed in the old timer did not prove useful. The stepper-relays were omitted because they generated noise and were too slow in performing certain functions. The line-frequency-dependent mechanical clock was also replaced because it was inaccurate and generated noise. The diode-matrix function-output board was condensed into

1. Supersaturation range:
2. Counting technique:
3. Accuracy:
4. Concentration range:
5. Operating principle:
6. Frequency of measurement:
7. Expansions per hour:
8. Data output:
9. Data processing:
10. Power requirements:
11. Number of units:
12. Physical Size:

Chamber:
Programming Unit:
Recorder:
Pressure Gauge:
13. Camera:
14. Film:
15. Drop counting:
$24 \%-280 \%$, subject to easy adjustment.
Direct photography.
$\pm 10$ percent when the drop counts are around 100 per cc.
Capable of operating at any concentration down to 100 per cc due to inherent dilution of the test sample.

Adiabatic expansion.
5 min .
24, approximately; main expansion is followed by the clearing expansion each time.
(i) role of the film shot during the experiment furnishes the record of the drop counts (ii) the $8^{\prime \prime}$ wide, direct print paper rolled out by the chart recorder gives the record of the pressure before and after expansion, volume of the expansion, and the volume of the sample vented in, (iii) the temperature of the chamber can be read in terms of the divisions of the meters mounted on the front panel.

For quick processing, the use of the WANG electronic calculator or any other digital computer is recommended.

115 volts, 60 cycles, 1.8 kilo-watts.
Three independent units, viz. (i) the main chamber, (ii) the programming unit, and (iii) the Model 800 direct-recording oscillograph supplied by the Midwestern Instruments. In addition, a Model 145 pressure gauge supplied by Texas Instruments is used for calibration purposes. An air tank maintained at 30 psi is used to pressurize the chamber and a suitable vacuum tank for expanding it.

| Length | Width | Height |
| :---: | :---: | :---: |
| $30^{\prime \prime}$ | $30^{\prime \prime}$ | $43^{\prime \prime}$ |
| $22^{\prime \prime}$ | $19^{\prime \prime}$ | $31^{\prime \prime}$ |
| $21^{\prime \prime}$ | $19^{\prime \prime}$ | $9^{\prime \prime}$ |
| $16^{\prime \prime}$ | $15^{\prime \prime}$ | $9^{\prime \prime}$ |

Motor-driven Nikon camera used with Micro-Nikkor auto 1:3.5, $f=55 \mathrm{~mm}$ lens; Shutter speed $1 / 30$ to $1 / 60 \mathrm{sec}$.
Kodak Linagraph Shellburst Easter Base 35 mm film.
Manual counting using mechanical counter.
several plug-in printed circuit boards. The new programmer has its own power supply which is mounted inside its chassis because the programmer timer's circuitry was vulnerable to external noise generated in other circuitry.

Certain ideas in the old timer proved to be beneficial and were incorporated into the design of the new timer. The same sequential times were experimentally found correct. External multipole relays with series-connected contacts were used because they reduced gap-arcing when controlling DC inductive loads such as the control valves. The general operating procedure of the old timer was also kept. Solidstate timing units are the basis for the new timer. The solid-state timing units consist of a unijunction and a S.C.R. combination which produce linear timing, give a high noise rejection, and are virtually temperature independent.

Instead of using the stepper-relay as the main controller which allowed the solid-state units to come briefly into the timing cycle, all solid-state units are used in a "round-robin" fashion. This arrangement is cyclic, more logical, and leads to easier design, layout, and construction of the timer. The cyclic timing is beneficial in decreasing the timing error.

A parallel-"T" phase shift oscillator is used as the time base for the interval time. This interval timer, in conjunction with the integrated circuits and numitron readouts, replaces the old line-frequency-dependent mechanical clock.

The new timer has an advantage over the old timer inasmuch as the time interval on each individual timer can be adjusted without having to wait for the timer to go through a complete cycle. The timer can
be manually set to go through any step or combination of steps desired for calibration or to check proper functioning.

The replacement of the diode-matrix, the clock, the stepper, the bell, and the excessive wiring with the solid-state circuitry and printed circuits eliminated the problems of magnetic fields, noise, and ground loops which interfered with the recording of galvanometer's traces.

The new timer greatly improves the performance of the equipment and is easier to operate.

## OPERATING PR.OCEDURE

The counter is set on an automatic cycle which performs the desired functions. A qualitative representation of this cycle is shown in Fig. 4 which represents the pressure in the chamber as a function of time. The pressure and time axes are not drawn to scale but rather to depict the necessary details in the chamber. The total time for one complete cycle is about five minutes which is realized in twenty different steps as shown in Table 2. Eight different supersaturations are realized by varying the opening-duration of the fast expansion valve. These are numbered as A through H in Table 2. The details of the functions operating during one complete cycle of the counter have already been discussed by Kassner et al. $(10,11)$. We would therefore, restrict ourselves to describing the routine operating procedure.

The counter cycle begins at Step \#14 when the piston of the chamber is bottomed out and the pressure in the chamber is below atmospheric pressure. The recorder bias is pre-calibrated and is set so that the fine pressure trace before venting in the test sample, is recorded. This is accomplished in Step \#15. During Step \#16, the vent valve (Fig. 3) opens for a set duration and the test sample is vented in. The time interval in Step \#16 is realized through a difference timer as indicated in Table 2. It may be varied in the range from less than 0.1 sec to 100 sec . Step \#17 provides sufficient time for the sample to get thermally stabilized and during \#18, the fine pressure trace is again recorded. The volume of the chamber therefore, remains constant during the venting in process and the pressure change measures the size of the test sample. The latter is an important factor in producing the desired sample - dilution. This counter is capable of handling very large concentration because of its dilution feature. The chamber stores about 15,000 cc of clean air for mixing with the test sample.

During Step \#19, the fast compression valve compresses the chamber to ready pressure. At Step \#20, the control of the piston is taken over by the servo motor through a servo bias which keeps regulating the ready pressure (Fig. 4). The regulation is achieved at different pressures through different servo biases. At Step \#1, the ready pressure and volume traces are recorded which represent position G of Fig. 4. During Step \#2, the fast expansion valve opens and $A$ represents the pressure after

TABLE 2 TIME INTERVALS AND FUNCTIONS DURING ONE COMPLETE CYCLE OF THE COUNTER

d, represents the difference time interval obtained from two timers.
$t$, these time intervals are adjustable from the front panel of the timer unit.


Fig. 4. Qualitative representation os the cloud chamber program.
expansion where the servo system holds the pressure constant at a slightly higher value in order to realize a well defined minimum. The fully developed droplets are now photographed and at Step \#7, the servo brings back the chamber to its ready pressure. During Steps \#1 to 4, the volume and pressure traces are recorded which help estimate the volume in the expanded position of the chamber and the pressure before and after expansion.

During Step \#8, the fast expansion valve again opens and a saturation ratio of about 3.8 is realized which is just short the value required for condensation on ions. This expansion is designated as clearing expansion in Fig. 4 which, indeed, is motivated to clean the chamber of residual nuclei (8). The servo regulates the pressure during Steps \#9 through 13 above atmospheric pressure. At Step \#11, the vent valve opens and a desired volume of air is expelled from the chamber. This operation is intended to maintain the pressure constant before vent in, which in turn governs the size of the sample intake. The time during which the vent valve opens for ventin and vent-out processes can be controlled through potentiometers mounted on the front panel of the timer unit.

The size of the sample vented in depends upon the pressure before vent-in and the time duration for the valve opening. The volume of the chamber stays constant during this process while during vent-out the pressure in the chamber is kept constant. Thus, thermodynamic coordinates of the chamber differ in vent-in and vent-out processes and therefore, for bringing the chamber to the same pressure after each cycle, different time settings for the openingduration of the vent valve are required.
$\mathrm{ff}, \mathrm{Pb}$ and Pa respectively represent the pressure in the chamber before and after venting in the sample. The nuclei concentration can be calculated as:

$$
N=\frac{n}{\left(P_{a}-P_{b}\right)} \frac{V_{e} e_{i}}{V_{c}} \frac{P_{r}}{T_{r}}
$$

Here $n$ is the number of drops per $c c, V_{c}$ and $V_{e}$ respectively are the total volume of the chamber and its volume after expansion; $T_{i}$ and $T_{r}$ are respectively the initial temperature of the sample in the chamber and in nuclei reservoirs; and $\operatorname{Pr}$ is the pressure in the nuclei reservoir which stores the sample. The saturation ration at which $N$ nuclei are activated can be calculated by knowing $\mathrm{T}_{i}$ and pressures before and after expansion.

## CONCLUSIONS

The refinements made in the UMR Absolute Aitken Nuclei Counter since 1967 have made it more versatile and reliable. The counter is capable of measuring concentration in a wide range because of its inherent sample-dilution feature. The direct photography is employed for measuring the concentration and the overall accuracy of the data is governed by the error in drop counts.

## ACKNOWLEDGMENTS

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## B. Cloud Condensation Nuclei

A DESCRIPTION OF THE ESSA-APCL PORTABLE THERMAL DIFFUSION CLOUD CHAMBER

## by

## P. Allee, ESSA, Boulder

The design of the ESSA-APCL thermal diffusion cloud chamber for counting cloud droplet condensation nuclei (CCN) is based upon that described by S. Twomey (Journal de Recherches Atmospheriques, 1963, pp. 101106).

The cylindrical chamber is 3 in . in diameter and 1 in . deep. All walls and top and bottom are lined with blotter paper that is wet during chamber operation. The base of the instrument is a sheet of Micarta, $16 \mathrm{in} . \times 16 \mathrm{in} . \times 5 / 8 \mathrm{in}$. The bottom of the chamber is a cylindrical block of solid aluminum $3 \frac{1}{4} \mathrm{in}$. high. The wall of the chamber is made from 1 in . thick plexiglass, open to a 3 in . diameter to form the aerosol chamber. Four holes are drilled through the chamber wall: (1) To let the light into the chamber that illuminates the droplets. (2) To let the illuminating light out of the chamber so there will be no reflection from or heating of the wall of the chamber. (3) An aperture to view and photograph the droplets with a reflex camera. (4) A hole behind the view seen by the camera that has a flat black paint at the far end to form a dark field against which the droplets are viewed. The angle between holes (2) and (3) is $70^{\circ}$. The holes that pass light are covered on the outside of the chamber with sections of microscope slides. The metal frames holding the glass to the chamber also hold tiny electric bulbs (G.E. No. 328, 6 v.) that furnish enough heat to keep the glass from being fogged by water vapor on the inner surface. The plexiglass is covered with black plastic electricians' tape to exclude extraneous light from the cloud chamber.

The top of the chamber is a copper block, topped with Micarta that holds nichrome heating coils so that the top surface of the chamber is warmer than the lower surface. "0" ring channels are cut in the aluminum and copper block surfaces and "0" ring gaskets are used to seal these blocks against the plexiglass chamber. Two thermocouples are buried, one in the aluminum block, one in the copper block, close to the blotter paper surfaces. One serves as a reference and a Weston galvanometer Model 440 (quite ancient) serves to measure their potential difference. One-half degree $C$ increments in temperature difference between the top and bottom surfaces of the chamber can easily be determined from the galvanometer.

A small diaphragm vacuum pump is used to draw the air sample into the chamber through holes in the aluminum block base.

The light source for illuminating the droplets is a G.E. No. 14936.5 v bulb. A resistance is normally kept in series with the bulb to prolong its lifetime. When a picture of the droplets is made a shorting switch across the resistor allows the bulb to attain its full light output.

The power supply for the bulbs and heating coils is a 6.3 v.a.c. filament transformer.

The light from the illuminator bulb passes through a 5.0 cm focal length, 3.0 cm aperture, f 1.4 camera lens and is so located that the image of the filament is focussed at the center of the chamber.

The camera is a Leica equipped with a reflex housing to view the droplets, so that the optimum time for taking the picture of the droplet concentration can be judged.

The film used is fine grained, ASA 400, and is developed in DK-11, a high contrast developer. G. Langer at NCAR is using a film with an ASA 2000 rating, but film as sensitive as this requires special handling in the dark room to prevent fogging.

The controls, visible on the accompanying photographs are: (1) LIGHT. On-off switch for illuminating light, with the BRIGHT push button above it to increase the light intensity at the time of photographing the droplets. (2) HEATER. On-off switch for the nichrome wire heater coil in the top of the chamber. A variable resistance controls on the front of the box adjusts the current to the heater coils so that a stable operational temperature can be maintained from $\Delta T=0^{\circ} \mathrm{C}$ up to $\Delta T=12^{\circ} \mathrm{C}$. (3) PUMP. On-off switch is located in the center of the top of the control box. The closing of the stop cock valve in the inlet tube to the chamber is sometimes necessary to avoid turbulence wi thin the chamber, especially when the chamber is hooked into a line carrying a continually moving air sample. (4) WINDOWS. On-off switch to control the small bulbs that keep the windows free of condensed moisture.

One other structure on the base is visible in the forward right hand corner. When the chamber is put on "standby", the top is stored on a circular plexiglass box that contains a very wet sponge. This allows heat to the to to be left on during the standby period of time without drying out the blotter paper lining on the top. Simultaneously the plexiglass
box top is placed over the chamber to keep the blotter paper there in a moist condition.


Fig. 1. ESSA-APCL Portable Thermal Diffusion Cloud Chamber.

CORNELL AERONAUTICAL LABORATORY
THERMAL DIFFUSION CHAMBER
by
W. Kocmond, CAL, Buffalo


The CAL thermal gradient diffusion chamber has been used for making measurements of cloud nuclei since about December, 1964 (1-4). The basic design of the chamber is patterned after that of Langsdorf, Wieland and also Twomey (5-7). Photogranhs of the chamber in its present configuration are shown in Figs. 1 and 2.


Fig. 1. CAL thermal diffusion chamber and temperature control unit.


Fig. 2. Chamber configuration during operation.

In brief, the unit consists of a cylindrical plexiglass chamber with upper and lower water reservoirs, a collimated light beam to illuminate a small volume within the chamber, and a polaroid camera for photographing droplets that have formed on condensation nuclei.

During operation, water vapor diffuses from the warmer upper surface to the lower reservoir, with the chamber supersaturation being a known function of temperature difference between the two reservoirs. A series of 10 thermocouples (five on each surface) is

WATER DROPLETS FORMED AT 0.3\% SUPERSATURATION 1050 DROPS $/ \mathrm{cm}^{3}$


WATER DROPLETS FORMED AT $0.9 \%$ SUPERSATURATION 4200 DROPS $/ \mathrm{cm}^{3}$


WATER DROPLETS FORMED AT $3.0 \%$ SUPERSATURATION 5000 DROPS $/ \mathrm{cm}^{3}$


Fig. 3. Enlarged photographs of droplets formed in thermal diffusion charber.
used to measure $\Delta T$. When the desired supersaturation has been achieved, an air sample containing nuclei to be investigated is drawn into the chamber at a continuous rate for several seconds. The air sample is allowed to reside in the supersaturated environment where, in a few seconds, droplet growth proceeds on the most active condensation nuclei. The growing droplets are illuminated by a 200 watt 0 sram lamp and photographed at $90^{\circ}$ to the light beam moments before sedimentation begins (this can be easily estimated after some experience in using this instrument). Enlarged photos of droplets formed in the chamber are shown in Fig. 3.

The number of active nuclei can be estimated from the photographs by using a transparent overlay having dimensions of $0.5 \mathrm{~cm} \times 1.0 \mathrm{~cm}$.

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AN IMPROVED AUTOMATIC CLOUD CONDENSATION NUCLEUS COUNTER
by
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The automatic cloud condensation nucleus counter used during the second IWCIN is functionally similar to the unit originally described by Radke and Hobbs (1969). The second generation unit incorporates a number of size, weight, and electronic improvements, but the principle change is the greatly reduced height of the thermal diffusion chamber.

Briefly the instrument operates as follows:

1) The thermal diffusion chamber is set to a given supersaturation, and an air sample is drawn in. It has been experimentally determined that the polydisperse nuclei result in a nearly monodisperse cloud of droplets in a short time.
2) The time necessary for the cloud to become monodisperse is measured by monitoring the $20^{\circ}$ forward scatter of a laser beam directed through the cloud. In the Mie particle size region the scattering "efficiency coefficient", $\mathrm{K}_{\mathrm{S}}$, oscillates through values larger than one. The resulting oscillations in forward scattered intensity provide both a measure of the relative degree of size uniformity and the mean size of the droplets. It was found that, for a wide range of natural and artificial aerosol types, the time for the cloud droplets to grow to the size corresponding to the first Mie peak $(\lambda=1.2 \mu \mathrm{~m})$ was nearly a constant. This supersaturation dependent "time constant" shows negligible variation at 1\% supersaturation, although there is sufficient variation at $0.1 \%$ to make the term, constant, questionable. Therefore, 0.1 to $0.2 \%$ supersaturation is near the lower limit of this technique's application.
3) Thus, at a given supersaturation it is known that at a given time the cloud droplets are nearly of a known, nearly identical, size. Since in this size region the light absorption is very much smaller than the light scatter, the number of droplets in the cloud can be expressed in terms of only the light scattering coefficient, $b_{S}$, with $K_{S}$ and the drop radius known

$$
N=\frac{b_{s}}{k_{s} \pi r^{2}}
$$

The light scattering coefficient is measured with an integrating nephelometer (Charlson et al., 1967) at the moment of cloud growth, when the drop radius and $K_{s}$ are known.

The sensitivity of the nephelometer is such that an unambiguous calibration of the device can be provided by the Rayleigh scattering of certain gases.
4) Following the measurement of concentration, the chamber is purged and the cycle is repeated. At $1 \%$ supersaturation the sampling rate is nearly five samples per minute.

The correctness of the various assumptions involved in this measurement procedure have been born out by repeated comparisons with droplet concentrations recorded both photographically and with a "stop-action" video tape system.

A schematic view of the unit is shown in Fig. 1. The laser and camera system are not shown.


Fig. 1. 1-Flash tube, 2-Opal glass, 3-Parson's optical block, 4-Sequencer, 5-Flash tube power supply, 6-Power supdly, 7-Photo tube, 8 -Amplifier, 9 -Recording system, $10-$ Collimating discs, and 11-Samnle nump.

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## MRI CLOUD CONDENSATION NUCLEI COUNTER

## by

Thomas Lockhart MRI, Altadena, California

The MRI Model 1521 Cloud Condensation Nuclei Counter (CCN) is a commercially manufactured configuration of the Radke CCN Counter described in detail elsewhere in this report. For over a year MRI has been manufacturing the Integrating Nephelometer which is an important part of the CCN Counter providing the automatic counting feature derived from the measured scattering coefficient.


DESCRIPTION OF NRL CCN EQUIPMENT
by
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## CLOUD CHAMBER

The heart of the equipment is a thermal-diffusion chamber as designed and used by Twomey (1963). The top plate is at room temperature and the bottom plate
is cooled to a predetermined temperature, as measured by thermocouples, to give the selected supersaturation. The chamber is 1.25 cm deep and 7.5 cm in diameter. The cylindrical wall of the chamber is glass, and the top and bottom plates are covered with wetted filter paper. A high intensity mercury arc with an appropriate lens system illuminates a volume .1 cm thick by .3 cm high and 1.5 cm long in the cloud chamber. This illuminated volume is viewed at a $90^{\circ}$ scattering angle by an 8 mm camera and a video camera system described below.

## VIDEO RECORDING AND MONITORING

A video system permits immediate playback and stop-frame counting of the cloud droplet concentration in the chamber. The lens optics of the TV camera have been chosen to provide a viewing width of 1.5 cm and a height of .3 cm , identical to that viewed by the 8 mm camera which is used simultaneously. The 12-inch monitor screen is marked off to identify these viewing dimensions for ease in counting the droplet images. The viewing volume is $.0675 \mathrm{~cm}^{3}$. A Sony model AV5000 video recorder is used with a Sony camera and monitor.

## PROVISION FOR PREHEATING OF AIR SAMPLE

Twomey (1968) describes a method for obtaining information on the possible composition of cloud nuclei. In this method the air sample under study is preheated to various temperatures and the number of nuclei which survive is measured as a function of the amount of heating. The preheating is obtained by providing a means for passing the air sample through five parallel channels. In each of the channels the air first passes through a quartz tube of .8 cm i.d. and 70 cm long around which is wound a heating coil, the five coils differing in the wattage consumes. After passing through the heating tube the air samnle passes into a 3 liter holding chamber, one chamber for each channel, for temporary storage. A sixth channel is provided in which no preheating is annlied to the air sample before storage in its holding chamber. The flow rate through each channel while refilling the respective chambers is $20 \mathrm{~cm}^{3} \mathrm{sec}^{-1}$.

## SAMPLING SEQUENCE

The air sample is introduced first to the room temperature conditioning chamber. After at least five minutes of flushing ( $2 \frac{1}{2}$ changes of air) the conditioning chamber is closed off with inlet and outlet valves. An aluminized mylar diaphragm lining the bottom of the chamber is then slightly pressurized from below to cause the sample to gently flow through the diffusion cloud chamber whenever a valve is opened to it. The diffusion chamber is flushed, then closed off to permit the preset supersaturation to be reached.

The cloud droplets formed in the predetermined volume in the light beam are photographed and simultaneously presented by the TV camera to the TV monitor and to the video recorder for later playback and stop-frame analysis. After the cloud droplets have fallen out, a preset vacuum can be connected to the diffusion chamber to give an expansion with a $1.21: 1$ pressure ratio to provide a supersaturation of $345 \%$ similar to that used in a Pollak counter. (This Aitken nuclei observation is useable to a concentration of only about $6,000 \mathrm{~cm}^{-3}$ without sample dilution provisions).

A new sample is then introduced to the diffusion chamber and the observation procedure repeated two more times (for a total of three observations for each filling of the conditioning chamber).


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CONDENSATION NUCLEI COUNTER, MODEL ECOM-ASL 001

## by

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The Condensation Nuclei Counter, Model ECOM-ASL 001, consists of four principal components: a thermal diffusion chamber, a collimated lighting system, a photo-recording system, and one or two conditioning chambers. The original design of this counter was supplied by Dr. S. Twomey of CISRO, Sidney, Australia, although minor innovations have been added from time to time; such as, a more sophisticated purging device, a more efficient pressurizing system, and a motordriven still camera instead of a movie camera. However, basically, the counter operates in the manner as his, now in use by the Naval Research Laboratory. Therefore, for a detailed description thereof, reference may be made to the section contributed by that laboratory.

Nevertheless, one feature of this counter needs to be brought out; that is, the use of a dual-channel Varian recorder for recording both the top-plate

temperature and the temperature difference between the warm plate and the cool plate on the bottom. As has been experienced, the amount of supersaturation in the thermal diffusion chamber can be estimated with confidence only when the ton-plate temnerature is accurately known, which often deviates from the ambient temperature considerably. With such a record in hand, an analyst can easily go back to it for the correct amount of supersaturation at the moment the cloud nuclei are being sampled, despite the fact that a preliminary setting of the desired supersaturation has been made. Only in this way can one be confident of the sampling results.

Three pictures of the counter are presented. Figure 1 is a front view of the counter, which comprises the motor-driven camera, the Fluke meter to measure temperature differences, the light tube, the control panel, and the purging device hanging on the left side. Figure 2 is a rear view of the counter, which comprises the light tube, the back of the thermal diffusion chamber to the left of the tube, and two conditioning chambers. Figure 3 is an enlargement of a $35-\mathrm{mm}$ negative of cloud nuclei. Not shown here is the Varian recorder, which is normally located to the left of the Fluke meter and which records the latter's output as well as the top-plate temperature.
(See figures on next page).


Fig. 1. Front view of the Condensation Nuclei Counter, Model ECOM-ASL 001 (the Varian recorder not shown).


Fig. 2. Rear view of the Condensation Nuclei Counter.


Fig. 3. A sample of cloud nuclei.

## NCAR CONTINUOUS CLOUD-CONDENSATION NUCLEUS COUNTER

## by

## G. Langer

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*The National Center for Atmospheric Research is sponsored by the National Science Foundation.

The NCAR continuous cloud-condensation nucleus counter, which differs completely from the usual thermal-gradient diffusion chamber, was built to meet the need for a small, simple counter that could be operated unattended for long periods, could be installed with a minimum of effort, would give an immediate and continuous readout, and would operate at high altitudes ( $-40,000 \mathrm{ft}$ ) without loss of sensitivity. Because of its ready availability, an acoustic sensor was used to count the cloud droplets. As a simple means of activating the cloud nuclei, the process of condensation in a controlled laminar jet was used. This precluded accurate supersaturation control, but since our interest was more in the overall population of cloud active nuclei, this compromise was acceptable. Also, it was felt that once the design was proven practical, more sophisticated means of activation could be used.

Figure 1 shows the counter. The cloud chamber is operated as a closed system. The air is circulated at a rate of 141 pm . It is filtered, humidified, and then enters the cloud chamber with a temperature of about $45^{\circ} \mathrm{C}$ and a dew point of $40^{\circ} \mathrm{C}$. The entrance nozzle produces a laminar jet that issues into an essentially open area. The supersaturation regime in such a jet has been fully described by Amelin (1967). The salient feature is that, as the jet enters the chamber, contours of constant supersaturation form around the jet. At a certain mixing ratio of incoming air to the sink air a maximum isoline of supersaturation is crossed. Beyond that point the supersaturation drops to a lower level; drops that have formed will continue to grow, but no new ones are generated, a situation that can be verified visually. The dilution of the sample is such that the presence of the water drops does not affect the supersaturation.

A sample is taken by adding a small stream of outside air to the main airstream circulating in the closed loop. Any cloud nuclei active below the maximum supersaturation are then activated and form
droplets. For the particular configuration used this maximum supersaturation is about $2 \%$. The cloud chamber has about a 15 sec hold up to allow the drops to grow until their diameters are over 20 microns for detection by the acoustic sensor. So far most of the work has been concentrated on making the instrument operable rather than on a detailed study of the activation process.

The counter has been flown to an altitude of $20,000 \mathrm{ft}$ to check the pressurization operation. Because of the closed loop flow system, its operation should be independent of altitude. This has been found to be true except for the leakage along the shaft of the pump presently used to pressurize the system. The sample is injected by a diaphragm pump which loses effectiveness with altitude, and a positive displacement type pump has been designed to replace it. The new pump will also reduce the possibility of aerosol losses or generation of particles. However, initial tests with an optical counter and a Rich Counter have shown that these are not problems with the present diaphragm pump. The present sampling lines are fairly long, and small nuclei are lost to some extent by diffusion.

The instrument's response to various aerosols will be tested against a thermal gradient diffusion device. This will show if nuclei are lost because activation time is too short. Better information on drop losses to the walls are also needed. So far tests with solid particles of low density to simulate water drops indicate a constant loss of $80 \%$.

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## C. Ice Nuclei

(1) Acoustic Counters

## LONG-CONE NCAR ICE-NUCLEUS COUNTER

## by

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The NCAR Counter (Langer, 1967) consists of a continuous-flow ( 14 lpm ) cloud chamber with an exit cone leading into an acoustic sensor (Langer, 1966) which counts ice crystals in the cloud. The acoustic sensor consists of a 1.5 mm diameter capillary about 6 cm long which produces a loud click when an ice crystal or other particle over 20 microns in diameter passes through at velocities near $100 \mathrm{~m} / \mathrm{sec}$.

The model shown in Fig. 1 is a commercial unit, made by E.G.\&G., Inc., Boulder, modified as described below to improve its operation. The $45^{\circ}$ taper of the exit cone was increased to about $22^{\circ}$ to reduce settling losses (Langer, 1969) of the ice crystals.

In the system that coats the chamber interior with glycol to prevent frosting, a polyurethane foamplastic filter was added ahead of the pump to catch particles large enough to stall the nump. In


Fig. 1. Long cone NCAR ice nucleus counter.
addition, the pump inlet, filter, and line to the pump from the tank were lowered to make the pump selfpriming.

The lid of the commercial chamber was replaced with a loosely fitted 10 cm adapter ring (and lid) that prevents choking of the glycol flow and raises the sample inlet about 7.5 cm above the glycol distributor. This slows the cooling of the air entering the cloud chamber and avoids extreme supersaturation and prevents frosting on the inside of the lid. A felt strip, which draws glycol up from the main felt liner by capillary action, prevents frost on the region of the adapter in contact with cold glycol flowing into the distributor head. A second layer of felt may be required around the lower edge of the glycol distributor if it is uneven since this can choke off glycol flow at some points. Finally, a hydrometer was incorporated into the system to monitor the glycol water content. The flow to the hydrometer should be very slow so that the glycol warms to the calibration temperature of the hydrometer. Glycol specific gravity should not be lower than 1.070 for operation down to -26 C .

To reduce hold-up time in transfer lines, the humidifier was moved from alongside the chamber to directly above the cloud chamber. The commercial humidifier provides poor vapor transfer and therefore must operate at relatively high temperatures. This, in turn, warms the incoming air and increases the heat load on the chamber. In the modified humidifier three concentric felt rings (as shown in Fig. 1) dipping into the water are used to increase the vapor transfer. The air passes through many holes in the felt, and is humidified to $90 \%$ r.h. at 22C. This provides a good cloud for operations below -15C. For operations at $-8 C$ the water temperature must be raised to 29C. Finally, the new humidifier has a by-pass to allow sample injection directly into the cloud chamber for sample flows of 51 pm or less.

Cloud condensation nuclei must be added to the sample air because if it is relatively clean some large water drops may form when the moist air condenses to form the supercooled cloud. These large drops can trigger the sensor and give erroneous ice crystal counts. Condensation nuclei are generated by atomizing a dilute salt solution. The atomizer provided with the commercial units tends to perform marginally. It uses a metal nozzle that is subject to corrosion, and often air by-passes the nozzle because it is loose in the fitting. Also, the commercial atomizer uses a pump that often does not provide enough flow, i.e., less than 2.51 pm .

The modified atomizer uses a flow of 3.5 lpm and a plastic nozzle. With the higher flow rate, a much weaker salt solution can be used than the usual $0.1 \%$. This eliminates possible nucleation by salt particles below -20C. Only 10 individual grains of salt per 500 cc are used. We also filter through a 0.8 micron filter the distilled water that makes up the solution. Before filling, the atomizer is thoroughly cleaned to eliminate nucleation by spurious dust particles at lower temperatures. This atomizer produces salt particles only in the range between 0.01 and 0.2 microns.

In one of the commercial counter models the humidifier salt solution atomizer system is renlaced by a special atomizer that produces a cloud of fine water drops directly. That is, a cloud is generated before the air enters the cloud chamber. This socalled cold-cloud atomizer has several disadvantages. Because it is difficult to humidify the atomizer air, the cloud tends to evaporate before reaching the chamber. If the water in the atomizer is heated to increase the humidity, spurious ice nuclei active at $-20 C$ and below, are generated. Also, particles are entrained from the sample air into the atomizer water by the scrubbing action of the atomizer spray. Some of these particles are re-entrained into water drops and may nucleate ice in them. Heating the water probably increases scrubbing efficiency. An apparent solution is to by-pass the sample air around the atomizer, but this gives a poor cloud since not enough drops reach the cloud chamber under these conditions.

If the above cold-cloud atomizer is used, frequent tests with clean sample air must be made to ensure that no nuclei originate in the atomizer. The humidifier salt-solution cloud-generating system should also be checked with clean air. The procedure is to place an absolute filter on the sample air stream to remove all aerosol particles. The details regarding the use of such a filter are given below in the discussion of how to locate leaks.

Many counters tend to leak at various points. The most common ones are the cone joint; joints in the humidifier and atomizer; and the seals around the viewing port, the illuminator nort, and the thermometer well. The extent of this problem usually goes unnoticed. A quick check, if leaks are suspected, is to apply positive pressure of 1 inch of water to the chamber by connecting it to a low-pressure compressed-air line with a by-pass to control the pressure. This air must be filtered with an absolute filter. If the cloud in the chamber clears when the salt atomizer is blocked off, no leak is present. To locate leaks, place at the intake an absolute filter that requires a negative nressure of less than
1.5 in of water in the cloud chamber; otherwise, glycol is pulled into the sensor from the reservoir. A manometer should be placed on all counters to monitor suction and to serve as a safety blow-out in case the air inlet is blocked. When the filter is in place, shut the atomizer off and pinch shut the hose leading to it. Run the counter at -22 C in the normal manner. If no leaks are present, after 5 minutes only a few cloud droplets or none will form in the chamber. Some screeching in the sensor may result because of condensation there. If the cloud persists, first check the absolute filter. Light a match and let the smoke drift into the intake. If there is a sudden increase in small drops, the filter is defective. If the filter is good and many drops are still seen, check further for leaks. Disconnect the atomizer and run it as a source of nuclei to be directed with a hose at suspected leak points. If a leak is present, the cloud in the chamber will suddenly intensify. To check along the cone joint, pull the insulation back and point the illuminator downward to reveal any cloud intensification.

When the modified counter is operating properly, i.e., no leaks, no frost, and good atomization (often a problem), counts at $-22 C$ with a filter should be fewer than $0.1 / \mathrm{min}$ (occasionally, higher count rates may still be observed because some absolute filters have a significant efficiency loss in the submicron range, and nuclei in this range penetrate the filter).

The drift of the ice crystals in the cloud chamber clearly shows that many fall onto the bottom of the chamber. This loss has been investigated (Langer, 1969) for an altitude of 1800 m by lining the cone with a static layer of supercooled sugar solution that records ice crystals as white spots. To correct for this crystal loss, counts at 1800 m are multiplied by a factor of 3.3 for the long cone and by 5.0 for the commercial one. At 5600 m the correction factor is 10 (Grant, 1970).

The new counters have been operated for as long as a month with just normal maintenenace. Water and glycol are added every 2 to 10 days depending on their reservoir capacities. Results from two machines operated in parallel (Langer, 1969) at the same temperature have consistently been in agreement.

## ACKNOWLEDGMENTS

Thanks are due to R. Husar, University of Minnesota, for providing the performance data on the salt atomizer.

The National Center for Atmospheric Research is sponsored by the National Science Foundation.

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# MODIFIED NCAR CONTINUOUS ACOUSTICAL <br> ICE NUCLEUS COUNTER 

by

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A modified NCAR continuous acoustical ice nucleus counter was operated (Fig. 1) during the IWCIN in Fort Collins, Colorado, from August 3 through August 21, 1970. This is a commercial unit produce by E.G.\&G., Bollay Associates. The basic instrument is described elsewhere (G. Langer, J. Rosinski and C.P. Edwards, 1966; R.L. Steele, C.P. Edwards, L.0. Grant, and G. Langer, 1967).


Fig. 1. Schematic diagram of continuous NCAR acoustical ice nucleus counter including modifications.
1-Teflon funnel, 2-Funnel edge heater, 3-150 watt heat lamp, 4-5 Gal. glycol reservoir, (below sensor level) 5-Submerged water pump, 6-Water heater, 7-Aerosol air pump, 8-Water reservoir for humidifier, $9-\mathrm{NaCl}$ solution reservoir for atomizer.

To improve the instrument's capacity for continuous operation in the field, the following changes have been incorporated:

1. The cloud chamber head (Fig. 2) has had a teflon funnel attached which, in inverted position, is connected to the air intake joining to its lower edge with the plastic foam lined chamber wall. This allows a good condensation runoff into the antifreeze-water-solution. A heating tape placed between the outer large funnel opening and the inner head wall prevents ice formation from small droplets while it remains slightly above freezing level.


Fig. 2. Modified cloud chamber head of NCAR acoustical ice nucleus counter
1-Air intake, 2-Impactor, 3-Humidifier water, 4-Foam filling, 5-Teflon funne1, 6-Heating device (around outer funnel edge and 7-Foam lined cloud chamber wall.
2. A semi-continuous water flow has been added to the humidifier system to maintain the water level inside the humidifier water heater within two limits resulting in a near-stable humidifier water temperature.
3. A semi-continuous NaCl solution flow has been added to the atomizer system resulting in a continuous NaCl aerosol flow into the cloud chamber to preclude the formation of large cloud droplets.
4. The antifreeze-water-solution reservoir has been enlarged to obtain prolonged unattended solution circulation within the antifreeze system within the proper density levels.
5. A heating lamp has been placed near the lower chamber to prevent ice formation at the inner rim of the trough.
6. All thermometers reaching into the cloud inside the cloud chamber are removed from the cloud chamber once the desired temperature levels are established inside the chamber. The thermometers are inserted only for short periods testing the temperature levels. This precaution is taken to eliminate ice from forming on the thermometer.

The instrument now can be operated for an unattended period of about 10 days; thereafter minor maintanance is necessary. No icing inside the cloud chamber has been observed during the past 12 months of normal continuous operation.

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AEROSOL SAMPLING AND DATA ANALYSIS WITH THE NCAR COUNTER
by
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## INTRODUCTION

In the latter part of 1968, B.F. Ryan and I found that the counts from the NCAR Counter increased markedly with the onset of rainfall (Ryan and Scott, 1969). The tentative explanation for this was that ice nuclei were concentrated near the rain envelope. It is possible that they were scavenged by the rainfall. Alternately, perhaps, the Bergeron precipitation mechanism was operating and we measured the particles responsible for the precipitation growth. The smaller raindrops presumably had evaporated and left the observed large concentration of particles.

In any case, it was desirable to continue the measurements and try to obtain an estimate of the number of ice nuclei in the raindrops. For this purpose, a differential sampling technique was devised. The raindrops were nebulized and evaporated; then the counts obtained from air containing the residual solid
particles were compared with the counts from the air alone. Before long it was apparent that there was an increase in count due to the rainfall. But, surprisingly, this increase sometimes occurred without rainfall (see Scott et al., 1969).

An explanation for this effect is difficult but it was probably associated with the orientation of the sampling systems and the containment of the outside air in the drop dispersing chamber. It may indicate that ice nuclei are occasionally large particles or that their activity is altered while in a closed chamber.

My object in coming to the Workshop is to look in detail at the effects of the sampling system on the count from an NCAR Counter. In support of this objective, an electronic analyzing system has been developed to analyze the output of the acoustical sensor and gain a maximum of information from the counter. Of course, these data may add to our knowledge of ice nuclei.

In particular, I hope to answer these questions:

1) Are a substantial number of natural ice nuclei large particles? Are nuclei deactivated during sampling? If so, the Workshop participants measuring natural ice nuclei should take care in their sampling procedures and in applying their results to the natural aerosol.
2) Does a multiplication of ice crystals or a recycling of particles occur in the NCAR Counter?

It may be possible to answer these questions by close examination of the pulse height spectra and time lag spectra of the acoustical pulses.

## SAMPLING

Samples of the natural aerosol in Fort Collins (designated "outside") will be taken directly from a vertical, $1 \frac{1}{2}$ inch aluminum tube 20 feet long which sticks approximately 12 feet above the roof of the Atmospheric Simulation Laboratory. As time permits, samples will be taken from the sampling duct (designated "duct").

A photograph of the apparatus is shown in Fig. 1.
The entire sampling system is shown in Fig. 2. Both tubes enter directly into the top of the cold chamber of the NCAR Counter. The sampling rate is $101 / \mathrm{min}$. Humid air with a salt aerosol is added to the sample air at a rate of $5 \mathrm{1} / \mathrm{min}$. The air is humidified by bubbling in water at a temperature of approximately $86^{\circ} \mathrm{F}$. Valves $\mathrm{b}, \mathrm{c}$, and d are pinch clamps; valve a is a sleeve and rubber cork. Alternately either $a$ and $b$ are opened ( $c$ and $d$ are closed) or $a$ and $b$ are closed ( $c$ and $d$ are opened).

## ANALYZING THE ACOUSTICAL PULSES

The pulses are recorded individually on a strip chart recorder using a peak sample and hold amplifier. The schematic diagram of the amplifier is shown in Fig. 3. Figure 4 is a picture of the amplifier. It is contained entirely within a small aluminum box attached directly to a 110 V.A.C. power plug.


Fig. 1. Apparatus.


Fig. 2. Diagram of the sampling system.

The amplifier is tuned to a frequency of 2500 Hz and has a voltage gain of 250 at that frequency. The output of the microphone sensor on the NCAR Counter is a damped harmonic signal of approximately 2500 Hz . The final stage is coupled to a peak-sample-and-hold circuit that automatically holds pulses of amplitude greater than 0.5 volts. The circuit is designed so that high frequency electronic noise merely raises the voltage threshold and hence has a minimum effect on the recording of the pulses.

An example of the readout of the amplifier is shown in Fig. 5. Bunching of the pulses as shown creates errors and small signals are not recorded

## TYPICAL RESULTS

The pulse heights and the times between pulses are measured, tallied and plotted as a function of their relative frequency of occurrence. Figures 6a and 6 b show some results.

It is interesting that the histogram of Fig. 6a is bimodal. As a result, the average time between pulses is not the most frequent.

(a)

(b)

Fig. 6. Histograms of the time between pulses (a) and Pulse heights (b). August 10, 1970, 1653-1703 hrs., chamber at $-20^{\circ} \mathrm{C}$; natural aerosol at Ft. Collins.

## ACKNOWLEDGMENTS

Thanks are due to Gerhard Langer for his willing help and the loan of an NCAR Counter during the period of the Workshop.
immediately following larger ones. Also, the circuit is not entirely linear. Nevertheless, for counts less than 10 per minute, the amplitudes of the pulses are reproduced to better than $20 \%$.

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## (2) Millipore Filters

ICE NUCLEI MEASUREMENT WITH MILLIPORE FILTERS
by
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The procedure consists mainly of passing a known volume of air through a millipore filter, coating the suction side with Vaseline, placing the filter on a brass plate with the Vaseline side down. The brass plate is placed in a diffusion chamber under controlled temperature and supersaturation. Ice crystals which grow on the surface of the filter are revealed by a light beam and may be counted.

The diffusion chamber is cooled from the bottom by circulating a mixture of glycerin and water. The brass plate which carries the filter is rested on and cooled by three thermoelectric coolers connected in series. On the base of the chamber ice formed from a thin water film acts as a water vapor source. Temperature of the filters and the ice surface is measured by two fine thermocouples which are connected to a two-channel recorder for a continuous temperature recording. A diagram of the chamber is shown in Fig. 1. For further detail see Alkezweeny, 1970: The use of millipore filters for ice nuclei measurement.

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## ACKNOWLEDGMENTS

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Fig. 1. The diffusion chamber (insulation is not shown).
1-Air gap, 2-Transparent 1id, 3-Permagum for sealing, 4-Brass sheath, 5 -Brass plate, 6-Ice surface, 7-Trays with filter, 8-Thermocouple to a two-channel recorder, 9 -Circulating cold glycerin and water, and 10 -Thermoelectric cooler.

## APPARATUS FOR DETECTION OF ICE NUCLEI ON FILTERS

by
E.K. Bigg


Detection of ice nuclei deposited on membrane filters was discussed by Bigg et al. (1963). The equipment used here represents a modification of that method suggested by Stevenson (1968).[1]

The equipment used was designed to be as simple as possible. Four membrane filters on which an aerosol sample has been captured are placed on black brass discs covered with a thin layer of a viscous oil (Shell "Dentax 250"). The oil renders the filter transparent and seals the pores without wetting its upper surface and without the necessity of heating the filters as in the "Vaseline method." The filters are then inserted into an ice crystal developing chamber which consists of parallel plates of 1 cm thick aluminum spaced by 1 cm with strips of plexiglass 1 cm wide around their edges. The upper plate is hinged to the plexiglass for ease of inspection and manipulating the filters.

Humidity is controlled by varying the temperatures of the upper plate, the lower surface of which is coated with a layer of glaze ice less than 1 mm thick, and the upper surface of which is in contact with an electrical heater.

The temperature difference between the plates is recorded with a differential thermocouple whose output is recorded by an optical meter relay which turns off the electrical heating at a preset temperature difference.

Ten minutes are necessary for thermal equilibrium to be re-established, after filters are inserted an extra five minutes are needed to achieve the required humidity difference and about 20 minutes to grow the crystals to a suitable size for counting.

In most systems of this type the finite size and conductivity of temperature sensors lead to small errors in temperature. In this one the location of the sensors within the metal plates leads to small errors because the ice and filter surfaces are not in complete thermal contact with the plates. The onset of condensation must be used in preliminary experiments to check the humidity deduced from the temperature measurements.

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Fig. 1. Cross-section of diffusion chamber for detection of ice nuclei on membrane filters. 1-aluminum plate, 2 -thermistor, 3 -thermocouple, 4 -removable plexiglass tray holding 4 brass discs, 5 -ice, 6 -hinge, 7 -heater, 8 -thermocouple in aluminum plate, 9-styrafoam insulation.

## A THERMAL DIFFUSION CHAMBER FOR <br> ICE NUCLEI MEASUREMENTS



The thermal diffusion chamber described below was designed to provide conditions for the processing of membrane, $0.45 \mu$ filters. The details are given in Fig. 1 and in two photographs (Figs. 2 and 3 ). This apparatus is an improved and modified version of the previous instrument described by Gagin and Aroyo (1969).[1]

The chamber consists of two horizontal, thermally regulated aluminum plates separated by a cylindrical metal wall 2 cm high. The upper plate consists of a sheet of glaze ice embedded in a space occupied also by two wire meshes, 3 mm apart. This arrangement provides an improved thermal conductivity in the ice layer and hence a sensitive control of the temperature of the ice surface. The lower plate is a polished metal surface on which the filter trays are put for processing. Two aged thermistors are used to measure the temperatures of the filters and ice surface. The regulation of these temneratures is governed by two separate thermoelectric units, operated by DC power sources the polarity of which is altered by simple bridge circuits commanded by two separate platinum wire resistors. The resultant temperature fluctuations at the ice and filter surfaces are of the order of $\pm 0.05^{\circ} \mathrm{C}$. Any temperature can be selected independen $\bar{t} l y$ for each plate, within the range of $0^{\circ} \mathrm{C}$ to $-30^{\circ} \mathrm{C}$. The primary cooling of the whole system is carried out by an ordinary refrigeration system acting also as a heat sink for the lower and upper thermoelectric unit. The regulation of temperature together with the introduction of heat conducting walls provides a basis to the


Fig. 1. The thermal diffusion chamber. 1-camera adaptor, 2-microscope, 3-thermistor elements, 4 -thermomodule, 5 -observation window, 6 -ice surface, 7 -filter plates, 8 -thermomodule, and 9 -heat sink.


Fig. 2. Thermal diffusion chamber for IN measurements (upper part).
assumption that a linear temperature gradient exists in the space between the two thermally controlled surfaces which is nearly the same as that maintained on the walls. The ratio of the ice surface diameter to the separation distance between the two plates ( $7.5: 1$ ) allows the assumption that steady state conditions are quickly attained such that the water vapor


Fig. 3. Thermal diffusion chamber for IN measurements (general view).
pressure anywhere at the chamber is at equilibrium with that of the ice surface. The humidity stratification can now be easily defined, requiring only accurate enough measurements of both the filter and ice surface temperatures. The ice surface is the major and warmest vapor source in the chamber. The coldest surface in the chamber is the filter surface and hence this is where the maximum supersaturation exists.

The perspex tray holder which covers the lower plate and also the edges of the filter plates together with the preparation of the filters with vaseline [2] reduces the vapor losses to a minimum, this leaving the growing ice crystals on ice nuclei as the only sinks for water vapor. These losses are easily compensated for by diffusion from the ice surface thus providing a constant supersaturated atmosphere for the processing of the filters.

Two more separate chambers exist for observation and introduction of the filters trays. The microscope has five different, easily interchangeable, magnifications, thus making the count of high concentration rather convenient through the use of five different fields of view. The lowest magnification has a field of view $60 \%$ that of the total effective area of the filter.

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## NCAR ICE NUCLEUS ANALYZER

by

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The NCAR ice nucleus analyzer was designed to "develop" membrane filters used in the study of ice nuclei by the Stevenson method and by the direct deposition of drops; in the study of freezing nuclei by the Vali droplet freezing method; and in the study of the nucleating activity of rain, hail, and snowflake residues. For these studies it is necessary to watch with a microscope the changes taking place.

The new device is shown in Fig. 1. The use of the observation window and the application of droplets required a flow of air of controlled humidity to prevent heat from the warmer window from affecting the water saturation over the observation area, to allow drops to settle on the sample, and to allow sublimation of hail slices and snowflakes to expose residues.


Fig. 1. NCAR ice nucleus analyzer.

The air, which flows at a rate of 31 pm , is first dried to a dew point of -50 C and then filtered before entering the flow distribution grid where it passes between a series of parallel plates covered with wet blotting paper. These plates serve as an ice surface upon freezing and saturate the air with water vapor at the vapor pressure of ice and cool the air to the plate temperature before it enters the observation area. There it passes over the sample, which may be heated or cooled with respect to the air temperature by a thermoelectric element contained in the temperature control block, which in turn rests on the base plate. This controls the water vapor saturation with respect to the sample.

A sample is placed on a brass plate 6.5 mm square and 1 mm thick, which is previously coated with a

Vaseline layer 1.5 mm thick and then heated to give a smooth Vaseline surface. After the plate has cooled, the membrane filter sample is placed on the Vaseline surface, and the plate is heated to 49C. The Vaseline just penetrates the surface of the membrane and then cools. For white membranes Vaseline penetration is complete when the filter is transparent; for black membranes, the surface turns shiny. For studies of hail slices, snow flakes, or droplet freezing a clean (unexposed) filter is used. The brass plate is brought into good thermal contact with the temperature-control block by a coating of glycerin on the block. Airflow is begun, the lid shown in Fig. 1 is opened momentarily, and the sample is inserted. By means of the temperature-control block the sample temperature is then adjusted to give the desired effect. The temperature of the humidifier and sample enclosure is controlled by the freon compressor cooling the base plate. This system responds fairly slowly.

The performance of the humidity control is verified by the following observations. The temperature of the sample surface is measured with a thermister mounted onto the edge of the temperature-control block. It measures accurately as verified with thermocouples placed on a filter and on the block. When the temperature of the sample surface is slightly above that of the humidifier, an ice crystal will slowly sublime. The onset temperature of water saturation on the sample surface was verified with a piece of aluminum foil placed on the Vaseline surface. When the block was cooled to the temnerature at which the humidified air passing over it would be at water saturation, condensation on the foil was observed. However, a problem is encountered if some ice crystals develop early and grow relatively large by the time later ones are nucleated. In that case an ice-and-condensation-free "halo" in which further nucleation is suppressed develops around each large crystal. This can lead to low counts especially with natural ice nuclei.

It has been found that membrane filter samoles as received have a varying ice nuclei count. Below -20 C this renders their use impractical. To check on the background count a special filter holder, shown in Fig. 2, is used. The annular area serves as the reference area to subtract out the blank count from the central deposit. The complete separation of the reference area from the air flow is necessary because, even if no flow is allowed directly through the annular area, particles are deposited there by turbulence, diffusion, and electric effects.

To produce a cloud of droplets to pass over the sample it was found most convenient to simply breath into the manifold in the lid. This cooling manifold is wiped with glycerin to orevent frost. Cloud droplets form as the air is cooled in the manifold, which widens out into a slot with a baffle to spread the cloud. Some of the drops can be observed with the microscope to fall on the sample. Breathing is adjusted to give nearly complete coverage of the sample with drops. Usually two to three slow puffs are sufficient depending on the temperature. Comnlete coverage can be verified by watching the droplets fall onto black filters. For white filters, the contrast is too poor to allow observation of the drops. Since the ice crystals become visible in a few seconds, one applies a few puffs, and, if no new crystals appear, interaction is complete. So the crystals


Fig. 2. Modified membrane filter holder to provide blank reference area.
will grow, the temperature of the filter for this developing process is kept above the ice saturation. Originally (Langer, 1970) an atomizer was used to provide the drops, but this system lacked the fine control possible by breathing. The droplet technique is used to develop for AgI (Langer, 1970), phloroglucinol, and other artificial agents which act principally by contact with supercooled drops.

Some thought must be given to the selection of the sampling filter pore size. Some AgI smokes are quite fine and may penetrate into the filter pores or even pass through them. So far it appears that a $0.22-\mathrm{mi}$ cron pore filter is adequate.

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(3) Mixing Chambers

A BRIEF DESCRIPTION OF THE OPTICAL COLD-CHAMBER SYSTEM

## by

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## THE EXPERIMENTAL APPARATUS

The optical cold-chamber system consists of three separate units: (1) aerosol generator, (2) air preparation system, and (3) microscope and coldchamber assembly.

The generator has been designed to produce aerosols of low AgI mass output such that the effluent can be" channeled through the system and exhausted through a laboratory hood. The components of this system are diagrammetrically sketched in Fig. 1. The solution if gravity fed through an atomizer connected to a pressure pump into the rear of the combustion chamber. The combustion is supported by a propane flame, the temperature of which is approximately $1300^{\circ} \mathrm{C}$ at the hottest measurable position. Additional air is brought in through the rear portion of the combustion chamber through open ports, and excess pressure which might result from large fuel injections can be relieved through a side port and exhaust fan not shown in sketch. The aerosol is constantly drawn through the combustion chamber, hose lines, and condensation tanks through the exhaust pump and into the exhaust hood.

Two flow meters are in the system, one in the gravity feed line for measurement of solution flow rate and the other on the exhaust end of the condensation tanks to permit calculation of dilution of the aerosol. Attached to the combustion chamber is a Büchner funnel which permits trapping of the aerosol in fine fiberglass filter paper, or, without the filter paper, the aerosol passes freely through the lines and can be sampled by insertion of a syringe as shown in the figure. For most experiments the solution flow is kept at $0.7 \mathrm{ml} / \mathrm{min}$ with the capability of $1 / 2$ to $2 \mathrm{ml} / \mathrm{min}$ range, and a maximum output of $1 / 2 \mathrm{gm} \mathrm{AgI} / \mathrm{min}$. The exhaust pump brings extra air into the system through the ports, mixes it with the


Fig. 1. Schematic diagram of low output aerosol generator. 1-Combustion chamber, 2-Syringe sampling, 3-4-Condensation tanks, 5-Flow meter, 6-Exhaust pump, 7-Pump, 8-Propane bottle, 9 -Air intake ports, 10-Flow meter, 11-Gravity feed solution reservoir, 12-to hood.
combustion products, resulting in a flow of 400 to $600 \mathrm{ml} / \mathrm{sec}$. This results in a particle density of $10^{5}$ to $10^{10}$ particles/ $\mathrm{cm}^{3}$ depending upon the solution flow. The approximate AgI density upon sampling is $10^{-7} \mathrm{gm} / \mathrm{cm}^{3}$.

Even with the low AgI output of this system it is difficult to directly obtain particle solution densities of less than $10^{8} / \mathrm{cm}^{3}$ without diluting the initial solution. However, dilution can be accomplished in the syringe following sampling, though with less accuracy. This generator has been designed specifically for small chambers requiring sample volumes of less than $100 \mathrm{~cm}^{3}$. All types of solutions may be used in the generators; however, isopropylamine and ammonia solutions cause slow deterioration of the hoses and copper or brass components.

As modified for use at the Workshop, we have removed the Buchner funnel and condensation tank assembly so that only the propane burner, combustion chamber, and gravity feed system remain.

The generator is mounted at the base of the CSU vertical wind tunnel where the air flow is such as to sweep the generator effluent directly into the tunnel with negligible loss. Three tunnel dilutions are possible:

1) Natural stack up draft of 4000 cfm ,
2) Minimum fan up draft of $63,200 \mathrm{cfm}$, and
3) Maximum fon updraft of $134,500 \mathrm{cfm}$.

From a mean AgI particle radius of $0.04 \mu$ (obtained by X-ray diffraction) the minimum and maximum particle densities (in units of $\mathrm{cm}^{-3}$ ) for the above three dilution flows are $1 \times 10^{5}$ and $4.1 \times 10^{5}$; $6.6 \times 10^{3}$ and $2.6 \times 10^{4}$; and $3.1 \times 10^{3}$ and $1.2 \times 10^{4}$, respectively.

The air preparation system is a device designed to start with dry nitrogen separated into two systems, one a humidifying circuit and the other a dry nitrogen circuit, and to mix them in the proper proportions before exit from the system to achieve the desired dew point for whatever purpose is desired. Knowing the temperature of the humidifying system and the vapor pressure of the dry nitrogen used (which does not have to be known accurately) the final dew point can be calculated for various ambient temneratures. This has been completed and has resulted in a series of charts from which one can determine the dew point desired for a given flow rate needed for his experiment. Part of the system includes a means for direct dew point determination. This is based on the direct observation on the bulb of a thermometer of condensation at the dew point read directly from the thermometer. A sensor chamber is built into the system which permits the mixed air to flow over an insulated thermometer, the lower one-half of the bulb being in contact with a copper tube which is temperatured controlled by a resistance heater and liquid nitrogen source. Condensation is observed by a microscope tube focused directly on the surface of the thermometer bulb. Thus at any time the dew point of the air being used can be checked directly, providing a small quantity of liquid nitrogen is available.

There does not seem to be a noticeable effect on habit or nucleation efficiency in the data collected so far which might suggest that the lack of oxygen in the system has affected these parameters. Thus in the experiments being performed at this workshoo the atmosphere consisting of pure nitrogen and water vapor (with a small amount of oxygen possibly entering the system by diffusion) is assumed to behave in the same manner as the natural atmosphere.

A diagrammatic sketch of the optical chamber is given in Fig. 2. In this system nitrogen vapors are passed through a double-walled aluminum chamber surrounding the plastic sample well. The nitrogen vapors upon leaving the chamber are recirculated through a plastic shield housing the microscope objective such that no condensation can occur on the lenses or cover glass surface of the chamber. This latter feature also assists in the insulation such that no further insulating materials are needed on the chamber.


Fig. 2. Essential components of optical cold stage assembly. S-Supporting plate, TC-Thermocouples, OA-Inlet of the prepared air, and BO-Outlet of the prepared air.

Housed in the cooling chamber is a plastic well chamber which has removable upper and lower surfaces for cleaning and sample mounting. The chamber is provided with an inlet and outlet (A and B, Fig. 2) for the prepared air, an injection needle for injecting of the aerosol sample (C, Fig. 2), and a thermocouple (T.C.) placed in contact with the surface upon which the aerosol particles settle. A low- and highpower magnification is available with the system as well as Polaroid or $35-\mathrm{mm}$ photography.

The size of the particles activated within the chamber depend upon the settling time allowed before the run is actually started. For AgI spherical particles it requires exactly 1 hour for the $.04 \mu$ radius mean size particle to settle the 4 mm to the chamber floor. Particles smaller than this should theoretically be swept out the exhaust line when the activating air flow is started. Because of a few degrees temperature gradient formed during the prerun cooling smaller particles may be thermally precipitated but no quantitative data is presently available on this effect.

This chamber is designed to measure activity of artificial nuclei from generators using acetone or isopropylamine solutions and only where concentrations on the order of $10^{3}$ or higher can be sampled. Inadvertant contamination results in a "background" or "blank run" count of $100 \mathrm{~cm}^{-3}$ or less.

## PROCEDURE

The settling surface in the sample well and the walls of the well are cleaned with ammonia solution and acetone and the entire cold-chamber assembly placed on the microscope stage. With dry nitrogen circulating through the chamber the system is cooled to the desired temperature and the nitrogen is shut down at any temperature between 0 and the run temperature. At this time the aerosol generator is placed in operation and the sample obtained through a small syringe. The chamber temperature need not be exact at this point but within one or two degrees of the run temperature. The aerosol is then injected into the sample well through the syringe needle and the aerosol allowed to settle for a specified period. During this period one must cool or warm the chamber to reach the desired run temperature. At this time the prepared moist air (which can be flowing through the flow meter during the cooling period but disconnected from the chamber) is allowed to pass through the chamber and initiate the nucleation process. For runs at $-20^{\circ} \mathrm{C}$ the prepared air generally has a dew point of +5 to $-2^{\circ} \mathrm{C}$, this yielding nearly saturated conditions at the sample surface but insufficient water to freeze and plug the air line. The resulting crystals are permitted to grow and are photographed and counted for efficiency calculations. A very interesting aspect of all experiments performed with this system is that a maximum number of nuclei are formed when the dew point is sufficiently high (such as $+5^{\circ} \mathrm{C}$ ) as to provide water for all available nuclei. For dew points of air around $-5^{\circ} \mathrm{C}$ for run temperatures of $-20^{\circ} \mathrm{C}$ not all nuclei are activated. Diffusion of water vapor to the chamber walls undoubtedly depletes a large share of the vapor pressure of the entering air.

## CLOUD SETTLING CHAMBER FOR ICE NUCLEI COUNT

## by

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## INTRODUCTION

In February of 1960, the Japanese Cloud Physics Group tried to compare ice nuclei concentrations with use of their individual ice nuclei counters at Hirosaki, Japan. Seven kinds of equipment were used for this purpose taking air samples from the same polyethylene bag. Even though it could not be explained satisfactorily, there were great discrepancies in reduced concentrations of ice nuclei counted. The only possible explanation was that the amount of moisture supplied to the counter is one of the major factors controlling the number of crystals formed in the chamber. This conclusion encouraged us to develop an ice nuclei counter in which the humidity in the chamber can be controlled. This paper presents the counter we developed for this purpose in 1961.

## DESCRIPTION OF EQUIPMENT

Even though in the past nine years the detail of equipment has been improved, the essential princible has not changed. So in this paper only the improved equipment is shown in Fig. 1.


Fig. 1. Apparatus

A plastic tube is mounted on top of an isothermal chamber in which temperature is controlled by adjustment of refrigerant pressure. On top of this tube an electric skillet is placed upside down. A sponge whose temperature can be controlled by adjusting the skillet temperature is glued to the skillet surface.

When wet, the sponge diffuses moisture downward which condenses to water droplets at less than 1 cm below the sponge. Figure 2 shows the size distributions of the water droplets taken at 5 cm below the sponge for various wet sponge temperatures. Condensation occurs in the part of the plastic tube which is near room temperature. The water droplets sedimentate exactly at Stokes' speed without significant changes of size and concentration. If any freezing nucleus hits the droplets, they freeze into ice crystals, drop into a sugar solution placed at the bottom of the chamber and grow to visible size in it, while the supercooled water droplets which do not freeze in the chamber fall into the sugar solution and dissolve in it without making ice crystals. The sugar solution is made from mixing 100 parts water with 130 parts of sugar by weight. The temperature of the sugar solution is lowered to -12 to -15 C .


Fig. 2. Size distributions of fog droplets at 5 cm below wet sponge at several different wet sponge temperatures.

RELATION BETWEEN TEMPERATURES OF WET SPONGE AND FOG DROPLETS

The warmer the wet sponge, the more water vapor it releases; consequently it results in the formation of more droplets under the sponge. Figure 3 shows the relationship between the wet sponge temperature and droplet concentration. It is noted that the curve seems to be flat at the sponge temperatures higher than $40^{\circ} \mathrm{C}$.

## RELATION BETWEEN WET SPONGE TEMPERATURES AND ICE NUCLEI DETECTED

Diluted AgI smoke was used to obtain the relation between wet sponge temperatures and ice nuclei detected. The relation is shown in Fig. 4. It shows that the plots are very scattered on the figure. It must be due to AgI decay in the AgI storage bag. So the times for taking samples are also shown in the figure. The figure shows that the number of ice nuclei detected changes with temperature of the wet sponge. Also it shows a constant value of ice nuclei at the sponge temperatures around $40^{\circ} \mathrm{C}$. This is the reason the temperature of wet sponge was adjusted to $40^{\circ} \mathrm{C}$ for normal counting of ice nuclei.

## PROCEDURE AND FURTHER REMARKS

The chamber wall is coated with glycerin before cooling. At the time of IWCIN, a CSU expansion type


Fig. 3. Relation between temperature of wet sponge and fog droplets concentration at 5 cm below the wet sponge.


Fig. 4. Relationship between sponge temperature and ice nuclei detected (AgI). The numbers on vertical coordinate show the concentration of ice nuclei in 20 cc air of diluted AgI smoke. Numbers on graft are observed time.
counter is used as an isothermal chamber with no expansion cooling. Air to be measured is flushed through the chamber with a vacuum pump.

For various chamber temperatures, the temperature of sugar solution is controlled by use of a piece of plastic ring underneath the sugar solution tray.

Temperature distribution in the chamber is shown in Fig. 1. According to the figure, the temperature varies only about $0.4^{\circ} \mathrm{C}$ between the center of the chamber and 1 cm out from the side wall. Even though the humidity condition near the wall may be different from the center because of glycerin coating at the wall, it seems to be similar to the temperature distribution in the chamber. The temperature of the sugar solution is slightly higher than that of air, so that slight convection would be expected. However, the temperature of air above the sugar solution is warmer by only $0.5^{\circ} \mathrm{C}$ than that of middle center. So probably such a slight convection would cease at most 3 cm from the bottom.

The wet sponge (temperature is adjusted to $40^{\circ} \mathrm{C}$ ) is placed on the plastic tube for $3 \frac{1}{2}$ minutes (or 210 sec ) to allow water droplets to sweep the nuclei in the air inside and crystals in sugar solution to grow to visible size. The depth of the chamber used in the workshop is 30 cm , so that a $10 \mu$ water droplet arrives at the sugar solution tray in 100 sec .

The most significant feature of this counter is that the humidity in the chamber is maintained at 100 percent wi th respect to $10 \mu$ diameter water droplets. (The humidity with respect to flat water is estimated $100.012 \%$ ). The air in the chamber is not mixed with air outside of the chamber, because of lower temperature in the chamber. So this is neither a mixing type chamber nor diffusion type chamber, because it has been designed as an isothermal chamber. Only water droplets are supplied into the isothermal chamber. To form a lot of water droplets, enough condensation nuclei near the wet sponge are needed. Such condensation nuclei are enough for formation of water droplets during $3 \frac{1}{2}$ minutes. Falling of many cloud droplets can be observed from outside of the plastic tube.

There must be two kinds of ice nuclei in natural clouds. One type is ice nuclei which collide with droplets that then freeze into ice crystals; another type is that in which the particles condense to water droplets first, then freeze into ice crystals. The former ones must be colliding with the water droplets supplied by the wet sponge (see Fig. 5). The latter may be active in the chamber because of $100 \%$ humidity with respect to $10 \mu$ diameter water droplets, which is slightly higher ( $0.012 \%$ ) humidity than water saturation.

The advantages of this device to measure ice nuclei concentration are: 1) reasonable principle of system, 2) low cost and easy machine operation. The disadvantages are: 1) it takes time to cool air to be measured and sugar solution, 2) only intermittent measurements are planned, 3) not automatic counting. Actual operations of the machine for long running were successful (Ohtake, 1964).

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Fig: 5. Relationship between fog droplets concentration and ice nuclei detected
$x$ measured values at Hirosaki
0 measured values at Hirosaki
(adjusted for decay of AgI)

- measured values at Sendai (adjusted for decay of AgI)


## CSU ISOTHERMAL CLOUD CHAMBER <br> by

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The Colorado State University (CSU) isothermal cloud chamber, originally designed by Professor Roger L. Steele, is a large vol me ambient pressure chamber primarily intended for the study of artificial ice nuclei. Figure shows the isothermal cloud chamber and control panel.

The chamber basically consists of a closed cylinder 4.5 ft in diameter by 5 ft high and constructed of $1 / 4^{\prime \prime}$ aluminum. Cooling is accomplished by a twophase Freon flow in a system of 1 " tubes welded to the outside of the aluminum shell at $6^{\prime \prime}$ spacings. These features together with a $6^{\prime \prime}$ polyurethane insulation were designed to maintain a temperature variance

of $\pm .1 \mathrm{C}$ on the chamber walls. An outer steel shell provides mechanical support and protection. Inside the chamber a black velvet liner, about $5^{\prime \prime}$ from the walls, was originally installed to reduce frost and shattering of ice crystals from the walls but turned out to be important for maintaining a cloud with acceptable density gradient. The effective experimental volume within the liner is $1.4 \mathrm{~m}^{3}$ with $.94 \mathrm{~m}^{2}$ bottom area.

An ultrasonic nebulizer (Monaghan 670) produces cloud droplets of approximately $5 \mu$ mean diameter which are carried by a glass fiber filtered, cont inuous stream of air ( 40 liters $/ \mathrm{min}$ ) into the cloud chamber. A precooler mounted in the center below the chamber lowers the temperature of the incoming cloud to nearly chamber temperature; the fog subsequently rises slowly in the center of the chamber and is then distributed throughout the volume. The continuous flow exist through a number of small openings in the sides of the chamber.

Without changing the droplet size considerably, the 1 iquid water content (LWC) can be varied from . 3 $\mathrm{g} / \mathrm{m}^{3}$ to about $4 \mathrm{~g} / \mathrm{m}^{3}$. The LWC is determined by measuring the dew point of a cloud air sample aspirated through a heated probe (where all the 1 iquid is evaporated) into a Cambridge dew point hygrometer. The LWC is then calculated from dew point and cloud temperature. The LWC is highest in the center of the chamber, decreases slowly to about $70 \% 6^{\prime \prime}$ from the walls, and to about $30 \% 1$ from the walls.

Temperatures throughout the system are measured by thermocouples and are recorded continuously. Depending on LWC and temperature, the vertical temperature gradient is negligible inside the experimental volume (i.e., inside the velvet). Controlling equipment can keep the chamber temperature constant with time to within $\pm .2 \mathrm{C}$ over the range of 0 to -22 C .

Ice nuclei samples (usually carried in large syringes) are injected into the cloud through a small opening in the side of the chamber. The number of ice crystals developing in the chamber is determined by sampl ing settling crystals on microscope slides placed near the bottom of the chamber, halfway between wall
and center. The slides are then removed through the side opening to be examined microscopically in a small cold box. Experiments have verified that the position of the slides in the chamber results in a representative sample from which the total number of crystals can be calculated.

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DESCRIPTION OF THE GOETZ AEROSOL SPECTROMETER

## by

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## THE INSTRUMENT

The Goetz Aerosol Spectrometer (Goetz et al., 1960; Goetz et al., 1962) is essentially an ae $\frac{\text { alo }}{\text { a }}$. centrifuge which consists of an electric motor driven rotor in the form of a cone (A), (Fig. 1). The conical rotor carries on its surface $2^{\frac{1}{2}}$ turns of a double helix (B) that forms three of the four walls of two independent, identical channels (C) of uniform (parallelogram) cross-section. The channels are sealed by an exactly fitting sampling foil (D) which is held airtight in position by a conical cun ( $E$ ).


Fig. 1. Cutaway view of the G.A.S. centrifuge. Arrows indicate air flow. A-Conical rotor, B-Conical helix, C-Channel, D-Sampling foil, E-Cup, F-Entrance orifice, G-Exit orifice, H-Removable jet orifice.

As the cone is rotated on its central axis, the helix imparts momentum on the air, causing it to follow down the channels in a continuous laminar flow. If a suspended particle enters the grooves of the helix, the specific gravity difference between the air and the particle causes the latter to fall out against the foil which can be microscopically inspected on removal.

Size dispersion of a polydispersed aerosol deposit on the foil comes about from the different fallout rates (Stokes law) of the various sized particles. Particles with a large aerodynamic diameter $D$ are deposited along a short strip on the foil, while small particles are deposited over a greater length; the deposits overlap since they all start near the channel entrance. Hence, the particle concentration along the foil is related to the particles' cumulative aerodynamic diameter size distribution. This distribution is given by

$$
\mathrm{dN} / \mathrm{dD}=\mathrm{dC} / \mathrm{dl} \cdot \mathrm{dl} / \mathrm{dD} \cdot \mathrm{~A}(\mathrm{D}) / \mathrm{S},
$$

where $\mathrm{dC} / \mathrm{dl}=$ gradient of the particle concentration along the deposit length 1 ,
$d 1 / d D=$ slope of the curve relating 1 and $D$,
$A(D)=$ areal deposition function of monodispersed particles, and
$S \quad=$ sampling rate.
It is possible to find the size distribution by just microscopically enumerating the particles along the foil after both $\mathrm{dl} / \mathrm{dD}$ and $\mathrm{A}(\mathrm{D})$ have been established by calibration of the Spectrometer with monodispersed particles of known size.

## CALIBRATION OF THE INSTRUMENT

Contrary to the earlier verdicts of the Spectrometer's inability to size polydispersed aerosol accurately (Stöber, 1964; Rabbe, 1966; Green and Lane, 1966; and Baust, 1967), Gerber (1970) found proper operation after removal of the baffle from the instrument's inlet tube and for specific operating conditions of the instrument (rotor rpm, R; exit orifice size, 0 ; given as $R / 0$ ).

Monodispersed polystyrene latex particles were used to calibrate the Spectrometer for a large selection of R/0. Those which gave the best accuracy were used to find $\mathrm{dl} / \mathrm{dD}$ and $\mathrm{A}(\mathrm{D})$ (Fig. 2). Figure 2 shows that the three $R / 0$, which must be used independently, give an operating range from 350 A to $20,000 \mathrm{~A}$.


Fig. 2. Collecting foil deposit length $l_{D}$ vs aerodynamic diameter $D$ of monodispersed latex particles (dashed lines; effective particle density is $1.0 \mathrm{gm} \mathrm{cm}^{-3}$ ). The area of the foil covered by particles of size $D$ is given by $A(D)$. The operating mode of the Spectrometer $D / R / 0=$ particle diameter , A/ rpmx $10^{-3}$ /exit orifice diameter, mil.

The Spectrometer was also calibrated for thermally produced AgI particles ranging from 200A to 1500A (Fig. 3; Gerber et al., 1970).

The experimentally measured resolving power of the Sepctrometer for the chosen $R / 0$ is given in Fig. 4.

The flow rates for $18 / 16,18 / 30$, and $6 / 80$ are respectively $0.49 \mathrm{Lmin}^{-1}, 2.50 \mathrm{~L} \mathrm{~min}^{-1}$, and 3.95 L $\mathrm{min}^{-1}$.


Fig. 3. The deposit length $I_{D}$ of AgI particles (Katz-Davis generator) as a function of their projected diameters (0). The latex particle calibration points (■) are used to find the equivalent aerodynamic diameter of spherical AgI particles ( $\Delta$ ) as well as the effective density $2.06 \mathrm{gm} \mathrm{cm}^{-3}$ and shape factor 2.17 of the generated AgI particles. The Spectrometer operating mode is $18 / 16$.

## SIZING ICE NUCLEI

Allee, Gerber, and Weickmann (1968, 1969) developed a technique which utilizes the Spectrometer to size ice nuclei. The suspended nuclei are deposited as usual on the polished chrome foil which upon removal from the instrument is exposed to a water vapor environment at the desired sub-freezing temperature. Under these conditions the active nuclei grow into visible ice crystals, (Fig. 5). The nucleating mechanism is either sorption or freezing depending on the chosen vapor pressure over the hydrophobic foil. To find the diameter of the nuclei, only the location of the corresponding ice crystals need be determined since the coordinates of the foil are related to particle size through the calibration curves of either Fig. 2 or Fig. 3.

The outstanding advantage of the Spectrometer sizing technique over all others is that it is capable of discriminating between the sizes of active and inactive nuclei. The fact that all ice nuclei generators have efficiencies much less than $100 \%$ makes this ability indispensable.


Fig. 4. The relative dispersity $\sigma / D$ of monodispersed particles for the optimum operating conditions of the GAS as a function of $D$. The contribution due to the Brownian diffusion of the particles is given by $B$.


Fig. 5. A portion of the flattened Spectrometer collecting foil on which active AgI particles have been grown by sorption at -20 C to visible ice crystals. The two independent deposits start at the top and progress diagonally across the foil in alternate strips.

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(4) Rapid Expansion Counter
$\frac{\text { SLOW ACTIVATION AUTOMATIC }}{\text { ICE NUCLEUS COUNTER }}$
by
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## INTRODUCTION

To estimate in the laboratory the number of ice nuclei which are active under natural atmospheric conditions it is essential that the method for testing should simulate the natural atmosphere not only in regard to temperature, humidity and water content but also in regard to the time dependence of each of these parameters. Traditionally, cloud chambers of various designs have been used but all have had to compromise to some extent, particularly in regard to the relative humidity and the duration of the required experimental conditions. The result has been that although each chamber yields self-consistent results, it is uncertain whether any chamber truly simulates atmospheric conditions.

The chamber described in this report is designed to continuously and automatically count ice nuclei meanwhile enabling one to study the effect of varying the relative humidity to which the incoming sample of air is exposed. To meet this requirement the following innovations have been made:

1. In most chambers the walls are coated with glycol which absorbs water vapor so rapidly that only by intense humidification can a visible cloud be maintained in the chamber. The present chamber uses a silicone oil wash on the walls which should enable a visible cloud to be maintained without exceeding the relative humidity of a naturally growing cloud.
2. The cloud is generated by placing a constant temperature water vapor source in the upper part of the chamber or by expanding the incoming air at a controlled rate.
3. Two identical chambers are to be operated simultaneously using a common supply of atmospheric aerosol. While one chamber is run under standard conditions, the second will be used to study the effect of various humidification procedures on the number of ice crystals formed.

## GENERAL PRINCIPLES

Atmospheric air is humidified, then cooled by passage through a ten-liter cold chamber in which the nominal residence time is about one minute. Ice crystals which form within the chamber fall onto a continuous moving belt coated with supercooled sugar solution (Fig. 1). The ice crystals are carried out of the chamber by the moving belt and after they have grown to a suitable size, they are counted by an optical sensor which continuously scans the belt. After allowing the belt to warm up, thus melting, the ice crystals, the sugar solution is replaced by contact with a roller-coater. The belt is then ready for its next passage through the cold chamber.

The whole apparatus is contained in a portable aluminum case $36^{\prime \prime} \times 24^{\prime \prime} \times 30^{\prime \prime}$ high and is powered by 12 -volt batteries, thus providing for field use.

## EXPERIMENTAL DETAILS

1. For washing the chamber walls, silicone oil (Dow Corning DC-330) is pumped at a rate of 3 ml per second via a perforated copper tube located above the jacketed part of the chamber, i.e., above freezing level when drained along the wall of the chamber. The oil is collected in a gutter, pumped through a bed of silica gel where it is dried, then recirculated. There is a slow accumulation of a sludge of ice and oil in the gutter which requires that the gutter be defrosted once every hour by passing current through an appropriate heater attached to the wall a procedure which takes about two minutes. If the defrosting is not carried out soon enough, the silicone oil overflows the gutter where it is collected in a drain and carried clear of the belt. This outer drain is warmed gently at all times to prevent frost formation on its outer walls.

By washing the chamber walls with silicone oil, one eliminates any background count due to the detachment of ice crystals from the chamber walls, but one does not completely eliminate the accretion of ice on


Fig. 1. Diagrammatic section of the ice nucleus counter.
1-Sample air inlet, 2-Silicone oil distributor, 3-Wires to heater, 4-Humidifier, 5-Jacketed chamber walls, 6-Deforstable gutter, 7-Warmed surface, 8-Overflow drain, 9-Silica gel, 10-Sponge filter, 11-Gear pumps, 12Flow splitter, 13-Reversible motor, 14-Heat exchanger with dry ice, 15-Thermistor, 16-Drain out, 17-Scanner head, 18-Mylar belt, 19-Mirror surface, 20-Developing plate, 21-Base plate, 22-Drive roller, 23-Tension roller, 24-Roller coater, and 25-Sugar solution.
the walls. It is therefore necessary to periodically inspect the chamber and defrost the walls when accretions become serious.
2. The temperature of the chamber and base plate is maintained constant by sending a proportion of the coolant through a heat exchanger, the proportion being governed by the position of a motoroperated sleeve valve. A thermistor which senses the temperature of the coolant supply, forms one arm of a bridge circuit, and the out-of-balance signal from which actuates the reversible motor.

The inlet temperature of the coolant, which is the nominal temperature of the chamber, is constant to $+1 / 4^{\circ} \mathrm{C}$. The coolant temperature rises not more tha $\bar{n}$ two degrees during this passage through the chamber jacket.

The temperature of the developer plate is maintained constant by a duplicate control system.
3. The method for automatically counting the ice crystals on the belt makes use of the fact that when an ice crystal is interposed between a light source and a phototransistor, the light reaching the latter is reduced to about $10 \%$ of its original value. The resulting increase in resistance of the photo-
transistor is ample to trigger a counting circuit which not only records each crystal as a spike on a recorder chart, but records the integrated count by means of taller spikes representing "tens," "hundreds," and "thousands."

The feasibility of the optical arrangement and the response of the counter have both been checked experimentally and found satisfactory. The mechanical arrangement which has been developed for optically scanning the belt is as follows. The belt, which must be transparent, is fabricated from Mylar. At the scanning line the belt is in good contact with a cooling plate which has a highly-polished plane surface. The scanning head which is located above the belt incorporates both the light source and the phototransistor, arranged in such a way that when no ice crystals are present, the scanning beam is directed through the belt and reflected directly onto the phototransistor by the polished plate.

By means of a mechanical device the head is caused to scan across the belt at constant speed. In order that each crystal should intercept the beam once only, the time between successive scans equals the time required for the belt to move forward one crystal diameter.

Clearly, it is vital for all the ice crystals to be the same size. This is accomplished by adjusting the composition of the sugar solution so that no appreciable ice crystal growth takes place while the belt is traversing the cold chamber. Then, as the belt traverses the "developer plate," which is kept at a lower temperature than the chamber, all ice crystals will be enlarged to the same extent, by the time they have been conveyed to the scanning line.

## CONTINUOUS ICE NUCLEI COUNTER

## MRI-Mode1 1511

by
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## GENERAL DESCRIPTION

The MRI Model 1511 Continuous Ice Nuclei Counter shown in Fig. 1 is a fast-response device embodying many unique features including an optical detection system designed to respond only to ice crystals. Its compact size and low power requirements make the Model 1511 well suited for airborne as well as mobile ground and laboratory use.

Figure 1 shows schematically the basic makeup of the instrument. In operation, the air sample is first precooled to about $0^{\circ} \mathrm{C}$ and humidified in a preconditioning chamber. The sample air then passes through an expansion nozzle into the nucleating chamber where ice crystals form and grow in the water-rich environment of a supercooled cloud. Controlled adiabatic expansion reduces the temperature of the sample air from $+2^{\circ} \mathrm{C}$ to that of the test temperature, which may be as low as $-25^{\circ} \mathrm{C}$ (see Fig. 2). The expansion nozzle is kept ice free by a resistance wire heater embedded in the nozzle, and the test chamber wall is kept ice


Fig. 1. Schematic presentation of continuous ice nuclei (IN) counter.
free by a continuous coating of a non-hygroscopic fluid. This fluid also serves to maintain the wall at test temperature. All component cooling is achieved by a solid $\mathrm{CO}_{2}$ refrigeration system which requires no external power.

The instrument occupies approximately three cubic feet and requires an electrical power input of only 150 watts at 24 volts DC. Basic data output is in the form of pulses, which may be processed in various digital and analog forms.

The standard package includes a four-digit decade counter and a digital display module which provides a real-time four-digit visual presentation of either counts per liter or total counts over a selected period of time.

The front of the instrument is shown in Fig. 3. A11 sperating controls and instrument readouts are located on the front of the instrument with the exception of the expansion nozzle adjustment which is located on top.

The operating temperatures are controlled by servo-driven pumps. The control is not perfect because of the large thermal inertias and inaccuracy in sampling a large volume at one point. The fluid temperature in the nucleation chamber is adjusted by a control on the front panel. The temperature of the preconditioning chamber is set with a trimpot on the servo board, S2. (In making adjustments, one must allow for considerable backlash inherent in the trimpot.)

The instrument detects ice crystals ontically. Because of the small size of the ice crystals and the losses due to polarizing, the amount of light reaching the photodetector is very small. The photoelectric detection circuit must, therefore, have the lowest possible noise level and a high order of stability.


Fig. 2. Relation between expansion, cooling, and vacuum in nucleation chamber.

The instrument utilizes a very sensitive photomultiplier tube with closely regulated supply voltage. Moderate frequency shaping of the photomultiplier signal reduces the spurious counts which are inherent in all photomultipliers. A conventional operational amplifier receives the shaped signal, amplifies it, and impresses it on a voltage comparator.

The photomultiplier gain can be adjusted over a very wide range by the screwdriver adjustment in the high-voltage power supply. This is available through an access hole in the lower rear panel. The signal-to-noise ratio is practically constant between 1000 and 1700 volts. For quietist performance, the face of the photomultiplier tube must not contact anything.

The counting level of the voltage comparator may be adjusted by a trimpot on the side of the counting amplifier circuit board. Sufficient amplification is available to produce continuous counts on background noise alone.

The counts are made visible by the digital panel display. The counts are also fed to the small connector on the panel for connection to a recorder.

The Nixie panel display can be used in several ways. With switches in COUNT and MANUAL position, it will count consecutively. The reset button will return the display to zero. With switches in STORE and AUTOmatic position, the count will be internal and will be displayed at the end of a desired interval. This interval can be adjusted by the panel potentiometer. With a sample air flow of $\frac{1}{4}$ liter per second, a 4-second setting of the sample interval will give the proper count for each liter of sample air through


Fig. 3. Front view showing location of operating controls and instrument readouts.

1-Expansion nozzle adjust, 2-Humidifying chamber, 3-Flow meter, 4-Flow control, 5-Vacuum gage, 6-Counter, 7-Auto/manual select, 8 -Store/count select, 9 -Internal adjust, 10 -Reset, 11 -Wall fluid temp. control, 12-Temperature check selector, 13-Temperature gage, 14 -Wall fluid heat, 15-Observation window, 16-Lamp, 17-Electronics, 18-Servos, 19-Vacuum, 20-Power, 21Sample input, 22-Access for external cooling, 23-Recorder output, 24-28 VDC Input, 25Panel removed, 26-Drier simp jar.
the instrument. The electronic board labeled READOUT conditions the signal for the digital display, which does its own dividing.

The recorder signal is produced by the COUNT board. Each individual count causes a small signal. Each tenth count causes a somewhat larger signal. Each hundredth count is larger, and each thousandth count even larger. This makes possible with only one channel on an analog recorder to record all counts from units to thousands.

## POWER SUPPLY

This instrument is intended for operation on 24 volts ( 12 cells) of lead-acid storage battery or from an aircraft power supply of 28 volts $\pm 4$ volts. This device requires about 6 amperes of current. The critical circuits and optical system lamp are regulated so that any voltage above $23 \frac{1}{2}$ volts and below 32 volts is suitable. Polarity must be observed.

Provisions are made for illuminating the nucleation chamber while the instrument is operating. A
light through the optical port on the rear of the instrument will illuminate the chamber so that it can be viewed through the port on the front panel. This method can be used to check the wall coating or to observe the cloud as it passes through the chamber.

## NCAR MODIFICATION OF BIGG-WARNER EXPANSION CHAMBER COUNTER <br> by

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The NCAR instrument is a modification of the U.S. Weather Bureau version of the Bigg-Warner counter, a generally well designed instrument with, however, a serious defect. If a sample is taken from a chamber, duct, or outdoors, it must be passed through the pressurization pump. This can lead to contamination of the pump by particles lost from the sample, which in turn results in a low total count for the original results sample and in spurious counts (caused by particles shed by the pump) for later samples. Because of the virtues of the Bigg-Warner counter -its convenience for obtaining temperature spectra and its positive response to ice crystals -- we decided to retain the basic design and modify it.

Figure 1 shows the NCAR modifications. The chamber is now flushed with sample air by an auxiliary pump at 101 pm for 3 min . A nearly absolute filter which must withstand sudden pressure changes and remove all particulate matter is installed between the pressurization pump and the chamber. If desired, the chamber exhaust can be collected in a balloon or plastic bag and vented into a hood.

Before any modification is attempted, all tubing connected to the chamber must be replaced because it may continue to shed active particles.

A properly modified unit will produce zero counts when flushed only with the pressurization pump even at $-25 C$. One should, however, be careful to verify that evaporation has not rendered the sugar solution inactive by dropping some frost into the solution which should give an immediate response. A pressurization pump that has been badly contaminated with AgI may release iodine and continue to produce a few counts despite a good filter. Such pumps should be replaced.

The above modification introduces a correction, in addition to the altitude effect, since the pressurization air dilutes the sample. The count is simply multiplied by the factor:
atmospheric pressure + pressure due to pressurization atmospheric pressure

We normally operate the chamber at -10C. The sugar solution that works best at this temperature consists of 425 gm of sugar and 60 drops of $25 \%$ aerosol OT wetting agent in 650 ml of water. The water should be hot to dissolve the sugar.

The chamber walls must always be checked for frost. The appearance of "dry" spots on the wall are


Fig. 1. NCAR modification of the Bigg-Warner counter.
indicative of incipient frost. The two inlets in the chamber wall must be coated frequently with glycerine using a cotton swab. Numerous counts opposite the inlets mean that frost in the lines is being blown out on expansion. It helps to wipe the pan edge with glycerine. This prevents water that condenses on the cold pan when it is outside the chamber from turning to ice that nucleates the solution.

With these modifications and precautions the counter has proven to be a valuable research tool. Most data taken with the unmodified counter are doubtful.

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## by

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## INTRODUCTION

The USWB version of the Bigg-Warner expansion counter has been described previously (Warner, 1957; USWB, 1959; Kline and Brier, 1961). An apparatus description will not be repeated here.

The instrument has been evaluated by Reinking (1970a, 1970b). Some of the basic characteristics of the instrument determined in these investigations will be outlined here.


## PERFORMANCE CHARACTERISTICS

All current techniques for routine field measurements of nucleus concentrations have many limitations as well as advantages. No existing instrument meets the requirements necessary to be an absolute reference unit. Precision (consistency) and accuracy (representation of nature) of the various devices must be known to interpret the measurements correctly. In this study previous and new observations have been combined to establish the advantages and limitations of the Bigg-Warner rapid expansion instrument in relation to the collection and activation of the ice nuclei, and the growth, detection and counting of the nucleated ice crystals. Some of the principal results of this study are outlined here in terms of eight criteria for ice nucleus counter evaluation:

[^1]1. Mechanical reliability and convenience of operation are strong points of the instrument. Simplicity is emphasized in construction and operating procedures.
2. Time continuity of measurements. The expansion unit is the only device which can be used to obtain successive counts at different nucleating temperatures at 10-12 minute intervals. The expansion-produced temperatures do not require changing and stabilizing the (constant) wall temperature. Measurements are immediately available for interpretation in the field.
3. Pre-nucleation effects on particles. Pump heating of sampled air will tend to destroy the activity of preactivated or preconditioned ice nuclei. Stokes law computations indicate that particles larger than about $2-3 \mu$ will settle out during a 5 -minute pre-expansion period allowed to stabilize temperature and humidity of the air sample. Computations with standard diffusive deposition equations show that roughly $10 \%$ of particles with $d \leq 0.01 \mu$ will be lost by diffusion to the walls. Theories of size effect on nucleation indicate that the sedimentation loss of nuclei is most significant. On the other hand, in an aged aerosol population, few particles larger than $2-3 \mu$ will remain.
4. Pressure characteristics. Pressure transducer measurements show that any particular overpressure is repeatable to at least $+2 \mathrm{mb}( \pm 0.05 \mathrm{C})$. The air is overpressured from ambient pressure. Variations of the latter can cause a maximum deviation of $\pm 0.5 \mathrm{C}$ and a normal deviation of $\pm 0.1 \mathrm{C}$ from the theöretical nucleation temperature. Expansion rate is the same for all overpressures (Fig. 1). Observer effects on decompression time from a given overpressure are negligible (Fig. 1) and are not expected to affect nucleation temperatures.


Fig. 1. Total air sample decompression time, $t_{R}$, as a function of overpressure, and of corresponding temperature drop for a pressure elevation of 840 mb .
5. Temperature and moisture characteristics and the nucleation process. A 5 minute preconditions period is necessary to bring the air sample to a steady state temperature. The moisture level at the end of this period regulates the cloud formed on
expansion. Cambridge hygrometer measurements of dew point up to this time using initially dry air ( $T_{d}=-6.8 C, e / e_{W}=0.11$ ) and initially extremely moist air $\left(T_{d}=+25.8 C, e / e_{W}=1.0\right)$ show that moisture diffusion to the walls and evaporation from the sugar solution greatly reduce initial moisture differences (Fig. 2). In almost all situations the final pre-expansion saturation ratio will be between 0.7 and 0.8 .


Fig. 2. Saturation ratio with respect to water as a function of time elapsed during the preconditioning period. Wet air cases are indicated by $W$, dry by d. Dashed curve $=$ ice saturation.

Temperature uniformity in the chamber after expansion is shown in Fig. 3 for a theoretical $T=$ -20C. Without compensation, a count underestimate of $4.5,8$ and $18 \%$ for $T=-14,-20$, and $-26 C$, respectively, would result from portions of the chamber being warmer than T. Supersaturations (SS's), droplet nucleation and growth, and cloud duration during and after expansions also influence ice nucleation and the nucleus count. Cloud durations in the chamber are not extremely different for moist and dry air (Fig. 4). This is due to the


Distance From Center (cm)

Fig. 3. Cross-sectional chamber map of nucleation temperatures actually attained on expansion to a theoretical temperature of -20 C .


Fig. 4. Cloud duration $t$ in the expansion chamber. (a) Very moist intake air, $T=23 C$.
(b) Dry air, $T_{d}=4 C \quad(P=840 \mathrm{mb})$.
equalization of moisture content during preconditioning of the air sample. The cloud persists for a time substantially longer than the maintenance of the minimum nucleation temperature ( -5 s ). SS's greater than in natural clouds do occur in the chamber, but the following evidence shows that their effects on nucleus count are less influential than often assumed. The expansion proceeds at a finite rate; it is not instantaneous (Fig. 1). Natural condensation nuclei are present in the air sample. Droplets form at very low SS's and continue to grow throughout the expansion, thus reducing the SS. A realistic estimate of maximum peak SS is roughly $50 \%$. Mossop and Jayarweera (1969) found that SS has little effect on freezing (AgI-NaI) nuclei, whereas a significant effect on sublimation (AgI) nuclei was observed for $T \geq-15 C$, but not for colder temperatures. Alkezweeny (1970) observed that at -18C an increase of SS from $1 \%$ to $22 \%$ increases the count by $1.5-3 x$, but SS's above $22 \%$ in the $22-60 \%$ range produce little increase in (sublimation) nucleus count. These observations apply to the expansion count. A further factor that will cause the SS to play a subordinate role is the short presistence time of SS's in the chamber. The nucleus count is influenced by the duration of SS (Alkezweeny, 1970; Fletcher, 1962). The post-expansion duration of SS is $<1 / 10$ the time that the cloud remains within IC of the minimum nucleation temperature $\mathrm{T}_{\mathrm{f}}$. (Determined by measuring time to completion of cloud development in the chamber as revealed by laser beam extinction). The measured post-expansion supersaturation durations of 0.1 to 0.3 sec agree with estimates by Mason (1951). A further experiment has shown that the time lag in nucleation leads to activation of a significant number of "post-expansion nuclei." These activate in the absence of supersaturations.

The net effect of SS, temperature and the resulting cloud may be examined by comparing mean expansion and mixing chamber spectra. In both types of chambers, SS's exist and increase toward colder temperatures. This increase is somewhat greater in expansion units at least at the colder temperatures. The expected relative position and slope of the nucleustemperature spectra if SS's were the dominant factor, and the supersaturation in the expansion chamber were greater for larger expansions and greater than in
the mixing chamber is illustrated in Fig. 5a. The expected relative spectra if persistence of both minimum temperature and cloud are the dominant factors (both are less in expansion than in mixing chambers) is shown in Fig. 5b. An extensive series of comparisons by Kline and Brier (1961) resulted in mean spectra as in Fig. 4b. The mixing curve-expansion curve separation varies with mixing technique, but mixing chambers generally read about $2-5 x$ higher. The evidence is that SS in the expansion unit partially compensates for the temperature nonuniformity effect but is not the dominant factor in determining the nucleus count.


Fig. 5. (a) above, and (b), below. Expected nucleus temperature spectra under conditions specified in the text.
6. Detection of nucleated ice crystals. Computations of crystal growth and evaporation times related to settling velocity and cloud durations show that crystals nucleated when $T_{f} \leq-20 C$ will definitely survive for a time sufficient to settle into the sugar solution for detection. For $T_{f} \leq-17 C$, detection will be marginal for crystals other than those formed and settling from relatively near the chamber base. Thus, at -14C as few as $1 / 3$ of the crystals may be detected. Partial compensation is provided by the facts that (1) due to the temperature distribution in the chamber, a greater fraction of crystals will be produced in the lower part, (2) the average natural nucleus concentration at -14C is below the counter's minimum detectable level ( $0.1 / \ell$ ), so when average-or-below conditions apply no crystals will be detected with or without the undercount, and (3) the calculations were made assuming crystal evaporation rate was equal to growth rate; actually the evaporation is slower.
7. Contamination of the air pump without nucleating material will occasionally occur. It is estimated that observed contamination has occurred at the level of about $10^{10}$ particles of which only a minute fraction is released with any one measurement. Maximum possible contamination due to an upwind seeding operation is estimated at $10^{6}$ nuclei, assuming all nuclei passing through the pump are collected. Even with this impossible collection "efficiency," the $10^{6}$ level is $10^{4}$ lower than observed. In a great number of field seeding cases, the measured nucleus count has been observed to quickly retreat to levels comparable to the pre-seeding background concentrations after seeding has been stopped. It is very unlikely that the observed contamination is due to pump collection of seeding material dispersed upwind. All evidence indicates that the pump contaminates only when a quite active nucleant is handled in large quantities in the immediate vicinity of the instrument. This means that contamination may be controlled with precautions. The contamination factor must be watched very carefully.
8. Comparisons. Intercomparisons of several rapid expansion counters resulted in count correlation coefficients $\geq 0.95$ for paired series (Kline and Brier, 1961). The natural cloud is the one absolute reference available for calibrating ice nucleus counters. Observed ratios of natural cloudproduced ice crystal concentrations to ice nucleus concentrations measured by the expansion method at corresponding temperatures for individual unseeded cases were $1.3,3.3,1.7,0.5$, and 0.33 . For cases seeded with AgI, ratios were $1.0,0.8,1.0$, and 0.2 . Cloud top temperatures for the various cases ranged from - 15 to -26C (Hindman, 1967; Grant, 1968; and Reinking and Grant, 1968). The agreement is good. Counts obtained with the expansion unit are certainly within a range of magnitudes that have direct physical meaning in terms of cloud processes.

## CONCLUSION

A more comprehensive version of the evaluation has been completed by Reinking (1970b). The results of the study provide the basis for distinguishing between instrument effects and natural effects on the nucleus count. Utilization of their results will greatly enhance reliability of interpretation of data obtained using the expansion technique. Further definition of the performance of expansion counters will emerge from interpretation of the data obtained during the IWCIN. As with all current ice nucleus measuring devices, improvements are in order. A number of improvements have been incorporated in a new CSU Expansion Counter.

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THE COLORADO STATE UNIVERSITY RAPID-EXPANSION ICE NUCLEUS COUNTER
by

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## INTRODUCTION

The Colorado State University (CSU) rapidexpansion ice nucleus counter is of the cloud chamber design utilizing the expansion principle to achieve the heterogeneous nucleation of ice crystals, and the supercooled sugar solution method to detect those crystals that form. The CSU counter incorporates several modifications to the basic design of the instrument built by Warner (1957) and modified by the U.S. Weather Bureau (U.S.W.B., 1959; Kline and Brier, 1961). The most important specific changes are:

1) Direct monitoring and control of the nucleation temperature achieved by expansion. The temperature drop on expansion is not controlled indirectly by pressure as in previous instruments; therefore it is not necessary to assume that the temperature

[^2]drop on expansion bears a specific (moist adiabatic) relationship to pressure drop. Electronic monitoring results in greater temperature repeatability.
2) A flushing system in which the sample air is pulled rather than pushed through the chamber. Use of this system permits the sample air pump to be placed after the nucleation chamber in the line of flow so as to negate effects of pump contamination sometimes observed in the USWB unit.
3) A thermal insulated lid (still transparent for viewing cloud and crystals) to produce a more uniform vertical temperature profile in the chamber.
4) Improved accuracy in wall temperature monitoring.

The CSU instrument may be used to obtain nuclei concentrations for nucleation temperatures in the $-12^{\circ} \mathrm{C}$ to $-30^{\circ} \mathrm{C}$ range. It is suitable for measuring concentrations of natural "background" nuclei as well as artificial nuclei used for cloud seeding. Even with improvements, all-important simplicity of operation has been maintained. This makes the instrument suitable for routine field operation.

## GENERAL DESCRIPTION

A schematic of the ice nucleus counter is presented in Fig. 1. The atmospheric sample is drawn in through the intake hose through Valve 1 into chamber by vacuum from the vacuum pump through Valve 2. The chamber is sealed and the sample is preconditioned to the temperature of the walls (normally -8C). A valve then is opened to allow flow of air into a vacuum. This action produces the expansion which results in cooling of the sample air to pre-set minimum temperature and simultaneous formation of a supercooled cloud. Moisture for the cloud is that contained in the original air sample plus that evaporated from the sugar solution, and minus that absorbed by glycerine which is coated on the walls to prevent frost. The ice nuclei act in the cloud to form ice


Fig. 1. Diagram of the Colorado State University rapid expansion ice nucleus counter. 1-Cloud chamber, 2-Sugar tray, 3 -temperature drop controller, 4-Lid, 5-Insulation, 6-Wal1 temperature sensor, 7-Refrigerating compressor, 8 -Vacuum tank, and 9-Vacuum pump.
crystals which settle into a tray of supercooled sugar solution where they grow to a size which can be counted visually. The cloud nucleation temperature achieved is the sum of the wall temperature and the temperature drop on expansion. The number of crystals counted is taken as a direct representation of the number of nuclei active at that cloud temperature, per the 10 liters chamber volume. Each observation takes approximately 12 minutes.

Refrigeration System. Refrigeration of the cloud chamber is accomplished with a standard freon22 system (see Fig. 1). The refrigerant circulates through coils encircling the outside of the copper chamber so as to provide a uniform wall temperature. Wall temperature is monitored in the -6 C to -11C range by a thermistor mounted in contact with the outside bottom of the chamber. The wall temperature can be cooled from room temperature to the normal sample precooling temperature of -8 C in approximately 15 minutes. Supercooling of the sugar solution is accomplished by placing the solution container in thermal contact with the bottom of the chamber. The refrigerant compressor is operated continuously throughout the period in which measurements are taken.

Air Intake and Expansion System. The chamber is flushed and the air sample is obtained by pulling new air through the chamber. An air pump positioned "downstream" from the chamber pulls air in through Valve 1 (Fig. 1), through the chamber, and out Valve 2. After five minutes of flushing, Valves 1 and 2 are electronically closed to seal in the air sample. The air sample remains at ambient pressure throughout a 5 -minute preconditioning period. Meanwhile, oneration of the pump is continuing to draw air from the vacuum tank. After the sample preconditioning period, and with the pump still in operation, only Valve 2 is opened electronically to allow expansion of sample air into the established vacuum. The air pump is kept in operation at all times.

The temperature drop occurring with the expansion is controlled by means of a thermistor and an associated valve-control bridge circuit. Before the expansion, the reference temperature dial (a reference resistor in the bridge) is set to the desired minimum temperature to be achieved on expansion. The thermistor resistance rises as the air temperature drops during expansion. This provides a monitor of temperature throughout the course of the expansion. Calibration against a resistance thermometer is such that the instant the dropping air temperature in the chamber reaches the desired minimum, the thermistor resistance achieves a value equal to that of the reference resistance. This balances the bridge and triggers a relay which automatically closes Valve 2. This stops the expansion; the temperature drods no further.

It is important to note that the thermistor does not monitor the actual temperature of the air inside the chamber. Rather, it follows a temperature in the exhaust tube which is proportional to the actual temperature in the chamber. However, through use of the existing proportionality, the temperature calibration has been done in terms of actual chamber temperature. Calibrations are particular to the individual counters. The temperature, pressure, and humidity characteristics of the chamber before and after expansion are similar to those for the Bigg-Warner expansion chamber as
described by Reinking (1970). However, the duration of the expansion is longer in the new counter (of the Order of 0.3 s instead of 0.07 s as occurs in the old expansion instrument).

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(5) Droplet Freezing

DROP FREEZING INSTRUMENT

## by

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## INTRODUCTION

While most ice nucleus counters observe the concentration of these particles in the air, it is most important to know their concentration in
precipitation because it is these nuclei that have actually participated in the formation of the rain or snow. The instrument described here has been successfully used to obtain freezing nucleus concentrations in precipitation as a function of temperature. In addition, some efforts have been made toward the use of it in observing sublimation and contact nuclei.

Freezing nuclei and contact nuclei are differentiated according to the temperature of the water droplet. If the droplet is initially above $0^{\circ} \mathrm{C}$ when the nucleus and droplet come together and nucleation occurs when the system is supercooled then we observe a freezing nucleus. If the droplet is below $0^{\circ} \mathrm{C}$ when it and the nucleus collide and nucleation occurs upon collision or shortly after we have contact nucleation. From a molecular viewpoint both of these are the same.

To determine the concentration of freezing nuclei the work of Vali (1968) has been followed. An array of water drops are placed on a cold stage and cooled at a constant rate. The relation between the temperature and the number of unfrozen drops is then used to determine the nucleus concentration spectrum. It is assumed that the concentration $n$ of freezing nuclei effective at a supercooling $T_{s}$ or less (i.e., between temperatures $-T_{S}$ and $0^{\circ} \mathrm{C}$ ) is given by

$$
n=n_{0} \exp \left(\alpha T_{s}\right)
$$

where $n_{0}$ and $\alpha$ are constants. The probability $P$ that a drop of volume $V$ contains at least one freezing nucleus effective at $T_{s}$ is, from Poisson's statistics,

$$
P=1-\exp (V n)
$$

and therefore

$$
n=-\frac{\ln (1-P)}{V} .
$$

Now $P$ is given by the fraction of the drops in the array which are frozen at $-T_{S}$. Hence,

$$
\begin{equation*}
n=(-1 / V) \ln \frac{N\left(T_{s}\right)}{N_{0}} \tag{1}
\end{equation*}
$$

where $N\left(T_{S}\right)$ is the number of unfrozen drons at temperature $-T_{S}$ and $N_{0}$ the total number of drops in the array. The quantity $n$, defined by Eq. (1) is the probability that at least one freezing nucleus active at some temperature above $-T_{\mathrm{S}}$ will be found in a drop of volume $V$ and it is termed the cumulative spectrum. The number of freezing nuclei per unit volume which become active in a particular temperature interval is called the differential spectrum.

These same principles can be applied to sublimation and contact nuclei but changes in experimental procedure are necessary. If the array of precipitation drops are evaporated, the nuclei are left behind as a residue. The surface with this residue can then be subjected to a supersaturation whereupon sublimation will occur on the appropriate particles. If, on the other hand, supercooled droplets are allowed to fall onto the residue, contact nucleation may occur. Under these conditions, $V$ in Eq. (1) is the volume of the evaporated drop, and $N\left(T_{s}\right)$ of the number of residual locations with no ice crystals, and $N_{0}$ is the total number of residual locations.

## INSTRUMENTATION

This apparatus consists of three basic sections: (1) the environmental control; (2) the cold stage; and (3) the associated electronics and data recording system. These are integrated into Fig. 1. Each is described separately.


Fig. 1. Drop freezing instrument.

1. The environmental control. Filtered air is passed through a radiator, a humidifier and then over the cold stage. Its temperature is controlled by regulating the temperature of a methanol bath which is circulated through the radiator. This air temperature can be varied between $-30^{\circ} \mathrm{C}$ and $+30^{\circ} \mathrm{C}$ and will fluctuate less than $.2^{\circ} \mathrm{C} / 24 \mathrm{hr}$ at $-10^{\circ} \mathrm{C}$. The cold air is humidified to saturation by passage through damp towels hung in the air passage. A flowmeter on the input monitors the rate of flow of the air.
2. The cold stage. The basic part of this section is an Ohio Semitronics TA-6F thermoelectric cooling unit. It is $4 \times 6$ inches and capable of a temperature difference of about $30^{\circ} \mathrm{C}$ between the cooling surface and the heat sink. This heat sink is kept at a constant temperature by bringing the methanol from the radiator and circulating it through the heat exchanging fins of the sink.

Attached to the cooling surface is an aluminum vacuum manifold. This manifold has a large number of small holes drilled in the top and all connected to a common connecting tube. Applying a vacuum to this connecting tube allows a thin sheet of plastic film ("Handiwrap" or "Saran Wrap") to be held down tightly and uniformly to the cooling surface. This eliminates the use of greases to hold the clean surface to the substrate in good thermal contact. A thermistor is imbedded in this aluminum manifold for sensing the temperature of the sample.
3. Associated electronics and data recording. Two YSI Thermilinear thermistors are used to monitor the temperature -- one in the cold plate and one in the air. The output from these are used to control
the temperature of the cooler. A block diagram of the control circuit is shown in Fig. 2. The cold plate thermistor voltage is compared either with the air temperature or a ramp voltage and, through a control circuit, kept equal to it by adjusting the current passing through the thermoelectric cooler. The reversing relay provides for changing the direction of this current flow.

This circuitry allows us to keep either a constant sample temperature $\left(-30^{\circ} \mathrm{C}\right.$ to $\left.+50^{\circ} \mathrm{C}\right)$, a constant temperature difference between the cold plate and the air $\left(-30^{\circ} \mathrm{C}\right.$ to $\left.+30^{\circ} \mathrm{C}\right)$ or a constant cooling rate $\left(+4^{\circ} \mathrm{C} / \mathrm{min}\right.$ to $\left.-4^{\circ} \mathrm{C} / \mathrm{min}\right)$. The air temperature which is controlled by the methanol is regulated separately.

The phase of the water is recorded on a video tape recorder which has the advantage of instant replay and stop action. Originally the water was viewed using the crossed polaroid lenses and reflected light but this has been found to be unnecessary. A voice channel allows coordination of freezing events with the temperature or any other parameter of interest.


Fig. 2. Temperature control circuit (block diagram). 1-Thermoelectric cooler, 2-Direct current thermometer, 3-Air thermometer, 4-Amplifier, 5-Amplifier, 6-Ramp, 7-Comparator, 8-Control, 9 -Power supply for T.E.C., and 10 -Reversing Relay.

## PROCEDURES

Because experimentation in utilizing this instrument for observing sublimation and contact nuclei has not been completed, only the procedure for determining the freezing nucleus concentration will be outlined. It is quite straight forward. With the cold stage temperature at about $+10^{\circ} \mathrm{C}$, a clean sheet of polyethylene film ("Handiwrap" manufactured by Dow Chemical Corp.) is placed on it and the vacuum pump turned on. A disposable syringe and 26 gauge needle are rinsed with a small portion of the sample to be investigated and then used to make an array of 100 drons on the smooth plastic surface. The individual drops are about 0.25 inches apart to prevent any interaction. After the drops have been made, a cover is put over them and they are cooled at a rate of about $1.5^{\circ} \mathrm{C} / \mathrm{min}$. Their phase is monitored by the television camera and audio notes of the temperature are made at each whole degree Celsius.

At the end of the run, the temperature of the plate is returned to +10 ; the sample, film, syringe, and needle are discarded and a new run prepared.

Collecting procedures. This equipment is normally used to measure the concentration of freezing nuclei in precipitation samples. Since the nuclei used during the workshop were aerosolized, a technique was
used to transfer the nuclei into filtered, distilled water. This was done by passing a measured volume of aerosol-containing air through a membrane filter in a conventional manner. The next step was to turn the filter over and flush the particles back off the filter with water. This water was then used for analysis. By knowing the amount of water used to reverse flush the filter, and the volume of air sampled, one can determine the number of nuclei per liter of air.

Two correction factors need to be considered. First is the background nucleus concentration in the wash water. Figure 3 shows the mean nucleus spectrum as calculated from a number of different runs. These values should be subtracted from the basic nuclei/ gram spectra. The second factor is that the flushing may not get all the nuclei off of the filter. Vali has suggested the calculated concentrations are a factor of 6 too low because of this.

Neither of these factors have been included in data.


Fig. 3. Freezing nucleus concentration in distilled water.

AN AUTOMATED DROP FREEZER SYSTEM FOR DETERMINING THE FREEZING NUCLEUS CONTENT OF WATER
by
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## INTRODUCTION

It has been shown by Vali (1966) that the population of freezing nuclei present in samples of water

can be determined by using the drop freezing technique. This technique involves the placement of equally sized drops on a thermally controlled surface and monitoring when the drops freeze together with the temperature, rate of cooling, etc. The newly designed and fabricated Vali-Knowlton dron freezing system utilizes these principles and when operated in the auto-mode automatically monitors and records on magnetic tape ( $\frac{1}{2}$ inch IBM compatible) the germane parameters; when the system is operated in the photomode, the freezing events and temperatures of freezing are recorded on 16 mm movie film. The auto-mode is utilized for the detection of freezing nuclei present in samples of water which can be systematically placed on the cold stage; the photo-mode is used for detecting freezing nuclei present in drops not placed in the systematic fashion on the cold stage, i.e., single snow or ice crystals which have been captured and melted. The final nucleus concentration as a function of temperature can be obtained by processing the data from the magnetic tape utilizing the proper software and a digital computer. The spectra can be obtained from the photographic data by simply viewing the film and selecting the required information. When the system is operated in the auto-mode it is not difficult to obtain the nucleus spectra for 30 samples in an 8 -hour period.

## SYSTEM DESCRIPTION

The Vali-Knowlton System (see Fig. 1) consists of six basic units which are (1) cold stage assembly, (2) detector, (3) temperature controller, (4) digital controller-multiplexor, (5) magnetic tape recorder and (6) power supply assembly.

1. Cold stage assembly. The cold stage assembly provides the working surface on which the sample drops are placed for observation. The working surface of the cold stage consists of a copper block which is thermally bonded to a set of thermoelectric coolers. The remaining side of these coolers are attached to the top of a larger copper block which serves as the intermediate stage. The bottom of the


Fig. 1. Drop freezer system.
intermediate stage is attached to an additional set of thermoelectric coolers which are in contact with a forced air heat exchanger. The cold stage assembly is shown on Fig. 2.

The intermediate stage is kept at approximately -10C by supplying a controlled amount of current to the intermediate stage thermoelectric coolers. Each of the copper blocks has a thermistor embedded in it to facilitate determination of their temperatures.
2. Detector. When the system is operated in the auto-mode the drops of equal size are placed on the working surface in an 11 by 11 matrix. The drops are covered with a transparent plastic box and illuminated. When the drops freeze, they change from a clear to an opaque appearance. Light from the frozen drops is passed through a lens and focused on an $11 \times$ 11 matrix of phototransistors. The frozen state of the individual drops is detected by the phototransistors and an electrical signal is generated and recorded on the digital magnetic tape.


Fig. 2. Cold stage assembly.
In order to insure registration between the drop placement and the detector matrix a similar array of miniature incandescent lamps is projected through the same lens, thereby indicating the proper placement of each drop on the working surface. When the drops have been placed on the working surface the lamp location matrix is slipped to one side and the detector matrix is placed in the same position over the lense. The physical arrangement of the detector is shown on Fig. 3.

When the system is operated in the photo-mode, a 16 mm movie camera is mounted on one side of the cold stage. The drop freezing events and the temperature of the events are incrementally recorded at any selected rate.
3. Temperature controller. The temperature controller provides the means by which the cold stage temperature can be programmed to operate in a number of different modes. The voltage produced by the network associated by the thermistors embedded in the


Fig. 3. Detector assembly.
copper blocks is amplified and used to control the temperatures. The temperature controller provides a voltage corresponding to the desired temperature, which is compared to the voltage from the thermistor voltage amplifier. The difference between these voltages is transmitted to the power drivers which control the current to the appropriate thermal coolers thereby producing an observed temperature of the working surface equal to the selected temperatures.

These are three modes in which the temperature controller operates which are a) preset, b) cool and c) warm. When the preset mode is selected, the temperature controller produces a voltage determined by a precision potentiometer located on the control panel. The voltage provided from this potentiometer is used by the feedback temperature control loop to warm or cool the working surface temperature to the preset temperature which will be maintained to $+0.1^{\circ} \mathrm{C}$.

Operation of the system in the cool mode requires the temperature control to produce a voltage corresponding to $0^{\circ} \mathrm{C}$ and thus, the working surface is cooled to $0^{\circ} \mathrm{C}$ at a maximum rate. When the working surface achieves a temperature of $0^{\circ} \mathrm{C}$, the temperature controller generates a linearly decreasing
voltage corresponding to a selected temperature which the feedback temperature controller uses to control the working surface temperature. The cooling rate can be preselected on the control panel utilizing a precision potentiometer. The rate remains linear to within $\pm 0.2^{\circ} \mathrm{C}$ from $0^{\circ} \mathrm{C}$ to $-28^{\circ} \mathrm{C}$. When the temperature of the working surface reaches the selected cold limit the temperature controller terminates the linear voltage decrease and the feedback temperature controller maintains the selected working surface temperature.

When the system is onerated in the warm mode the temperature controller produces a voltage corresponding to the ambient temperature or a temperature selected by the warm limit potentiometer on the control panel.

A block diagram of the temperature controller is shown on Fig. 4.

A typical temperature time profile as presented on Fig. 5 is produced when the system is operated in the various modes.


Fig. 4. Temperature controller.


Fig. 5. Temperature-time profile.
4. Digital controller-multiplexor. The digital controller-multiplexor processes the various signals necessary for digital recording. This portion of the
system consists of an oscillator, controllermultiplexor, clock, temperature counter, display, and record check. The oscillator is a precision 100 kHz square wave unit which produces the base line for all timing and control signals. The controller counts the oscillator signal, decodes it to produce all the timing signals necessary for the complete system operation. Typical of the signals which are produced are the inter-record gap and step and record pulse for the recorder, transfer and reset pulses for the temperature counter, reset pulses for the multiplexor, end of tape pulses for the end of tape indicator on the control panel, etc. The multiplexor serializes all digital data into the proper format for recording on the digital magnetic tape. This unit contains a 32 bit ring counter which produces the 326 -bit character record once each second. The clock receives 1 -second pulses from the controller and counts them up to 24 hours and decodes the counters for the time display and multiplexor. The clock is setable to any desired time using thumb wheels on the control panel. The temperature counter converts the analogue temperature voltage to a digital frequency and counts this frequency for a 1 -second period to obtain the digital representation of the temperature. The temperature counter contains the necessary decode for display and recording of the temperature. The analogue voltage representing temperature covers the range of $0-5$ volts corresponding to a temperature range of $-50^{\circ} \mathrm{C}$ to $+50^{\circ} \mathrm{C}$. The voltage to frequency conversion corresponding to this temperature range is 0 to 2000 cps linear to $0.1 \%$. The display unit decodes the digital signal from the clock and temperature counter and displays them numerically utilizing rear projection screen indicators.

The record check is a digital system used to interrogate the multiplexor output to the recorder. It is a final check on the data transmitted to the recorder to determine if it is correct. The record check displays the 6-bit character selected by the proper push-button on the control panel. All 32 characters are checked by selecting the appropriate push-buttons. The digital portion of the digital controller-multiplexor uses Amelco high noise immunity integrated circuits operating at +12 volts. A block diagram of the digital controller-multiplexor is shown on Fig. 6.


Fig. 6. Digital controller multiplexor.
5. Magnetic tape recorder. A magnetic digital stepping tape recorder receives the signals from the digital controller and produces the proper signals to drive the magnetic tape transport and thus produce the permanent magnetic tape record. The recorder also generates an inter-record gap and an end of fil mark on the tape, making it IBM compatible. The recorder uses $\frac{1}{2}$ " magnetic tape at 500 bpi and records at 100 cps. The recorder produces odd parity character and has an odd parity echo check capability. A block diagram of the incremental tape recorder is presented in Fig. 7.


Fig. 7. Incremental tape recorder.
6. Power supply assembly. The power supply assembly supplies the regulated power necessary for the thermoelectric coolers, the temperature controller and for the digital controller. The assembly consists of five individual power supplies. Two of these are 7 volt -- 30 amp units for the working surface thermoelectric coolers; one unit is a 12 volt -- 30 amp power supply for the intermediate stage thermoelectric cooler and two units are $\pm 12$ volt -- 6 amp supplies used for the digital electronics and temperature controller.

The power supply assembly also contains forced air heat exchangers necessary to cool the power supplies and power transistors.

## SYSTEM SPECIFICATIONS

1. Power consumption -- 1200 Watts.

Linear cooling rate -- linearity $\pm 0.2^{\circ} \mathrm{C}$.
Preset temperature control limits $\pm 0.1^{\circ} \mathrm{C}$.
Cooling rate range is 0 to $6 \mathrm{deg} / \mathrm{min}$.
Preset temperature range is $+30^{\circ}$ to $-40^{\circ} \mathrm{C}$.
Cold limit range is $0^{\circ} \mathrm{C}$ to $-30^{\circ} \mathrm{C}$.
Warm limit range is $0^{\circ} \mathrm{C}$ to $+30^{\circ} \mathrm{C}$.
Time for intermediate stage "lock on" approximately 10 minutes.
9. Clock resolution 1 second.
10. Clock accuracy 3 seconds ner day.
11. Temperature display resolution $0.05^{\circ} \mathrm{C}$ (relative accuracy).
12. Temperature display accuracy $0.2^{\circ} \mathrm{C}$.

## ACKNOWLEDGEMENTS

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USE OF THE VALI-KNOWLTON (UNIVERSITY OF WYOMING) NUCLEUS SPECTROMETER AT THE IWCIN

by<br>G. Vali University of Wyoming Laramie

For the purposes of aerosol measurements, the system will be operated to analyze membrane filters. The filters will be exposed in the usual manner and will be processed in three different ways:
a. The filters are saturated with Vaseline or oil and are placed at the bottom of a small diffusion chamber. Vapor is supplied by an ice surface which is situated at close spacing above the filter surface. Supersaturation is controlled by controlling the temperature difference between the filter and the ice surfaces. The filter is kept at the desired conditions for periods of 30 to 45 minutes after which the number of ice crystals which developed on the surface are counted.
b. Water droplets of millimeter size are placed at regular spacing on the filter surface. From the freezing temperatures of these drops while the filter is gradually cooled, and by taking into account the fraction of filter surface covered by the water drops, the population of freezing nuclei on the filter are evaluated. In this manner, nonsoluble nuclei active in bulk water are detected.
c. As an alternative to $b$ above the filter can be reverse-flushed with clean water and the numbers of nuclei on the filter deduced from the concentrations detected in the rinse water. The details of this elutrition technique have been described in the proceedings of the Lannemezan Workshop.

In addition to the aerosol measurements, the system will be operated to determine the nucleus contents of water samples (precipitation) to compare with other similar methods.

The instrument itself consists of four main parts: cold stage, ice plate, optical detector, and control and recording system*. Figure 1 shows a general view of the apparatus. Both the main cold stage and the ice plate are cooled by double stage thermoelectric coolers with forced air heat removal. Temperature measurements are made by the use of thermistors. The temperature of the main cold stage is programmable from the control panel. The temperature of the ice plate can be controlled either independently or can be slaved to the cold stage in order to maintain a fixed temperature difference. Figure 2 illustrates

[^3]

Fig. 1. General view of the Vali-Knowlton nucleus spectrometer.


I CENTIMETER

Fig. 2. Cross-sectional view of the diffusion chamber for filter processing.
the physical arrangement of the diffusion chamber and Fig. 3 shows the pattern of temperature variations of the cold stage and of the ice plate during the processing of a filter. Care is taken to maintain conditions of sub-saturations throughout the periods preceding and following the actual period of development. During development, supersaturations can be maintained with accuracies of approximately $0.5 \%$. The crystal count is obtained after the filter has been warmed to a temperature slightly below $0^{\circ} \mathrm{C}$ where no additional nuclei will be activated during the observation. This is especially important since when the ice plate is removed the filter is momentarily exposed to the room air which undoubtedly creates very high supersaturations. The optical detector is used for the automatic detection of the freezing of water drops. When operating in this mode, that is analyzing water samples, the relevant system parameters and the freezing of the water drops are recorded on digital magnetic tape. This tape is computer compatible so that all data handling and analysis can be done on the computer to provide the final output in the form of a nucleus spectrum.


Fig. 3. The sequence of temperature variation of the filter ( - ) and of the ice surface (---) for the processing of a filter in the diffusion chamber. $T_{0}$ is the temperature of development and $\Delta T$ is the temperature difference between filter and ice which produces the desired supersaturation.

## SECTION VI

## OTHER WORKSHOP ACTIVITIES

SUMMARY OF OTHER ACTIVITIES DURING THE SECOND IWCIN

by<br>\section*{Joseph Podzimek}

1) On the recommendation of the Steering Group some of the operators of the apparatus tested (Whitby, Langer, Grant, Scott) enabled other participants attending the IWCIN without an instrument to help them during the workshop. This cooperation contributed much to the wider knowledge of the working methods and to the estimation as to what degree does the accuracy of the measured nuclei characteristics depend on the experience of the operator.
2) During the first days of the IWCIN the chemical formation of aerosol in Bag B leading to the high rise of Aitken nuclei during some minutes has been observed. Prof. K.T. Whitby and Dr. R. Husar using the MAAS in a systematic way were studying the possible sources of these nuclei and came to the following conclusions:

For the chemical aerosol formation at least two requirements seem to be necessary; the presence of unfiltered atmospheric air and the presence of duct tape (probably evaporating the alpha pinene). There was also some indication from the first runs during the preparation week that cooling reduced the aerosol formation. The final result of all the investigations was the improvement of the storage in Bag B, which was reached after removal of all duct tape (and any material emitting the terpene vapor) and cooling of the air outside of Bag B.
3) The Organizing Committee and the Steering Group of the 2nd IWCIN organized an informal meeting to discuss a) terminology as applied to nucleation and b) proposed mechanisms for that nucleation process leading to the eventual formation of ice. The convenor of the meeting, Prof. M.L. Corrin, submitted the following summary of the discussions which took
place Thursday evening, August 20, 1970, at the CSU Atmospheric Science Building.

It was proposed that nucleation terminology be at present restricted to a statement defining the initial and final states and that nucleants be described as (1) deposition-VS (2) condensation - VL (3) freezing - LS (4) vaporization - LV (5) melting S1 and (6) sublimation - SV. It was also suggested that terms such as "contact nucleation" be precisely defined by users. Other classifications were suggested. Considerable discussion followed with regard to the proper usage of such terms as "heterogeneous nucleation," "homogeneous nucleation," "phase," etc. It was a general feeling of the group that it should at present make no recommendations regarding nucleation terminology; it was recognized that a better understanding of nucleation processes is essential.

A number of short presentations concerning the mechanisms of nucleation were presented by participants. These included the concept of phase transitions in the water film adsorbed on solids and the existence of a two-dimensional ice-like phase on which ice nucleation occurs, experiments dealing with history effects observed with organic nucleants and the freezing of water droplets, results of a study of the adsorption of water vapor on "pure" silver iodide at relative humidities in the saturation region, and a progress report on the adsorption of water vapor on three organic nucleants. There was vigorous discussion on the part of many of the participants. Some of the additional topics discussed included (a) history effects, (b) the chemical nature of nucleants as related to nucleation mechanisms (c) electrical effects and (d) possible changes in mechanisms with altering ambient conditions. It was pointed out that (a) experiments designed to fully understand the mechanisms of ice nucleation may depart markedly from conditions in a cloud and (b) possibly many mechanisms, some of which may occur simultaneously, are involved in the ice nucleation process.
4). Thanks to the initiative of the IWCIN participants (Langer, Robertson, Vali) using the drop freezing method the apparatus based on this method were tested August 13, 1970, using a Soil Suspension. The description of these experiments, submitted by Prof. G. Vali is the following:

Sample preparation. Approximately 0.1 g of soil was put into 50 g of distilled water. The soil originated from the grassy area just north of the Cloud Simulation Laboratory. Most of the soil was allowed to settle out over a period of a few hours. A syringe was then filled with water near the top of the container. A diluted sample was also prepared by using water from the syringe to dilute with distilled water in the ratio of 1:10.

Testing. The samples were tested by three different arrangements of drop freezing: Robertson's, Langer's, and Vali's. The cumulative nucleus spectrum for the sample was deduced from the numbers of drops frozen. The range of detectable concentrations was extended by using the diluted sample.

Results. See attached graph (Fig. 1).
5) On the proposal of Dr. H. Gerber, the simultaneous measurement of the size distribution of the AgI aerosol used during the 2nd IWCIN with the MAAS and the Goetz centrifuge was made August 20,1970 . This comparison should give the aerodynamic diameter size distribution and the mean effective density and shape factor of the particles.


Fig. 1. Freezing nuclei in water suspension of CSU dust sample Aug. 13, 1970.

## SECTION VII

## SUMMARY OF THE WORKSHOP

It will be many months or years before analyses of data collected during the second IWCIN will be completed. A brief summary of workshop activities prepared immediately after its completion appeared appropriate. Such a summary was made and presented to the opening session of the American Meteorological Society Conference on Cloud Physics held on the campus of Colorado State University immediately following the workshop. J. Podzimek described the Conference, U. Katz and K. Whitby described the laboratory facilities, R.E. Ruskin and W.C. Kocmond summarized the Condensation Nuclei Investigations and E.K. Bigg summarized the Ice Nuclei Investigations. This section of the report includes the presentation of Drs. J. Podzimek, Ruskin and Kocmond, and Bigg. The presentations describing the facilities are included earlier in this report in Section IV.

## CONFERENCE DESCRIPTION

by
Joseph Podzimek
I feel very honored speaking at the beginning of the Conference on Cloud Physics of the American Meteorological Society in Fort Collins and, on behalf of the President of the International Commission on Cloud Physics of the IAMAP IUGG, Dr. Helmuth Weickmann who is not able to attend this opening session, I would like to wish you full success for this carefully prepared conference.

It is a great pleasure for me to inform you on the request of the Organizing Committee of this conference about the general scope of the 2nd International Workshop on Condensation and Ice Nuclei, which just ended. The more detailed information about the laboratory facilities and the activity during the Workshop will be presented by Dr. Katz, Prof. Whitby, Dr. Ruskin and Dr. Bigg, the members of the Steering Group of the Workshop.

One may ask why we started to organize the Workshops on Condensation and Ice Nuclei, the first of which took place in 1967 in Lannemezan, France, thanks to the enthusiasm and extreme diligence of Prof. H. Dessens. In this connection let me only mention the era of Aitken pocket counters and of Scholtz counters fifty or forty years ago. At this time the world wide measurements were organized using these counters and the results contained in the monographs of Burckhardt - Flohn or Landsberg were interpreted in meteorology. Later Junge, Kohler and others came to the conclusion that these measurements have little to say to the processes governing the formation of clouds or fogs. The situation became much more complicated 30 years later after the discovery of sublimation and ice nuclei. We started to ask to which degree the processes inside the apparatus simulate the conditions in the clouds and we wanted to know more about the reproducibility and accuracy of the measurements. Many of the institutions were asking advice about which one of the offered apparatus to buy for a special aim and which is the best for airplane
or field measurements. The Subcommittee on Nucleation decided in 1966 to organize the workshop though being aware that we cannot give an unambiguous judgement of a certain type of apparatus used for different purposes, but we were sure that publishing almost all the original data from a testing of apparatus made under well controlled laboratory conditions, we can contribute to the improvement of the designs of these instruments and facilitate the choice of apparatus bought by other institutions.

What are the main experiences we made during the Lannemezan meeting 1967? The main conclusion drawn from the comparisons of the measurements using natural nuclei are seen in the slides representing some of the evaluation of the data from the measurement of Aitken nuclei, cloud condensation nuclei and ice nuclei. The first slide (Fig. 1) shows nice agreement between two Gardner's counters operated by Hogan and Slob but not so perfect response between Gardner's counters and GE counter. Relatively good agreement was shown by the apparatus measuring cloud condensation nuclei presented by Jiusto, Gagin and Auer. In Fig. 2 we see that the correlation of the data measured at the supersaturation $0.1 ; 0.3 ; 1.0$; $3.0 \%$ is close. However, there is very weak correlation between Aitken nuclei counters and Jiusto's chamber working at the supersaturation of $1.0 \%$. Speaking about ice nuclei counters and looking at the comparison of the data in Fig. 3, we can easily draw a conclusion that the apparatus based on the same principle and having the same dimensions gave comparable results. It is nicely seen from the ice nuclei measurements, i.e., with the aid of the mixing chamber of the Soulage-Admirat type. On the other hand, there is a big difference between the apparatus based on different principles like the SoulageAdmirat chamber and the Langer-Rosinski acoustic counter. In general, the highest concentrations were measured in the 300 liter expansion chamber


Fig. 1.


Fig. 2.


Fig. 3.
operated by Mossop and in all mixing chambers of Soulage-Admirat type. Unfortunately, the number of measurements made simultaneously at the same temperature or humidity is too low, this is seen in the table (Fig. 4) showing some of the possible correlation factors of the apparatus used. The testing of the apparatus using the artificial nuclei did not appear to

| IN | No. <br> Cases | Correl. <br> Factor |
| :---: | :---: | :---: |
| Admirat (b) -Admirat (I. floor) | 7 | 0.85 |
| Admirat-Benesch | 5 | 0.60 |
| Admirat-Podzimek | 4 | 0.80 |
| Admirat-Langer | 11 | -0.30 |
| $\quad$ CCN | 16 | 0.91 |
| Jiusto-Auer | 8 | 0.86 |
| Jiusto-Gagin |  |  |

Fig. 4.
give ambiguous results. All the laboratory tests were made in a partially controllable environment. The generated particles were stored in a mylar bag and supplied to the apparatus in a large syringe. Some measurements support the idea that we did not succeed in storing well-controlled aerosol particles for any extensive time.

Knowing all the imperfections of our workshop 1967 we tried to improve first of all the control of the laboratory rooms and of the aerosol particles used for testing the apparatus. Our program in Fort Collins was originally based on the assumption of the use of a natural aerosol and of a possibly monodisperse artificial aerosol and of the storage of aerosol particles in such a way that it will be possible during one hour to keep aerosol in a well controlled state. The condition of a monodisperse aerosol is connected with the difficult technique of the generation of these particles and is limited by the production rate of aerosols in a unit of time. On the other hand, we have better control of the processes leading to the time changes of the size spectrum of particles stored in a vessel and generally speaking of the stability of physical parameters inside the chambers to be tested. The use of two level storage of aerosol particles, which was necessary for taking the samples of aerosols at much higher concentrations in an automatically working analyzing device, evoked many questions relating to the invariability of size spectra of aerosols during an extended time in both storage bags, in the ducts and in the sampling tubings.

Bearing all these facts in mind, we established our program during the 2nd International Workshop on Condensation and Ice Nuclei in Fort Collins. Following this program we wanted to check the stability of size spectra of stored aerosols, to get the information of the size spectra of all particles used for the testing, which were natural nuclei, NaCl particles, teflon, phloroglucinol and AgI particles generated in different ways. Testing and comparing the apparatus we try to deduce the humidity characteristic of cloud condensation nucleus counters using the supersaturations $0.3 ; 0.75 ; 1.0 \%$, the temperature characteristic of the ice nucleus counters using mainly temperatures $-12 ;-16$; and $-20^{\circ} \mathrm{C}$ and in size characteristic of all counters using well defined size spectrum of nuclei. The final result of our

Task should be the deduction of some correlation factors between the apparatus and the checking of the reliability of the apparatus and reproducibility of their data. To what degree we succeeded in this task you will judge after the presentation of preliminary reports and after reading the report of this workshop which will appear probably at the beginning of the next year.

In closing this communication on behalf of the Subcommittee on Nucleation, allow me to thank the National Science Foundation for its financial support of this Workshop; the Colorado State University for hosting all participants in its facilities for three weeks and represented in the Organizing Committee by Prof. L. Grant; Prof. R. Steele, the chairman of the Organizing Committee; and to all its members and to the members of the Steering Group, as well. I admired the enthusiasm and diligence of Dr. Katz and the staff of the Simulation Laboratory, and Prof. Whitby and Dr. Husar, as well, who contributed much to the running of all the laboratory work. Last but not least, many thanks to more than forty participants of the Workshop, who showed an admirable discipline submitting more than 500 daily reports in time and bringing all the 29 apparatus tested in gear. In the name of all the participants of the Workshop, I would like to invite you to come in the evening to the Cloud Simulation Laboratory and look at the facilities and the instruments tested.

THE PARTICLE SIZE SPECTRA OF ATMOSPHERIC AEROSOLS MEASURED ON AUG. $10,11 \& 12$ DURING THE IWCIN

## by

## K. T. Whitby and R. B. Husar

The size spectra of atmospheric aerosols used during the atmospheric aerosol portion of the IWCIN were measured with the MAAS. This paper presents data in graphical and tabular form for the aerosol sampled directly from outside during the time when Bag $B$ was being filled and sampled directly from Bag B during the time the aerosol was being used by the workshop participants.

It will be noted that the size distribution data for runs $13,15,16,17$, and 18 are not included here. These runs were either tests with clean air in Bag B or consisted of runs not relevant to the participants of the workshop. Some runs are not as complete as others because we were occupied with other duties or because of various problems in switching the sampling lines from outdoors to Bag B. However, enough good data was obtained to indicate the general character of the size distributions measured and the magnitude of the aerosol changes experienced in Bag B due to chemical production of aerosol and due to losses by settling and diffusion.

## COMMENTS ON THE DATA

Runs 11, 12, 14, 19, and 20 show typical comparisons of data during the days when there was considerable generation of chemical aerosol in Bag B from the vapors produced by the duct tape. The large increase in aerosol number for sizes below approximately $0.1 \mu \mathrm{~m}$ was apparently caused by chemical formation of aerosol
that resulted from a reaction of something in the unfiltered atmospheric air and vapors given off by the duct tape used to seal the cracks in the tank. As seen from Run 11, the number concentration of these aerosols rose very rapidly and then decayed for sizes smaller than about $0.02 \mu \mathrm{~m}$. However, the number concentration increased with time for sizes on the order of $0.06 \mu \mathrm{~m}$. The number concentration in the 0.1 to 1 mm range remained practically unaffected by the bag filling operation and by diffusion and settling in the bag and distribution system. However, the number concentration of particles larger than 1 m decreased by progressively greater amounts in the 1 to $10 \mu \mathrm{~m}$ range until the total change at 10 um was about a factor of 8 from the direct outdoor sample to the last sample taken from Bag B.

For Run 21 it will be noted that the number concentration measured direct is smaller than that from Bag B for some sizes. The reasons for this are not known exactly but it is suspected that the aerosol sampled direct was too far removed in time from the time period when the bag was being filled.

During runs $24,25,26$ and 27 the amount of chemical aerosol production in Bag B had been reduced. Run 24 probably presents the best comparison of the direct with bag $B$ samples unaffected by the chemical formation of aerosol in Bag B. It will be noted that there is about a factor of 2 loss of aerosol smaller than $0.01 \mu \mathrm{~m}$ and larger than $1 \mu \mathrm{~m}$. In the size range from 0.02 to $0.8 \mu \mathrm{~m}$ the losses are less than a factor of 2 .

## COMPARISON OF ATMOSPHERIC SIZE SPECTRA MEASURED IN FORT COLLINS WITH THOSE MEASURED IN OTHER PLACES

The. Particle Technology Laboratory at the University of Minnesota has been studying the aerosol size spectra in urban atmospheres for a number of years. We therefore are interested in how the size spectra measured in Ft. Collins compares with previous data obtained in Minneapolis, Los Angeles and other places.

In Figure 1 we have compared a typical volume particle size distribution measured in Ft . Collins measured direct from the atmosphere with a measurement from Los Angeles having about the same total mass concentration. Also shown is a distribution obtained by Junge in the South Atlantic during the 1970 Meteór expedition.

This particular form for plotting the distribution has the property that the area under the curve in a given log size interval is directly proportional to the mass in that size range. This makes it easy to compare the relative volume or mass in the different size ranges.

Several things may be noted from this plot. First is that there is a pronounced minimum at about $1 \mu \mathrm{~m}$. Second is that the mass in the range below $1 \mu \mathrm{~m}$ and above $1 \mu \mathrm{~m}$ is relatively independent, suggesting that in general the principal sources of mass in the $D_{p}<1 \mu \mathrm{~m}$ is different than for the range above $1 \mu \mathrm{~m}$.

Further, it is easily seen that the mass distribution for Ft. Collins is quite different than that for Los Angeles and more like that obtained by Junge in the South Atlantic except that the absolute mass


Fig. 1. Volume size distributions measured in three locations.
concentration is higher. The particular L.A. distribution was selected because its $\phi$ was about the same as the average $\phi$ observed in Ft. Collins. Whereas in Los Angeles 50\% or more of the aerosol mass is usually below $1 \mu \mathrm{~m}$, in Ft. Collins $10 \%$ or less was usually below 1 mm . One of the rather striking features of the Ft. Collins aerosol was the existence of the mass mode at about the upper sizing limit of our OPC.


Run 11
August 10, 1970

| $D_{p}-\mu m$ | 9:30 | 9:47 | 9:59 | 10:10 | 10:21 |
| :---: | :---: | :---: | :---: | :---: | :---: |
| . 0075 | 24900 | 1000000 | 618000 | 4.37000 | 328000 |
| . 01 | 11700 | 1000000 | 618000 | 437000 | 295000 |
| . 015 | 5500 | 649000 | 526000 | 392000 | 279000 |
| . 02 | 3380 | 315000 | 358000 | 300000 | 254,000 |
| . 03 | 1840 | 58100 | 121000 | 132000 | 129000 |
| . 04 | 1260 | 13300 | 394,00 | 53700 | 60700 |
| . 06 | 897 | 2750 | 8100 | 14,000 | 18500 |
| . 08 | 661 | 1330 | 2400 | 4380 | 6260 |
| . 1 | 495 | 81,4 | 1080 | 1540 | 2239 |
| . 125 | 363 | 615 | 678 | 810 | 1071 |
| . 15 | 281 | 464 | 514 | 513 | 647 |
| . 2 | 158 | 277 | 275 | 252 | 281 |
| . 3 | 48.1 | 80.4 | 82.3 | 72.6 | 64.6 |
| . 6 | 4.21 | 3.72 | 4.00 | 3.34 | 3.51 |
| . 72 | 2.35 | 1.92 | 2.02 | 1.75 | 1.86 |
| . 88 | 1.63 | 1.30 | 1.39 | 1.16 | 1.25 |
| 1.02 | 1.25 | 1.03 | 1.04 | . 9174 | . 9787 |
| 1.3 | . 8272 | . 7677 | . 7064 | . 6915 | . 6604 |
| 1.66 | . 6217 | . 5992 | . 4604 | . 5706 | . 5217 |
| 2.02 | . 1.574 | . 4171 | . 3357 | . 42152 | . 31447 |
| 2.46 | . 3285 | . 2898 | .24,81 | . 3234 | . 2460 |
| 3.13 | . 21.94 | . 2162 | . 1613 | . 2617 | . 1919 |
| 1.12 | . 161,7 | . 1149 | . 0779 | .14,21 | . 0966 |
| 5.55 | . 1111 | . 0724 | . 0455 | . 09149 | . 06447 |
| 6.6 | . 0777 | . 0421 | . 0251 | . 060 ! | . 0506 |
| 8.55 | . 0391 | . 0141 | . 0072 | . 0128 | . 0374 |
| 10.78 | . 0170 | . 0030 | . 0034 | . 0043 | . 0119 |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 12
August 10, 1970

| $D_{p}-\mu \mathrm{m}$ | $10: 55$ | $11: 06$ | $11: 16$ | $11: 27$ | $11: 39$ |
| :---: | :---: | :--- | :--- | :--- | :--- |
| .0075 | 347000 | 829000 | 656000 | 421000 | 289000 |
| .01 | 90900 | 766000 | 616000 | 406000 | 289000 |
| .015 | 20100 | 332000 | 440000 | 330000 | 269000 |
| .02 | 9790 | 96100 | 226000 | 223000 | 204,000 |
| .03 | 33.20 | 16300 | 50000 | 70700 | 82500 |
| .04 | 3050 | 7760 | 15300 | 24100 | 33500 |
| .06 | 1510 | 4010 | 5070 | 6850 | 9780 |
| .08 | 1050 | 2430 | 2760 | 2970 | 3890 |
| .1 | 756 | 1550 | 1630 | 1680 | 1980 |
| .125 | 558 | 1050 | 1120 | 1060 | 1170 |
| .15 | 428 | 765 | 789 | 768 | 818 |
| .2 | 254 | 411 | 435 | 391 | 426 |
| .3 | 80 | 119 | 131 | 108 | 116 |
| .6 | 5.31 | 7.42 | 7.59 | 6.81 | 6.93 |
| .72 | 3.22 | 4.01 | 4.13 | 3.48 | 3.57 |
| .88 | 2.32 | 2.77 | 2.81 | 2.29 | 2.34 |
| 1.02 | 1.83 | 2.19 | 2.13 | 1.65 | 1.75 |
| 1.3 | 1.35 | 1.53 | 1.477 | 1.10 | 1.13 |
| 1.66 | 1.10 | 1.14 | 1.09 | $.84,89$ | .8413 |
| 2.02 | .8630 | .8021 | .7162 | .6443 | .6064 |
| 2.46 | .6655 | .5715 | .4387 | .4800 | .3966 |
| 3.13 | .4983 | .4255 | .3187 | .3528 | .3051 |
| 4.12 | .3260 | .2562 | .1855 | .1915 | .1766 |
| 5.55 | .1940 | .1553 | .1281 | .0881 | .1043 |
| 6.6 | .1357 | .0911 | .0762 | .0838 | .0626 |
| 8.55 | .0591 | .0374 | .0277 | .0285 | .0328 |
| 10.78 | .0243 | .0140 | .0192 | .0136 | .0157 |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 14
August 10, 1970

| $D_{p}-\mu m$ | $13: 51$ | $14: 36$ | $14: 47$ |
| :---: | :---: | :---: | :---: |
| .0075 | 11,200 | 321000 | 226000 |
| .01 | 10300 | 307000 | 220000 |
| .015 | 3510 | 253000 | 210000 |
| .02 | 3510 | 168000 | 165000 |
| .03 | 3510 | 62400 | 73200 |
| .04 | 2570 | 21100 | 32200 |
| .06 | 1210 | 5330 | 8910 |
| .08 | 997 | 2220 | 3160 |
| .1 | 893 | 1160 | 1400 |
| .125 | 597 | 750 | 828 |
| .15 | 437 | 533 | 570 |
| .2 | 248 | 71.1 | 296 |
| .3 | 84.9 | 5.92 | 127 |
| .6 | 4.14 | 2.41 | 6.10 |
| .72 | 2.37 | 1.46 | 2.63 |
| .88 | 1.70 | 1.07 | 1.62 |
| 1.02 | 1.35 | .7200 | 1.23 |
| 1.3 | .9779 | .5540 | .8889 |
| 1.66 | .7455 | .4008 | .6851 |
| 2.02 | .5545 | .2881 | .5170 |
| $2.1,6$ | .4077 | .2251 | .3349 |
| 3.13 | .3196 | .1353 | .2366 |
| 4.12 | .1834 | .0851 | .1043 |
| 5.55 | .1170 | $.04,68$ | .0613 |
| 6.6 | .0681 | .0213 | .0460 |
| 8.55 | .0370 | .0089 | .0111 |
| 10.78 | .0174 | .0055 |  |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 19
August 11, 1970

| $D_{p}-\mu \mathrm{m}$ | $9: 20$ | $9: 40$ |
| :---: | :---: | :---: |
| .0075 | 17100 | 319000 |
| .01 | 10900 | 281000 |
| .015 | 6530 | 241000 |
| .02 | 4100 | 171000 |
| .03 | 2160 | 51600 |
| .04 | 1320 | 16000 |
| .06 | 1020 | 3380 |
| .08 | 753 | 1380 |
| .1 | 603 | 749 |
| .125 | 497 | 571 |
| .15 | 416 | 452 |
| .2 | 248 | 260 |
| .3 | 96.2 | 103 |
| .6 | 16.6 | 14.1 |
| .72 | 9.09 | 7.41 |
| .88 | 5.99 | 4.99 |
| 1.02 | 4.37 | 3.72 |
| 1.3 | 2.77 | 2.46 |
| 1.66 | 2.05 | 1.84 |
| 2.02 | $1.41_{4}$ | 1.26 |
| 2.1 .6 | .9843 | .9264 |
| 3.13 | .7200 | .6830 |
| 4.12 | .3872 | .3847 |
| 5.55 | .2187 | .2089 |
| 6.6 | .1315 | .0770 |
| 8.55 | .0566 | .0332 |
| 10.78 | .0294 | .0200 |

Run 20
August 11, 1970

| $\mathrm{D}_{\mathrm{p}}-\mu \mathrm{m}$ | 10:10 | 10:20 | 10:30 | 10:40 |
| :---: | :---: | :---: | :---: | :---: |
| . 0075 | 12500 | 499000 | 429000 | 310000 |
| . 01 | 12500 | 457000 | 417000 | 298000 |
| . 015 | 9740 | 134000 | 273000 | 257000 |
| . 02 | 4.460 | 37200 | 123000 | 162000 |
| . 03 | 2310 | 7540 | 20300 | 16000 |
| . 04 | 1550 | 3710 | 5970 | 12800 |
| . 06 | 1200 | 1770 | 2270 | 2930 |
| . 08 | 851 | 1140 | 12140 | 1330 |
| . 1 | 64,6 | 769 | 661 | 824 |
| . 125 | 54,2 | 580 | 605 | 592 |
| . 15 | 4.04 | 460 | 4.64 | 441 |
| . 2 | 208 | 267 | 277 | 241 |
| . 3 | 85.7 | 96.2 | 105 | 98.9 |
| . 6 | 11.9 | 13.0 | 12.4 | 13.6 |
| . 72 | 6.1:2 | 6.32 | 6.01 | 5.47 |
| . 88 | 4.27 | 3.87 | 3.71 | 3.22 |
| 1.02 | 3.11 | 2.75 | 2.66 | 2.19 |
| 1.3 | 1.92 | 1.72 | 1.71 | 1.32 |
| 1.66 | 1.40 | 1.23 | 1.23 | . 9021 |
| 2.02 | . 9809 | . 8540 | . 8102 | . 5604 |
| 2.1.6 | . 6783 | . 5702 | . 5157 | . 3617 |
| 3.13 | . 504.7 | .1,102 | . 3477 | . 21,64 |
| 4.12 | . 2694 | . 2004 | . 14.64 | . 1119 |
| 5.55 | . 1770 | . 1008 | . 0702 | . 0638 |
| 6.6 | . 09714 | . 0528 | . 0511 | . 0374 |
| 8.55 | . 0519 | . 0153 | . 0230 | . 0068 |
| 10.78 | . 0234 | . 0051 | . 0060 | . 0051 |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 21
August 11, 1970

| $D_{p}-\mu \mathrm{m}$ | $11: 50$ | $13: 40$ | $14_{4}: 00$ | $14: 20$ |
| :---: | :---: | :---: | :---: | :---: |
| .0075 | 174,00 | 52700 | 197000 | 163000 |
| .01 | 13200 | 45900 | 117000 | 14,2000 |
| .015 | 104,00 | 31800 | 54,400 | 59800 |
| .02 | 6170 | 22500 | 334,00 | 27800 |
| .03 | 2970 | 12100 | 16500 | 8950 |
| .04 | 1590 | 6810 | 9150 | 8950 |
| .06 | 815 | 3050 | 4040 | 4330 |
| .08 | 563 | 1730 | 2080 | 2940 |
| .1 | 409 | 1100 | 1160 | 2130 |
| .125 | 301 | 772 | 737 | 967 |
| .15 | 238 | 572 | 536 | 505 |
| .2 | 14,0 | 301 | 272 | 209 |
| .3 | 49.3 | 89.2 | 79.5 | 76.5 |
| .6 | 5.01 | 7.17 | 6.52 | 15.9 |
| .72 | 2.53 | 3.06 | 2.81 | 9.16 |
| .88 | 1.58 | 1.75 | 1.58 | 6.04 |
| 1.02 | 1.11 | 1.24 | 1.11 | 4.19 |
| 1.3 | .6643 | .7634 | .6949 | 2.17 |
| 1.66 | .4523 | .5583 | .4843 | 1.24 |
| 2.02 | .3043 | .4234 | .2979 | .6340 |
| 2.46 | .1902 | .3021 | .1787 | .2911 |
| 3.13 | .1255 | .2251 | .1217 | .1421 |
| 4.12 | .0634 | .1149 | .0519 | .0668 |
| 5.55 | .0298 | .0694 | .0238 | .0413 |
| 6.6 | .0204 | .0434 | .0132 | .0192 |
| 8.55 | .0111 | .0243 | .0047 | .0098 |
| 10.78 | .0051 | .0157 | .0009 | .0072 |

Run 22
August 11, 1970

| $D_{p}-\mu m_{1}$ | $15: 10$ |
| :---: | :---: |
| .0075 | 40800 |
| .01 | 31500 |
| .015 | 28700 |
| .02 | 23800 |
| .03 | 15600 |
| .04 | 10200 |
| .06 | 4780 |
| .08 | 24,00 |
| .1 | 121,0 |
| .125 | 707 |
| .15 | 473 |
| .2 | 21,6 |
| .3 | 57.4 |
| .6 | 5.28 |
| .72 | 2.52 |
| .88 | 1.51 |
| 1.02 | 1.01 |
| 1.3 | .6132 |
| 1.66 | .4357 |
| 2.02 | .321 .3 |
| 2.46 | .2191 |
| 3.13 | .1536 |
| 4.12 | .0928 |
| 5.55 | .0600 |
| 6.6 | $.01,26$ |
| 8.55 | .0281 |
| 10.78 | .0119 |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO. CCM)
Run 23
August 11, 1970

| $D_{p}-\mu \mathrm{m}$ | $17: 00$ |
| :---: | :---: |
| .0075 | 292000 |
| .01 | 219000 |
| .015 | 63000 |
| .02 | 32100 |
| .03 | 13500 |
| .04 | 7160 |
| .06 | 3320 |
| .08 | 1790 |
| .1 | 990 |
| .125 | 605 |
| .15 | 411 |
| .2 | 208 |
| .3 | 59.4 |
| .6 | 4.06 |
| .72 | 1.98 |
| .88 | 1.23 |
| 1.02 | .8979 |
| 1.3 | .6136 |
| 1.66 | .4481 |
| 2.02 | .2902 |
| 2.46 | .2021 |
| 3.13 | .1511 |
| 4.12 | .0821 |
| 5.55 | .0460 |
| 6.6 | .0230 |
| 8.55 | .0115 |
| 10.78 | .0077 |

Run 24
August 12, 1970

| $D_{p}-\mu \mathrm{m}$ | 9:20 | 9:40 | 10:00 | 10:20 |
| :---: | :---: | :---: | :---: | :---: |
| . 0075 | 88000 | 31700 | 29800 | 31400 |
| . 01 | 58600 | 29000 | 29800 | 28200 |
| . 015 | 46900 | 28000 | 29800 | 27100 |
| . 02 | 37700 | 24,700 | 25500 | 23100 |
| . 03 | 21,200 | 15200 | 16500 | 15000 |
| . 04 | 12900 | 9670 | 10800 | 10100 |
| . 06 | 6650 | 5070 | 5670 | 5340 |
| . 08 | 3710 | 2810 | 3100 | 2820 |
| . 1 | 2210 | 1610 | 1720 | 1530 |
| . 125 | 1390 | 1020 | 1060 | 921 |
| . 15 | 908 | 701 | 712 | 595 |
| . 2 | 449 | 339 | 267 | 293 |
| . 3 | 120 | 92.6 | 83.3 | 88.0 |
| . 6 | 6.78 | 5.31 | 4.55 | 3.49 |
| . 72 | 4.11 | 2.99 | 2.52 | 1.94 |
| . 88 | 3.11 | 2.06 | 1.73 | 1.31 |
| 1.02 | 2.56 | 1.59 | 1.35 | 1.02 |
| 1.3 | 1.89 | 1.07 | . 9421 | . 7323 |
| 1.66 | 1.55 | . 7881 | . 7213 | . 5630 |
| 2.02 | 1.23 | . 5736 | . 5587 | . 12226 |
| 2.46 | . 9655 | . 3928 | . 4000 | . 3170 |
| 3.13 | . 7983 | . 21,17 | . 2885 | . 2294 |
| 4.12 | . 4932 | .11,26 | . 1562 | . 0974 |
| 5.55 | . 3460 | . 0970 | . 0860 | . 0553 |
| 6.6 | . 2285 | . 0647 | . 0481 | . 0396 |
| 8.55 | . 1191 | . 0132 | .014,0 | . 0226 |
| 10.78 | . 0557 | . 0043 | . 0060 | . 0094 |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 25
August 12, 1970

| $D_{p}-\mu \mathrm{m}$ | $11: 30$ | $11: 50$ | $12: 10$ |
| :---: | :---: | :---: | :---: |
| .0075 | 59800 | 39200 | 36100 |
| .01 | 39300 | 32100 | 27500 |
| .015 | 27500 | 23200 | 21500 |
| .02 | 18900 | 17400 | 17200 |
| .03 | 9630 | 9630 | 9710 |
| .04 | 5880 | 5750 | 2570 |
| .06 | 3020 | 2840 | 1590 |
| .08 | 1740 | 1620 | 843 |
| .1 | 1150 | 937 | 575 |
| .325 | 780 | 476 | 390 |
| .15 | 563 | 225 | 199 |
| .2 | 272 | 71.3 | 60.8 |
| .3 | 82.8 | 4.48 | 3.37 |
| .6 | 6.15 | 2.54 | 2.01 |
| .72 | 3.57 | 1.80 | 1.48 |
| .88 | 2.56 | 1.39 | 1.16 |
| 1.02 | 2.04 | .9872 | .7685 |
| 1.3 | 1.47 | .7643 | .5574 |
| 1.66 | 1.13 | .5494 | .4004 |
| 2.02 | .8553 | .3774 | .2740 |
| 2.46 | .5187 | .2906 | .2072 |
| 3.13 | .3536 | .1681 | .1251 |
| 4.12 | .2370 | .1055 | .0928 |
| 5.55 | .1834 | .0677 | .0681 |
| 6.6 | .0804 | .0243 | .0140 |
| 8.55 | .0200 | .0153 | .0026 |
| 10.78 |  |  |  |

Run 26
August 12, 1970

| $D_{p}-\mu m$ | 13:40 | 14:00 | 14:20 | 14:40 |
| :---: | :---: | :---: | :---: | :---: |
| $\begin{aligned} & .0075 \\ & .01 \\ & .015 \\ & .02 \\ & .03 \\ & \hline \end{aligned}$ | $\begin{array}{r} 96700 \\ 52500 \\ 21900 \\ 13500 \\ 6080 \\ \hline \end{array}$ | $\begin{array}{r} 50100 \\ 50100 \\ 34000 \\ 20600 \\ 8600 \\ \hline \end{array}$ | $\begin{aligned} & 75900 \\ & 63000 \\ & 46000 \\ & 31000 \\ & 13000 \\ & \hline \end{aligned}$ | $\begin{aligned} & 75400 \\ & 65600 \\ & 42800 \\ & 31700 \\ & 13200 \\ & \hline \end{aligned}$ |
| .04 <br> .06 <br> . 08 <br> . 1 <br> .125 | $\begin{array}{r} 4020 \\ 2390 \\ 1620 \\ 1070 \\ 771 \\ \hline \end{array}$ | $\begin{array}{r} 4430 \\ 2380 \\ 1620 \\ 1060 \\ 731 \\ \hline \end{array}$ | $\begin{array}{r} 5790 \\ 2700 \\ 1580 \\ 991 \\ 677 \\ \hline \end{array}$ | $\begin{array}{r} 6490 \\ 2650 \\ 1460 \\ 834 \\ 546 \\ \hline \end{array}$ |
| .15 .2 .3 .6 .72 | $\begin{aligned} & 601 \\ & 303 \\ & 95.5 \\ & 7.72 \\ & 4.36 \\ & \hline \end{aligned}$ | $\begin{aligned} & 522 \\ & 286 \\ & 81.5 \\ & 6.83 \\ & 3.69 \end{aligned}$ | $\begin{aligned} & 483 \\ & 250 \\ & 77.3 \\ & 5.54 \\ & 2.74 \end{aligned}$ | $\begin{gathered} 370 \\ 191 \\ 55.7 \\ 4.01 \\ 1.96 \end{gathered}$ |
| $\begin{aligned} & .88 \\ & 1.02 \\ & 1.3 \\ & 1.66 \\ & 2.02 \end{aligned}$ | $\begin{aligned} & 3.15 \\ & 2.52 \\ & 1.85 \\ & 1.48 \\ & 1.18 \end{aligned}$ | $\begin{aligned} & 2.54 \\ & 1.97 \\ & 1.47 \\ & 1.19 \\ & .9302 \\ & \hline \end{aligned}$ | $\begin{aligned} & 1.83 \\ & 1.41 \\ & 1.02 \\ & .7945 \\ & .5698 \\ & \hline \end{aligned}$ | $\begin{aligned} & 1.27 \\ & 1.01 \\ & .7170 \\ & .5523 \\ & .4085 \\ & \hline \end{aligned}$ |
| $\begin{gathered} 2.46 \\ 3.13 \\ 4.12 \\ 5.55 \\ 6.6 \\ 8.55 \\ 10.78 \end{gathered}$ | $\begin{aligned} & .9166 \\ & .7404 \\ & .4872 \\ & .3209 \\ & .2187 \\ & .1064 \\ & .01,89 \end{aligned}$ | $\begin{aligned} & .6830 \\ & .5383 \\ & .34,13 \\ & .2374 \\ & .1540 \\ & .0796 \\ & .0149 \end{aligned}$ | $\begin{aligned} & .4383 \\ & .3468 \\ & .2060 \\ & .14,68 \\ & .1060 \\ & .0600 \\ & .0391 \end{aligned}$ | $\begin{aligned} & .2885 \\ & .2174 \\ & .1026 \\ & .0583 \\ & .0302 \\ & .0043 \\ & .0030 \end{aligned}$ |

CUMULATIVE (GREATER THAN) DISTRIBUTION FOR ATMOSPHERIC AEROSOL (NO./CCM)
Run 27
August 12, 1970

| $\mathrm{D}_{\mathrm{p}}-\mu \mathrm{m}$ | 15:20 | 15:30 | 15:50 | 16:10 |
| :---: | :---: | :---: | :---: | :---: |
| . 0075 | 18200 | 504,00 | 209000 | 167000 |
| . 01 | 161,00 | 27800 | 138000 | 151000 |
| . 015 | 9990 | 13700 | 36800 | 63200 |
| . 02 | 7280 | 9160 | 14,000 | 194,00 |
| . 03 | 3690 | 4250 | 5920 | 6480 |
| . 04 | 2470 | 2580 | 34.50 | 3650 |
| . 06 | 1650 | 14,80 | 164,0 | 1700 |
| . 08 | 1050 | 896 | 957 | 956 |
| . 1 | 693 | 563 | 576 | 523 |
| . 125 | 483 | 371 | 367 | 321 |
| . 15 | 337 | 264 | 277 | 209 |
| . 2 | 166 | 14,0 | 137 | 99.5 |
| . 3 | 46.1 | 35.7 | 38.8 | 11.0 |
| . 6 | 3.42 | 1.62 | 1.46 | 1.53 |
| . 72 | 2.24 | . 8928 | . 7940 | . 8949 |
| . 88 | 1.74 | . 5987 | . 54443 | . 6038 |
| 1.02 | 1.43 | . 4643 | . 4323 | . 4621 |
| 1.3 | 1.07 | . 2991 | . 3140 | . 3183 |
| 1.66 | . 8953 | .24,09 | . 2536 | . 2370 |
| 2.02 | . 7477 | . 1868 | . 1919 | . 1672 |
| 2.4 .6 | . 6004 | . 1336 | . 1451 | . 1123 |
| 3.13 | . 49440 | . 1004 | . 1140 | . 0817 |
| 4.12 | . 3140 | . 0591 | . 04.98 | . 0523 |
| 5.55 | . 2170 | . 0289 | . 04.13 | . 0285 |
| 6.6 | . 1468 | . 0162 | . 0345 | . 0209 |
| 8.55 | . 0813 | . 0098 | . 0089 | . 0085 |
| 10.78 | . 0400 | . CO 44 | . 0030 | . 0017 |

SUMMARY OF CONDENSATION NUCLEUS INVESTIGATIONS AT THE 1970 INTERNATIONAL WORKSHOP

ON CONDENSATION AND ICE NUCLEI

## by

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## ABSTRACT

Intercomparisons were made of five cloudcondensation nucleus (CCN) instruments of the thermaldiffusion chamber type and one continuous mixing jet type at the International Workshop on Ice and Condensation Nuclei at Fort Collins, Colorado in August 1970. Six commercial Aitken nucleus or total-aerosol counters of the expansion type were compared, along with one research type Wilson cloud chamber. Some important conclusions from the experiments conducted were as follows: (1) Only the thermal diffusion cloud-chamber type of instruments provided reliable CCN measurements with agreement within $\pm 30 \%$ of each other. (2) The only particle-counting types of diffusion chambers which gave reliable counts were those using high-level illumination from a 200 -watt Hg lamp and a detector sensitivity equivalent to an $f: 1.5$ lens © $1 / 40 \mathrm{sec}$ and ASA 125 film . (3) The University of Washington automatic CCN instrument using integration of scattered light, agreed well with the particle-counting type diffusion chambers, (however, the commercial version of this instrument was not satisfactorily operable during the workshop). (4) The NCAR continuous mixing-jet type of condensation nucleus instrument appeared to subject the sample to excessive supersaturation, causing its readings to vary from $1 / 25$ to 8 times the comparative values from the thermal-diffusion chamber type CCN instruments. (5) The commercial Aitken nucleus counters agreed with each other within a factor of 2 , but at concnetrations above $10,000 \mathrm{~cm}^{-3}$ averaged $1 / 2$ of the values from the research Wilson cloud chamber, probably because of vapor depletion in the commercial instrument without sample dilution. (6) The Gardner expansion-type counter operating at low expansion ratios to simulate $1 \%$ supersaturation apparently subjects the sample to $30 \%$ supersaturation. In the case of some types of nuclei the error thus introduced into the Gardner counter is then compensated by an error of the order of 10 in the optical detection system because the fog droplets grow so slowly that they are smaller than the optics were designed for. The resultant data relating to fairly non-active nuclei tested at the workshop were in error by a factor of 17 too high. Data already in the literature using expansion-type counters to measure increases in what were purported to be CCN in pollution are unreliable and may be much as 17 times too high when the pollution contains many fairly non-active Aitken nuclei, as is frequently the case.

## INTRODUCTION

During the International Workshop on Condensation and Ice Nuclei (IWCIN), held at Fort Collins,

Colorado during August 1970, comparisons were made among six cloud condensation nucleus (CCN) instruments and among seven total aerosol or Aitken nucleus counters, in addition to the ice nucleus instruments reported in a separate summary. The Aitken nuclei were generally between two and a thousand times more numerous than the $C C N$, which are only those condensation nuclei which are activated at slight supersaturations characteristic of clouds (typically less than 1\%).

## THERMAL-DIFFUSION CHAMBER TYPE CCN INSTRUMENTS

The only type of CCN instrument found to be satisfactory at this workshop employed the thermal diffusion principle used by Langsdorf (1936) and Wieland (1956), with the present types patterned generally after Twomey (1963). Five of the six CCN instruments were of this design. In this type of instrument two horizontal wet surfaces are spaced 1 to 2 cm apart. The surface of the bottom plate is maintained approximately 3 to 5C colder than the top plate in order to produce the desired low supersaturations.

Diffusion of heat and water vapor from the warmer toward the colder surface produces linear temperature and vapor pressure gradients. Because the saturation vapor pressure vs temperature curve is concave upward the actual water vapor pressure exceeds the saturation vapor pressure (because of the linear gradients) between the two plates and continually produces a supersaturation which is a maximum near the midpoint between the plates. One water droplet forms on each of those nuclei present which are activated at the low supersaturation. Figure 1 shows examples of these droplets in a welldefined light beam across the center of the space between the plates. These droplets may be counted individually or the number may be indicated by detecting the integrated light scattered by the cloud of droplets. Four of the instruments at the IWCIN employed photographic recording of the individual droplets for subsequent counting and one of these, shown in Fig. 2, was additionally equipped with a video camera, monitor, and recorder to permit real-time observation of the peak concentration of droplets and to permit, with stop action, counting on the TV screen.

The fifth instrument employed a nephelometer incorporating a wide-angle light flash with photoelectric detection of the integrated light scattered from all droplets illuminated within the viewing path of the detector as described by Radke and Hobbs (1969). The time of flash was preset after laboratory experiments to determine the time to reach a pre-selected droplet size at each supersaturation.

Each of the two types of droplet detection has its advantages and limitations. Counting of droplet images has the advantage of being more nearly independent of droplet size and growth rate. But if the light level is marginal, there may be loss of detection of smaller droplets or lack of discrimination between images and film imperfections. Three of the instruments were equipped with a 200 -watt 0 sram highpressure Hg arc lamp and large lens system as shown across the upper left in Fig. 2. These were the only instruments capable of providing all data as absolute counts per known volume without recourse to previous calibrations. Even with this high level of illumination ( $-500 \mathrm{im} \mathrm{cm}^{-2}$, (Twomey, 1970)) detection of

## WATER DROPLETS FORMED AT 0.3\% SUPERSATURATION 1050 DROPS $/ \mathrm{cm}^{3}$



## WATER DROPLETS FORMED AT $0.9 \%$

 SUPERSATURATION 4200 DROPS $/ \mathrm{cm}^{3}$

## WATER DROPLETS FORMED AT 3.0\% SUPERSATURATION 5000 DROPS $/ \mathrm{cm}^{3}$



Fig. 1. Examples of droplets in CAL thermal-diffusion cloud chamber illuminated by high intensity light beam.
droplets down to $0.6-0.7 \mu$ is possible only when using an imaging sensitivity equivalent to about $1 / 40$ sec at $\mathrm{f}: 1.5$ with ASA 125 film . Camera data taken with this sensitivity by NRL indicated that $15-20 \%$ of the droplets had not been visible with the video system as used at the workshop. This instrument with the video readout and the other thermal diffusion CCN instruments including the integrating nephelometer type were generally in agreement within $\pm 30 \%$.

## INTEGRATING NEPHELOMETER TYPE AUTOMATIC CCN COUNTER

The integrating nephelometer type of detection has the advantage of automatic and real-time readout with its chief disadvantage being the requirement for a large sampling volume in order to provide sufficient light scattering intensity at low nucleus counts. The sample flow requires precautions in temperature conditioning of cold samples, such as aboard aircraft, to avoid excessive supersaturations when a cold sample is


Fig. 2. NRL CCN instrument; thermal diffusion chamber in front of camera (at operator's hand) video camera on opposite side of chamber, video monitor and recorder below; upper left, light source optics; four flat cylinders visible across top are chambers for temperature conditioning of the sample.
brought into contact with the warmer wet surface of the top plate of the diffusion chamber. This precaution is more difficult to obtain when the sample flow is large. This requirement of warming the incoming sample air is further accentuated when the upper plate is heated above room temperature (as opposed to cooling the lower plate to provide the necessary $\Delta t$ between plates).

A further concern with any automatic CCN instrument is the variable time to reach a given supersaturation when the sample air is at different initial relative humidities (Twomey and Davidson, 1970). It should be emphasized, however, that the workshop data discussed below indicate in general, remarkably good agreement among all of the thermal diffusion chamber type CCN instruments including this automatic one with its added convenience and potential for use by observers who are less specialized than is required for the present image-counting types. As with most new developments, problems with this automatic type of instrument are not yet all solved, as demonstrated by the unsuccessful attempt to get a commercial model into operation at the workshop.

## NCAR CONTINUOUS COUNTER FOR CONDENSATION NUCLEI

Only one instrument intended for $C C N$ measurement at the workshop did not use the thermal-diffusion cloud-chamber method of obtaining the small supersaturations necessary to simulate natural cloud conditions. This instrument is an experimental development model from NCAR using a continuous jet of warm moist air impinging into cold air. The resultant supersaturation maximum has been calculated by Langer to be approximately $2 \%$ assuming that the incoming air is at $90 \%$ relative humidity. The cloud droplets
formed on the nuclei are allowed to grow to 20 or 30 u before they are passed through an acoustic detector. In laboratory tests droplets were found to collect on the walls. To correct for this, the droplet count is multiplied by 5. In its present stage of development this instrument gave readings $1 / 3$ to $1 / 25$ of the thermal-diffusion chamber readings with NaCl , and 8 times too high for less easily activated nuclei such as Teflon. For natural nuclei it varied from 1/15 to 3.6 times the thermal-diffusion averages. From these data it appears that the maximum supersaturation is 5 to 10 times higher than calculated. Wall or other losses are 3 to 8 times the adjustment of 5 times the count, as now being used. These data also suggest that measurements in plumes from pollution sources where the nuclei may be fairly inactive as compared to natural nuclei could be systematically biased by factors of 5 to 8 too high in the plumes. On the other hand, this instrument has the advantages of simplicity for unattended operation and presents an automatic readout in real time. It is hoped that further development can make it a useful CCN instrument, that is, useable in the 0.3 to $1 \%$ supersaturation range, as opposed to the present probable 10 to 20\% supersaturation.

## COMPARISON OF THERMAL-DIFFUSION CHAMBER CCN DATA

In general, the agreement among cloud nucleus measurements at the IWCIN in natural aerosols was excellent. Measurements made with each of the thermal-diffusion instruments were usually found to lie within $\pm 30 \%$ of the mean. Where significant differences did occur they cquid usually be ascribed, in part, to the different techniques of particle detection employed by each instrument. However, agreement among the instruments was less satisfactory for artificial aerosols such as NaCl , AgI and Teflon than for natural nuclei. Even with the artificial nuclei the data were normally less than a factor of two from the mean.

In Fig. 3, nucleus concentration for a typical natural aerosol experiment are shown for the three specific supersaturations employed at the workshop. The agreement among the data for the five thermaldiffusion cloud chambers compared in this example is quite good. In this experiment note that the NCAR continuous jet mixing type read about one-third of the diffusion chamber average extrapolated to $2 \%$, which was the theoretical supersaturation of the NCAR instrument. The deviations of the various diffusion cloud chambers were generally greatest at low supersaturations. The spread in the data is about a factor of two at $0.3 \%$ supersaturation, probably caused mainly by the different procedures used for detecting droplets in the various thermal-diffusion chambers. For example, some instruments photograph droplets at shorter exposures or smaller lens apertures, resulting in a lower count particularly at low supersaturation where the droplets are small and less uniform in size. Where the droplets are larger and also more uniform, differences in methods of droplet detection are less important and counts are generally in close agreement.

## MINIMUM NUCLEUS SIZE FOR ACTIVATION

The ease of activation of CCN is dependent on several factors, including solubility, hygroscopicity, size, and for insoluble materials the wettability or contact angle of a water drop on the material. For nuclei of uniform chemical composition, their activity is a function of nucleus size. Natural nuclei contain


Fig. 3. Spectrum of CCN activated at three supersaturations - average of all thermaldiffusion chambers operated at IWCIN. The NCAR continuous instrument reading is not included in average.
some nuclei which are easily activated, some nonwettable, and some which are mixtures of various materials, a small but varying fraction of which are soluble. For the natural nuclei some small soluble nuclei can be more active than larger nuclei which may contain a small soluble particle surrounded by inactive particles which have become attached by natural coalescent processes as discussed by Twomey and Severynse (1964).

At the workshop several experiments were run using artificially generated nuclei of nominally uniform composition. In these cases the minimum size of nuclei which could be activated at each supersaturation used in the CCN instruments could be determined by finding on the Whitby size distribution curves the particle size corresponding to the number of CCN counted. Results of these comparisons are presented in Fig. 4. Slight adjustment of these curves will be needed if the Whitby size distributions are later corrected for changes in the aerosol sizes during the time of each experiment. Even with the limitations on accuracy the four top curves (using averages of the five diffusion-chamber instruments) provide a comparison of the sizes activated using artificial nuclei of Teflon, AgI pyrotechnic, and NaCl . The apparent diameter of the natural nuclei as shown is probably too large because of the likelihood that the Whitby size distribution included large particles which were inactive as nuclei. Therefore the CCN counts from the diffusion chamber must include some nuclei which were smaller than was indicated by the simple method of comparison with the size distributions. The bottom curve is included to present
data on the smallest sizes indicated by any of the instruments. On the basis of this curve at $0.3 \%$ and $1 \%$ supersaturation the smallest average particle diameter of NaCl that was activated was $0.07 \mu$ and


Fig. 4. Apparent sizes of $C C N$ as determined from Whitby size distributions of the aerosol samples measured.
$0.04 \mu$, respectively. These values are about twice the sizes that are predicted from theory and found in other experiments (Twomey, 1965).

This workshop provided probably the first comparative data on the ease of activation of AgI produced by LW-83 pyrotechnic for functioning as $C C N$ in conjunction with its use as ice nuclei. This aerosol was found to contain large numbers of very active CCN, possibly from some contaminants other than the pure AgI; but in any case, it is important in weather modification to know that this type of pyrotechnic device was found to produce many particles that were nearly as active in producing cloud droplets as NaCl .

The top curve in Fig. 4 shows the relative activity of a less wettable aerosol, Teflon. This powdered material had a size range of about 0.01 to $2 \mu$ diameter when it was mechanically aerosolized and injected into the IWCIN sampling system.

Since there has been some evidence that insoluble, nonwettable particles can behave as cloud nuclei (Jiusto and Kocmond, 1968) it was thought that Teflon would provide a test of this evidence. The results were not conclusive in this regard, but were of great value for identifying the causes of errors in using other than diffusion chambers in attempting to measure CCN.

Although the bulk Teflon material when pressed to form a plate with a flat surface had a contact angle in excess of $150^{\circ}$, the nuclei that were intro-
duced into the diffusion chambers were found to produce some cloud droplets at supersaturations as low as $1.0 \%$. According to the Volmer theory of nucleation these particles should not be active until supersaturation in excess of $200 \%$ are reached. The unexpected ease of activation is probably due to unknown characteristics of the surface chemistry of these presumedly hydrophobic particles. (The manufacturer did not disclose whether detergents were used in the grinding process). Additional tests should be conducted with other nonwettable materials with better known surface characteristics to either support or refute these results.

## AITKEN NUCLEUS MEASUREMENTS

In addition to the Whitby particle size analyzers six instruments provided data on the total number of aerosol particles or Aitken nucleus counts of the same sample air being measured by the CCN and ice nucleus instruments. The Aitken nucleus instruments were built by commercial suppliers with the exception of one from the University of Missouri at Rolla (UMR) described by Kassner et al. (1968). The general operation of all of the Aitken counters is by an adiabatic expansion of saturated air with the resultant cooling producing a supersaturation of several hundred percent. The UMR Wilson cloud chamber instrument is a research tool designed to minimize most of the errors of the commercial instruments by
(1) producing accurately known supersaturations,
(2) determining accurately the number of droplets formed (hence the number of nuclei), and (3) providing the proper amount of sample dilution to keep the number of droplets in the counting field large enough to provide a statistically valid sample, yet small enough to control vapor depletion caused by condensation on large numbers of droplets.

Commercial instruments provided electrical readout from photoelectric detection of light extinction or scattering from the cloud formed after expansion. All of the commercial instruments agreed quite closely with each other, but all apparently suffered from vapor depletion at the higher concentrations. At concentrations above $10^{4} \mathrm{~cm}^{-3}$ the UMR Wilson chamber often showed counts 2 to 3 times higher than the commercial instruments.

## gardner counter at low expansion ratio

The workshop also provided probably the best opportunity to date to study the readings of a Gardner counter operated at a low expansion ratio and compare these condensation nucleus readings to CCN counts from diffusion chambers. For this study a Gardner counter was operated at a vacuum of $1 \frac{1}{2}$ inches mercury, the pressure difference which had been found empirically with natural nuclei to give meter indications most nearly equal to the counts of CCN as determined from diffusion chambers operating at $1 \%$ supersaturation. Figure 5 from Austin Hogan's IWCIN data shows that most natural nucleus readings fell within factors of $1 / 2$ to 2 times the CCN count, only one case being a factor 6 too high with natural nuclei.

Calculations indicate that the supersaturation at $1 \frac{1}{2}$ inches vacuum should be about $30 \%$ at the altitude of Fort Collins. One explanation advanced for the meter readings at this supersaturation being as low as the CCN counts at $1 \%$ is that the droplets are smaller (growing more slowly) at $30 \%$ supersaturation


Fig. 5. Counts of CCN from diffusion chamber at $1 \%$ supersaturation compared to Aitken nucleus readings from Gardner counter operating at $1.5^{\prime \prime} \mathrm{Hg}$. vacuum.
than they would be at the normal 250 to $300 \%$ supersaturation for which the optical detector is calibrated, hence a lower reading than should have occurred with the number of droplets being actually much higher than the CCN count. If this explanation is correct, then condensation nuclei which are less easily activated than most natural CCN should be activated in the Gardner counter at $30 \%$ but not by the diffusion chamber at $1 \%$ supersaturation. In this case the Gardner counter should read too high. At the IWCIN, Teflon nuclei provided data to test for this effect. The result as shown in Fig. 5 was a Gardner counter reading of $1000 \mathrm{~cm}^{-3}$ as compared to 60 in the diffusion chamber.

Further, if the explanation above is correct, the Gardner counter readings should be too low when using NaCl nuclei which are more easily activated than the average of the natural nuclei used in the empirical adjustment of the expansion ratio to give a reading equal to the number of CCN at $1 \%$ supersaturation as measured by a diffusion chamber. However, in this case some compensation in the Gardner optics would be achieved by the faster growth rate of the cloud droplets. This faster growth is often observed in the thermal diffusion chambers when counting NaCl nuclei. The one data entry for NaCl shown in Fig. 5 indicates a reading $1 / 3$ that of the diffusion chamber CCN count at $1 \%$ supersaturation.

If these results are accurate, they indicate that when pollution aerosols are less hygroscopic or
less wettable than natural $C C N$, the Gardner counter at a low expansion ratio could be systematically biased by factors of up to 17 too high when measuring supposedly CCN in the pollution as compared to nearby unpolluted areas.

## CONCLUSIONS

(1) Five thermal-diffusion chamber type of CCN counters provided quite consistent agreement with each other both with natural and artificial nuclei ranging from easily activated nuclei ( NaCl ) to nuclei with less than normal ease of activation (Teflon).
(2) The principal source of errors in the diffusion chamber readings found at the IWCIN was due to insufficient illumination levels and/or insufficient sensitivity in droplet imaging.
(3) An automatic thermal-diffusion chamber type of CCN instrument using an integrating nephelometer consistently produced CCN data in good agreement with the image-counting types of manual instruments.
(4) The NCAR continuous mixing-jet condensationnucleus instrument produced unacceptably large errors, on the low side with soluble and the high side with less easily activated nuclei. These results indicate the need for further development to reduce the supersaturation and increase the detection efficiency in this instrument.
(5) All of the commercial counters of Aitken or total condensation nuclei agreed well with each other under the variety of conditions available at IWCIN. All of these instruments read low by a factor of two as compared to a research type Wilson cloud chamber when vapor depletion was appreciable (with high nucleus counts) in the commercial instruments.
(6) Gardner counters operating at a low expansion ratio and thus purportedly measuring CCN may read up to 17 times too high for nuclei which are less easily activated than natural nuclei. Gardner Aitken nucleus counters at a low expansion ratio should not be considered to give CCN measurements. Particularly susceptible to misleading conclusions are data of this type from pollution where the condensation nuclei may be less easily activated than the average of natural CCN.

## ACKNOWLEDGMENTS

Much of the data analysis for this IWCIN summary was accomplished by Dr. Larry Radke. The productivity of the workshop was greatly enhanced by the leadership and hard work of Drs. Josef Podzimek, Lewis Grant, Ulrich Katz and his staff, and Roger Steele, chairman of the Organizing Committee. Special mention is made of Prof. Ken Whitby and Rudolf Husar for their imaginative techniques of producing nuclei for testing at the workshop.

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## REPORT ON THE ICE NUCLEUS WORKSHOP FORT COLLINS, AUGUST 1970

## by

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## SUMMARY

Estimates of the concentration of natural ice nuclei in 17 experiments, of active nuclei in 7 thermally generated silver iodide experiments and in a variety of other artificial aerosols made during the Workshop are considered in this report. The main emphasis is on a comparison of the reproducibility of measurements and the proportion of nuclei detected with different techniques.

Results from any one experiment were usually insufficient to draw useful conclusions on either point so that it was necessary to combine the experiments in groups. For the thermally generated silver iodide experiments, theoretical curves of the concentration of nuclei expected as a function of temperature were constructed from the known size distributions and Fletcher's (1969) theory of nucleation. This allowed measurements made at different temperatures to be included in the analysis. It was found that several systems had quite good reproducibility in their measurements and these were then chosen as reference methods for studying the natural aerosol results. A fortunate circumstance was that according to these techniques, the concentration of ice nuclei was relatively constant throughout all experiments. This again allowed many results to be combined to give statistically useful results.

A close control over humidity seems to be the essential feature for reliability and this was much more critical for artificial than for natural nuclei. No instruments using high supersaturations can be considered to give quantitative estimates of nucleation in natural clouds without first having been calibrated for the appropriate aerosol against one of the humidity-controlled systems.

The measuring systems which performed best were:
(a) For all nuclei
(i) The Goetz aerosol spectrometer used by Gerber.
(ii) The settling-cloud chamber used by Ohtake.
(iii) Membrane filters, using the method demonstrated by Gagin.
(b) For high nucleus concentrations only
(iv) C.S.U. $1 \mathrm{~m}^{3}$ isothermal chamber, used by Carlson
(v) The miniature sedimentation chamber used by Davis.

There is some doubt about the behavior of the C.S.U. chamber used with mixed artificial aerosols.

## AIM OF THE REPORT

Concentrations of natural ice nuclei as a function of temperature are required as background information for studying the initiation of the ice phase in supercooled clouds, while concentrations of ice nuclei introduced during seeding are also required in order to estimate their effect on the formation of precipitation in clouds. In either case an essential feature of the measurement is that conditions which are important to the activation of the relevant nuclei in clouds should be maintained also in the measuring system. Though simple in principle, this is extremely difficult to achieve in practice and all our instruments for measuring concentrations of nuclei represent some compromise between convenience and accuracy. Ideally we require a system in which there are only slight errors from experimental causes and in which the apparent concentrations are a fixed proportion (preferably 100\%) of the number of nuclei which would be active in a natural cloud of a specified type in the absence of multiplication mechanisms. Unfortunately we have no standard method to tell us what the true number of nuclei is and few instruments which have been checked for reproducibility of measurements with aerosols of different types.

The most important function of this Workshop has been to provide information on relative concentrations measured by the various instruments using the same aerosol and environmental conditions and to compare the reproducibility of measurements from each.

A glance at the results of any experiment such as the one shown in Fig. 2 is sufficient to show that this information is essential, for the scatter in individual estimates is appalling. Values were usually grouped in a 100:1 range but extremes were as great as $10,000: 1$. While high accuracy is not
required for many purposes, it certainly needs to be better than this! It is suggested that to be considered reliable an instrument working under these conditions should record half or more of measurements within a $3: 1$ range and at least three-quarters in a 10:1 range. It is the aim here to find which systems can achieve even these generous limits of reproducibility.

## THERMALLY-PRODUCED SILVER IODIDE AEROSOLS

For the natural aerosols used, we can have no idea what concentration a perfect instrument would have measured. It is therefore better to consider first the seven experiments numbered $38-44$ using thermally-produced silver iodide aerosols for which Whitby and Husar provided size distribution curves. Fletcher's (1969) theory of the active fraction of silver iodide particles, which incorporates the idea of a distribution of areas of active sites on the particles, can then be used to produce an estimate of the number of nuclei to be expected as a function of temperature. The estimate may be wrong, for it relies on Fletcher's empirically determined parameter " $\gamma$ " (which measures the width of the distribution of areas of active sites on the particles). However, it forms an objective basis for comparison for all. systems used at any temperature in any of the 7 experiments, allowing statistically meaningful numbers of results to be combined.

Figure 1 shows the expected concentrations as a function of temperature for each of the experiments numbered $38-44$. A " $\gamma$ " 0.8 was assumed. For the small sedimentation chamber used by Davis the curves had to be recalculated, for particles smaller than about $0.1 \mu$ diameter were not captured in it. Gerber's experiments with the aerosol spectrometer, which are reported separately, showed that the size of particles detected decreased as the humidity increases. Strictly, we should calculate the curves individually for each method according to the working humidity, but objectivity would thereby be lost. Since the value of " $\gamma$ " chosen is appropriate to mixing chamber results (which have a wide range of supersaturations relative to water), it therefore discriminates against the low supersaturation devices. The aim is to show that in spite of this there are some methods working at low supersaturations which are still amongst the most satisfactory performers.

Experience with sodium chloride aerosols and the more reliable cloud nucleus counters in the preceding part of the Workshop showed that the concentration of aerosols decreased during the experiment to as low as $50 \%$ of its initial value. This has not been corrected for and adds to the apparent scatter but it is plain from the good performance of some systems that it does not greatly influence the result.

Examination of the results of individual experiments in this series such as number 38 illustrated in Fig. 2 for $-16^{\circ} \mathrm{C}$ working temperature showed much greater decreases with time recorded by some instruments. A key to the instrument numbers used in this diagram will be found in Table 1 and in the description at the end of this report. Since those operating under more cloud-like conditions did not show more than the expected decrease, it suggests that the inability to detect the aging aerosol is an inherent


Fig. 1. Expected concentration of ice nuclei as a function of temperature for experiments $38-44$ using known size distribution and Fletcher's (1969) theory with $\gamma=0.8$.
defect of these systems and should not be corrected for in producing an estimate of reliability.

A11 recorded concentrations measured at a particular working temperature were translated into ratios of the observed to expected active fraction of silver iodide particles using Fig. 1, or the special calculations for the mineature sedimentation chamber. In Table 1 the methods of measurement have been placed in order according to the degree of consistency in this ratio.

Since changes in aerosol concentrations with time were not corrected for, the first six entries (five methods) listed can be considered relatively satisfactory. Of these, Gagin's filter method and Ohtake's cloud chamber covered the widest range of temperature and times and contained by far the most measurements; they should therefore be considered relatively better than the results indicate.

The percentage of particles detected is also important. Logarithmic means of the percentage ratio of observed to expected particle concentrations are shown in Table 2.

Note that all the methods giving consistent results are also amongst those detecting the highest percentage of particles, with the exception of Gerber's measurements at water saturation. Presumably if he had measured instead at $101 \%$ R.H. his results would have been comparable.


Fig. 2. Apparent decrease in concentration recorded in the course of experiment 38 at $-16^{\circ} \mathrm{C}$. A key to the instruments represented by the numbers will be found in Table 1.

There is a marked difference between the estimates of the deposition methods used at water saturation and Ohtake's chamber, or the CSU chamber. Capture of nuclei by water drops could cause the difference, but since this also occurs due to the growth of droplets of condensed water on foils or filters when water saturation is exceeded a more likely explanation is that the transient supersaturations created during injection of the sample into the cloud chambers influences the results.

Table 2 illustrates some other interesting points. (i) There is a $20: 1$ ratio in the mean estimates of the two apparently identical rapid expansion Bigg-Warner type cold chambers, the NCAR model used by Carnuth and the CSU model used by Reinking and others. This was caused by the much faster apparent decay of activity of the aerosol in the CSU model. The implication of this is that the estimate of concentration given by rapid expansion chambers is not necessarily related at all to concentrations measured at water saturation for this type of aerosol. In fact, this type of aerosol is not used in practical applications and there is no evidence of a similar phenomenon with the common cloud seeding materials. However, it is an unexpected aspect of this method's behavior which clearly should be investigated.
(ii) There is an 8:1 ratio between mean concentrations estimated by the two acoustic counters of Langer and Schnell for which sufficient measure-

TABLE 1 Maximum Percentage of Occasions on Which the Ratio of the Observed to Expected Active Fraction of Silver Iodide Particles Fell Within Any 3:1 or Any 10:1 Range

| Key to Nos. in Fig. 2 | Measuring System | Observer | 3:1 | 10:1 |
| :---: | :---: | :---: | :---: | :---: |
| 1 | C.S.U. isothermal chamber | Carlson | 83* | 100* |
| 3 | Goetz aerosol spectrometer (100\% R.H.) | Gerber | 75* | 100* |
| 5 | Membrane filter | Gagin | 71 | 92 |
| 4 | Sedimentation chamber | Davis | 70 | 90 |
| 3 | Goetz aerosol spectrometer (high supersaturation) | Gerber | 56* | 77* |
| 2 | Settling-cloud chamber | Ohtake | 48 | 72 |
| 7 | Membrane filter (100\% R.H.) | Bigg | 43 | 70 |
| 6 | Membrane filter | Alkezweeny | 37 | 70 |
| 16 | M.R.I. continuous counter | Dunsmore | 37 | 65 |
| 7 | Membrane filter (110\% R.H.) | Bigg | 33 | 60 |
| 8 | Membrane filter | Vali | 31 | 62 |
| 9 | Membrane filter (droplet method) | Langer | 29 | 58 |
| 13 | NCAR long-cone acoustic counter | Langer | 29 | 58 |
| 10 | Rapid expansion counter (B-W type) | Carnuth | 26 | 50 |
| 11 | Rapid expansion counter ( $B-W$ type) | Reinking | 26 | 38 |
| 12 | Rapid expansion counter (CSU type) | Vardiman | 23 | 36 |
| 14 | NCAR acoustic counter | Schnell | 22 | 43 |
| 15 | NCAR acoustic counter | Scott: Insufficient results |  |  |
| 18 | Droplet freezing | Robertson: Not determined |  |  |

[^4]ments were available. Although the CSU model operated by Schnell detected a much higher percentage of particles, the greater reliability of Langer's long cone model would make it a preferable instrument. Far more measurements were recorded by these two automatic instruments than by any other system.

The drop freezing method used by Robertson which was under development during the Workshop was not really perfected and gave apparent concentrations quite unrelated to those of the nuclei. However, a demonstration by Vali and Robertson of the number of drops frozen as a function of temperature using water containing nuclei shows that the method is capable of useful and reproducible results. The difficulty is apparently simply one of getting the aerosol into the water to be frozen without losing it or dissolving it.

## OTHER SILVER IODIDE AEROSOLS

Production of ice nuclei by pyrotechnics or solutions containing sodium or ammonium iodides have much more practical relevance than those just considered. Unfortunately, the aerosols contain an unknown proportion of particles other than those of silver iodide so that we cannot legitimately use the aerosol distribution to estimate the number of active nuclei. If the obviously incorrect assumption is made that all the particles were silver iodide the
results from different experiments at different temperatures can be combined. It would be out of the question to use these for an estimate of reliability but it is instructive to use it to compare relative percentages of the nuclei detected.

Table 3 shows the logarithmic means of percentage ratios of observed to expected nuclei assuming that all the particles were silver iodide, for experiments $50-53$. These included one pyrotechnic device, two experiments using the burning of a silver iodide, sodium iodide, acetone solution, and one using ammonium iodide in place of the sodium iodide.

The ratios observed are in much the same order as before for the deposition methods (Goetz aerosol spectrometer, sedimentation chambers and filters), Ohtake's settling cloud chamber and the acoustic and MRI counters and are about one-fifth to one-tenth of those shown in Table 2, which we might have expected from the assumption that all of the particles were silver iodide.

The expansion chambers all record as many, or more nuclei than before, which also might have been expected since the curious apparent decay phenomenon was absent. The reproducibility of their results was comparable with those from the best methods. The really surprising and quite unbelievable result is that of the CSU chamber which recorded an actual

TABLE 2 The Detected Percentage of Particles Estimated from Fig. 1 to be Active Ice Nuclei

| Method | Observer | Percentage |
| :--- | :--- | :---: |
| (a) Deliberately high supersaturations |  |  |
| Goetz aerosol spectrometer | Gerber | 150 |
| Sedimentation chamber | Davis | 135 |
| Membrane filters | Bigg | 30 |
| (b) Normal operation |  | 59 |
| Settling cloud chamber | Ohtake | 36 |
| CSU isothermal chamber | Carlson | 33 |
| NCAR acoustic counter | Schnell | 21 |
| Rapid expansion (B-W type) | Carnuth | 13 |
| Membrane filters (101\% R.H.) | Gagin | 12 |
| Membrane filters (droplet method) | Langer | 8 |
| Membrane filters (101\% R.H.) | Alkezweeny | 6 |
| Membrane filters (100\% R.H.) | Bigg | 6 |
| Goetz aerosol spectrometer (100\% R.H.) | Gerber | 4 |
| NCAR long cone acoustic | Langer | 3 |
| Membrane filters (101\% R.H.) | Vali | 1 |
| Rapid expansion (B-W type) | Reinking | 1 |
| Rapid expansion (CSU type) | Vardiman | 1 |
| MRI continuous | Dunsmore | <<l |
| Droplet freezing | Robertson |  |
| NCAR acoustic | Scott: Insufficient results |  |
| University of Denver continuous mixing | Evans: No results |  |

TABLE 3

| Method | Observer | Percentage |
| :--- | :--- | :---: |
| CSU isothermal chamber <br> University of Denver, continuous <br> mixing chamber <br> Sedimentation chamber <br> Goetz aerosol spectrometer <br> (high supersaturation) | Carlson | 45 |
| Rapid expansion chamber (CSU type) | Evans | 34 |
| Rapid expansion chamber (B-W type) | Davis | 31 |
| Rapid expansion chamber (B-W type) | Gerber | 25 |
| Settling cloud chamber | Vardiman | 22 |
| NCAR acoustic | Reinking | 18 |
| Membrane filters | Ohtake | 9 |
| Membrane filters | Schnell | 6 |
| Membrane filters (droplet method) | Alkezweeny | 6 |
| Goetz aerosol spectrometer (100\%) | Langer | 2 |
| All other methods | Gerber | 2 |

increase in the percentage detected. As sugges ced before, the effects of supersaturation during sample injection require very close scrutiny.

## NATURAL NUCLEI

If the mean concentrations of ice nuclei recorded at, say, $-20^{\circ} \mathrm{C}$ are shown for each experiment in a time sequence as in Fig. 3, some instruments show changes as large as $100: 1$ while others indicate that no changes larger than $4: 1$ occurred. Remembering Table 1, in which only five methods appeared to be capable of recording more than half their measurements in a $3: 1$ range on an aerosol changing in concentration by less than $50 \%$, this is not surprising. Of those five instruments, the CSU chamber and Davis' sedimentation chamber were not used in the natural aerosol experiments. We are therefore left with Gagin's filter method, Gerber's aerosol spectrometer and Ohtake's settling cloud chamber. Because the aerosol spectrometer was operated at high supersaturations in all but two experiments it may have responded quite differently to nuclei present on different days and cannot be used as a standard without further investigation. The other two methods each showed a total range of variation of less than $4: 1$ at $-20^{\circ} \mathrm{C}$.

It is therefore a reasonable procedure in order to provide statistically useful comparisons of reproducibility and performance to suppose that the ice nucleus population was constant throughout all experiments and to combine all the results.

In order to do this for different temperatures, Gagin's and Ohtake's results were separately plotted and logarithmic mean concentrations deduced as a function of temperature. It is very important to note that for each the concentration varied with


Fig. 3. Mean natural ice nucleus concentrations recorded at $-20^{\circ} \mathrm{C}$ during the Workshop. A key to the instruments represented by the numbers will be found in Table 1.
temperature in exactly the same way. Together with the mean results for most of the other methods these, and the adopted standard of reference (taken as the logarithmic mean between Gagin's and Ohtake's mean results) are shown in Fig. 4. The slope of this reference line is a reasonable compromise between those derived for all the methods used and is in good agreement with a large body of previous experience. From this reference line, extrapolated where necessary, the ratio of observed to reference nuclei was calcualted for each measurement. Measurements at $-16^{\circ} \mathrm{C}$ which were below the threshold of detection were combined as a total count in a total volume for all experiments. Table 4 shows the maximum percentage of occasions on which the ratio of observed to reference nuclei occurred within any $3: 1$ or any $10: 1$ range.

This time two expansion chambers, the NCAR longcone acoustic counter, two additional filter methods, and the droplet freezing technique, as well as the reference methods, gave acceptable results.

The logarithmic means of the ratios of observed to reference concentrations are expressed as percentages in Table 5.

Once again Gerber's results show the importance of avoiding high supersaturations if realistic concentrations are to be obtained, but the spread in values is not as great as for silver iodide.

It should be emphasied that for the acoustic counters the correction factor allowing for detector losses has been omitted. If the usual factors had been included two of these counters would have been near the middle of the group. The rather large


Fig. 4. Mean (logarithmic) dependence of concentration on supercooling for various instruments. A key to the numbers is given in Table 1.
spread of values given by Schnell's counter appeared to be due to two separate modes of operation one giving high and one low concentrations. The reason for this was not at all obvious but has been observed with other instruments of this type. Scott's gave consistently very low concentrations but comparable with those reported in the literature by Nagamoto et al. (1967) and Hobbs and Locatelli (1970). The above comparisons show clearly that such concentrations are instrumental in origin. Until their cause is discovered it should not be assumed that this type of counter gives results which relate to the effective nuclei present.

Langer's acoustic counter on the other hand gave a high degree of reproducibility and a slope of the $\log$ concentration-supercooling line which agreed exactly with the reference line's. It evidently represents a considerable improvement over the commercially produced models for counting natural nuclei.

DISCUSSION OF THE PERFORMANCES OF THE INDIVIDUAL TECHNIQUES USED

## 1. CSU large isothermal chamber

While this instrument appeared to be one of the most successful for the thermal silver iodide experiments, its use at only one temperature and at one time in each experiment biased the results heavily in its favor. The inability to take readings quickly, or at different temperatures, or for low concentrations of nuclei and its lack of portability make it of limited use. Its behavior with mixed silver iodide aerosols was surprising, the results being improbable and disagreeing with those made by other methods. The transient supersaturations occurring during sample injection are suggested as possible sources of trouble.

## 2. Ohtake's settling cloud chamber

This simple instrument had a remarkably consistent performance in all stages of the Workshop and is one of the few which deserved a strong recommendation as a standard method. Its most appealing feature is that conditions in a natural cloud are quite closely modelled. Fallout and transient supersaturations during sample injection appear to be possible defects, although its behavior was much more what would be expected than that of the CSU chamber with mixed aerosols.

## 3. The Goetz aerosol spectrometer

This was perhaps the most interesting technique demonstrated at the Workshop. The comparison of apparent concentrations at low and high supersaturations relative to water has greatly helped in understanding the reasons for the alarming discrepancies between different systems and its unique ability to distinguish the size of the active particles is an enormous asset. Used in conjunction with a range of supersaturations and temperatures it will yield much valuable information on ice nucleation. In its present form it is more valuable as a research tool than as a field instrument but its consistently reproducible results recommend it as another standard method.

TABLE 4 Estimates of Reproducibility in Experiments With Natural Nuclei

| Method | Observer | $3: 1$ | 10:1 |
| :---: | :---: | :---: | :---: |
| Settling cloud chamber | Ohtake | 82 | 100 |
| Membrane filters | Gagin | 72 | 100 |
| NCAR long-cone acoustic | Langer | 66 | 91 |
| Membrane filters | Alkazweeny | $63^{*}$ | 96* |
| Goetz aerosol spectrometer | Gerber | 63 | 93 |
| Rapid expansion counter (B-W type) | Reinking | 56 | 86 |
| Membrane filters | Bigg | 54 | 94 |
| Rapid expansion counter ( $\mathrm{B}-\mathrm{W}$ type) | Carnuth | 52 | 74 |
| Droplet freezing | Robertson | 48 | $78$ |
| Membrane filters | Vali | 44 | 69 |
| Rapid expansion (CSU type) | Vardiman | 38 | 73 |
| NCAR acoustic | Scott | 38 | 54 |
| NCAR acoustic | Schnell | 33 | 55 |
| MRI continuous counter | Dunsmore | 32 | 56 |
| Membrane filters | Langer | 29 | 55 |

*At $-20^{\circ} \mathrm{C}$ only

TABLE 5 Percentages of the Reference Nuclei Detected

| Method | Observer | Percentage |
| :--- | :--- | :---: |
| Goetz aerosol spectrometer <br> (high supersaturation) | Gerber | 300 |
| Membrane filters | Gagin | 146 |
| Goetz aerosol spectrometer (100\% R.H.) | Gerber | 117 |
| Settling-cloud chamber | Ohtake | 85 |
| Rapid expansion counter (B-W type) | Reinking | 70 |
| Rapid expansion counter (CSU type) | Vardiman | 62 |
| Membrane filter | Bigg | 59 |
| Membrane filter | Vali | 58 |
| Membrane filter | Alkazweeny | 49 |
| MRI continuous counter | Dunsmore | 32 |
| Membrane filters | Langer | 19 |
| Rapid expansion counter (B-W type) | Carnuth | 18 |
| NCAR long-cone acoustic | Langer | 14 |
| NCAR acoustic | Schnell | 13 |
| Droplet freezing | Robertson | 6 |
| NCAR acoustic | Scott | 2 |

4. Miniature sedimentation chamber

This novel device is simple in principle and obviously effective in practice. In these experiments it was run at supersaturations relative to water greater than the $1 \%$ which is regarded as a maximum in natural clouds and it hence overcounted the effective nuclei, but this is not an inherent defect of the system. Used at the appropriate humidities and allowing a longer settling time in order to catch all the nuclei effective at the temperature of interest it could be used as a standard method.

## 5. Gagin's filter method

The easily controlled humidities in Gagin's elegant ice nucleus detector combined with its remarkably good performance in all experiments are strong recommendations for its adoption as a standard method. It does not seem by comparison with the other deposition methods or Ohtake's chamber that an appreciable proportion of the nuclei are lost through being buried in the filter. The simplicity of the filter method in field use, and the ability to detect low concentrations of nuclei are considerable advantages.

6-9. Filter methods other than Gagin's
Three of the four remaining filter developing systems (used by Alkazweeny (6), Bigg (7), and Vali (8)) were similar in principle to Gagin's but gave generally lower and more erratic concentrations. They performed much better with natural nuclei than with silver iodide, showing that the difference was due to a less precise control over humidity, which is much more critical with silver iodide. In the fourth method, due to Langer (9), a stream of humidified air was passed over the filters to activate the nuclei. The very erratic results showed that this was even less precise in achieving the required humidity. Used in the "droplet method" for the silver iodide aerosols a supersaturation was probably often caused at the filter surface leading to higher counts but again without sufficient control for good reproducibility.

All these methods are potentially good but clearly need increased attention to precision in regulating the humidity.

## 10-12. Rapid expansion counters

The ease with which temperature spectra can be obtained, their simplicity and portability are great advantages. Their performance with natural nuclei appeared to be good, but the extraordinarily poor performance of two of the three with thermally generated silver iodide is a very disturbing feature of their operation. The possibility needs examination that they achieve a realistic count by a fine balance between gains due to high supersaturations and losses due to short sensitivity times. Then, if the balance were not achieved for some nucleus types, results like those for thermal silver iodide might be obtained. Throughout, the Bigg-Warner type of counter from NCAR operated by Carnuth (10) behaved with reasonable consistency, the two CSU models (11, the B-W type operated by Reinking and 12, the CSU model operated by Vardiman) being most affected by the troubles referred to.

Summarizing, these instruments will remain useful for field operations, but possibly giving unreliable results in some circumstances. The technique cannot be recommended for calibrating other instruments.

## 13. NCAR long-cone acoustic counter

The remarkably good performance of this counter with natural nuclei and its fair results with artificial nuclei show it to be a great improvement on the earlier commercially produced acoustic counters. The ability to obtain a printout of the total count at chosen intervals and to operate continuously are enormous assets and make it extremely useful for many operations. In principle, the high transient supersaturations which must occur during the cooling of the sample will always be a difficulty in quantitative studies.

## 14, 15. Other NCAR acoustic counters

The apparent operation in two counting modes by Schnell's counter (14) and the very low counts of Scott's (15) make it seem doubtful whether measured concentrations bore any worthwhile relation to actual nucleus concentrations. The performance of Schnell's counter with silver iodide aerosols was considerably better and its measured concentrations with the complex aerosols surprisingly high. As in all these instruments there are great advantages for qualitative work but their limitations must be clearly unders tood.

## 16. Met. Research Inc. continuous counter

Instrumental difficulties during the experiments may have been partly responsible for the poor reproducibility of results from Dunsmore's equipment. The system, however, used high and uncontrolled supersaturations and could only be considered useful for crude qualitative work.
17. University of Denver continuous mixing chamber

Unfortunately this counter did not become fully operational until the last days of the Workshop when its performance appeared to be reasonably satisfactory. Like the acoustic counter its main use should be for tracking plumes of silver iodide and not for quantitative work, because of the wide range of supersaturations and temperatures within it.

## 18. Droplet freezing technique

The droplet freezing method used with natural aerosols appeared to give reasonably reproducible answers but detected less than 10\% of the nuclei. The situation was far worse with silver iodide and evidently many of the nuclei dissolved during the washing process. It is an interesting method with obvious drawbacks.

Robertson has submitted revised estimates of ice nucleus concentrations after subtracting the "background concentration" and using a correction factor to allow for the nuclei retained by the filter. These procedures have substantially increased the effective concentrations. They have been omitted in this treatment because the policy laid down by the
organizers was to use only data submitted during the Workshop and to ignore correction factors, whose value can be assessed by comparison with results from other techniques.

## CONCLUSIONS

The great achievement of this Workshop has been to provide some reasonably clear guidance on which techniques have the best performance and why. This should be a great help in designing and calibrating future instruments since there are several available systems sufficiently reliable to give quantitative answers to the order of accuracy we are usually concerned with.

The results suggest that we should look very critically at the design principles of all instruments in use and in particular to the degree of percision with which they model cloud conditions. There
is simply no virtue or sense, for example, in obtaining the highest possible nucleus count from silver iodide generators if it is achieved by using supersaturations which are never found in the atmosphere.

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List of Participants of the 2nd IWCIN, Fort Collins - 1970

| Name | $\begin{gathered} \text { Identific } \\ \text { No. } \end{gathered}$ | ion Affiliation | Address | Apparatus | Outlet \# on duct |
| :---: | :---: | :---: | :---: | :---: | :---: |
| A. Alkezweeny | 1 | MRI | 464 W. Woodbury Rd. Altadena, Cal. | Membrane filter | 1 |
| P. Allee | 2 | U.S. Dept. of Commerce | Boulder, | Cond. nuclei counter (Twomey) | 14 |
| D. Alofs | 3 | Uni. of Missouri at Rolla | Rolla, Mo. |  |  |
| B. BenAri | 4 | Hebrew University | Jerusalem, Israel |  |  |
| A. H. Auer | 5 | Uni. of Wyoming | Laramie, Wyoming |  |  |
| E. K. Bigg | 6 | CSIRO | Sydney, Australia | Membrane filter | 5 |
| D. N. Blair | 8 | So. Dakota School Of Mines \& Tech. | Rapid City, So. Dakota |  |  |
| R. S. Bonner | 9 | U.S. Army Electronics Command | White Sands Missile Range | Cond. Nuc. counter (Twomey type) | 10 |
| W. Carnuth | 11 | Forschungsstille der Fraunhoerges | West Germany | Rapid exp. i.n. counter | 25 |
| $Y^{\prime}$. Chen | 12 | So. Dakota School of Mines \& Tech. | Rapid City, So. Dakota |  |  |
| B. L. Davis | 13 | " | " | Cond. \& i.n. chamber | 8 |
| J. E. Dinger | 14 | Naval Research Laboratory | Washington, D. C. |  |  |
| H. Dunsmore | 16 | DRI | Reno, Nevada | Cont. cold chamber | 11 |
| L. F. Evans | 17 | Uni. of Denver | Denver, Colorado | Cont. cold chamber | 9 |
| L. Facy | 18 | Meteorologie Nationale | France |  |  |
| N. Fukuta | 19 | Uni. of Denver | Denver, Colo. | Cont. cold chamber | 9 |
| A. Gagin | 20 | Hebrew University | Jerusalem, Israel | Membrane filter | 7 |
| H. Gerber | 21 | Atm. Sci. Laboratory | Ft. Monmouth, N. J. | i.n. spectrometer | 26 |
| G. Goyer | 23 | NCAR | Boulder, Colorado |  |  |
| L. 0. Grant | 22 | CSU | Ft. Collins, Colo. |  |  |
| T. J. Henderson | 24 | Atmospherics, Inc. | Fresno, California |  |  |
| A. W. Hogan | 26 | SUNY | Albany, New York |  |  |
| R. Husar | 27 | Uni. of Minn. | Minneapolis, Minn. | to aerosol monitering system | 24 |
| U. Katz | 28 | CSU | Fort Collins, Colo. |  |  |
| W. C. Kocmond | 29 | Cornell Aeronautical Laboratory, Inc. | Buffalo, New York | Cond. Nuc. counter | 4. |
| G. Langer | 30 | NCAR | Boulder, Colo. | Cont. nuc. counter Membrane filter i.n. NCAR i.n. counter | $\begin{aligned} & 21 \\ & 23 \\ & 27 \end{aligned}$ |
| T. Lockhart | 31 | MRI | Altadena, Cal. | Cond. Nuc. counter | 6 |
| Mr. Loittiere | 32 | Meteorologie Nationale | France |  |  |
| R. D. H. Low | 33 | U.S. Army Electronics Command | White Sands Missile Range, New Mexico |  |  |
| C. Magono | 35 | Hokkaido Uni. | Hokkaido, Japan |  |  |
| J. Neumann | 61 | Hebrew University | Jerusalem, Israel |  |  |
| T. Otake | 37 | CSU | Ft. Collins, Colo. | Mixing i.n. counter | 15 29 |
| J. Podzimek | 40 | Inst. Physics of the At SUNY | CAS, Prague, Czecho Albany, New York | vakia | 30 |
| H. Pruppacher | 41 | Uni. of California | Los Angeles, Cal. |  |  |
| L. Radke | 42 | Uni. of Washington | Seattle, Washington | Cond. nuc. counter <br> (light scattering) | 18 |

List of Participants of the 2nd IWCIN, Fort Collins - 1970

| Name Id | Identific No. | Ion Affiliation | Address | Apparatus | Outlet \# on duct |
| :---: | :---: | :---: | :---: | :---: | :---: |
| R. F. Reinking | 43 | San Jose State College | San Jose, Cal. | Rapid exp. i.n. Counter | $\begin{aligned} & 13 \\ & 17 \end{aligned}$ |
| C. Robertson | 44 | Uni. of Washington | Seattle, Washington | Membrane filter | 28 |
| J. Rosinski | 45 | NCAR | Boulder, Colorado |  |  |
| R. E. Ruskin | 47 | Naval Research Laboratory | Washington, D. C. | Cond. Nuc. counter (Twomey type) | 16 |
| V. K. Saxena | 48 | Uni. of Missouri at Rolla | Rolla, Mo. | Wilson chamber (cond. nuc.) | 20 |
| V. J. Schaefer | 49 | SUNY | Albany, New York |  |  |
| W. Scott | 50 | Uni. of Washington | Seattle, Washington | NCAR i.n. counter | 22 |
| G. Soulage | 52 | Faculte des Sciences de Clermont-Ferrand | France |  |  |
| R. Serpolay | 51 | Faculte des Sciences | de Brest, France |  |  |
| R. L. Steele | 54 | DRI | Reno, Nevada |  |  |
| A. C. Tebelak | 55 | Uni. of Missouri | Rolla, Missouri |  |  |
| G. Vali | 56 | Uni. of Wyoming | Laramie, Wyoming | Membrane filter | 3 |
| H. K. Weickmann | 57 | ESSA | Boulder, Colo. |  |  |
| K. T. Whitby | 58 | Uni. of Minnesota | Minneapolis, Minn. |  |  |
| T. A. Wojciechowski | i 59 | Naval Research Laboratory | Washington, D. C. |  |  |
| A. Schnell | 60 |  | Leadville, Colo. | NCAR i.n. counter | 12 |
| L. Vardiman | 62 | CSU | Fort Collins, Colo. | Rapid exp. i.n. counter | $\begin{aligned} & \text { 13, } \\ & 17 \end{aligned}$ |
| Carlson | 63 | CSU |  |  |  |
| Brown | 64 | CSU |  |  |  |
| C. Todd | 65 |  |  |  |  |
| L. Mendenhall | 66 | USAF Air Weather Servic |  |  |  |
| J. Moorhead | 67 | CSU |  |  |  |
| R. Cheng | 68 |  |  |  |  |
| M. Newchurch | 69 | CSU |  |  |  |
| D. Dahl | 70 | CSU |  |  |  |
| C. Swain | 71 | CSU |  |  |  |
| P. Squires |  |  |  |  |  |
| J. Hallett |  |  |  |  |  |



Numbers on the photo correspond to the "Identification Number" in the List of Participants.

## ACKNOWLEDGEMENTS

Acknowledgement for the planning and operations of the 2nd IWCIN workshop lies essentially with the participants themselves, since the workshop was operated on a voluntary basis with each participant responsible for his own equipment and observations. The workshop report, however, would be remiss without several special acknowledgements since without these contributors and efforts, the workshop could not have been held or the quality would have been considerably reduced.

Prime acknowledgement is made to the National Science Foundation, which supported the workshop under NSF Grant No. GA-13416. Additiona1 support was provided by NSF for the operation of the laboratory which serviced the workshop under Grant No. GA-20341. The NSF contribution was noteworthy since it represented an appreciation of the importance and need for comparing instrumentation used in the measurement of atmospheric nuclei.

The efforts of the chairman of the WMO Cloud Physics Committee, Dr. Helmut Weickman, and of the chairman of the sub-committee on Nucleation, Dr. J. Podzimek, were critical to the success of the workshop. Basic credit for the actual organization of the workshop during the preceding two years properly lies with the organizing committee and particularly with its chairman, Professor Roger L. Steele. These efforts deserve particular credit since, in a loosely organized cooperative venture by interested researchers, the initiative of this group was absolutely essential. Additional specific acknowledgement is made to Dr. Joe Podzimek for his day to day supervision and compilation of data, and to Dr. Podzimek and Hans Pruppacher for collecting and initial editing of papers prepared by the participants. Special acknowledgement is also made to Dr. Ulrich Katz, David Dah1 and the other members of the C.S.U. laboratory for their efforts in preparing and operating the supporting facilities.

Perhaps the most unique addition to this workshop was the participation of Dr. Ken Whitby and Rudy Husar of the Particle Technology Laboratory of the University of Minnesota. The sincere thanks of all participants are extended for their participation in providing data on aerosol size and concentration distributions for the aerosols used in the respective comparisons.

## WORKSHOP DATA <br> INTERPRETIVE COMMENTS

This section of the report lists in tabular form most data collected during the Second International Workshop on Condensation and ice nuclei. Fifty-five specific experiments were made during the Workshop. The data in this section consequently have been organized by experiment, 1 through 55 . However, no experimental data was available for experiment numbers $5,6,15,16,17$, and 18 . For each experiment the following information is presented in the following order:
A. Purpose of the experiment
B. Description of the experiment
C. Prevailing weather
D. Description of aerosol used
E. Observational data (AN,CCN, IN) for experimental
sample (exp. data)
F. Observational data collected at time of experiment but not from experimental sample or from times other than experiment times (other data).

For a schematic of the sampling system refer to the figures on page 18. Nuclei data is organized to systematically present data according to chronological listing of numbers assigned to the respective instruments. Since numbers, 1-39 were assigned to IN counters, $40-49$ to CCN counters, and $50-60$ to AN counters, observation of the respective nuclei consequently always follow in this order. Table 1 is provided as an index to show the machine number, operator, opérator number, type apparatus, and sampling duct number for each instrument. The use of numbers was required since all data have been computer processed. Table 2 serves as a key to notations used in the data tables. The aerosol distribution shown with each experiment was measured by the Minnesota Aerosol Analyzing System unless specified otherwise. All times are Mountain Standard Time.

Frequently special comments were made by the respective operators regarding certain aspects of their observations during specific experiments. These have been printed out in the data tabulations immediately following that participants observations for the experiment. Certain participants made special comments that applied to all of their observations. These are listed below:

Hogan: Vacuum pressure (in. of mercury) for mach \#54 is listed in the supersaturation columns and indicated by an asterisk.

Langer: Humidities and supersaturations are estimated for machine \#14.

Otake: Supersaturations are estimated for Aitchen counter.

Robertson: Two versions of data collected were presented. Unadjusted data supplied during the workshop are listed under "Other Data" adjusted data supplied subsequent to the workshop are shown under "experiment data".

Schnell: Supersaturations are estimated.

TABLE 1
KEY TO OPERATOR, MACHINE, AND SAMPLING CODES

| Machine No. | Operator | Operator Number | Type Apparatus | Duct \# |
| :---: | :---: | :---: | :---: | :---: |
| 3 | Gerber | 21 | Goetz-centrifuge | 24 |
|  | Gerber | 21 | 1 | 26 |
| 11 | Alkazweeny | 1 | Muf-IN (membrane filter) | 1 |
| 12 | Bigg | 6 | " ${ }^{\prime \prime}$ | 5 |
|  |  | 6 |  | 9 |
| 13 | Gagin | 20 | " | 7 |
| 14 | Langer | 30 |  | 23 |
| 15 | Vali | 56 | " | 3 |
| 16 | Robertson | 44 | Droplet freezing muf-IN | 28 |
| 21 | Vardiman | 62 | CSU Rapid expansion chamber - IN Bigg-Warner | 13 |
| 22 | Reinking | 43 |  | 17 |
| 22 | Grant | 22 | Bigg-Warner " " " | 17 |
| 22 | Brown | 64 |  | 17 |
| 23 | Carnuth | 11 | " " " $"$ (NCAR) IN | 25 |
| 27 | Carlson | 33 | " " " | 22 |
| 31 | Langer | 30 | Acoustical - IN (NCAR long cone) | 27 |
| 32 | Schnell | 60 |  | 12 |
| 33 | Scott | 50 | " | 22 |
| 36 | Evans | 17 | Continuous IN counter | 9 |
| 37 | Dunsmore | 16 |  | 11 |
| 38 | Davis | 13 | Non-continuous $\frac{\text { IN }}{\text { " }}$ counter | 8 |
|  | Chen | 12 |  | 8 |
| 39 | Ohtake | 37 | Cloud settling " " ESSA-ADCL Thermal Diffusion chamber-CCN | 15 |
| 41 | Alee | 2 |  | 14 |
| 42 | Kocmond | 29 | Non-continuous thermal diffusion chamberCCN |  |
| 43 | Bonner | 9 | " " " " | 10 |
| 44 | Radke | 42 | " " " | 18 |
| 45 | Ruskin | 47 |  | 16 |
|  | Wojciechowski | 59 | " " " " | 16 |
|  | Dinger | 14 | " " | 16 |
| 46 | Langer | 30 | NCAR Cloud cond. (Continuous acoust)-CCN | 21 |
| 50 | Ohtake | 37 | Gardner expansion AN counter | 15 |
| 51 | Allee | 2 | Gardner 961 - AN | 14 |
| 52 | Allee |  | " 911 " | 14 |
| 53 | Saxena | 48 | " Counter - ${ }_{\text {" }}$ AN | 20 |
|  | Tebelak | 55 | " | 20 |
|  | Alofs | 3 | " " | 20 |
| 54 | Hogan | 26 | Gärdner counter 961 - AN | 2 |
| 55 | Allee | 2 | ESSA Pollak Counter - $\overline{A N}$ | 14 |
| 56 | Saxena | 48 |  | 20 |
|  | Tebelak | 55 |  | 20 |
|  | Alofs | 3 | E-one 102 Aitken counter - AN | 20 |
| 58 59 | Hogan Hogan | 26 |  | 2 |
| 60 | Hogan | 26 | Gardner 814 - AN | 2 |

TABLE 2
KEY TO SAMPLE SOURCE CODE

| CODE | EXPLANATION | CODE | EXPLANATION |
| :---: | :--- | :---: | :--- |
| A | First large bag | H | Copper Pipe |
| B | Second large bag | I | Air between the actual big bag <br> and container |
| C | Third large bag | J | Room observation |
| D | Fourth large bag | K | Outside observation taken from <br> pipe leading to roof |
| E | Fifth large bag | L | Special backgrounds--soil samples, <br> water samples, etc. |
| F | Sixth large bag | Small bag direct (syringe) | M |




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[^1]:    *Present affiliation San Jose State College, San Jose, California.

[^2]:    *Present affiliation San Jose State College, San Jose, California.

[^3]:    *A detailed description of the apparatus is available in Information Circular No. 64 of the Natural Resources Research Institute, University of Wyoming, Laramie, Wyoming 82070.

[^4]:    *Fewer than 10 measurements.

