Vol.6 No.2 Advances in Atmospheric Sciences May 1989

A Study of Supersaturation Spectra of Deposition Ice Nuclei(1)

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Received June 25, 1988

ABSTRACT

Ice nuclei–supersaturation spectra in the form of a Power Law ($N_i = RS$; B and γ are empirical constants) have been expressed since 1973 when the curve was first introduced independentally by Gagin and Huffman. Experiments performed with a thermal gradient diffusion chamber in order to investigate the validity of the power curve. The results show that a linear curve fit to the data is as good as the power curve. The linear curve has the coefficients of correlation between 0.75 and 0.93 whereas the power curve fit to the same data has the coefficients between 0.82 and 0.93. The data reported by other workers, Zamurs and Jiusto and Zamurs et al., exhibit the same trend.

I. INTRODUCTION

It is well–established now that a significant fraction of dust particles suspended in the troposphere behaves as nuclei for ice formation in natural cold clouds. These nuclei are known as ice nuclei (IN) and are considered to be operative in clouds in three modes, namely, deposition, freezing and contact modes. Recently a new mode of nucleation called condensation–freezing mode was found active in certain cold clouds (Cooper and Saunders, 1980) and was tested in the laboratory for its efficiency (Hussain et al., 1984; Al–Naimi et al.,1985) .

Over the past many years, several equipments, called ice nucleus chambers / counters were designed and constructed for the measurement of concentration of natural ice nuclei. The purpose of such studies was mainly to understand the microphysics and precipitation mechanisms of cold clouds. The major IN chambers / counters are mixing chambers, expansion chambers, drop–freezing counters and thermal–gradient static and flow chambers (Hussain et al., 1984; Bigg, 1957; Bigg et al., 1963; Stevenson, 1968; Gagin, 1973; Huffman, 1973; Ohtake, 1976; Zamurs et al.,1977). These devices were designed to simulate cloudy conditions suitable for activation of ice nuclei. The temperature and supersaturation in expansion and mixing chambers could not be controlled accurately because of their large size. These parameters were controlled with much better accuracy in the subsequently designed thermal–gradient chambers. It was possible to achieve temperature and supersaturation in these chambers over the range found in natural cold clouds.

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(1) A part of the work presented in this paper was carried out by one of the authors (K. Hussain) at University of Manchester Institute of Science and Technology, P.O. Box 80, Manchester M60 1QD (ENGLAND).

On the basis of the data obtained from the mixing and expansion chambers, Fletcher (1962) introduced the following empirical relation

$$N_i = Ae^{(\beta \Delta T)},$$

where β and A are empirical constants having respectively the dimensions of 8^{-1} and l^{-1}. N_i is the concentration of IN per litre active at a temperature T warmer than T_0, and $\Delta T = T_0 - T$. T_0 is the temperature at which 1 IN per litre is active. The curve is based on the data obtained at water saturation. Experimental data gathered from all over the world by running an old version of thermal–gradient chamber (Bigg and Stevenson, 1970) were also found to obey, to some extent, the N_i versus T empirical relationship. Figure 1 shows some of the data and the error of measurement by vertical bars. It was reported that the IN were active in the freezing mode. Subsequent studies, however, showed that the IN activated by thermal–gradient chambers were deposition nuclei and not freezing nuclei (Hussain, et al., 1984; Huffman, 1973; Zamurs, 1977).

IN data obtained over a range of temperature and supersaturation with thermal–gradient chambers were showed to observe a power curve of the form

$$N_i = BS_i^{\gamma},$$

where S_i is percent supersaturation with respect to a plane surface of ice. The empirical constant B has dimensions of l^{-1} and the other constant γ is dimensionless. The equation is valid at a temperature at which IN get activated.

The IN data obtained from the subsequent experiments with new versions of the thermal–gradient chambers (Hussain et al., 1984; Zamurs et al., 1977) obeyed satisfactorily the empirical power curve. The mode of nucleation in all these experiments was found to be the deposition mode. The relationship therefore represents supersaturation spectra for deposition ice nuclei.

Experiments were done to investigate the nature of the supersaturation spectra for ice nuclei. A thermal–gradient chamber was employed for the purpose and the results of the study are reported in this paper. We find that a linear curve fits to the data as good as the power curve.

II. EXPERIMENT AND RESULTS

In order to obtain IN–supersaturation spectra for the ice nuclei active in the deposition mode, a static diffusion thermal–gradient chamber was employed. The device has been described in detail elsewhere (Hussain et al., 1984; Hussain et al., 1988). A brief account of the chamber is given here.

The static diffusion chamber (SDC) is similar in principle to the chambers reported earlier (Huffman, 1973; Zamurs et al., 1977). It is a metallic, rectangular box which consists of two horizontal aluminium plates separated by a vertical wall of adjustable height. The bottom plate is 1.6 cm thick with the dimensions of 14 × 14 × 0.6 cm on one side and 18 × 18 × 1 cm on

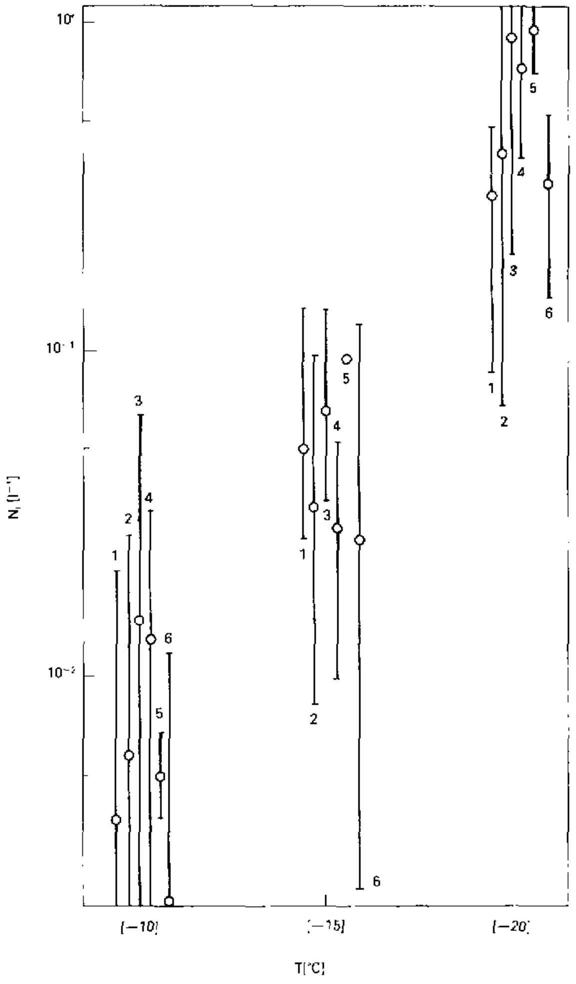

Fig.1. Ice nucleus versus temperature spectra at water saturation. (After Bigg and Stevenson, 1970).1: Europe; 2: North America; 3: Asia; 4: Africa; 5: South America; 6:Australia.

the other side. This plate has four recesses into which fit four brass discs each of 4.7 cm diameter. In the running mode, the brass discs carry filters which are then loaded in the bottom plate recesses. The bottom plate is called the filter plate. A thin perspex mask (14 × 14 cm) having holes of diameter 4.6 cm covers the filter plate except the filters. The idea behind the use of the mask is to avoid vapor losses at the filter plate. The top plate (16 × 16 × 1 cm) has a depression of 0.1 cm depth and a glaze ice is grown in the depression before taking measurements with the equipment. This plate is called the ice plate. Figure 2 shows the photographs of the ice and filter plates. The vertical perspex wall rests in a groove on the filter plate and encloses the 14 × 14 cm bottom surface. The top and bottom plates are clamped together and a rubber strip placed between them makes the chamber air-sealed.

Thick aluminium plates with channels milled in them were attatched to the outsides of the top and bottom plates. Chilled methanol solution from a NESLAB cooling system cools both the plates. The chamber is placed in a deepfreeze whose temperature is kept colder than the required temperature of the SDC. Thick sheets of polystyrene cover the plates to insulate the chamber from heat losses.

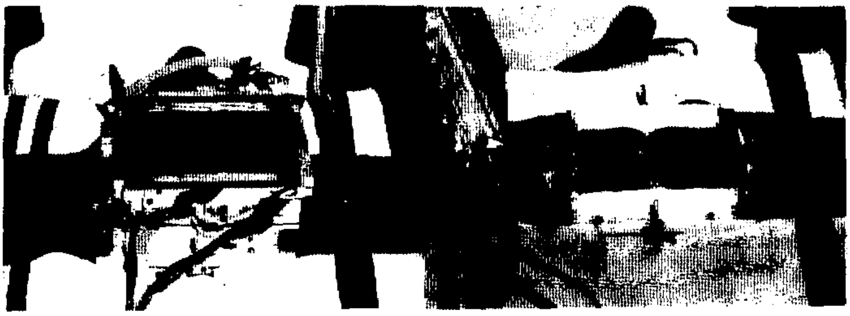

Fig.2. Photographs of the ice plate (a) and the filter plate (b).

Thermoelectric modules were embedded between the ice plate and the plate with cooling channels. The modules can warm the ice plate when required. Two aged thermistors embedded in the top and bottom plates close to the ice and filter surfaces measure and control temperature to an accuracy of ±0.05 °C.

In order to simulate cloudy conditions in the SDC, the ice and filter plates were subjected to a thermal gradient by warming the ice plate. In this way a known calculated supersaturation was produced in the SDC with an accuracy of ± 5%.

Several types of blank membrane filters were tested for the background count before being used for the measurements of IN concentrations. In a number of blank filters of different make, the Sartorius SM 13306 filters were found to give the minimum background count of 2.3 ± 0.2 per filter. The IN data obtained in subsequent experiments on processed filters were cured for the background count.

A large number of Sartorius filters were sampled for natural aerosol each for a volume of 100 litres. The sampled filters were kept in clean perspex boxes before processing. Experiments showed that the wall height of 0.7 cm gave more counts than the other heights. The sampled filters were therefore processed with a wall height of 0.7 cm.

In the running mode, the filter plate temperature was kept at –16 °C. The sampled filters were processed in the SDC in batches of four filters over a range of supersaturation with respect to ice. Both power and linear curves respectively of the forms $N_i = BS_i^{\gamma}$ and $N_i = C + \delta S_i$ were fit to all the data. Here B, γ, C and δ are empirical constants. Linear curve was also fit to the earlier data (Zamurs et al., 1977; Zamurs et al., 1982). The power curve had already been fit to the earlier data. Tables 1 and 2 record the present and the previous ice nucleus data. A linear curve and the corresponding power curve taken from the present data are plotted in Figure 3 (a,b).

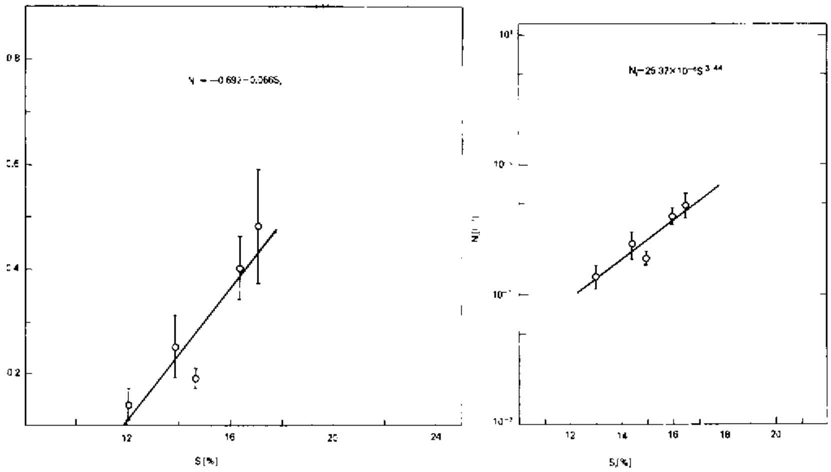

Fig.3. A linear ice nucleus versus supersaturation spectra (a) and the corresponding power curve fit to the data of the linear curve (b).

Table 1. Ice Nucleus Supersaturation Spectra (Present Study) (T = −16 °C)

Power curve			Linear curve		
$B(\times 10^{-6})$	γ	r'	C	δ	r'
29.65	3.48	0.90	−0.784	0.077	0.85
284.16	2.08	0.85	−0.067	0.010	0.86
25.37	3.44	0.93	−0.692	0.066	0.93
148.43	2.51	0.91	−0.230	0.024	0.93
32.26	3.16	0.82	−0.347	0.036	0.75
41.36	3.26	0.85	−0.297	0.038	0.89
11.71	3.47	0.84	−0.337	0.328	0.75
685.76	2.37	0.89	−0.639	0.071	0.91
9.28	3.40	0.86	−0.210	0.021	0.83
552.34	1.88	0.93	−0.075	0.011	0.91

Each curve in the present study was obtained from five data points; each point based on the average of counts of four filters processed simultaneously in the SDC. The earlier data were also obtained with a thermal–gradient diffusion chamber. The error of measurement in the present results was found to be 15.5% with a range of 21.0 to 33.9%. The previous data had an error of 21.7% with a range from 1.4 to 68.1%.

Table 2. Ice Nucleus Supersaturation Spectra (Zamurs et al., 1977; Zamurs and Jiusto, 1982) (T = −20 °C)

Power curve			Linear curve		
$B(\times 10^{-5})$	γ	r^1	C	α	r^2
385.55	2.12	0.94	−0.354	0.032	0.93
203.53	2.16	0.96	−0.188	0.018	0.95
180.84	2.56	0.96	−0.913	0.074	0.93
342.14	2.23	0.94	−0.394	0.037	0.95
266.90	2.48	0.99	−0.702	0.065	0.98
69.10	2.85	0.99	−0.697	0.060	0.98
96.73	2.90	0.97	−1.255	0.108	0.97
24.66	3.19	0.98	−0.809	0.069	0.98
131.80	2.82	0.97	−1.243	0.108	0.98
24.19	3.17	0.97	−0.764	0.065	0.98
16.41	3.47	0.97	−1.619	0.131	0.96
103.67	2.75	0.96	−0.840	0.071	0.94
56.10	3.22	0.99	−2.146	0.157	0.99
275.00	2.52	0.99	−0.875	0.071	0.99
84.32	2.93	0.99	−1.278	0.094	0.98
57.90	3.12	0.99	−1.626	0.119	0.99
61.35	3.07	0.99	−1.427	0.105	0.99
177.90	2.60	0.99	−0.745	0.060	0.99
105.02	2.77	0.99	−0.962	0.071	0.97
250.88	2.78	0.97	−1.649	0.139	0.99
26.13	3.13	0.99	−1.673	0.121	0.99
95.99	2.77	0.99	−0.731	0.057	0.99
311.03	2.41	0.99	−0.738	0.060	0.99
44.21	3.36	0.96	−2.256	0.175	0.99
18.28	3.43	0.99	−1.468	0.104	0.99

III. DISCUSSION AND CONCLUDING REMARKS

The research workers who measured IN concentration as a function of supersaturation with respect to ice. preferred to express IN versus supersaturation spectra in the form of a power law (Huffman, 1973; Ohtake, 1976). The main cause for adopting the power relation was probably due to the reason that cloud condensation nuclei (CCN) also obeyed a power law. The purpose of the present study was to investigate whether the ice nucleus data also observed linear trend with supersaturation or not. It is interesting to find that the linear curve fits to the data as good as the power curve. The coefficients of correlation for the straight curve vary between 0.75 and 0.93 whereas for the power curve they are between 0.82 and 0.93 (Table 1). The coefficients of correlation for the linear and power curves for the earlier data (Table 2) respectively are between 0.93 and 0.99 and between 0.94 and 0.99. The linear and power curve constants are therefore almost similar. It is understood that in natural clouds, the lower the sub–freezing temperature the more active is an ice nucleus at a supersaturation with respect to ice. The experiments performed in a simulated cloudy environment (for example, in a thermal–gradient chamber) exhibit this behavior of ice nuclei. The power curve demands that as the ice nuclei get activated at a sub–freezing temperature, there should be a sudden rise in the nucleability of activated ice nuclei. On the other hand, the linear curve demands that in a batch of ice nuclei activated at a sub–freezing temperature, the growth of ice crystals on the activated IN is not a sharp function of supersaturation with respect to ice. For example, a sub–micron size IN activated at a sub–freezing temperature may not be able to grow an ice crystal at an S_i while a micron size IN activated at the same temperature may grow crystal at the same S_i value. This reasoning is justified because size distribution and chemistry of IN play crucial role in their activation mechanisms (Pruppacher et al., 1978). IN of sub–micron size are difficult to activate as compared to IN of micron size. We cannot therefore specify a temperature at which a batch of IN gets activated. The different intercepts on the S_i–axis of N_i versus S_i plot show that some of the IN caught on the filter surface get activated earlier than the others at known temperature and supersaturation. Here again it is emphasized that activation of some of IN in a group dose not mean that the whole group has atarted activation. The above reasoning is justified in terms of a linear N_i versus S_i curve. The result needs to be further investigated.

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