VARIATIONS IN ATMOSPHERIC ICE NUCLEUS CONCENTRATIONS

by

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ABSTRACT

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Concentrations of ice nuclei at MacDonald College were measured at certain intervals during June 1967 to January 1968 with an NCAR-Bollay Acoustical Ice Nucleus Counter. By comparing 2 identical machines run concurrently at the same location, this instrument was found to be reliable and consistent in determining daily variations.

Concentrations during precipitation were studied to determine if they were related to the amount that fell. Little evidence for such a cause-effect relationship was found. However, large numbers of nuclei were associated with the downdraughts of several thunderstorms.

Continuous measurements during December 1967 and January 1968 failed to establish a definite extraterrestrial influence on ice nucleus concentrations. Increases occurred when winter winds blew from Montreal's industrial complex and an afternoon minimum with peaks in concentration in the forenoon and evening were observed in a study of the diurnal variations.

PREFACE

The work towards this thesis was done at MacDonald College of McGill University under the supervision of Professor R.H. Douglas. It is intended as a preliminary study of the atmospheric ice nucleus concentrations at MacDonald College as measured by an NCAR-Bollay Acoustical Ice Nucleus Counter.

I would like to thank my fellow graduate students and the staff of the Agricultural Physics Department for their inspirational help. Besides Professor Douglas, I am especially grateful to Dr. G. Vali for his stimulating and rewarding advice and his invaluable help in setting up the instrumentation. Specific gratitude should also be directed to Miss M. Candlish for her assistance in the preparation of some computer programs and in the drafting of thesis diagrams, and to Mr. P. Levert who photographically reduced several thesis drawings.

The loan of the ALHAS Ice Nucleus Counter by Dr. P.W. Summers of the Alberta Research Council was very much appreciated.

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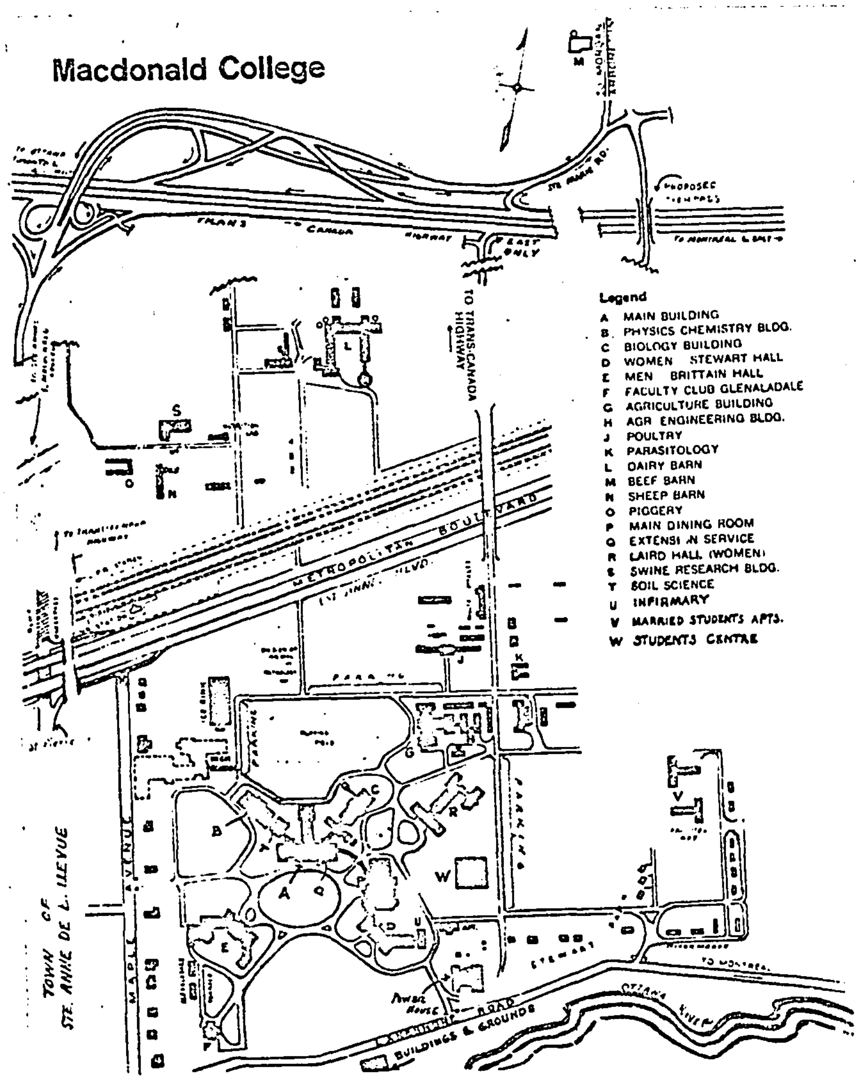
CHAPTER I

INTRODUCTION

Natural ice nuclei are considered important in atmospheric processes since they induce freezing in clouds thus initiating an efficient mechanism for cloud particles to reach a precipitating size. Consequently many studies have been undertaken to determine their chemical composition, their origin and their absolute concentrations. Unfortunately because of limited instrumental capability most of the above interrelated questions have many and sometimes directing conflicting answers. Comprehensive reviews on these ice nucleus studies have been made by Bigg (1961), Soulage (1961) and more recently by Mason (1968) and Dufour (1966).

In the following experiments an NCAR-Bollay Acoustical Ice Nucleus Counter was operated at MacDonald College of McGill University to determine ground level concentrations. The laboratory used was located in the Chemistry building (see map 1). Besides trying to obtain typical concentrations, experiments were undertaken to determine causes for fluctuations in continuous measurements. Such causes provide evidence suggesting possible sources and consequently probable nucleus chemical composition. Current theories provide three source regions, the ground, the stratosphere and outer space but little is known about the fraction of the natural concentration each forms.

There is always some doubt as to whether a point ice nucleus measurement is representative of its region. Several networks of stations have been set up and clarify this problem. Bigg and Miles (1964), using a filter technique, found their station network in Australia provided data which could be contoured. Kline and Brier (1961) observed that their downtown Washington D.C. station produced a mean count at -20C approximately 0.79 of those readings taken 8 miles west.



Map 1. Map of MacDonald College's grounds. The instruments were located on the southwest side of building B. From January 7 to 26, 1968 the ALHAS machine was in a laboratory on the northwest side of building B.

The twice daily measurements followed each other closely in pattern. Rosinski (1967) in comparing his Winter Park and Boulder stations, seperated by 45 miles, found an astonishing similarity in the monthly pattern of readings at 1400 hours taken at both -21C and -15C. Consequently if no local effects distort the count, the concentration at MacDonald College can be assumed representative of the immediate region. As Dufour mentions there is disagreement among scientists about whether the nucleus concentration varies with height in the first few kilometers. However it is assumed here that ground measurements are at least typical of the first kilometer.

The influence of a station's surroundings is critical. Effects of the natural vegetation, exposed fields of soil, nearby water bodies are important factors. MacDonald College borders a small city, St. Anne de Bellevue, along the Ottawa River and is mainly an agriculturally oriented institution where the immediate surrounding lands are farmed. Two four lane highways close by could have an effect on the count. Schaefer (1966) states that pure exhaust from an automobile has less than one ice nucleus per litre at -20C but under the influence of iodine vapour $5x10^6$ were observed. Since natural sources of iodine vapour are available, the possibility exists that ground measurements could be significantly affected by such a reaction. This effect will be assumed negligable and Schaefer's concentrations in pure exhaust would not significantly affect the values measured at MacDonald.

Cities with their industries, large transportation systems, and cold weather heating systems can produce pollutants which are potential nuclei sources. Dufour (1966) divides industrial sources into two categories according to results available in the literature. Organic residues such as soot, tar and fumes from machines such as planes and cars are not efficient ice nuclei but mineral residues such as silicates and metal oxides are. MacDonald College is not near any heavy industries but is approximately 20 miles west of Montreal, a city containing all varieties of factories. Consequently even though the winds are usually westerly, pollution from the city could influence the counts. Kline and Brier's (1961) values indicate the effect is not likely to be large.

CHAPTER 2

INSTRUMENTATION

2.1 Ice Nucleus Counters

The two major types of ice nucleus measuring techniques are chamber counters and air filtering devices. Since large variations in counting methods and operating procedures are contained within these two groups, good standard equipment is desired. Recent proposals for reference standards include the large CSF-NSF isothermal diffusion cloud chamber (Steele and Krebs, 1966) and a simple sugar bubble technique (Bigg, 1965). However even these methods have disadvantages.

Pulling air through membrane filters was a technique first develpped by Bigg et al (1961) and later elaborated on by Bigg et al (1963a), Mossop and Thorndike (1966) and Mossop et al (1966). Filters can be processed at leisure by cooling them usually to below -15C, humidifying them until ice crystals form and then allowing the crystals to grow in a sugar solution to a visually countable size. Unfortunately the above authors found that the humidification process is critical and the count does not increase linearly with volume sampled. However Stevenson (1968) has reduced these two problems considerably by processing the filters differently. The filter technique's main advantages are that it provides an integrated count over the filter's exposure and a station network is economically feasible with a central laboratory doing the analysis.

Mixing and expansion chambers are two variations for chamber counters. A mixing method (Bigg, 1957) simply cools an air sample in a cold box and an expansion counter (Warner, 1957) adiabatically cools a sample by expansion. Both techniques use visual or a sugar solution counting system. Because basically uncomplicated instruments of these types can only sample approximately 10 litres at a time with manual control necessary, they are unsutiable for atmospheric measurements. Large and rapid fluctuations can occur in the natural concentrations which require continuous monitoring. Consequently some automatic continuous mixing chambers have been developed. Those of Bigg and Meade (1959) and Hervier et al (1967) use photography with the sugar solution counting method to eliminate the observer. The NCAR-Bollay Acoustical Ice Nucleus Counter was developed using a new acoustical counting system proposed by Langer (1966). This mixing chamber requires limited maintenance during continuous operations.

Concentrations of nuclei with warm activation temperatures are the most interesting when considering cloud processes. Attempts are made to operate counters at the warmest possible temperature that one can observe reasonable numbers of nuclei. Since the mixing and expansion instruments listed have 10 litre chambers and natural background concentrations are from 1 to 10 nuclei per 10 litres, the normal operating temperature is -20C.

2.2 The NCAR-Bollay Acoustical Ice Nucleus Counter

The NCAR-Bollay Acoustical Ice Nucleus Counter has been manufactured in small but significant numbers and has gained recognition as a valuable machine. Langer et al (1967) describe its operation and application to tracking in the troposphere and Steele et al (1967) have calibrated it indirectly against the CSU-NSF isothermal diffusion cloud chamber. Although these papers describe the instrument adequately, a brief description with elaboration on a few important details follows.

Basically the machine (figure 1) continuously draws in air, usually at 13 litres per minute, humidifies and cools the sample to a desired temperature in a refrigerated chamber. A glycol-water mixture circulated gradually through the foam lining in the chamber prevents frost formation on the walls. After approximately a two minute mean residence time, the supercooled cloud including water droplets and ice crystals activated by ice nuclei are pulled through a small capillary. When a particle greater than the

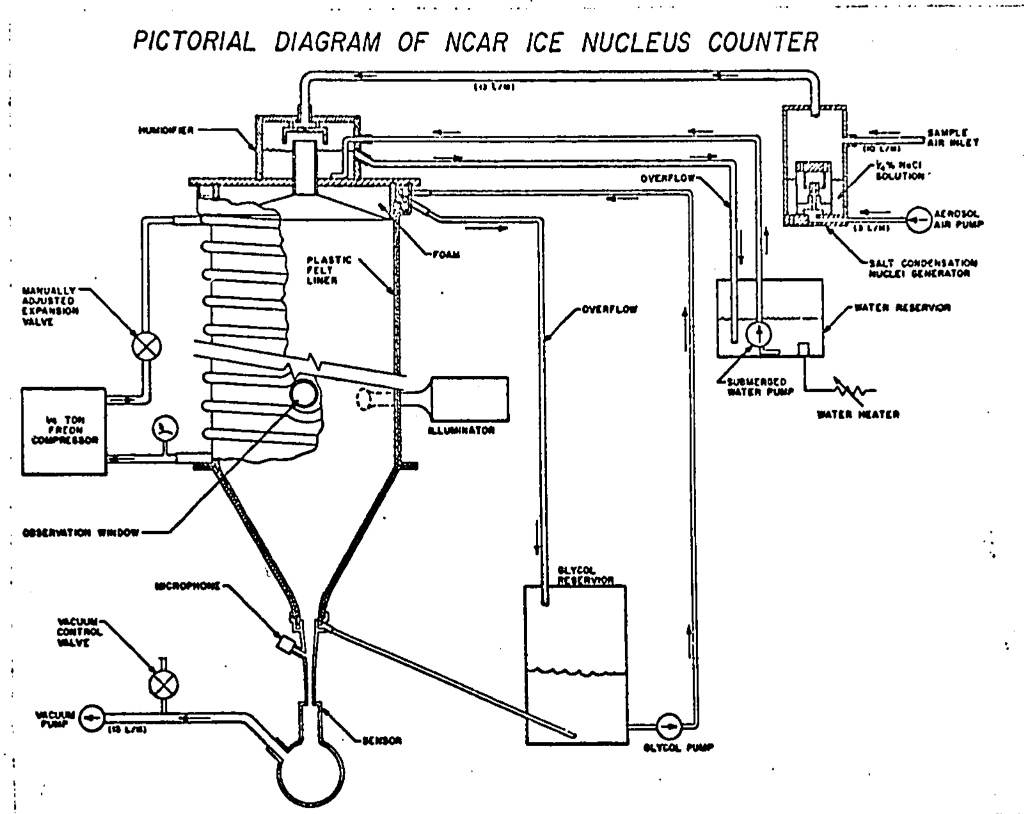


Figure 1. The NCAR-Bollay Acoustical Ice Nucleus Counter. (After Steele et al, 1967)

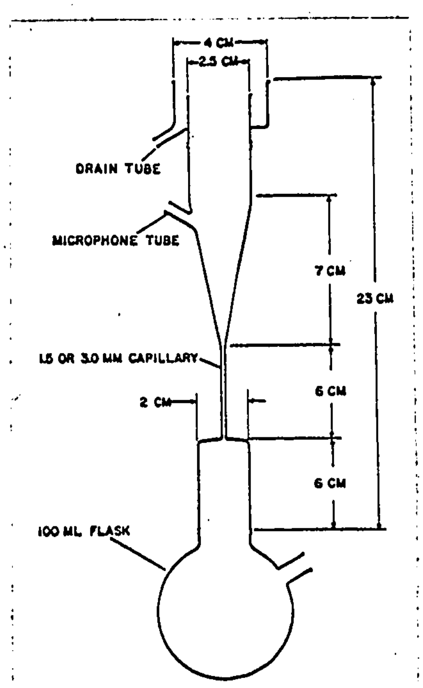


Figure 2. The acoustic sensor. (After Langer et al, 1967)

threshold size passes through this acoustic sensor (figure 2) an audible click results which is picked up by a microphone and counted by the rate computer. The exact cause for this click is unknown but a theoretical explaination is proposed by Dr. M.N. Plooster in a paper by Langer (1966). When a particle exits the capillary a sudden shift from laminar to turbulent flow causes a brief cessation of flow which results in a reflection of a shock wave up the capillary.

Since crystals grow to sizes greater than 40μ diameter, while the water drops remain considerably smaller, the sensor is designed for a threshold of 40μ. Langer (1966) has experimentally determined that when the sensor's suction setting is from 3 to 7 inches of mercury, the generated signal amplitude is not a function of size for particles greater than 40μ and below 10μ little or no response occurs. Particle density and shape do not determine the signal. Great care is taken to prevent particles other than ice crystals from being counted. An impactor at the chamber top removes all incomming airborne particles greater than 10μ. The aerosol generator, by bubbling filtered air through a salt solution of 0.25 per cent, introduces condensation nuclei into the incomming air to insure the formation of a dense cloud with drops smaller than 10μ. Because approximately 3 litres per minute goes through this aerosol generator, the counter samples 10 litres of atmospheric air every minute.

An NCAR-Bollay Rate Computer, model 102, was used as the electronic counting system. This instrument totalizes the crystals counted and automatically displays a reading in counts per minute on the most appropriate scale. The four scales have maximum readings of 10^1, 10^2, 10^3, and 10^4 counts per minute with the first, second, third, and fourth totalizing over 2 minutes, 12 seconds, 12 seconds, and 1.2 seconds respectively. The first two scales overload after 20 crystals are counted while the next two can reach 200. The sensor's microphone generates a sine wave pulse which according to Steele et al (1967) decays to one tenth of its original value in 5ms. To insure one crystal is not counted multiply, 5ms is taken as the dead or delay time setting on the rate computer.

Correctable errors occur because of this delay at counts greater than 10^3. Also to protect against false readings 50mv was taken as the threshold level on the incoming pulse. Since the rate computer's Rustrack chart output contains both the scale reading and the scale indicator, prolonged periods of unattended operation are possible.

Humidification is accomplished by passing the air over a heated water surface before introducing it into the chamber. The concentration measured depends on the water temperature. Morgan and Allee (1968) state that the optimum water temperature for silver and lead iodide is approximately 55C and 40C for chamber temperature ranges of -10 to -12C and -19 to -21C respectively. Heating the water less would decrease the count and greater temperatures would increase the supersaturations at the chamber top to destructive values for soluable deposition nuclei. Although Morgan and Allee's recommended values were not known during the experimental period, 40C ± 4C was taken as being the operational temperature between -19 to -21C. It is now evident that the allowable range of 8 degrees is large and could introduce an error of a factor two.

Temperatures in the instrument were measured with a thermocouple which when connected to a Varian recorder was accurate to approximately 0.25C. In figure 3 temperature profiles determined by running a thermocouple up the chamber's axis are plotted. During the measurement of these curves air was pushed through the chamber at the normal rate of 13 litres every minute. Using a thread trolley suspended at the top, the thermocouple was positioned in the chamber by pulling on threads near the glycol exit. Since the acoustic sensor was not connected, the values below 15cm are influenced by mixing and exposure to room air. The profile should be straight right to the bottom. Figure 3 shows a large temperature gradient near the top but below 55cm the profile flattens out. A position for taking a representative operating temperature could be and was taken approximately at the height to the observation window. Since ice crystals formed on the temperature probe if it remained in the chamber too long, the operating temperature was

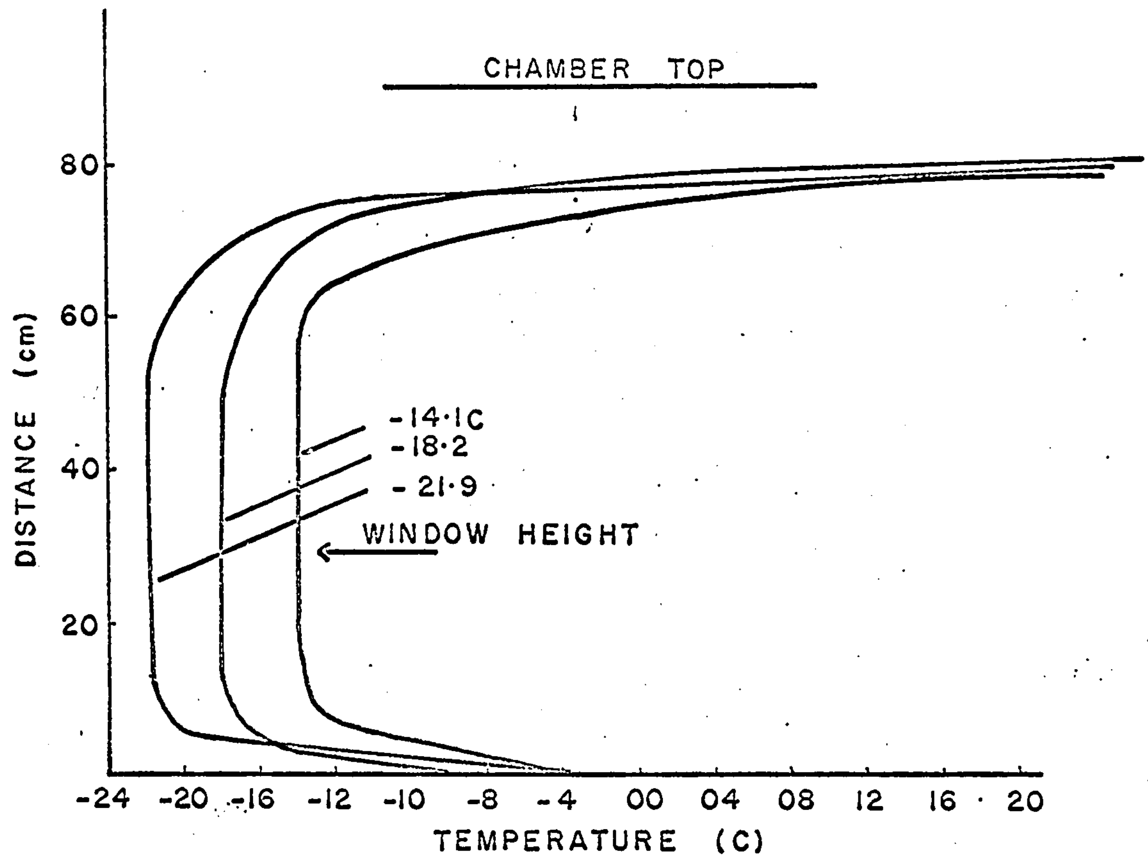


Figure 3. Chamber's vertical temperature profile for the indicated operating temperature. Distance is measured from the bottom of the chamber.

not continuously monitored. However when frequently checked during continuous runs, it remained remarkably constant and within 0.5C.

To consider the validity of peaks in atmospheric ice nucleus concentrations, the response of the NCAR-Bollay Counter should be known. Auer and Veal (1967) indicate that the machine can respond to sudden increases in 20 seconds. The decay in measured concentrations can be evaluated using a formula developed by Steele et al (1967). If perfect mixing occurs within the chamber then

$$C = C_o e^{-\left\{\frac{\dot{V}}{V}(t-t_o)\right\}}$$

defines the concentration C at time t where $\dot{V}$ is the flow rate, V is the chamber volume and C_o is the initial concentration and only source of nuclei. In such conditions C would decay by an order of magnitude in 5 minutes for $\dot{V}$ and V equal to 13 litres a minute and 28 litres respectively. However perfect mixing is a serious restriction since crystals fall to the bottom of the chamber quickly and the flow rate given would draw 28 litres in less than 2.5 minutes. Consequently 5 minutes is an upper limit for the time required for the concentraton to decrease an order of magnitude.

When Steele et al (1967) calibrated the NCAR-Bollay Counter against a modified Bigg-Warner Weather Bureau chamber, the mean ratio of counts as referred to this instrument was 0.36 with a standard deviation of 0.15. The Bigg-Warner machine, modified as a mixing chamber, gave excellent readings in comparison to a widely accepted standard, the CSU-NSF isothermal diffusion cloud chamber. Within the last 15 years estimates of the atmospheric ice nucleus concentrations were made only 2 or 3 times daily. However, cumbersome continuous machines showed that large and rapid fluctuations can occur in the natural concentrations. Since the NCAR-Bollay Counter reasonably agrees with non-continuous machines, it is an excellent instrument for continuously monitoring these variations. If the causes for most of these variations are determined, the natural sources of ice nuclei will be known.

CHAPTER 3

NUCLEUS ACTIVATION AND CRYSTAL GROWTH

3.1 Ice Nucleus Types

Four main ice nucleus activation processes should be considered possible in the NCAR-Bollay Counter. Two types of aerosols are only effective if they coagulate with the cloud drops. A freezing nucleus initiates crystalization if it can suspend itself inside a droplet while a contact-freezing nucleus causes freezing by simply touching a droplet. The third type consists of condensation and ice nuclei adhered together. Once condensation begins the ice nucleus will probably activate at a different temperature than it would if it coagulated with the drop. The fourth process consists of vapour directly converted into ice by a deposition nucleus* Most ice nuclei will be composed of particle aggregates containing liquid additives and this system will predetermine the activation temperature.

3.2 Processes Within the Chamber

If two populations of particles exist together such that one group (cloud droplets) are collecting the other group (ice nuclei) by collision or coagulation due to Brownian and turbulent motion, the fraction of the collectable aerosol (N_p) remaining at time t can be written as

$$\frac{N_p(o) - N_p(t)}{N_p(o)} = 1 - e^{-K' N_c t}$$

according to Wytlaw-Grey and Patterson (Byers 1965). N_c represents the number of collecting particles and K' the coagulation coefficient. Now

$$K' = K_B + K_T$$

where K_B and K_T are the coagulation coefficients for Brownian

* Name devised by J.E. McDonald (1958)
"Deposition"--a proposed antonym for "sublimation"
J. of Meteor. 15 245-247

Cloud Chamber Temperatures °C	Droplet Diameter,				Median Diameter, μ	Droplet Concentration cm^{-3}	Liquid Water Content g/m^{-3}	Super-saturation (Water) Percent	Super-saturation (Ice) Percent
	2.5	5	10	15					
-5.0	14	15	5	1	5.0	35	.014	.32	5.3
-9.5	26	34	10	1	4.9	71	.010	.40	9.9
-11.5	54	75	43	8	5.9	180	.040	.55	12.5
-16.0	82	100	33	3	5.0	218	.031	.62	17.5
-20.0	98	110	37	3	4.9	248	.035	.72	22.4

Table 1. A summary of parameters describing the cloud within the NCAR-Bollay Ice Nucleus Counter (After Auer and Veal, 1967)

and turbulent motion respectively.

$$K_B = \frac{RT}{3\eta N}\left[1/R_P + 1/R_C + A\ell\left(1/R_P^2 + 1/R_C^2\right)\right]\left(R_P + R_C\right)$$

where

$$\ell = \eta \pi^{1/2} / (2P\rho_a)$$

R =Gas Constant
η =coefficient of viscosity for air
ℓ =mean free path of air molecules
P =pressure
ρ_a =density of air
N =Avogadro's number
R_P, R_C =radius of nucleus, cloud droplet
A =0.9

Smoluchowski has shown (Byers, 1965) that

$$K_T = 4/3 \frac{\partial u}{\partial n}\left(R_C + R_P\right)^3$$

where $\frac{\partial u}{\partial n}$ equals the gradient of air velocity across the streamlines. Following Greenfield (1957) 30 (cm/sec)/cm is taken as an upper limit for $\frac{\partial u}{\partial n}$. If one assumes that the collecting group (cloud droplets) are just one size, it becomes very easy to determine the fraction of the ice nuclei collected by them as a function of time. The method used is identical to that developed by Greenfield (1957) in his paper on rain scavenging of radioactive particulate matter.

Auer and Veal (1967) have given representative values for some of the parameters within the NCAR-Bollay Counter. They are shown in Table 1. Values of R_c equal to 2.5×10^{-4} cm and a liquid water content of $0.04 gm/m^3$ were taken from this table giving N_c a value of 612. Following the above formulation $(N_p(0)-N_p(t))/N_p(0)$ was plotted against the radius of the collected particle for 1, 2, and 3 minute coagulation times. K_T is a negligable term for R_p less than 4×10^{-6} cm. The resulting graph (figure 4) should give a maximum for the number captured since an attempt was made to maximize the fraction by the appropriate choice of N_c and R_c. Very large drops are not sufficiently numerous to collect an appreciable fraction while very small drops

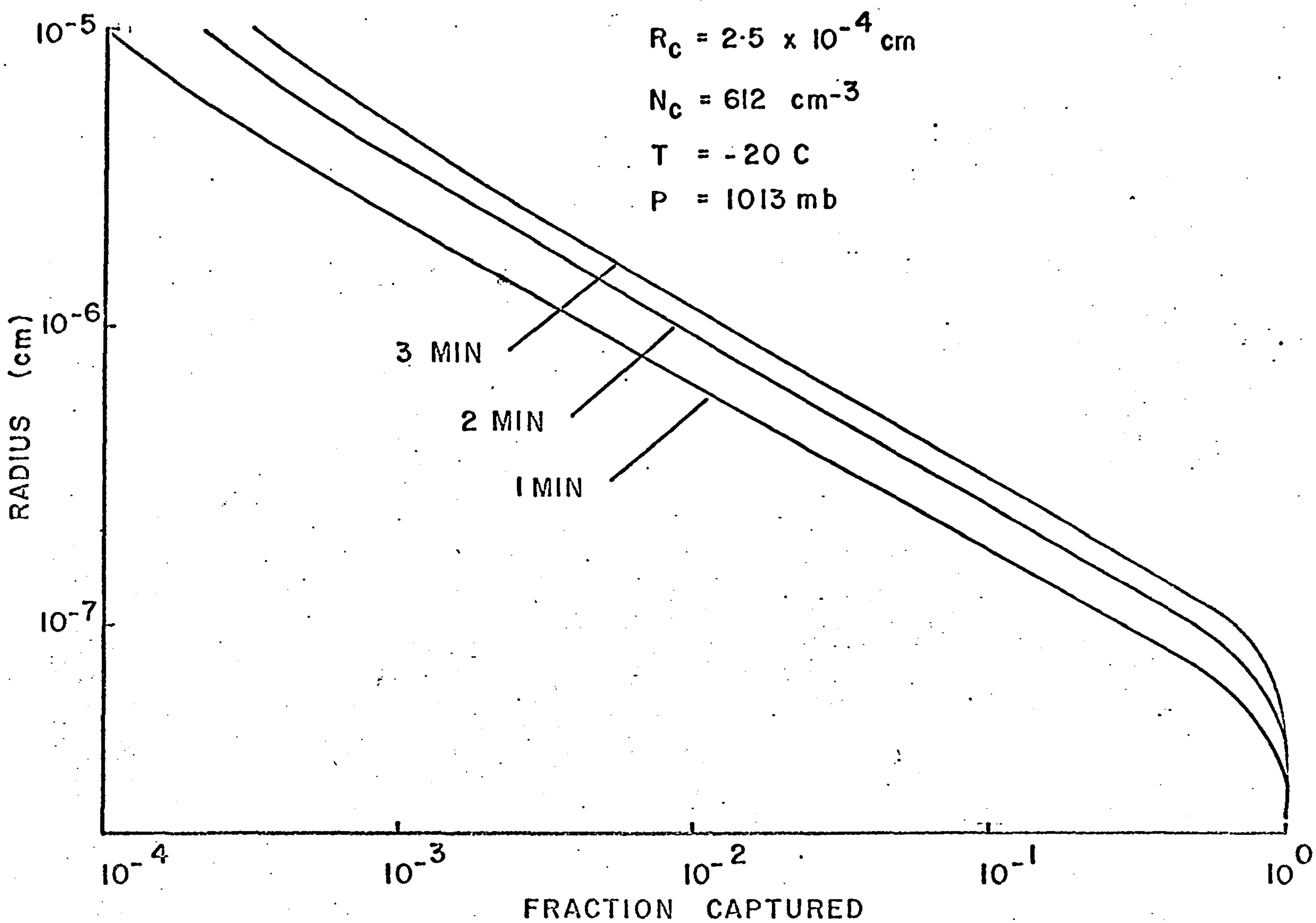


Figure 4. Fraction of an aerosol of one size captured by a cloud of drops 5×10^{-4} cm in diameter. Calculations for 1, 2, and 3 minute coagulation times are plotted.

or particles comparable in size to ice nuclei also cannot capture many.

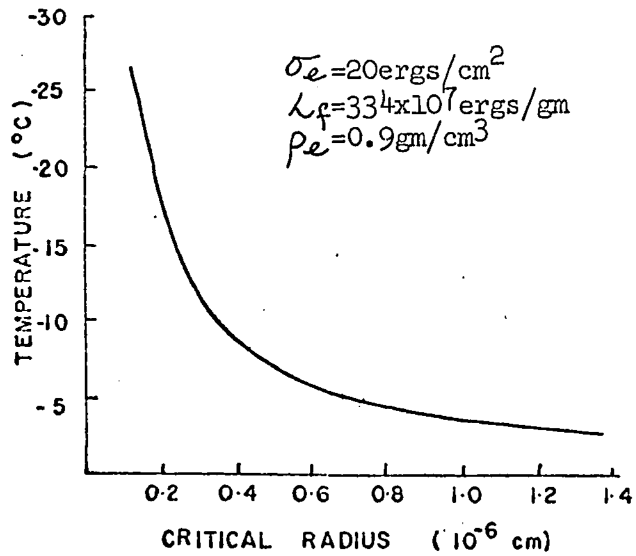


Figure 5. Critical radius for a freezing nucleus

Figure 5 is a graph of the critical radius for a freezing nucleus plotted against its activation temperature. It was calculated using

$$r^* = \frac{2\sigma_e T_0}{\rho_e L_f (T_0 - T)}$$

as given by Byers (1965) where r^* is the critical radius, σ_e is the specific surface free energy of the ice-water interface and L_f and ρ_e are the latent heat of fusion and the density of ice respectively. $T_0 - T$ is the supercooling required. Figure 4 shows that the NCAR-Bollay Counter only detects appreciable numbers of coagulating nuclei when their radii are less than $3x10^{-7}$cm. If freezing nuclei are as large as figure 5 indicates, they will not be efficiently counted by the Counter since they will never make contact with droplets. Junge (1963) shows that particles less than 10^{-6}cm cannot freely exist for more than a day in the atmosphere since they are captured by larger particles. Consequently if the Counter is recording reasonable concentrations of ice nuclei, there is evidence to suggest that it is not the freezing or contact-freezing process operating if Brownian motion and turbulence are the main coagulating mechanisms.

Langer et al (1967b) found in comparing the number of AgI particles generated, with the number activated in the NCAR-Bollay Counter, that the ratio was 1600 to 1. Edwards and Evans (1960) experimentally concluded that AgI particles would only act as deposition nuclei at 110 per cent relative humidity and thus were normally activated only when they collided with cloud droplets. Since 60 per cent of Langer et al's AgI nuclei were larger than 5×10^{-5}cm diameter, figure 4 successfully explains part of their observed discrepancy.

Vali (1966) suggests that most freezing nuclei in hail samples are smaller than 10^{-6}cm. If the NCAR-Bollay Counter is activating them, two possibilities arise. A continuous generator must exist releasing ice nuclei with a life time less than 24 hours (Junge, 1963) and some other process such as an electrical effect or Stefan flow is getting the nuclei into the drops. Or, condensation nuclei could capture freezing nuclei and the condensation-freezing mechanism would be the main process. These two possibilities are reinforced by the agreement Vali (1967) found between the NCAR-Bollay Counter and his droplet freezing technique. It must be remembered that if freezing nuclei are smaller than 10^{-7}cm, an efficient size for coagulation with cloud drops, then something is seriously wrong in figure 5.

Since Auer and Veal (1967) operated with their humidifier water temperature at 25C, it is probable that considerable errors are contained in their estimates of the chamber parameters. However, the discussion as to whether freezing nuclei are measured by the Counter is useful for determining what types of nuclei the chamber counts.

3.3 Crystal Growth

Ice crystals must grow to 40μ diameter particles in order to be counted by the acoustic sensor. Simple calculations were done using the standard crystal growth equation from Byers (1965). Thus

$$\frac{dM}{dt} = \left[\frac{4\pi C\,(S_e-1)}{\frac{L_s^2 m_\omega}{KRT^2}+\frac{RT}{D m_\omega P_e(T)}}\right]$$

C =capacitance= r for spheres, $2r/\pi$ for circular disks.
r =radius of crystal
$S_e - 1$ =supersaturation
L_s =latent heat of sublimation
R =gas constant
m_ω =molecular weight of water
$P_e(T)$ =saturation vapour pressure
T =temperature
D =diffusivity of water vapour in air
K =thermal conductivity of air
M =crystal mass

With appropriate changes in L_s, S_e, and $P_e(T)$ the above formula also specifies the growth rates of spherical water drops. Final radii of droplets and ice particles initially at 1μ in radius were calculated for 1 and 2 minute growth times in clouds at water saturation and supersaturations of 1 and 2 per cent. The range of supersaturations encompasses completely those in Table 1. Table 2 shows the calculation results for spherical drops and crystals, and ice disks one tenth their radii thick. Thicker disks would have final radii between the two crystal types calculated.

Phase	Water		Ice (C=r)		Ice (C=2r/π)	
Time (sec)	60	120	60	120	60	120
Supersaturation %						
0	1	1	20	28	59	83
1	5	7	21	30	61	86
2	7	9	21	30	62	88

Table 2. Final radius of water drops and crystals growing in various clouds at -20C. Initial radius is 1μ. Radius is given in μ .

At the usual chamber operating temperature of -20C, the majority of the crystals formed would be plates (Nakaya, 1954). According to Byers (1965) the circular disk's growth rate is representative of an hexagonal plate of similar dimensions. Table 2 shows that the water droplets cannot grow large enough to trigger the sensor but the crystals easily make the threshold limit.

CHAPTER 4

SUMMER DATA

4.1 Experimental Procedure

During June, July and August, the NCAR-Bollay Counter was run primarily in order to determine background levels and the variations in concentration before, during, and after precipitation. Special attempts were made to observe thunderstorms. The air inlet tube was located at an open window approximately 6 feet away from the machine (see map 1). Winds blew freely into the laboratory. This basement window of a three story building faced a large grass lawn with several cedars on the left and a paved road approximately 15 feet away. Cars did not affect the count (see Chapter 1 for a discussion of the ice nucleus concentration in auto exhaust) and the college road was not heavily travelled.

In figure 2 the top part of the acoustic sensor simply drains the glycol from the chamber's foam lining. Besides this straight drain, an elbow one designed for aircraft use is provided to allow the acoustic sensor to be positioned horizontally instead of vertically (figure 1). The straight tube was used in the summer even though operational difficulties were encountered if water drops fell off the chamber top into it. These drops could cause spurious peaks and an unusually high background count.

When the machine was operating frequent checks, at least every 6 hours, were made to determine if any ice had formed in the chamber and that the cloud was of a good visual density. These investigations included a complete halting of operations, making sure no water drops had fallen into the sensor, checking the chamber top for ice and cleaning it of water drops, and a final complete check of the inside chamber walls and glycol drain tube for ice. The acoustic sensor's capillary was cleaned daily with an acid and the glycol was changed often since it gradually collected water which raised its freezing temperature.

4.2 Summer Spectra

A plot of ice nucleus concentration against the chamber temperature will be called an ice nucleus spectrum. Normally an ice nucleus counter is run continuously at a single temperature making it necessary to be able to predict concentrations at other temperatures using a spectrum. During the summer a rough technique for measuring spectra with an NCAR-Bollay Counter was devised. The chamber was cooled not faster than 0.25 degrees C per minute while the Varian thermocouple recorder graphed the temperature variation thus allowing a direct comparison with the Rustrack recorder output. Slower cooling rates increased the probability that the natural concentration would change considerably and faster rates appeared to disrupt the chamber's temperature profile sufficiently to cause unusually low counts. As Morgan and Allee (1968) found, it is necessary to change the temperature of the chamber and humidifier together in order to obtain the best possible cloud. In these experiments compensating changes in the humidifier were not made when the chamber was cooled but no observable pattern directly associated with this approximation was observed.

In figure 6 MacDonald College spectra between -13C and -26C are plotted along with those of several other authors. In all cases a straight line was fitted to the set of experimental points but because of the considerable scatter this may be an oversimplification. Bigg (1961) has explained the physical significance of the spectrum's slope. Large numbers of nucleus types and broad size distributions among the varieties tend to produce straight line spectra. If one attempts to detect different types, the line's slope becomes important. For example, Bigg's curve (figure 6) indicated two nucleus varieties are present with only one above -23C.

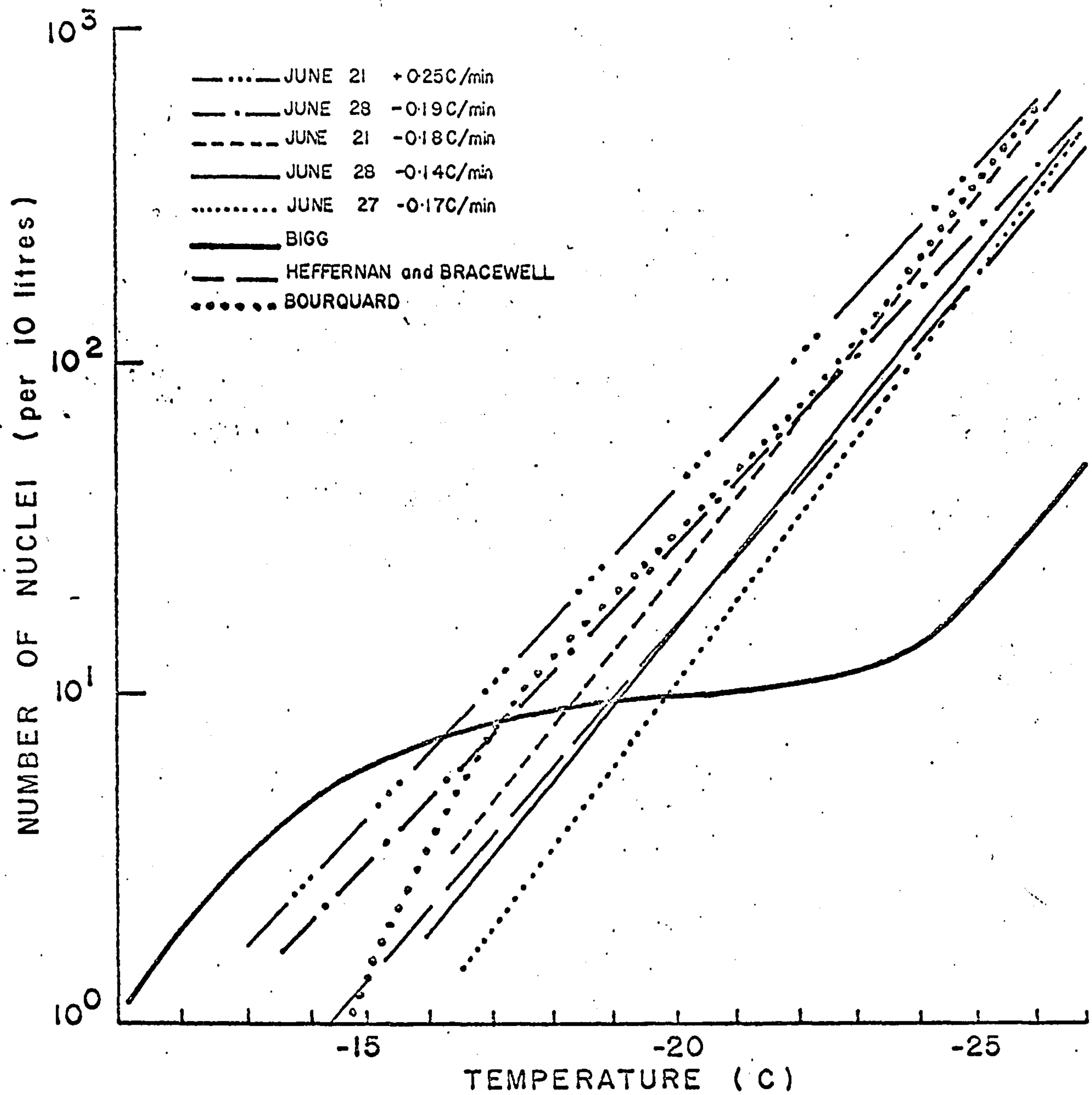


Figure 6. Spectra obtained during the summer. Three curves of other authors are drawn for comparison.

In figure 6 three curves are drawn for comparison including Bigg's (1961) tropical Australian station, Bourquard's (1963) mean Missouri (non operational morning) station curve and Heffernan and Bracewell's (1959) sample spectrum from Florida. Since the NCAR-Bollay values should be raised by a calibration factor 3 they would be above the comparison curves. However the spectra's slopes are comparable to the examples given here and are within the range Kline (1963) gives for his detailed study in the United States. Two sets of spectum curves on the same day are shown giving an idea of the possible daily scatter. One curve was calculated by warming the machine and in all cases the average cooling rate is indicated.

4.3 The Summer's Mean Concentration

During the summer the NCAR-Bollay Counter was run from 2 to 18 hours a day when it was convenient or when rain was expected. Usually the machine was operated in the late morning and early afternoon when it could be carefully observed. The overall average of 156 analysed hours of continuous measurements done between -14.6C and -21.9C was 14.7 nuclei per 10 litres at -21C. The concentrations were standardized to this temperature using the fairly constant spectrum slope indicated in figure 6. Case means were calculated by time weighing every rate computer reading. The mean is similar to those in other geographical area's (Bigg, 1965). Although the summer average is unexplainably lower than the mean concentration at -21C for the period in figure 6, spectra taken at odd and infrequent intervals throughout the summer all exhibited the same slope.

CHAPTER 5

ICE NUCLEUS CONCENTRATIONS AND RAIN

5.1 Literature Introduction

The relation between the occurrence of precipitation, the amount that fell and the concentration of atmospheric ice nuclei is a perplexing problem. Attempts have been made to prove the Findeisen-Bowen meteor hypothesis on the basis of rainfall singularities as well as ice nuclei anomalies on the predicted dates. The problem of the moon's influence is treated similarly. However the assumption must be made that the rainfall amount is dependent on the ice nucleus concentrations. Direct evidence for such an hypothesis is rare.

Gagin (1965) in Israel found no relationship between ice nuclei counts at -15C and rainfall. However on a plot of precipitable water and his defined effective nucleus concentration, he found it possible to discriminate between rainy and dry weather. He concludes that ice nuclei might initiate rain rather than determine the amount. Georgii (1960), in attempting to relate ice nucleus concentrations with the weather at his observation site, found that showers occurred on days with above average concentrations and continuous rain on days below average. Isono et al (1966) found "a close relation between the fallout rate of graupel pellets and snow crystals and the concentration of 'effective' ice nuclei which are active above temperatures of the cloud top." Bigg and Miles (1964) did find a relation between the concentration at -15C evaluated from a filter exposed for a day, and the daily rainfall amount. From a no rain case to 30mm a day the mean increased from 175 to 450 nuclei per cubic metre.

Isono and Tanaka (1966) found increases of ice nucleus concentrations underneath a thunderstorm in the downdraught. They suggest that the observed nuclei were entrained into the cloud, were activated, and subsequently placed in the downdraught only to have the droplet they eventually formed evaporate below the cloud base. This would suggest a thunderstorm filters out potential nuclei. Another possibility suggested was that the dry air forming the storm was unusually rich in nuclei. Preactivation and blown up dust were regarded as unlikely causes of the high concentrations. It is significant that one cannot safely conclude that the thunderstorm was caused or its intensity enhanced by the unusually high density of nuclei.

5.2 Storm Analysis Procedure

One of the main objectives during the summer and fall was to obtain variations in concentrations before, during, and after rain. The summer data were collected by the method described in section 4.1. However, from October 22 to November 1, a continuous run was attempted with variations from the summer procedure. The laboratory window was shut with an air inlet tube being placed 2 feet outside, 18 inches off the ground's surface. Unfortunately the instrument could only be checked at least once every 12 hours.

During precipitation periods when the machine was operating, the data were analysed in detail. For most storms every Rate Computer reading on the 12 second and 2 minute scales (0-10/min, 10-100/min) was tabulated and then time weighed over 7.5 minute periods. On October 26 and 28 the 7.5 minute means were obtained subjectively by a mental mean. Longer averaging periods (see figure 7) such as 15 and 30 minutes tended to obscure some of the detail while a shorter interval was not chosen because of a possible 2 minute delay in the response of the machine and timing errors.

Evaluation of rainfall rates was done using the campus tipping bucket data collected approximately 0.5 miles away. Errors will result because of this moderate separation when comparing with rapidly fluctuating concentrations in small disturbances such as thunderstorms. The beginning of rain has been taken as the time of the first bucket tip.

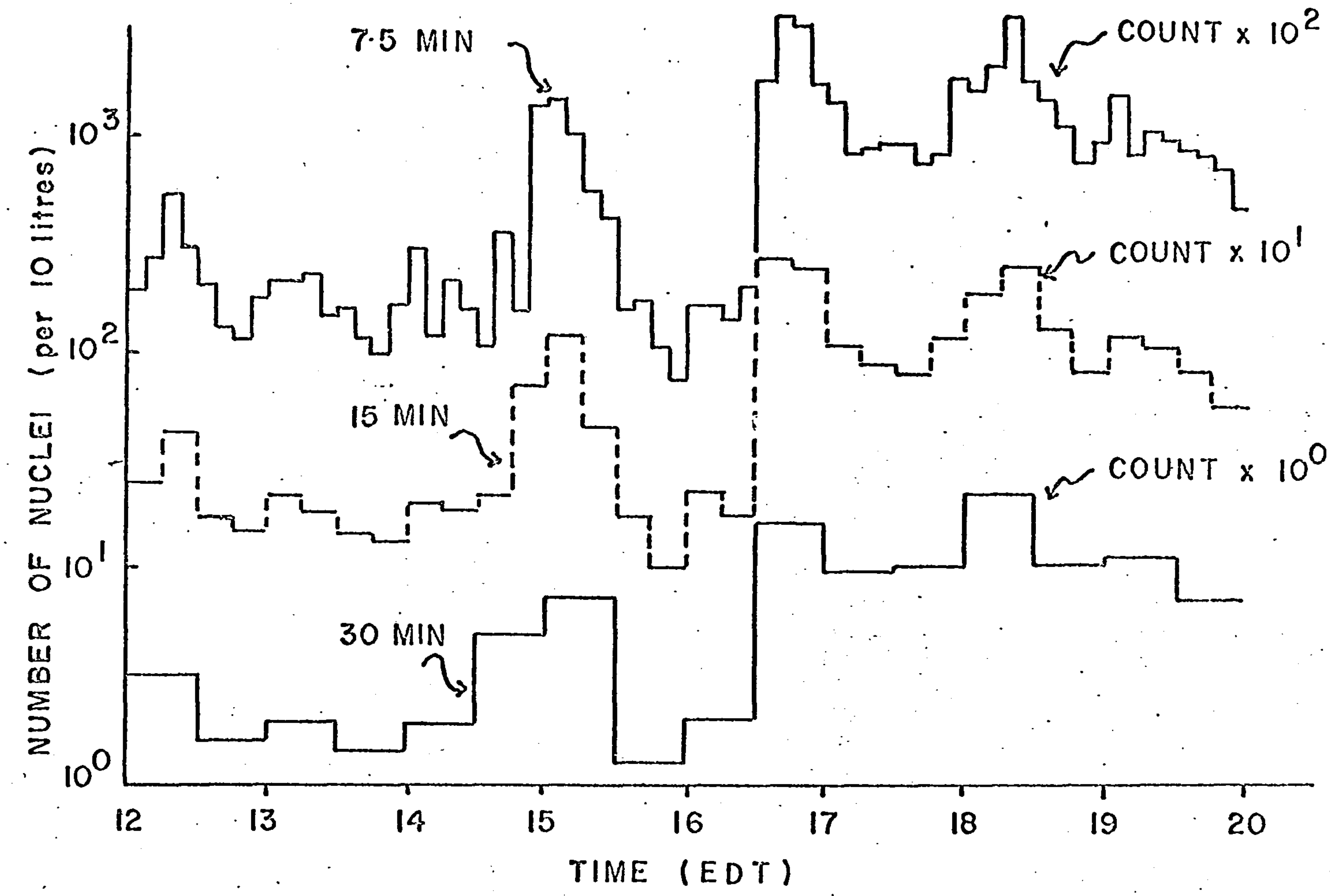


Figure 7. The storm of August 18, 1967. Concentrations of ice nuclei measured at -21C are drawn with 30, 15, and 7.5 minute means.

Continuous wind measurements at the observation site were needed. However the nearest wind measurements were taken more than 10 miles away at Montreal International Airport. Even though it is recognized that wind velocity magnitudes and changes are extremely important, because of this large separation, comparisons were not made. No other data were available from college instruments for correlations. The lack of radiosonde data made it impossible to estimate an 'effective' nucleus concentration, the number which would activate above cloud top temperatures.

5.3 Convective Rain Storms

On July 28 1967 a thunderstorm occurred associated with a front lying in the northeast-southwest direction. The count (figure 8a) remained steady from late morning to early evening with an increase occurring just after the main rainfall peak.

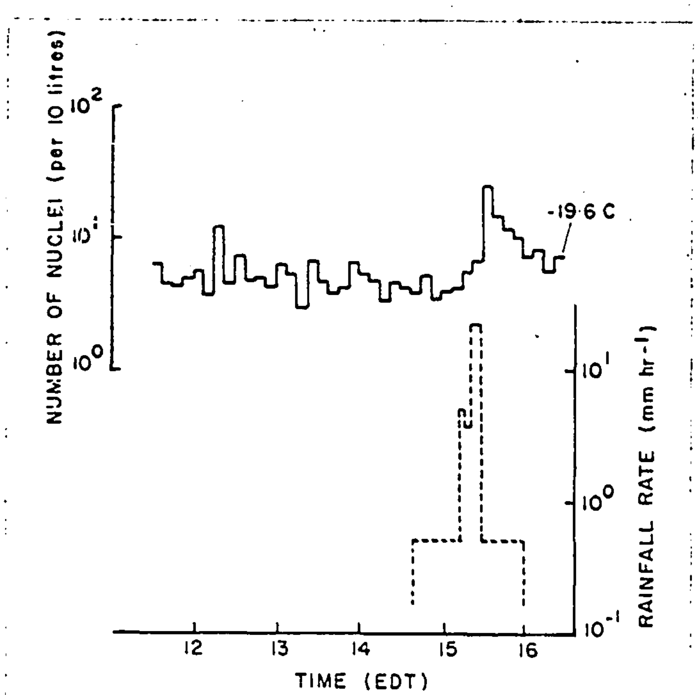


Figure 8a. Storm of July 28 1967. Ice nucleus concentrations at -19.6C and rainfall rate are plotted against time.

The peak's delay appears to be real and not associated with normal instrumental lag. In figure 8b an atmospheric nucleus spectrum, determined immediately after the storm, is plotted. The dashed lines represent the range of values while the solid line is the best straight line fit to the set of readings. Three spectra

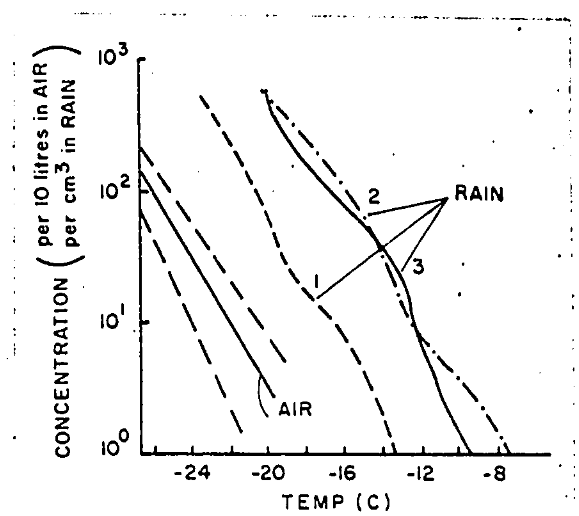


Figure 8b. An ice nucleus spectrum in air taken from 1625-1720 July 28, 1967. Freezing nucleus spectra from rain samples 1, 2, and 3 collected from 1520-1525, 1530-1545, and 1600-1617 respectively in the July 28 storm (see figure 8a)

of freezing nuclei in rain samples are also shown. They were analysed by the method developed by Stansbury and Vali (1965) and have been described in detail by Bishop (1968). The first, taken in the very intense rain, was much cleaner than the second and third collected during small rainfall rates. The marked similarity in the slopes of the air and rain spectra suggests that the two techniques are directly comparably. This is encouraging considering the rain technique counts only freezing nuclei while the NCAR-Bollay Counter is capable of activating many nucleus types. Also the nuclei in the rain samples were scavenged in the cloud while the NCAR-Bollay Counter measures the concentrations in the air at ground level.

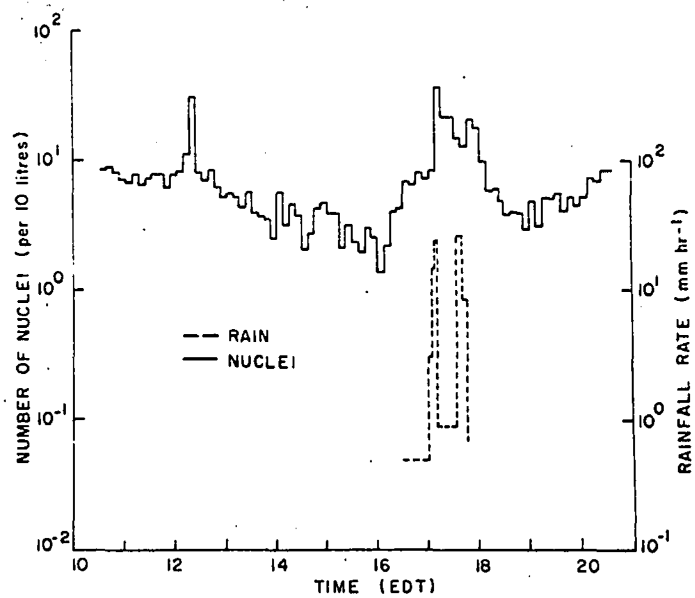


Figure 9. Storm of August 4, 1967. Ice nucleus concentration at -19.4C and rainfall rate are plotted against time.

On August 4 (figure 9) a storm occurred again with a front lying in the northeast-southwest direction. No thunder or lightning was observed but two distinct showers are recognizable. It was sunny until approximately 1600 when dark cumulus appeared. After the showers the cloud broke up and by 2000 blue sky was visible with thinly scattered relatively low cloud. Again the ice nucleus concentration decreased from 1000 until 1615 and then gradually increased with a sharp increase when the first shower began. The concentration decreased after the rain stopped.

On August 18 two storms occurred in the same evening with thunder and lightning observed with each. Both could be associated with a strong cold front with a notheast-southwest orientation. Forty mile an hour gusts, power failures and property

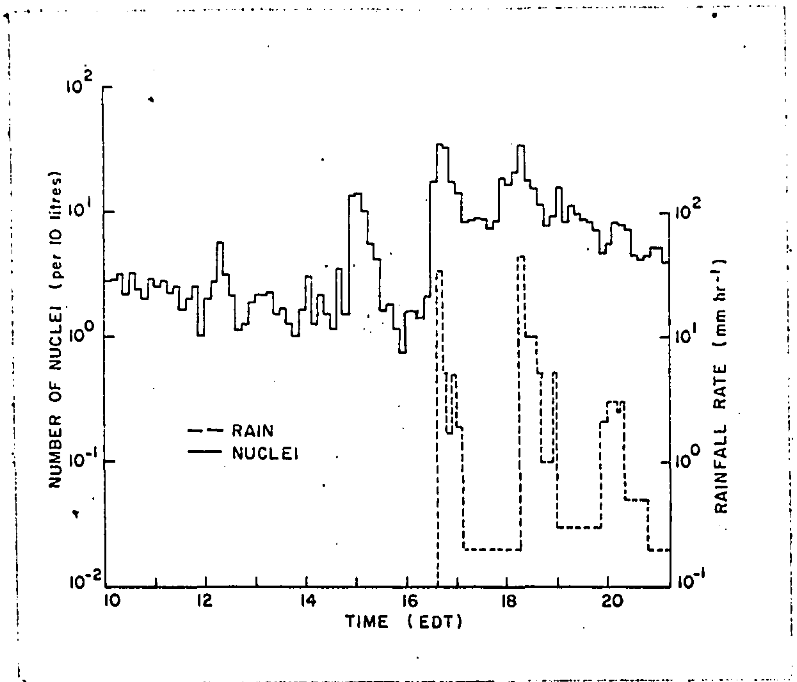
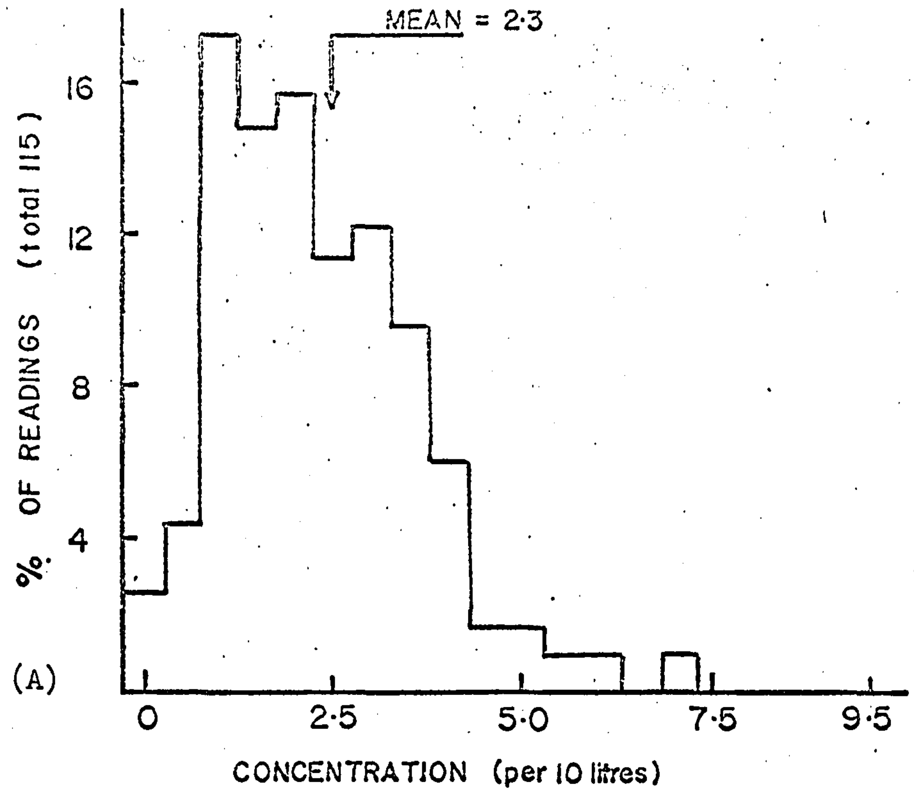


Figure 10. Storm of August 18, 1967. Ice nucleus concentration at -19.6C and rainfall rate are plotted against time.

damage due to winds and lightning were reported in Montreal. The ice nucleus counts (figure 10) generally decreased during late morning and early afternoon with a peak occurring at 1500. About 1625 the first storm increased the concentration 20 times. The original chart shows the count went from 1 to 20 counts per 10 litres (2 minute averages) in less than 4 minutes. After the rain almost stopped at 1710, the number levelled at 8.5 only to increase again before the second storm, near 1820. By 1910 the rain had stopped with the sky overhead clear and cumulus in the west. Another shower occurred later on in the evening. Because rain gauge data cannot accurately give beginning and end of precipitation, the clearing at 1910 is not shown in figure 10. Long rainfall rate bars indicate 0.01 inches of rain fell sometime within that time.

Histograms showing the frequency distribution of readings during raining and non-raining portions of August 18 are plotted in figure 11. Frequency distributions of Rate Computer readings are difficult to form because of the instrument's scale switching capability. Not only does this switching change the intervals between readout values but each scale averages its values over different time periods. All the readings for the non-raining portion (1000 to 1400 EDT) while just 6.3 per cent of the values for the raining



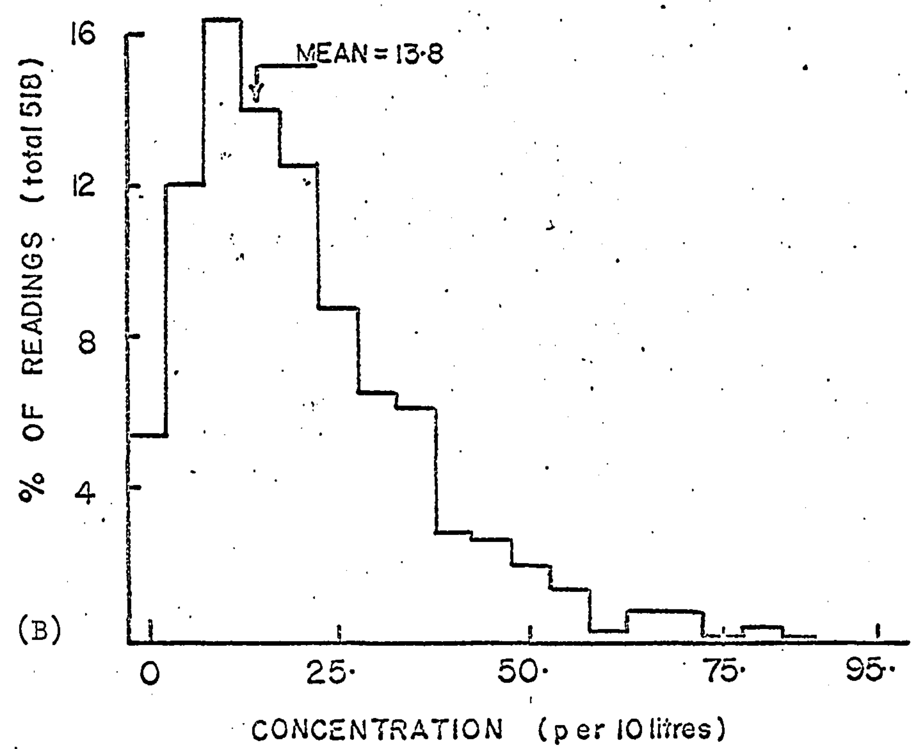


Figure 11. Histograms for August 18, 1967. A) Non-raining portion from 1000-1400 EDT with all readings used. B) Raining portion from 1630-1930 EDT with 6.3% of the readings omitted.

section (1630-1930 EDT) were registered on the 2 minute scale. Consequently only the first 12 second scale readings for the raining section are analysed in the histogram 11b. The percentages indicate the fraction of the total number of Rate Computer readout values giving that particular reading. Although the means between the two periods are almost a factor 6 apart, the distribution's shape in both cases is the same. It is not Poisson since such a distribution would have a peak at the mean. A more detailed discussion of the shape appears in section 6.2.

5.4 Continuous Rain Storms

Large fluctuations in nucleus counts during the continuous rain of August 9 are not observed. The concentration (figure 12) decreased from 1100 to a little after 1500 when the increase in rainfall rate did raise the concentration to a new level.

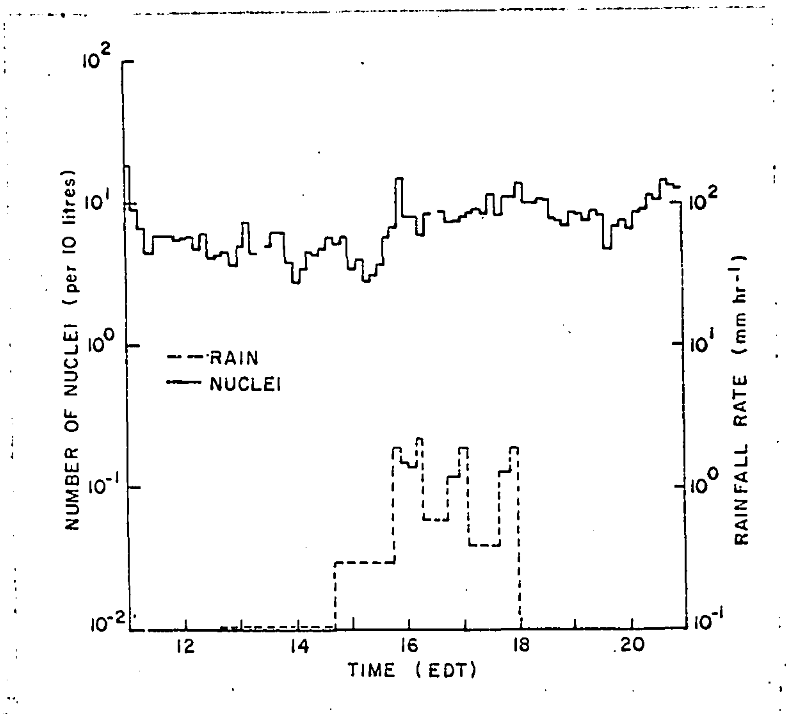


Figure 12. Storm of August 9, 1967. Ice nucleus concentration at -19.6C and rainfall rate are plotted against time.

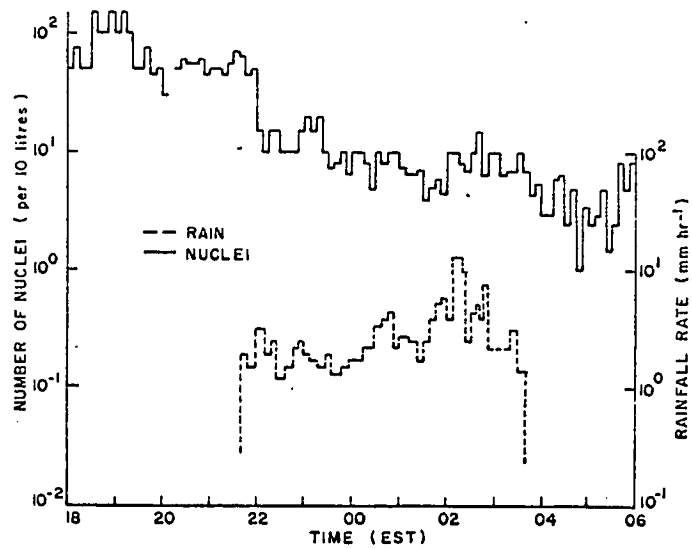


Figure 13. Storm of October 25 to 26, 1967. Concentration of ice nuclei at -21.1C and rainfall rate are plotted against time.

On October 25 to 26 when a cold front passed, associated with a strong low moving up from the southwest, rain occurred. As the rain started the concentration of ice nuclei, as shown in figure 13, dropped significantly. Before the storm began and after the storm ended the concentration was steady at the values indicated.

On October 28 (figure 14) a similar pattern in ice nucleus concentration occurred. Rain associated with an occluded front appeared to occur with a decrease in the number of nuclei. Here the changes were not as marked as in the previous case. During the October 25 to 26 and the October 28 storms, the air intake was a two foot tube placed approximately 18 inches off the ground. On August 9 the air intake was a tube placed at the open laboratory window. Consequently the change in the intake's position may have had some influence on the concentrations.

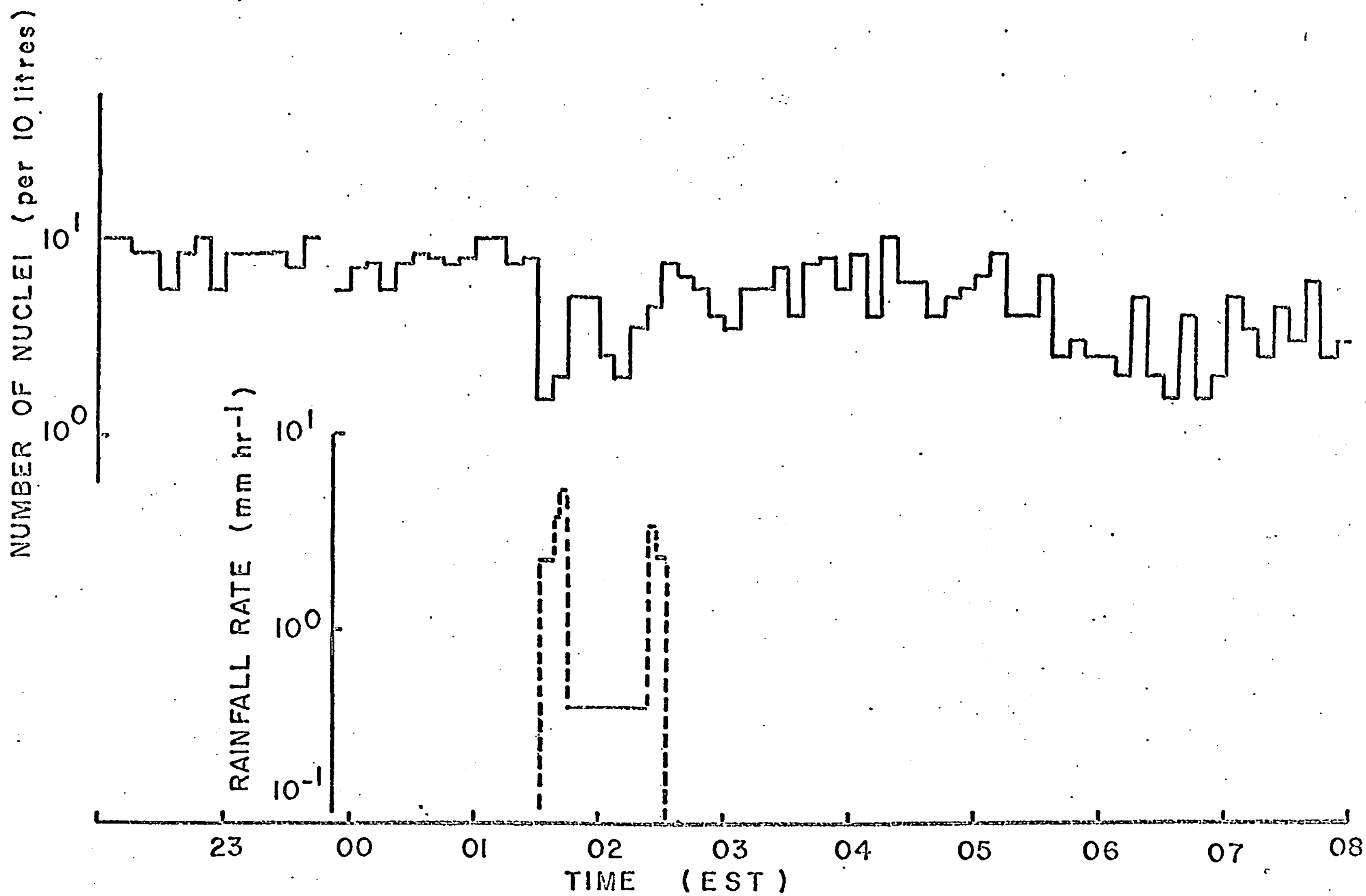


Figure 14. Storm of October 27 to 28, 1967. The concentration of ice nuclei at -21.2C and the rainfall rate are plotted against time.

5.5 Summary of the Observed Concentrations During Rain

A distinction between continuous and convective rain appears justifiable since different patterns are observed. Large and briefly sustained peaks occur in the convective but not in the continuous storms. The sharp increase in concentration associated with the onset of high intensity rain can be explained several ways as Isono and Tanaka (1966) have shown from their one observed storm (see section 5.1). Their conclusion that the increase occurred in the downdraught is logical for cases shown here since the jump is associated with the high intensity rain and comparable increases were not obtained in continuous rain. Unfortunately no other data were available at MacDonald to completely determine the time of the downdraught.

Isono and Tanaka eliminated the possibility of a blown-up dust source since dust swirls were not observed and the ground was wet. Isono (1959) has also stated that local dust sources do not affect nucleus counts. However, another possible dust effect is peculiar to the NCAR-Bollay Counter. Experimentally it was found that the impactor on the Counter (figure 1) can be overloaded and allow dust particles large enough to trigger the sensor through, but the overload concentration was not believed to be representative of atmospheric phenomena. Another source of inaccuracy could have been a mist of splashed-up particles entering the air when the heavy rain began. Attempts to induce such an effect using a water hose for a rain source failed. It seems probable that blown or splashed up dust did not induce the high concentration.

In the continuous rain examples interesting variations occur at the onset of rain. On August 9 the concentration increased while on October 25 and 28, when the intake was only 18 inches off the ground, the count fell. Whether the inlet's location accounts for the fall is not known. However because the count does decrease, it reinforces the argument that splash does not explain the increase in convective rain. No consistent washout pattern was observed in the continuous rain storms. Perhaps the variations detected are due simply to a change of air mass when a front passed.

The convective concentrations show a pattern similar to those Dingle and Gatz (1966) found in their 'well-organized' storms' precipitation samples. Their rain samples became cleaner in radionuclide and pollen concentration as the storm progressed. Bishop (1968) has studied the freezing nuclei concentration in rain samples, collected during the August 4, 9, and 18 storms described here, and has found evidence suggesting Dingle and Gatz's pattern is similar to that followed by freezing nuclei. However as figure 8b shows, the heavy rainfall produced rain with a lower concentration of freezing nuclei than the light rain.

It is a great temptation to attempt to relate ice nucleus concentration with rainfall rate even during individual storms. However the decision as to the time period over which to average the concentration and rainfall rate before comparing is difficult without a precise knowledge of wind patterns; if one assumes the air at the ground is instantaneously representative of that in the cloud, the result will be inaccurate. Figure 15 compares 7.5 minute averages for August 18 and 4 with all rainfall rates occurring with each average. A slight increase in concentration is observed for an increase in rainfall rate. However on October 25 and 28, the count decreased during precipitation obviously putting severe restrictions on figure 15. Too many variables are present for any simple comparison between rainfall rate and concentration to be valid. Individual storms cannot be lumped together since effective numbers will vary with different cloud top temperatures. Perhaps the long term comparison of daily rainfall and average daily concentration of Bigg and Miles (section 5.1) produces a realistic result but even it does not show dramatic interdependence.

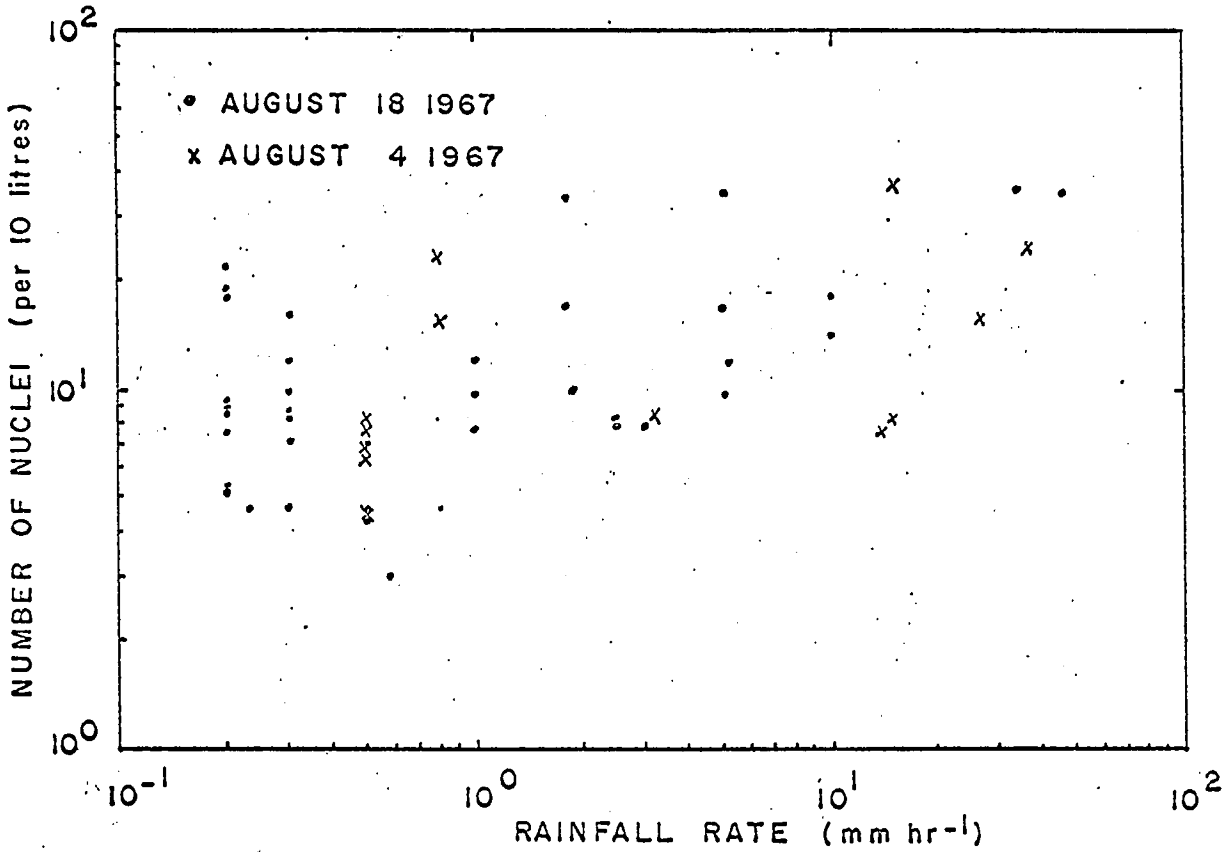


Figure 15. Relation between rainfall rate and ice nucleus concentrations at -19.5C for August 4 and 18, 1967.

CHAPTER 6

WINTER DATA

6.1 Objectives and Procedure

During the winter the ground is not exposed to the wind and consequently the possibility that a local dust source exists is remote. Also, during January several major Bowen rainfall anomalies exist which should be associated with ice nucleus peaks in concentration if the Bowen meteor theory is correct. The meteor showers occur in December with a lag of 28 to 30 days before the rainfall peaks. Consequently in order to evaluate the possibility of ground and meteor sources, the NCAR-Bollay Counter was run continuously at -21C from December 6 to January 26 with a break at Christmas. A second identical Counter, borrowed from the Alberta Hail Studies (ALHAS), was run under the same conditions from December 18 to January 5. This duplication was designed to estimate how real the absolute concentration and variations determined by a single machine were.

The outside air inlet was located approximately 10 feet off the ground and 3 feet away from a second story window. The instrument, connected to the inlet by a 1.5 inch plastic pipe, was located in a laboratory maintained at 20C. Since difficulties were previously encountered using a straight glycol exit tube, the elbow airplane designed tube was used. This effectively eliminated the problem of falling water drops getting into the acoustic sensor without changing the count. A great tendency for ice to form inside the glycol exit was reduced by directing a small blower on the outside of the tube keeping that surface ice free. Glycol was changed daily, the humidifier water pot was cleaned at least once a week and the sensor was cleaned daily with an acid. The instrument was checked thoroughly (see section 4.1) at least every 13 hours and usually three times daily at approximately 1000 to 1100, 1700 to 1800, and 2200 to 2400.

The operating temperature of the main instrument was measured 53 times during the period and was always within 0.25 degC of -21.3C. The ALHAS Counter strayed slightly out of this 0.25 degC range only twice in 21 readings. The time of measurement varied considerably and on several occasions two or three readings were made in a day.

The winter data were not analysed in as great detail as were the summer data. Subjective 15 minute averages, determined from the Rustrack charts by eye, speeded up the processing considerably. The possible averages were 0, 0.5, 1.0, 1.5,......,9.0, 9.5, 10., 15., 20., ..., 9.5., 100., 150., ..., 950., or the same as the readings the rate computer gave. Only periods where the full 15 minutes was available were tabulated. After checking the Counter, at least 15 minutes were taken as a warm-up period meaning at least 30 minutes of dead time are associated with each instrumental cleaning. Consequently usually 90 to 120 minutes a day were rejected for this reason. Other rejection reasons involved other cleanings, false data due to instrumental failure, or experiments designed for spectra, checks of inlets and flow rates, and rate computer checks. From December 6 to January 26 when the Counter was operating, 16.7 per cent of the time involved invalid counts.

6.2 General Description of the Continuous Run

Hourly averages for both the McGill (solid) and ALHAS (dotted) Counters are shown in figure 16 along with the outside air temperature readings obtained from Montreal's International Airport. Although points are not shown, no attempt has been made to smooth the curve. The daily weather summaries for December and January (Tables 3 and 4) have been extracted from the 'Monthly Meteorological Summary' prepared by the Meteorological Branch of D.O.T. for the airport. Wind velocity and direction will be discussed later.

Looking at the McGill curve (figure 16) it can be appreciated that several daily observations could easily give an incorrect

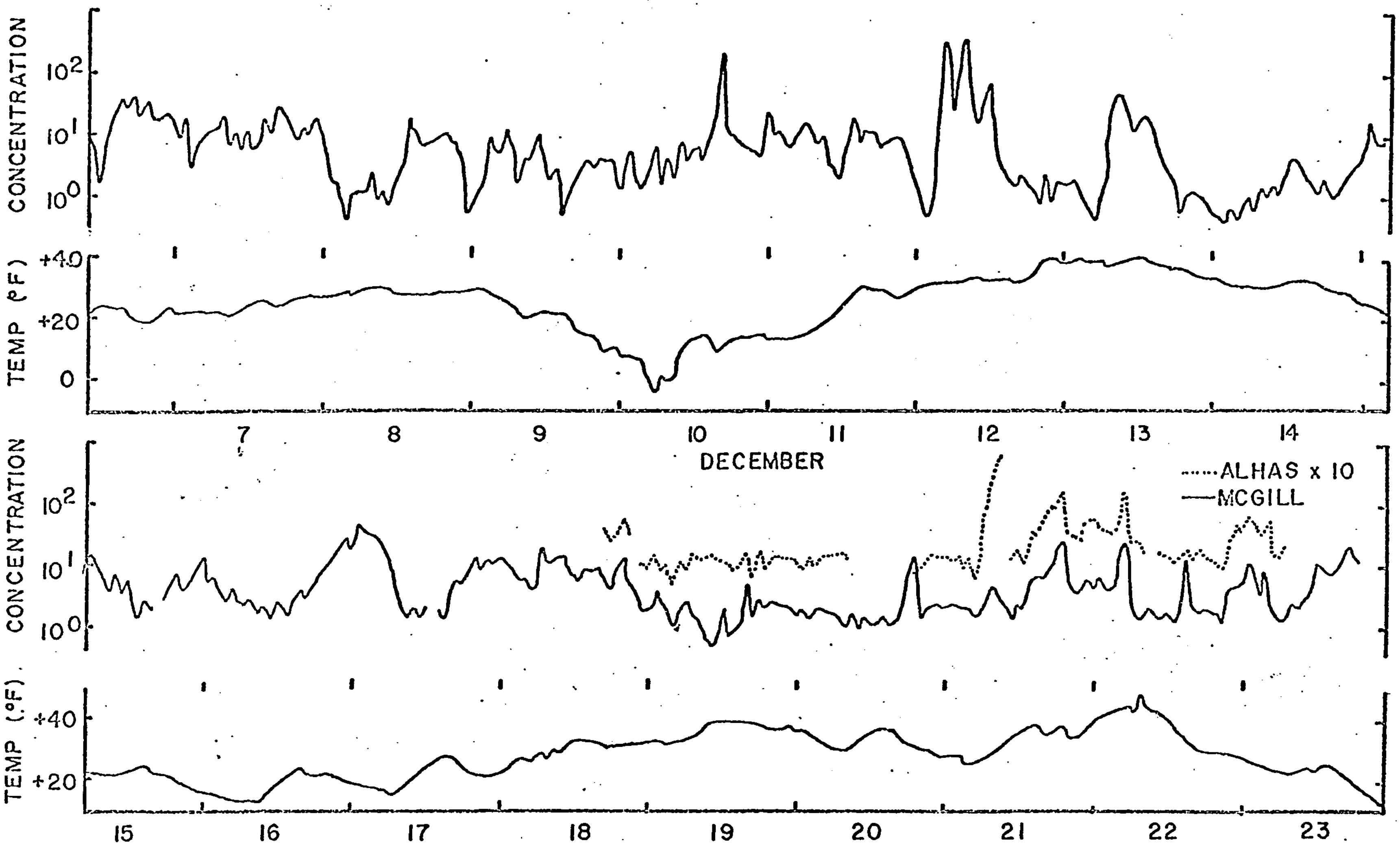


Figure 16., Part 1. Outside air temperature at Montreal International Airport and ice nucleus concentrations at -21C at MacDonald College from December 7 to 23, 1967.

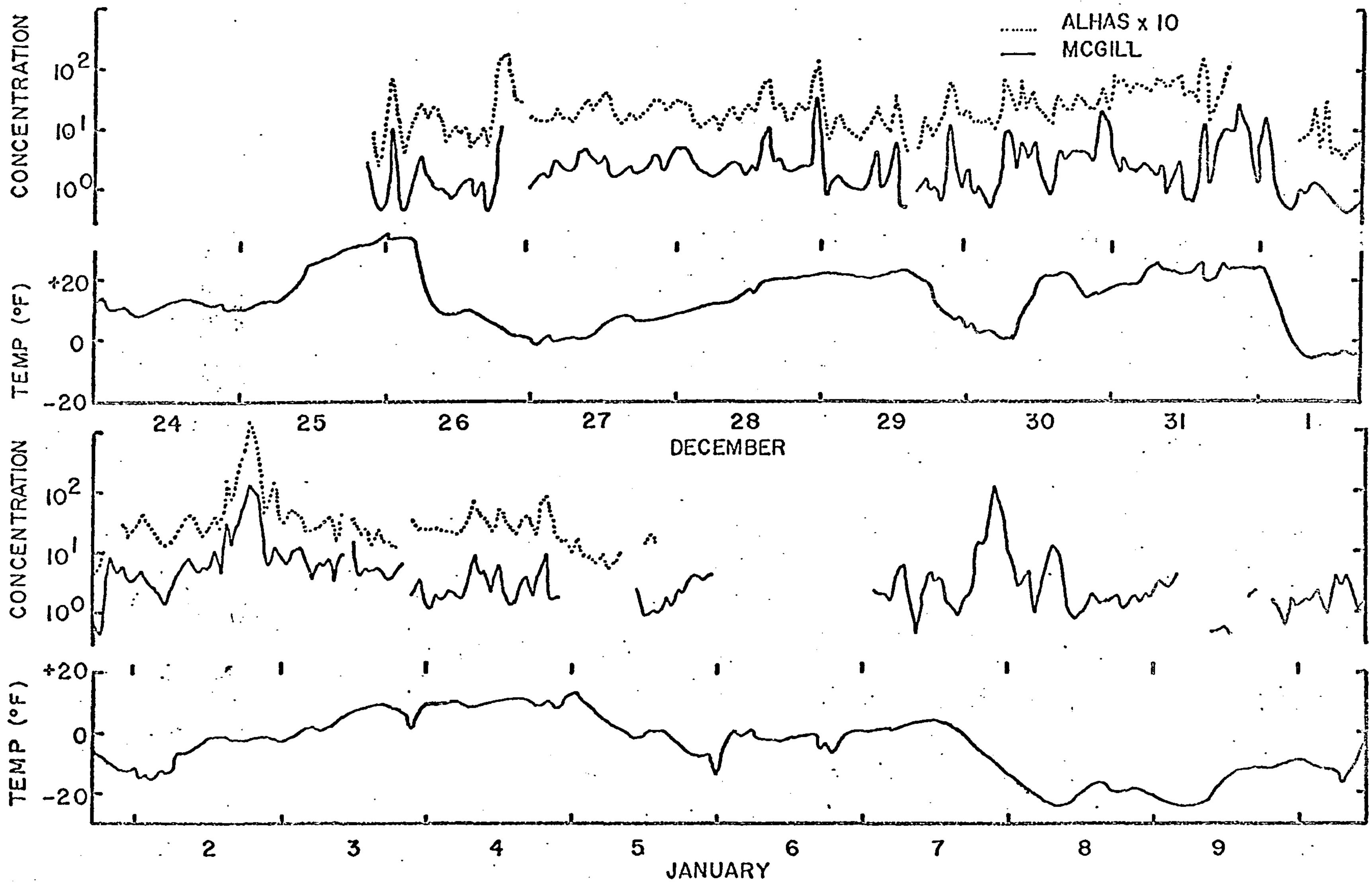


Figure 16., Part 2. Outside air temperature at Montreal International Airport and ice nucleus concentrations at -21C at MacDonald College from December 24, 1967 to January 10, 1968.

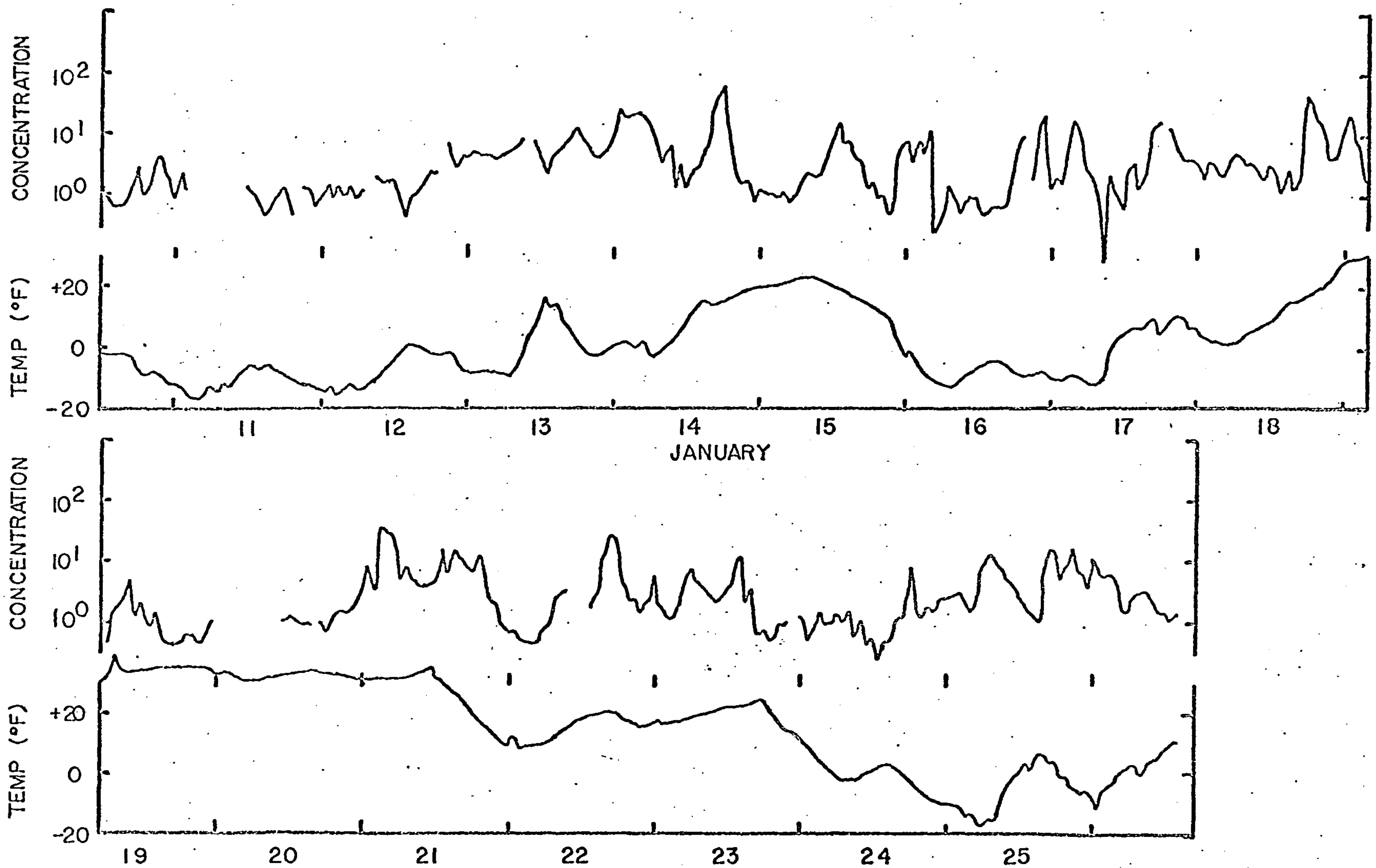


Figure 16., Part 3. Outside air temperature at Montreal International Airport and ice nucleus concentrations at -21C at MacDonald College from January 10 to 26, 1968.

TABLE 3

DAILY WEATHER SUMMARY FOR DECEMBER 1967 AT DORVAL, QUEBEC

1.	Sunny
2.	Sunny with cloudy periods
3.	Cloudy with snow. Clearing in the evening
4.	Cloudy with light snow in the afternoon.
5.	Cloudy. Clearing in the afternoon.
6.	Sunny. Gradually clouding in the afternoon.
7.	Cloudy. Snow end of the day.
8.	Cloudy. Snow and freezing precipitation most of the day.
9.	Cloudy in the morning, sunny rest of the day.
10.	Sunny. Fog early in the morning.
11.	Cloudy. Snow and freezing precipitation in the afternoon and in the evening.
12.	Cloudy. Freezing precipitation during the night. Rain the rest of the day.
13.	Cloudy with sunny periods.
14.	Cloudy with sunny periods. Snow late in the evening.
15.	Cloudy with sunny periods. Snow early evening.
16.	Sunny.
17.	Sunny with cloudy periods.
18.	Cloudy. Freezing precipitation early in the morning and late afternoon and evening. Thunder was reported in the city at 2100 E.S.T.
19.	Cloudy. Freezing precipitaton during the night.
20.	Sunny.
21.	Sunny in the morning. Cloudy rest of the day. Rain in the evening.
22.	Cloudy. Rain early in the morning.
23.	Sunny with cloudy periods.
24.	Sunny with cloudy periods.
25.	Cloudy. Snow in the afternoon and evening.
26.	Cloudy early morning. Rain and snow. Sunny in the afternoon. Clearing in the evening.
27.	Sunny in the morning. Cloudy in the afternoon and evening. Snow in the evening.
28.	Cloudy. Snow most of the day.
29.	Cloudy, clearing late evening. Snow in the morning.
30.	Sunny in the morning. Cloudy test of day. Snow in the afternoon and evening.
31.	Cloudy. Snow most of the day.

TABLE 4

DAILY WEATHER SUMMARY FOR JANUARY 1968 AT DORVAL, QUEBEC

1.	Cloudy early morning with snow. Sunny rest of the day.
2.	Cloudy. Light snow in the afternoon and evening.
3.	Cloudy. Light snow all day.
4.	Cloudy. Snow all day.
5.	Sunny and cold.
6.	Cloudy. Cold. Light snow in the morning.
7.	Cloudy in the morning with snow. Sunny in the afternoon and very cold in the evening.
8.	Sunny and very cold.
9.	Sunny in the morning. Cloudy rest of day.
10.	Sunny all day.
11.	Sunny all day.
12.	Sunny all day.
13.	Sunny all day.
14.	Sunny in the morning. Cloudy rest of the day. Freezing precipitation and snow in the evening.
15.	Cloudy with snow all day.
16.	Sunny all day.
17.	Sunny in the morning. Cloudy rest of the day.
18.	Sunny early morning. Cloudy with light snow rest of the day. Freezing precipitatation in the evening.
19.	Cloudy. Rain mix with snow in the morning and early afternoon.
20.	Cloudy all day. Snow shower in the morning.
21.	Cloudy with snow showers in the morning, clearing in the afternoon.
22.	Sunny in the early morning. Cloudy rest of the day. Light snow late evening.
23.	Cloudy all day with snow.
24.	Cloudy early morning with snow. Sunny rest of the day.
25.	Sunny all day. Ice crystals early morning.
26.	Cloudy all day. Light snow in the morning.
27.	Mostly cloudy all day.
28.	Cloudy with snow in the evening.
29.	Cloudy. Freezing precipitation and snow most of the day.
30.	Cloudy all day. Rain in the morning.
31.	Cloudy periods.

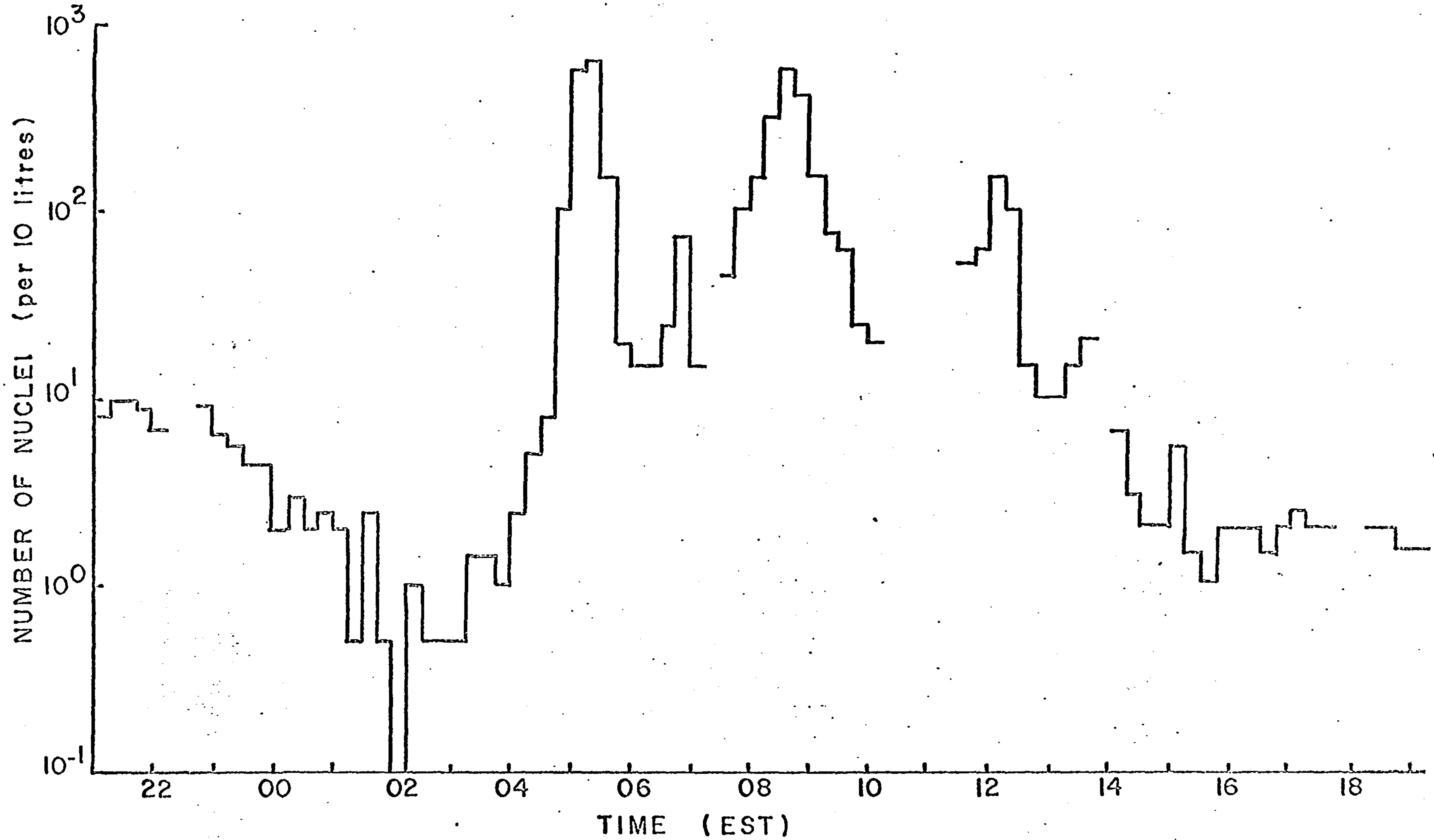


Figure 17. Ice nucleus concentrations at -21C from December 11, 2100 to December 12, 1900 EST. The gap near 1100 was due to a power failure while the other breaks are due to standard instrumental checks.

representation for that day. This is extremely important when considering the conclusions of authors who assumed good agreement and made significant conclusions about Bowen's meteor peaks in January. Concentrations of nuclei can vary up to three orders of magnitude during a day. Such a jump occurred on December 12 from 0200 to 0500 EST and is shown in detail in figure 17. On December 12 freezing rain began at the Montreal Airport at 2140, changed to light rain at 0500 and at least drizzle persisted until 1810. This station's pressure fell until 1400 and then started to rise. A wind shift simultaneously occurred with the passage of this trough. Only a station network would enable one to determine the primary cause of the jump. That is, was it a local effect due perhaps to the presence of freezing rain or was it due to a wind shift or another feature of the large scale circulation? Precipitation samples from the 12th were analysed by Bishop (1968).

No significant increase in concentration occurred when the outside air temperature was below 0C (figure 16) making it unlikely that pre-activated nuclei were abundant at those temperatures. This agrees with Bigg and Hopwood's (1963) measurements taken at McMurdo Sound in the Antartic. Increasing the dewpoint temperature (usually within 10F of the ambient temperature) does not have a significant effect on the concentration. An increased dewpoint might have changed the concentration by thickening the chamber's cloud, thus allowing more contact-freezing and freezing nuclei to coagulate with the cloud droplets. A visual and subjective evaluation of the cloud density did not show any variations due to outside conditions. The humidifier appears quite efficient from this evidence.

The influence of snow on the ice nucleus counts at -21C was not pronounced and consequently was not studied in detail. For example, according to the Montreal Airport record, almost continuous light snow fell from December 27 1700 to December 29 0800, and January 3 0100 to January 4 2200, and January 22 2200 to January 24 0500 without any great effect. A distinct snow shower occurred from 1120 to 1140 January 21 at MacDonald reducing visibility to less than 1000 feet, and increasing counts by a factor 5 for 4 minutes but otherwise had no immediate effect. The concentration gradually increased afterwards. Snow does not appear to have a significant influence when compared with the other large flucuations observed.

Figure 18 shows the frequency distribution of the first 24 possible averages for the period December 6 to January 26. One of the difficulties of forming figure 11 has been removed since all these averages were over 15 minute periods. Although the class interval changes for the values of 10., 15., 20., and 25. their percentages of the total number of readings have simply been lowered by a factor 10. This insures that the area under the bar is proportional to the percentage of the total number. Just 3.2 per cent of the readings have not been plotted due to a lack of space but they influence the mean of 6.3 nuclei per 10 litres enormously. As in figure 11, the mean is displaced from the histogram peak indicating a non-Poisson distribution. Bigg and Miles (1964) and Rosinski (1967) have observed similar patterns. The displacement of the mean shows that nucleus sources and sinks exist in the atmosphere.

The only available long term measurements taken with an NCAR-Bollay Counter at another location were those of Nagamoto et al (1967). Although the means are not explicitly given, it appears that the average from January 12 to March 10, 1967, uncorrected for calibration, would be 0.3 to 0.6 per 10 litres at -21C for their mountain Mona Loa station in Hawaii. These readings are much lower than the ones obtained at MacDonald but this could simply be due to their low humidifier water temperature of 28C.

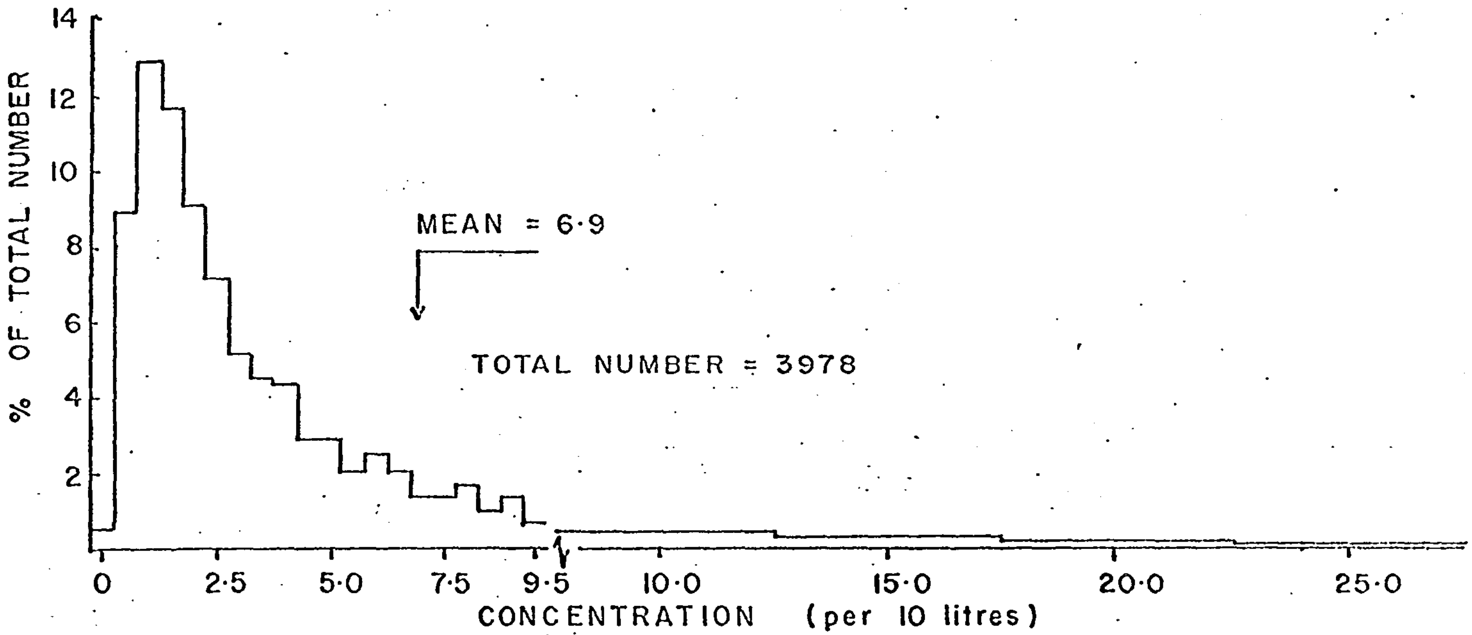


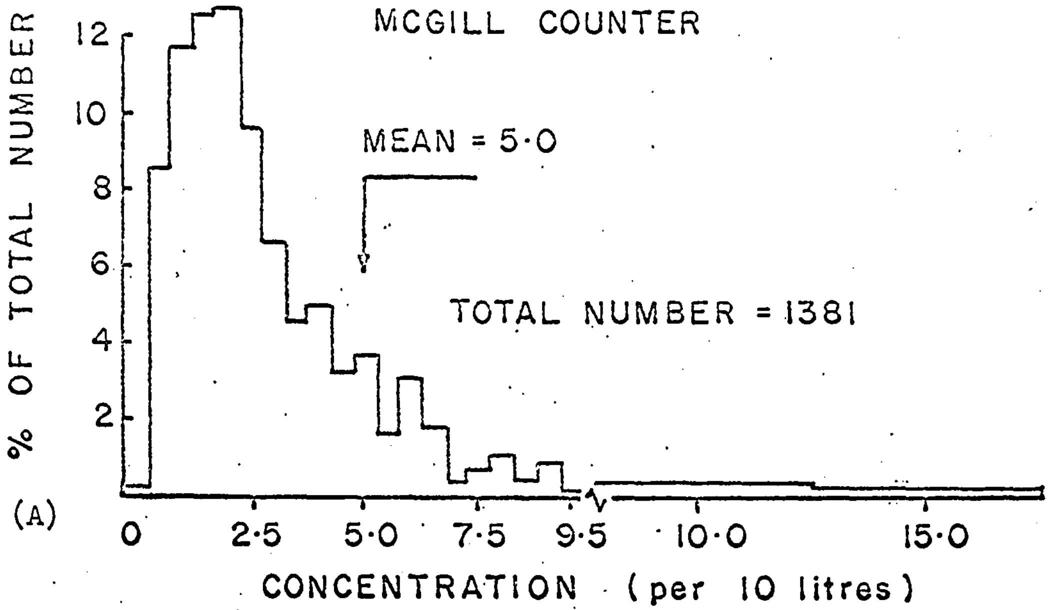
Figure 18. Histogram of 15 minute averages of ice nucleus concentrations at -21C from December 6, 1967 to January 26, 1968. The break between 9.5 and 10. indicates a change in class interval. (9.5 represents 9.25-9.75, while 10 represents 7.5-12.5 as determined on the Rate Computer's second scale) Percentages after 9.5 have been lowered by a factor 10 to keep the area proportional to the per cent of the total number. 3.2% of the total number of averages are larger than 25.

6.3 Comparison of the McGill and ALHAS Counters

Figure 16 shows the values obtained by the ALHAS machine as well as the McGill one over the comparison period of December 18 to January 5. Although poor agreement in absolute concentration is evident, the two patterns are comparable except for a few isolated peaks. Histograms (figures 19a and 19b) using all the readings of the McGill and ALHAS Counters in this period have the characteristic shape with means of 5.0 and 4.0 nuclei per 10 litres respectively. These two counters were run together for 1127 15 minute periods with a mean ratio as referred to the McGill instrument of 0.9.

Grant et al (1967) observed various flow patterns within the NCAR-Bollay chamber. The two dominant flow patterns produced concentrations differing by a factor 20 with the lower concentration mode existing 85 per cent of the time. Because of a lack of other types of ice nucleus counters at MacDonald, this effect could not be directly verified. However only 8 of the 1127 concurrent 15 minute means, obtained with the ALHAS and McGill Counters, differed by a factor 10 or higher. If changes in the chamber flow regime are induced internally, one would expect a larger number of widely differing values since there would not be any reason why simultaneous changes would occur in both instruments. If the changes are induced externally, the effect could not be observed with two identical counters. Because it was impossible to detect the chamber's flow pattern using only one observation window (see figure 1), concentrations are assumed to be independent of this variable.

To show how the two instruments compared over a major nucleus peak (January 2) and over a constant period (December 27), figures 20 and 21 were drawn using the original 15 minute extracted values. Remembering the limited range of values allowed in the extractions (see section 6.2), the two machines compare favourably in pattern if not in absolute concentration.



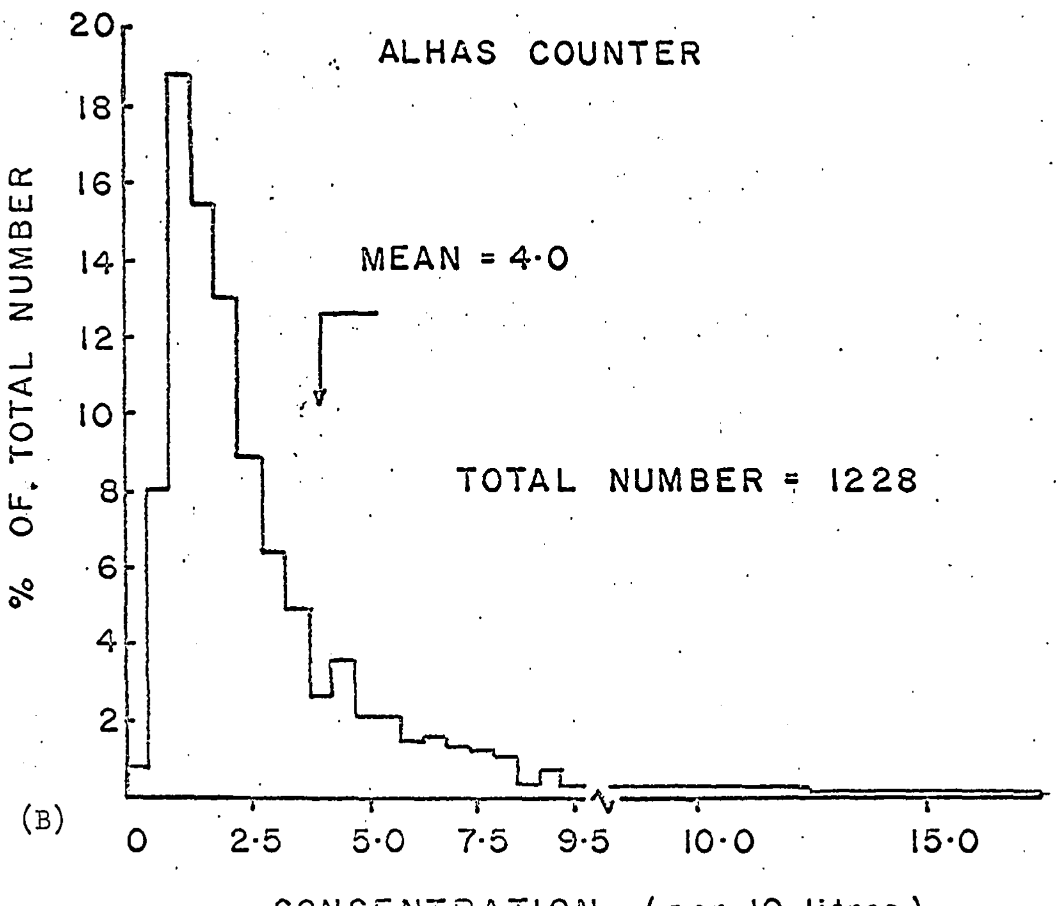


Figure 19. Histograms of readings from December 18, 1967 to January 5, 1968. See figure 18 for diagram format. A) McGill Counter, 2.5% of total number greater than 15. B) ALHAS Counter, 2.1% of total number greater than 15.

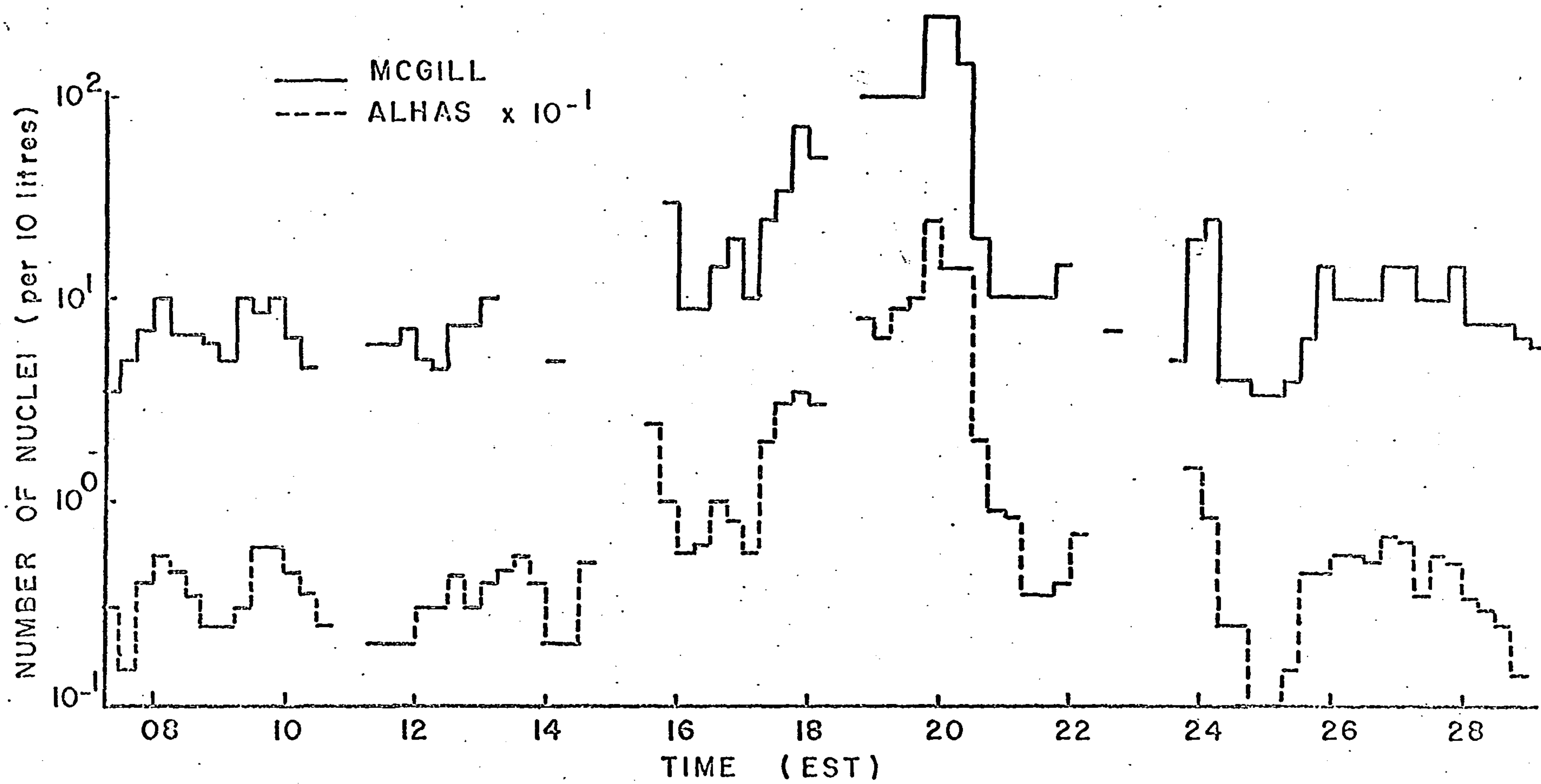


Figure 20. Ice nucleus concentrations measured by the ALHAS and McGill Counters at -21C for January 2, 1968. Both instruments used the downstairs air inlet tube.

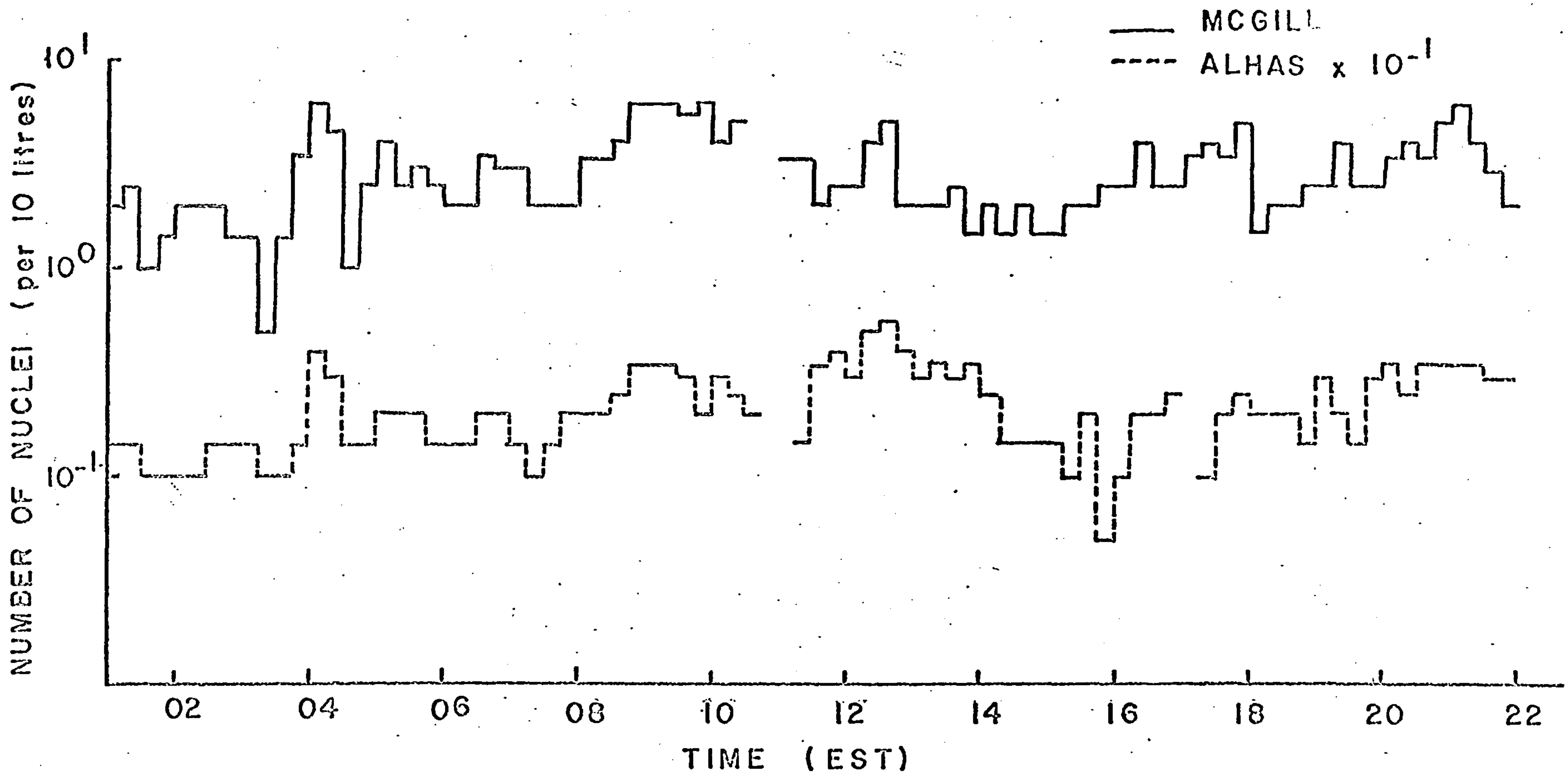


Figure 21. Ice nucleus concentrations measured by the ALHAS and McGill Counters at -21C for December 27, 1967. Both instruments used the downstairs air inlet.

Possible reasons for the differences include some not related to poor instrumental design. Because of a lack of acoustical sensors, McGill's machine was run with a sensor requiring a pressure setting of 150mm for a 13 litre per minute flow rate, while the ALHAS instrument used a 120mm sensor. Since the Rate Computer settings were identical, this may have induced a different count between the machines. Also the sample flow rate could have changed slightly because of insufficient control. However, no consistent ratio between the machines was observed with the daily average varying from 1.7 to 0.5 as referred to McGill's instrument.

On January 6 the ALHAS Rate Computer broke down making the Counter useless. Consequently the ALHAS chamber was moved upstairs to a northeast facing window. Periodically when the concentration appeared steady this Counter was operated with the McGill Rate Computer to determine if any error was caused in the readings due to the McGill's instrumental air intake or its immediate surroundings. Consecutive 15 minute averages downstairs and upstairs are compared in figure 22. Most of the points are within a factor 3 of each other and considering the natural fluctuation in consecutive 15 averages at one station (figure 21), the agreement is excellent. The main sampling inlet used for the McGill instrument is thus representative of the air around the building.

6.4 Winter Spectra

On 6 occasions when comparisons were made on the upstairs ALHAS Counter, a spectrum was run (figure 23). Spectra were not run on the McGill Counter downstairs since readjustment to the original operating temperature would have been difficult. The method of the spectrum calculation was different than the summer technique since time away from the McGill machine had to be minimized to obtain a reasonable continuous trace. Cooling was done in steps but no readings were used if the rate exceeded 0.25 degrees C per minute. During January it was found that concentrations tended to increase faster as the temperature was lowered and consequently the best curve was fitted by eye.

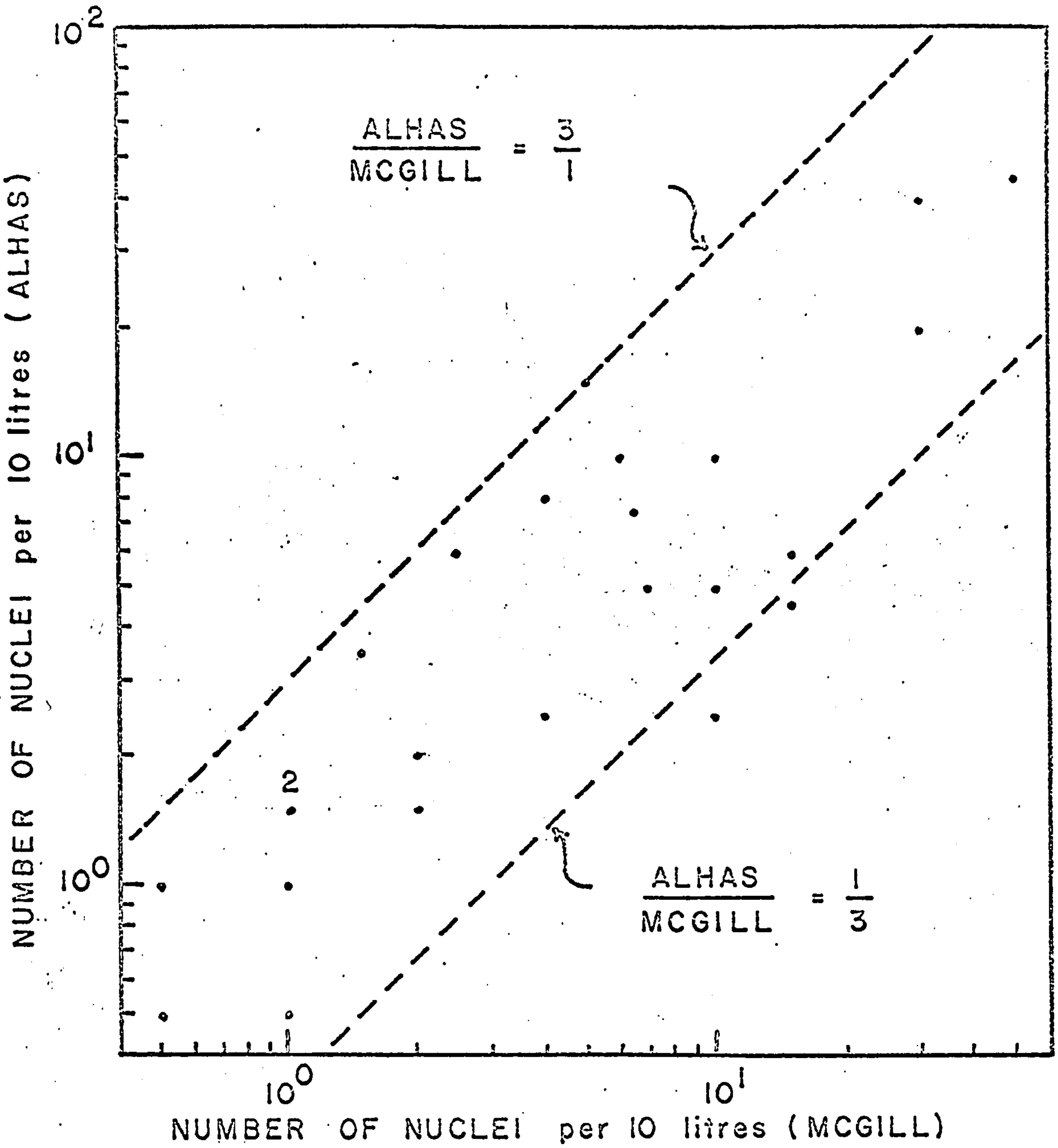


Figure 22. A comparison of the downstairs (McGill) air inlet and the upstairs air inlet (ALHAS). Each point represents 15 minute averages determined consecutively at each location. Diagram indicates downstairs air inlet is representative of the air around the building.

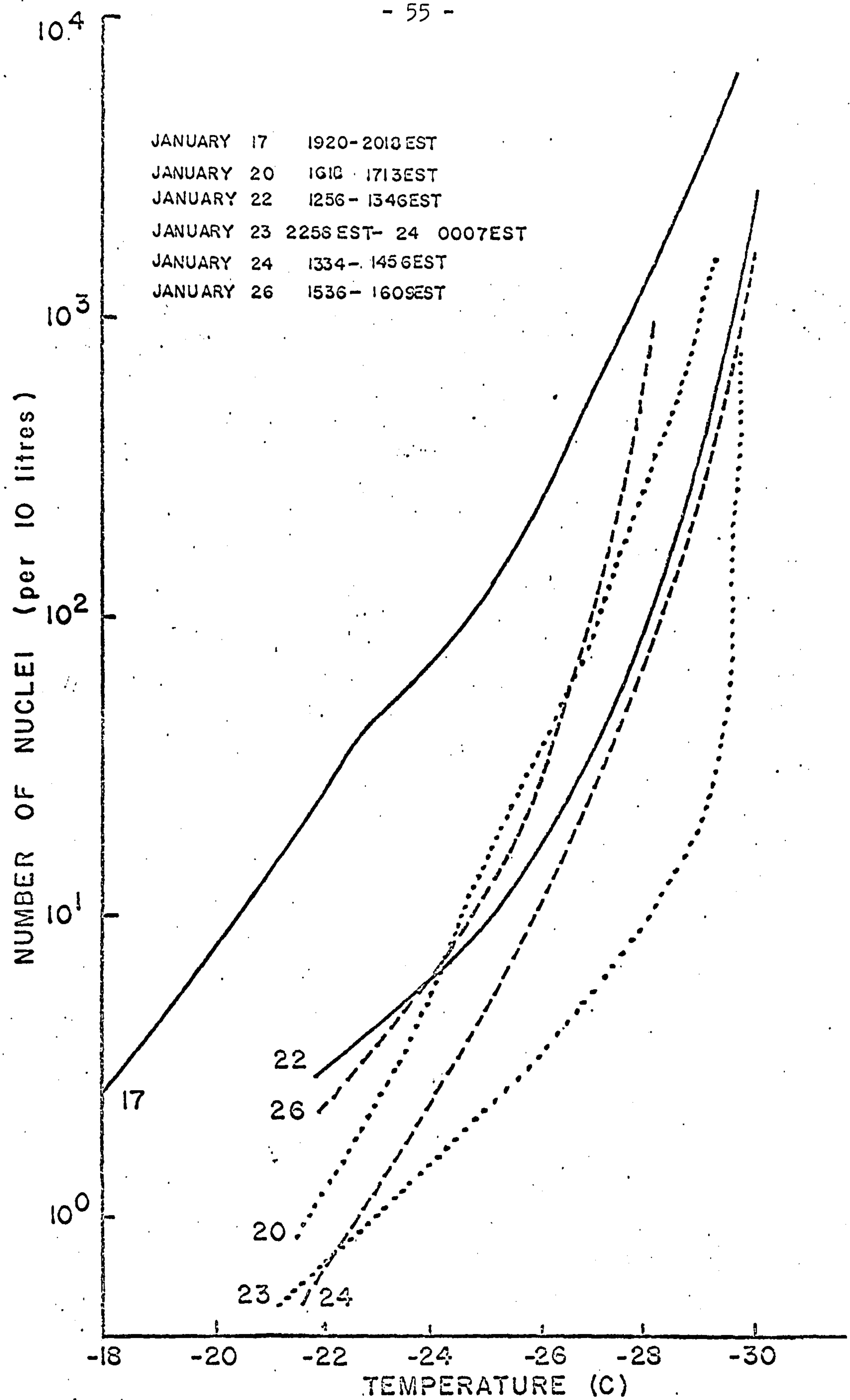


Figure 23. Winter Spectra.

On January 23 a spectrum taken during a snowfall shows a remarkable change in slope probably induced by scavenging of nuclei effective from -22C to -28C. Lower concentrations after and during snowfalls might then have appeared in figure 16 if the operating temperature had been decreased.

6.5 Winter Daily Cycle

Figure 24f shows the average daily cycle, composed of 15 minute means, for the complete winter period while figures 24a-e indicate patterns for portions. Sections of the winter period have been plotted because large concentrations dominate the mean patterns giving a distorted picture. Subdividing helps to clarify the significance of average values. A tendency for peaks to occur in the late afternoon or early evening as well as in the morning hours is shown in figure 24f. However between 0900 to 1600 only one peak occurs (figure 24a). A minimum during this time exists on December 6, 16, 17, 20, and January 10, 11, 12, 13, 16, 24, and 25 (see figure 16) which is most pronounced on sunny days when heating is significant. The afternoon temperature rises, the dewpoint rises with relative humidity falling and the count simultaneously falls. Convection mixing the nucleus ladden lower layer with air aloft, or another effect such as the relative humidity changing the nucleus surface properties, might be the cause.

Georgii (1959) and Bigg (1961) verify the forenoon maximum and the afternoon minimum described here. Bigg mentions that a peak in concentration can occur before sunrise due to an inversion formation trapping the nuclei below, or after sunrise due to an inversion breakup bringing in the nucleus ladden air from aloft. Both cases appear here. Nagamoto et al (1967), using an NCAR Counter at Mona Loa Observatory in Hawaii (considered above the inversion), found that a peak occurred in the afternoon with no morning and evening peaks visible. So far a reasonable explanation for the evening peak has not been found.

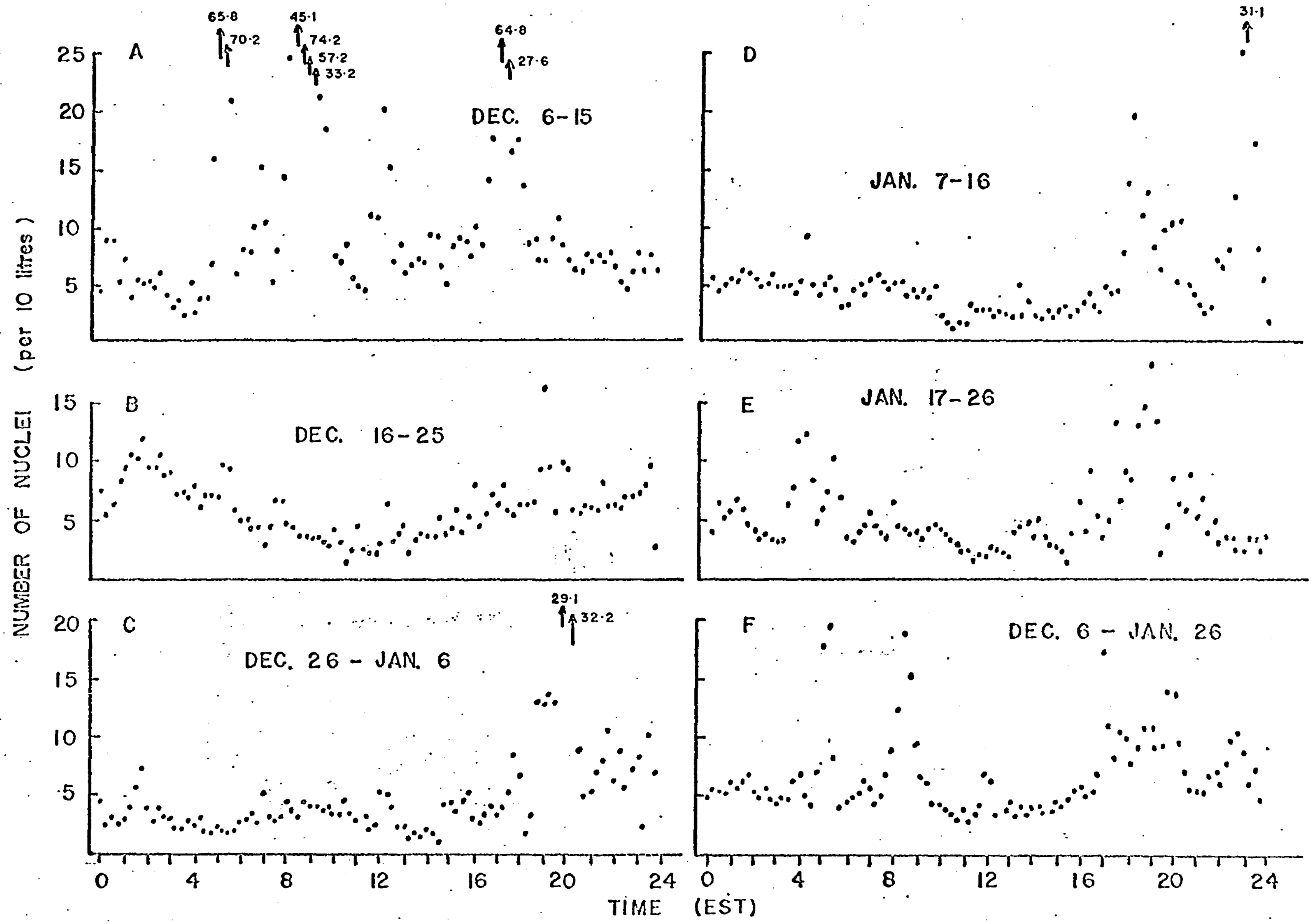


Figure 24. Winter daily cycle in ice nucleus concentration at -21C. A to E are formed from 15 minute averages for portions of the winter continuous run while F is the total mean for December 6, 1967 to January 26, 1968.

6.6 Correlation with Wind

Hourly values of surface wind direction and wind speed obtained from Montreal International Airport's records were compared with hourly means in ice nucleus concentration at MacDonald College from December 6, 1967 to January 26, 1968. A similar study was done from December 18 to January 5 and the results were the same.

Figure 25 examines how the concentration varied with wind direction. The number of hours each direction was recorded and average wind speeds (underlined) have been plotted beside each point. The predominance of northeasterly winds is probably helped by the orientation of the St. Lawrence Valley. Highest concentrations occurred with easterly and northeasterly winds, while westerly winds are associated with a relative minimum. Since the city is to the east, possibly industrial pollutants increased the concentration significantly when prevailing winds blew from this direction. This conclusion could be tested by running a comparable counter downtown while also recording values at MacDonald College. Unfortunately this was not done. Because the air inlet was located on the southwest side of the building (see map 1), dust might have been blown off the roof by easterly or northeasterly winds and thus increased the number of nuclei counted. However north's relative minimum would conflict with such a conclusion since this wind also sweeps across the roof. The possibility exists that the observed pattern is not caused by a local condition but is due to large scale atmospheric circulations.

Table 5 relates the concentration to wind speed. Strong winds do not appear to pick up nuclei and thus intensify the concentration. Similarly light winds do not show an increase that could be caused by trapping ground originating nuclei in the lower layer.

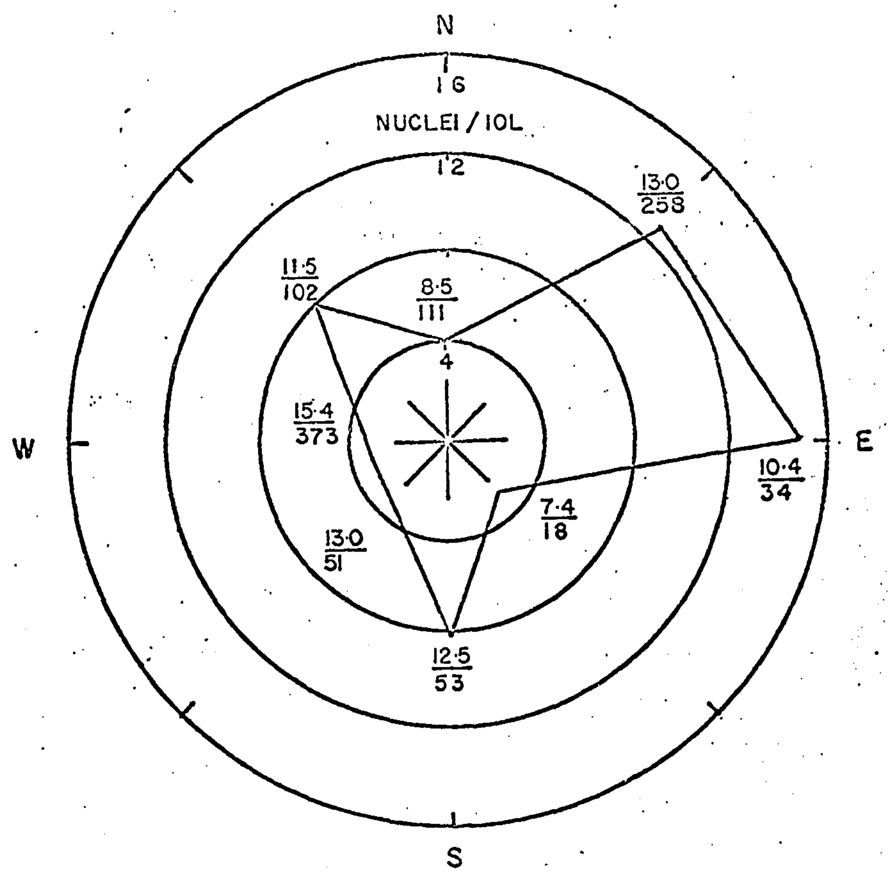


Figure 25. Concentration of atmospheric ice nuclei in relation to wind direction. Average Wind Speed (mph) / No. of hours used, is plotted beside each direction.

Interval (4mph)	Average Wind Speed (mph)	No. Hours in Average	Average Conc. (per 10L)
Calm	0.	46	5.9
1-4	2.9	99	6.4
5-8	6.6	176	4.7
9-12	10.6	245	5.9
13-16	14.4	233	9.5
17-20	18.3	163	9.4
21-24	22.5	63	3.2
25-28	26.1	42	6.3
29-32	30.5	13	3.5
33-36	34.4	9	3.1

Table 5. Concentration of ice nuclei in relation to wind speed

6.7 Extraterrestrial Influences

Many attempts have been made to prove the Findeisen-Bowen meteor hypothesis, summarized by Fletcher (1962), but collectively they have been unsuccessful because many contradictions arise between different studies. The best possible explanation for such a failure would be the tendency to take several measurements a day and call those representative. Two recent papers by Nagamoto et al (1967) in Hawaii, and Maruyama and Kitagawa (1967) in Japan eliminated this approximation and still produced conflicting results. Maruyama and Kitagawa, using 30 minute exposed filters and several observing sites, found that an increase in concentration occurred about 28 days after each shower. Nagamoto et al, using an NCAR-Bollay Counter, were not successful in finding a correlation between their counts and Bowen's rainfall singularities.

Figure 26 compares Bowen's rainfall pattern (Fletcher, 1962) with the daily mean in ice nucleus concentration found at MacDonald College. Unfortunately daily precipitation amounts for December and January were not available. The peak on January 2 (see figures 16 and 20), coinciding with a rainfall maximum, supports the meteor hypothesis. However, although some similarity between figures 26b and 26c exists, it is not striking. Until estimates of the possible increase due to meteors, its fluctuation properties and duration are known, little can be said about the meteor hypothesis. It is very easy to locate a peak in concentration if one allows a 2 day fluctuation in the time of a maximum as most authors do.

There exists some evidence that the moon exerts an influence on the ice nucleus concentration and rainfall amount. Brier and Bradley (1964) showed that a lunar cycle in rainfall occurred in North America and was homogeneous in phase across the continent. This indirect evidence requires the assumption that ice nucleus concentrations influence the rainfall amount. However, Bigg and Miles (1964) showed that a strong lunar cycle in nucleus counts very similar to that in rainfall exists in Australia with a maximum concentration twice the minimum. Yang (1966) found in a similar study that his data for Seoul was inconclusive. The moon's phases are marked on figure 26 but Brier and Bradley's cycle of 14.76 days, one half the lunar synodic month with maximums just before the first and last quarter, is not apparent. A more detailed study confirmed this observation. However it does not contradict the above authors' long term studies but merely indicates that such experiments are needed since many variables affect the ice nucleus concentration.

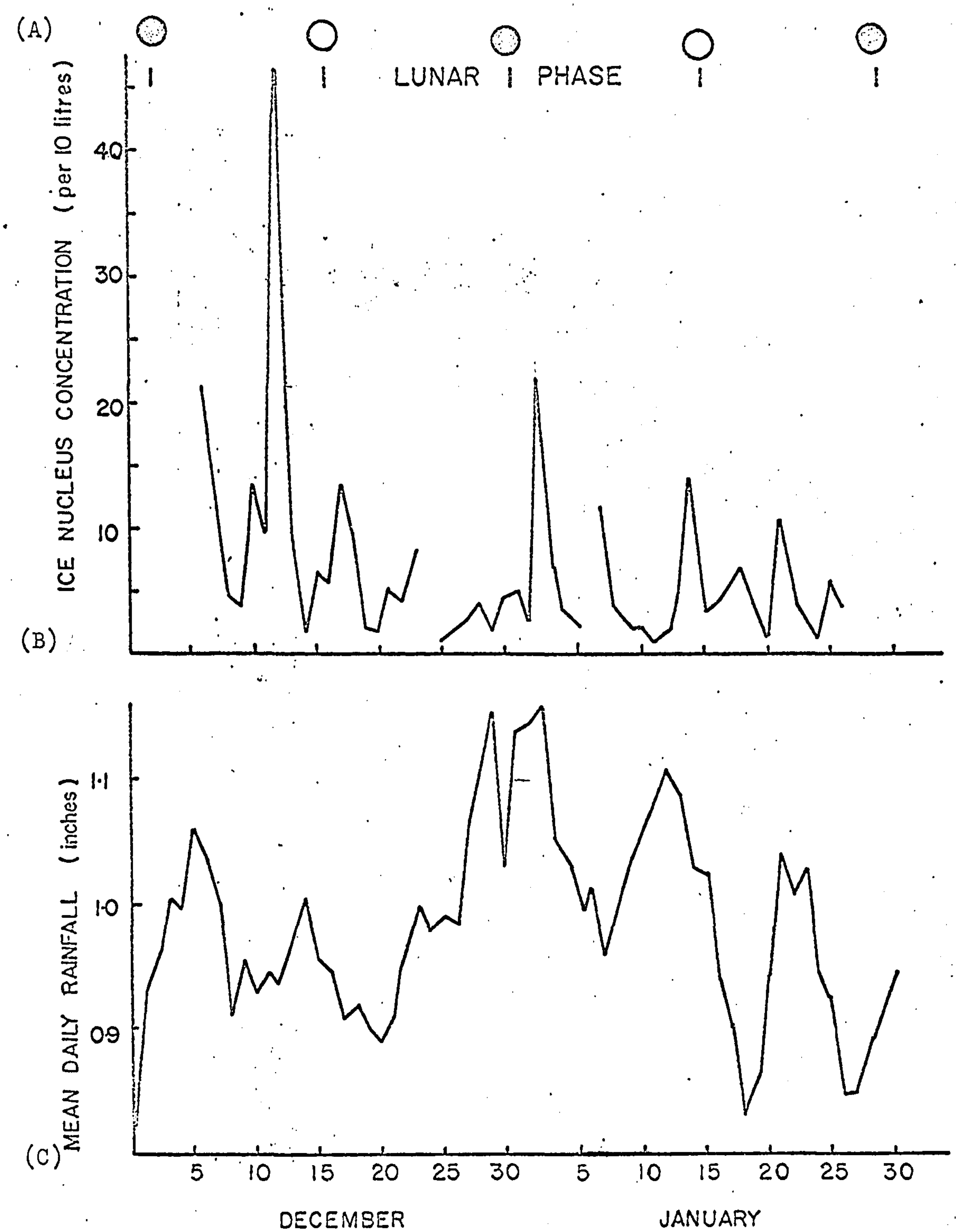


Figure 26. Extraterrestrial influences. A) Lunar phase. B) Daily averages in ice nucleus concentration at -21C. C) Bowen's daily rainfall values. (After Fletcher, 1962)

CHAPTER 7

CONCLUSIONS

The NCAR-Bollay Acoustical Ice Nucleus Counter's main asset is that it can operate continuously. Since calibration with excellent equipment indicates it reads a factor 3 low, and experiments described here show that two machines under identical operating conditions can differ considerably for short periods, the Counter's ice nucleus concentrations must be discussed with care. Variations in the counts can be produced by just the humidifier water temperature making direct comparisons with other authors' long term averages hazardous. Fortunately ice nucleus concentration patterns are easily reproduced. Some features of the ice nucleus concentration at MacDonald College are listed below with comments on possible methods for solving particular problems.

Although the summer mean concentration of 14.7 nuclei per 10 litres at -21C is higher that the winter mean of 6.9, it is not significantly greater if the differences in obtaining these values are considered. Schaefer (1954) found that from 1948 to 1953 at Mt. Washington, the winter counts were lower than those in summer.

Frequency distributions of counter readings are required for a complete understanding of average concentrations, since the histogram mean differs considerably from the modal concentration.

Ice nucleus spectra have a characteristic slope enabling estimates of the concentration at any temperature to be obtained as long as one point on the curve is known. It is interesting that the ice nucleus spectra in air and the freezing nucleus spectra in precipitation have the same slope.

Fluctuations in the nucleus counts during continuous rain should be studied to determine if the cause is an air mass change or a difference in the air's composition associated with the presence of rain. This problem could be studied best with a station network.

The concentration of ice nuclei appears to increase considerably in the downdraughts of convective storms.

No large concentration differences, indicating a washout mechanism, were observed at -21C after extended snowfalls. However this effect could possibly be observed at lower chamber temperatures.

Accurately determining the relation between the number of ice nuclei and precipitation amounts can only be done with precise cloud top temperatures. This data would allow estimations of effective concentrations. Ground stations are not desirable since they must be assumed representative of cloud air.

The daily cycle in winter concentrations at MacDonald College clearly indicates an afternoon minimum with a tendency for peaks in the early morning and early evening.

Winter winds from the northeast and east bring higher numbers of ice nuclei than those from the west. If pollution from Montreal is the cause for this peak then counters run simultaneously at MacDonald and downtown Montreal could easily verify this. However it is possible that large scale atmospheric circulations could directly explain the observed pattern. For example, when northeast winds are present, a low pressure system very often is located to the east.

Strong winter winds do not increase the concentration suggesting that a very local ground dust source does not exist.

The winter data provide little evidence to support theories which claim extraterrestrial influences. However, these theories require long term measurements since they probably do not directly explain brief ice nucleus peaks as shown in figure 16.

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