Tellus (1985), *37B*, 296–303

Ice nucleus measurements: effect of site location and weather

By R. AL-NAIMI and C. P. R. SAUNDERS, *Pure & Applied Physics Department, UMIST, P.O. Box 88, Manchester, M60 1QD, England*

(Manuscript received March 26; in final form July 5, 1985)

ABSTRACT

Filter samples of atmospheric aerosol were collected daily at a city and a rural site over an extended period. The filters were processed in an environment supersaturated with respect to ice in order to activate ice nuclei. The concentration of nuclei detected at the two sites was compared with reference to the local weather and air-mass trajectories. The city environment generally has a larger ice nucleus concentration than the rural site. However, fluctuations in the concentrations at the two sites are usually in phase, indicating a pre-dominantly common origin. With air-mass trajectories over industrial regions, the nucleus concentration is increased. Local weather has a profound effect; in particular, rain and snow cause substantial reductions in ice nucleus concentration.

1. Introduction

The objective of this study was to compare ice nucleus concentrations measured simultaneously at a city and at a rural site and to determine the effect of local weather upon the measured concentrations. British weather is controlled by a maritime environment which influences the hundred or so fronts that pass over annually. The air reaching the Isles from the west and north is relatively clear due to its ocean trajectory, while southerly and easterly winds normally carry more aerosols from their passage over industrial areas. The City of Manchester, which is surrounded by industrial regions, was one of the sampling sites. Its meteorological situation is described by Elson and Chandler (1978). It is bordered on the north, south and east by the Pennine Hills while to the west there is flat land extending about 60 km to the ocean and surrounded by industrial regions. The other sampling site was at the south-west foot of Great Dun Fell in Cumbria, a remote, rural, hilly, environment about 200 km north of Manchester.

Despite many recent advances, the sources, concentrations and behaviour of ice nuclei remain an area of debate. A discussion of ice nucleus origins is presented by Pruppacher and Klett (1978). Evidence has been obtained of the nucleation activity of soil particles (clay-silicates), which are found at the centre of ice crystals as well as of materials of biological origin; clay particles may well carry micro-organisms. There is evidence that lead particles from vehicle exhaust may form nucleating lead iodide, particularly at coastal city sites. However, there is also evidence of a reduction in the concentration of active ice nuclei downwind of cities, suggesting a deactivation effect of local pollutants. There is limited evidence of the effect of meteorological factors on ice nucleus concentrations. The nuclei are influenced by a number of atmospheric processes such as coagulation, sedimentation, advection, turbulent mixing and scavenging. The importance of these processes is not only a function of nucleus size and composition but is also affected by both large and small-scale meteorological conditions.

One of the modes of activity of ice nuclei is by the direct deposition of water vapour from the environment onto a nucleus which then grows as an ice crystal. This process requires supersaturation with respect to ice and so will take place in conditions both below and above water saturation.

Studies of such nuclei have been made on a worldwide basis and their concentration has shown great variability. However, there is agreement that more nuclei become active at colder temperatures. The standard method has been to trap aerosol particles on a membrane filter and to activate the nuclei on that filter in a static diffusion chamber in which low temperatures and a supersaturation are provided. Discussions of the results, the method and its limitations have been presented by Bigg (1963), Mossop and Thorndike (1966), Stevenson (1968), Gagin and Aroyo (1969), Lala and Jiusto (1972), Huffman and Vali (1973), Zamurs et al. (1977). In addition, details of the presently used technique are given by Hussain and Saunders (1984). Typically, a 47 mm diameter membrane filter having a pore-size of 0.45 μm is used to sample 100 litres of air. The filter is then placed in good thermal contact with a temperature-controlled surface and a heated ice-coated surface is located over the filter with a separation of a few millimetres. Typical processing temperatures of -10, -15 and $-20\,^{\circ}C$ have been used with the humidity set nominally at water saturation. Problems encountered have included the volume effect, whereby fewer nuclei are activated per unit volume for a larger sample volume. This effect has been attributed to competition for the available vapour on the filter surface. Attempts to correct the resulting concentrations in terms of the number of aerosol particles present have been made by Huffman and Vali (1973) and Hussain and Saunders (1984); Meyer and Gravenhurst (1976), used a low pressure chamber to reduce vapour losses to ice nuclei on the filter.

In the present studies, measurements were made of ice nucleus concentrations at two sites under various meteorological conditions. The convenience of the filter method led to its use but in an attempt to reduce the volume effect, only 25 litres of air were sampled per filter in 20 minutes. Samples were collected on a daily basis where possible, at a height of 20 m at UMIST in Manchester and at 2 m in Cumbria. Four filter samples were obtained at each of the sites at 1100 hours. The filters were subsequently processed in the static diffusion chamber in UMIST which has been described by Hussain and Saunders (1984). In order to activate as many nuclei as possible for comparative purposes, a low temperature of $-20\,^{\circ}C$ was used. Nominally, the chamber conditions were set at water saturation although this may not have been achieved because of competition for the available vapour on the filter surface.

In addition to the daily filter samples, particle concentrations in the size range 0.5 to 10 μm were measured at UMIST with an optical particle counter. Also, a Nolan-Pollak counter was used to obtain the concentration of condensation nuclei (CN). Surface meteorological variables were routinely recorded together with a record of precipitation at the filter sampling times. The 1200Z daily synoptic charts were used for weather system location analysis.

2. Results and discussion

About 20,000 l of natural air from each site were sampled and processed with the filter technique. The results were analysed in several ways with the focus mainly on the comparison of ice nucleus concentrations between Manchester and Cumbria and their correlation with some meteorological variables.

Fig. 1 shows the results of the comparison between Cumbria and Manchester for a period of 179 days of parallel filter samples at the two locations. Each sample point was the average of the data from four filters. The solid line is a best fit straight line with a coefficient of determination of 0.53. It can be compared with the 1:1 line which is also shown in the figure. The corresponding correlation coefficient was calculated to be 0.74. The results show that there are in general more ice nuclei in the city environment than the remote site. The mean values of concentrations detected at Manchester and Cumbria during the 179 day period were 0.76 l^{-1} and 0.71 l^{-1} respectively. A correlated t-test of the two samples revealed that the two populations are significantly different (99.9%). However, the differences in concentration between the two locations decrease with increased concentration which may be due to the presence of hygroscopic particles in the city environment causing vapour depletion over the surface of the filter samples when such particles are in high concentration. The results showing increased counts in the city may be compared with those of Zamurs and Jiusto (1982) who found that the small city of Albany had a count higher than both New York City and a remote mountain site.

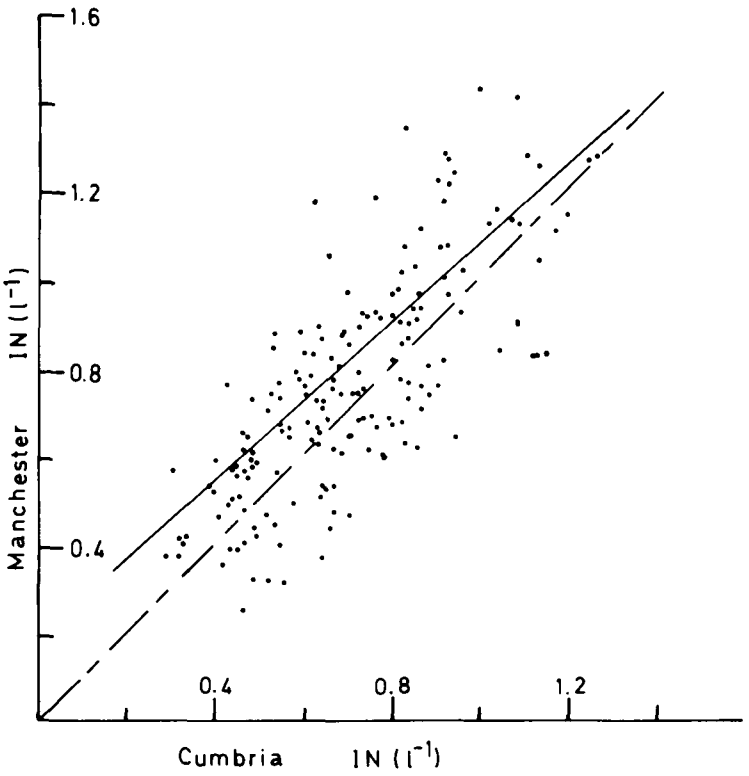

Fig. 1. Ice nucleus concentrations at Manchester and Cumbria.

They suggested that either the ice nuclei in New York City were being poisoned by other emissions or that local soil sources are effectively paved over in the city. Hussain and Saunders (1984) with only limited data, also noticed that Manchester had more ice nuclei than Cumbria.

Monthly averages of ice nucleus concentrations from Manchester and the remote site are shown in Fig. 2. In this figure it is clear that the variations of concentration at the two sites are generally in phase. Thus it seems that both sites are sampling the same air mass with nucleus concentrations being a function of the air mass origin and history. However, the figure shows that, in general, the concentration in the city is higher than that in Cumbria, which indicates that there is a superimposed local city effect. The general trend of the figure shows a maximum concentration in December and the months of spring (March–May) and a minimum in January–February and the summer months.

The concentration of condensation nuclei was measured in Manchester with a Nolan-Pollak counter during the period of ice nucleus measurements. The correlation coefficient between corresponding concentration values for one year of data was −0.06. This result is similar to that obtained by Georgii and Kleinjung (1967) and Zamurs and Jiusto (1982) and indicates no significant correlation between Aitken-sized particles and ice-nuclei. However, when the concentration data were averaged monthly, Fig. 2 confirms that in general, the condensation nucleus count bears an inverse relationship to the ice nucleus concentration. A similar result was explained by Hussain and Saunders (1984) in terms of competition for vapour on the filter surface when the particle concentration was high. There was a clear link between ice nucleus concentration and large particle sizes. For example, for particles in the 1.4 to 3 μm size range, the monthly correlation coefficients with ice nucleus concentration were always positive

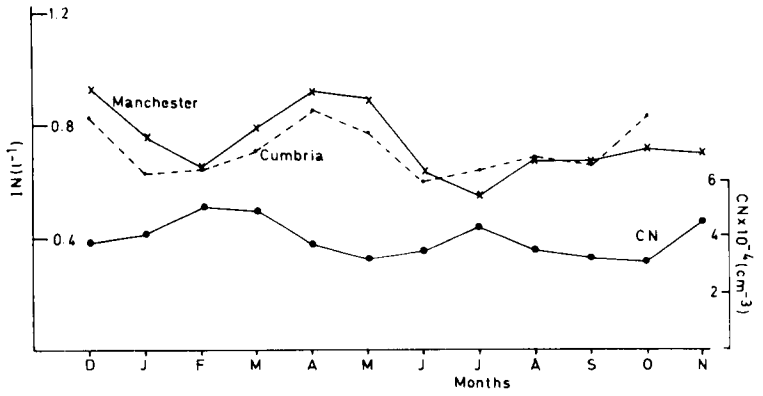

Fig. 2. Monthly averages of ice nucleus concentrations at Manchester and Cumbria together with condensation nucleus concentration at Manchester.

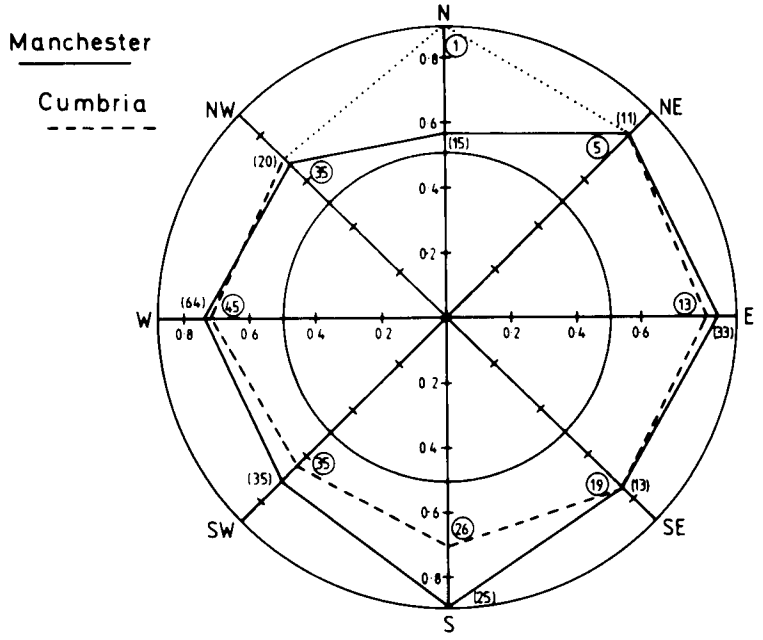

Fig. 3. Ice nucleus concentrations/l at Manchester and Cumbria. The bracketed and circled numbers are the number of samples for each corresponding wind direction. (——) Manchester data; (– – – –) Cumbria data.

with a maximum value of +0.61 in November, a minimum of +0.09 in July and a mean value of +0.34 ± 0.19. There is the possibility that the ice nuclei are very small and become attached to larger particles in the atmosphere.

Fig. 3 shows an analysis of the ice nucleus concentration from filter samples obtained at the two sites as a function of the local wind direction. The number of days of each wind direction at each site at the sampling times are shown. The concentrations plotted are averages over the appropriate number of days. Again, the city air has the higher concentration for most wind directions. The nature of the distribution of nuclei with wind

direction at the two sites also suggests that both sites were sampling the same air mass. The lowest ice nucleus concentrations are associated with northerly winds; the one occasion of north wind at the remote site is not significant. The highest differences in counts at the two sites were measured with southerly winds. This presumably reflects the pollution from the heavily industrialized Midlands about 150 km south of Manchester. Choularton et al. (1982) have reported that the highest concentrations of 0.7–1.4 μm particles occurred at Manchester when the airmass had traversed the Midlands.

Although the influences of local weather conditions on ice nucleus concentrations are likely to be complex due to the many processes affected, the concentration was found to be associated with some clearly recognizable events. Hussain and Saunders (1984), from their more limited data, observed that ice nucleus concentrations tended to be higher at or just before the arrival of a frontal system and tended to fall behind cold fronts. The decrease of concentration behind cold fronts has also been observed by Schnell et al. (1980). Fukuta and Tomlinson (1980) have reported that the count decreases sharply during snowfall.

Further evidence of the above effects was found in the present study. Examples can be seen in Fig. 4 which shows the daily values of the nucleus concentration at the two sites for the period from 1st December 1982 to 13th October 1983. The precipitation effective at the sampling time on both sites is indicated on the figure. The arrival of a frontal system can be seen to have an effect on the concentration. On 7th, 14th and 23rd December, 10th and 17th January, 1st March, 13th April, 5th and 25th May, 8th June, 14th July, 13th, 21st and 23rd September and 4th October, frontal systems were about to sweep across the British Isles. On all these days there were peaks in the nucleus concentration. In most of the examples, the following days show a sharp decrease in concentration, especially behind the cold fronts. The more pronounced decreases at the passage of the cold fronts are possibly not only due to the precipitation scavenging of the nuclei but also to the replacement of the more polluted air by cleaner air behind the cold fronts. This phenomenon seems to be in a good harmony with a report by Sheih et al. (1983). They found that the concentration of aerosol particles (<1.5 μm), in both warm and cold front cases, tends to increase just before precipitation, to decrease during the period of rain, and then to remain low behind the cold front.

The precipitation itself also has an effect on the nucleus count. As shown in Fig. 4, in general the presence of rain or snow caused the count to drop. A good example can be seen in the month of December where Cumbria recorded a lower concentration than Manchester through the month except on the 22nd when there was snow falling in Manchester but none in Cumbria. The low level of ice nucleus concentrations associated with snowfall could be due to the thorough cleaning of the atmosphere by snow. Numerous other observations made outside the fixed daily measurements during periods of snowfall corroborate the above hypothesis. Magono et al. (1974) found that one snow crystal had collected more than 6000 aerosol particles of diameter greater than 0.2 μm.

3. Conclusion

The average condensation nucleus concentrations in the city in the present work agrees closely with that observed by Zamurs and Jiusto (1982) in semi-urban Albany. They also observed a lower concentration of ice nuclei at a remote site in agreement with the present work. The particular conditions that led to a low concentration in New York City, namely nucleus poisoning and possible suppression of sources by cover-up, were evidently not appropriate to the Manchester environment. Manchester is probably more akin to Albany in that it is surrounded by open country. The inverse relationship between ice nucleus and condensation nucleus concentrations, when averaged on a monthly basis, may be due to reduced activation of ice nuclei on the filter when the competition for vapour is high. The objective of this work was to compare data from two sites and as the city had a larger concentration of ice nuclei than Cumbria, the effect of the vapour depletion would have been to under-estimate the city concentration more than the rural concentration.

The filter samples at the remote site revealed lower deposition nucleus activity than in the city. However, the variation of monthly averages of the nucleus concentrations obtained at the two sites was generally found to be in phase, which suggests that the nuclei at both sites are part of the same air

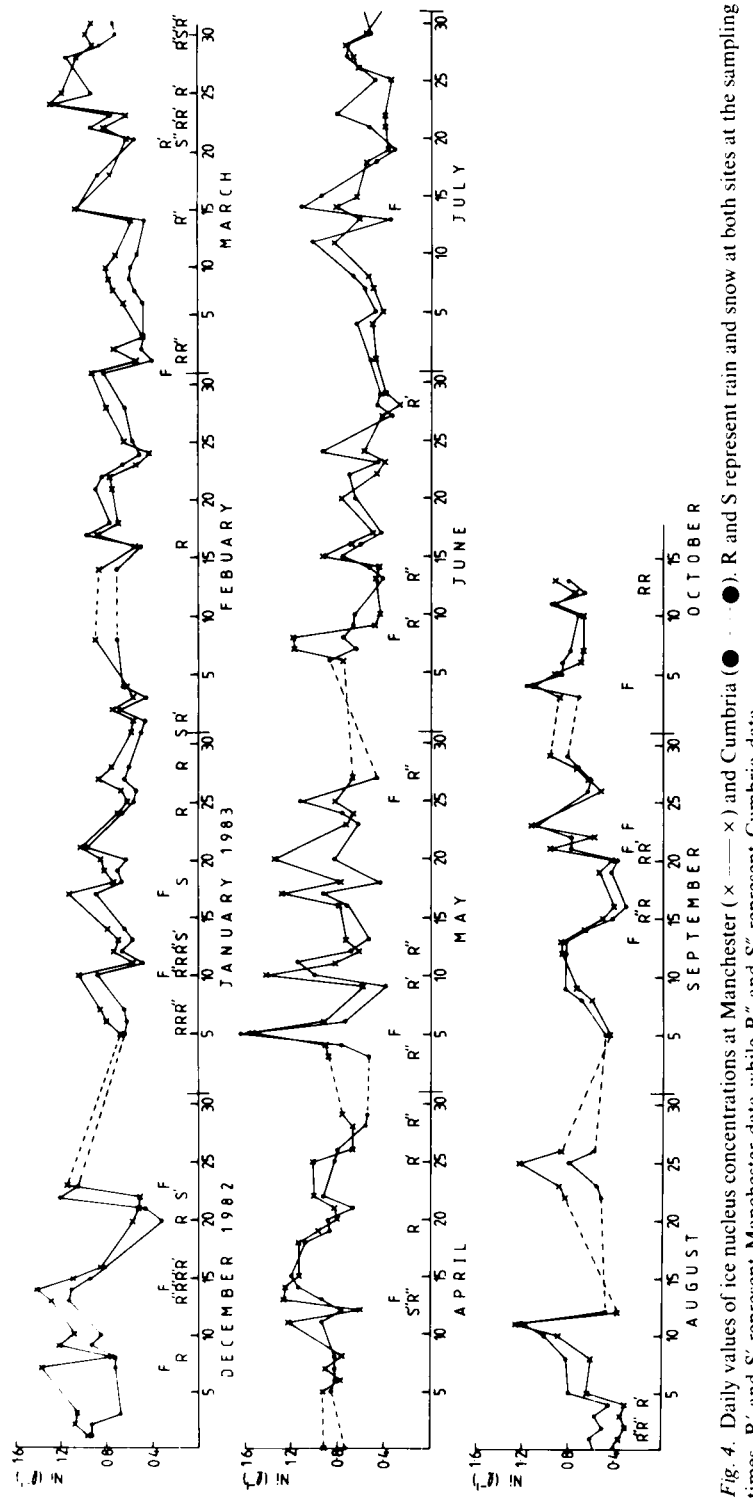

Fig. 4. Daily values of ice nucleus concentrations at Manchester (× —— ×) and Cumbria (● ····· ●). R and S represent rain and snow at both sites at the sampling times. R′ and S′ represent Manchester data while R″ and S″ represent Cumbria data.

mass and so have experienced the same trajectories. The difference between the two concentrations is most noticeable when the wind lies between the south-east and the west. The higher count in the city agrees with the finding of Choularton et al. (1982) that the highest concentration of aerosol particles occurred in Manchester when the air mass had traversed the heavily industrialized Midlands to the south of Manchester. The variation of ice nucleus concentration through the year is small, which indicates a constant source. A strong possibility is that the nuclei are of organic origin as pointed out by Vali (1968). Experiments by Schnell and Vali (1976) have shown that some ice nuclei in soils are produced by decomposing natural vegetation. The nuclei of organic origin may provide the fairly constant background levels while local industrial sources provide the larger variations in ice nucleus concentration.

The determination of size or the size spectrum of ice nuclei represents another unsolved problem in this field. Allee et al. (1968) with their Goetz spectrometer technique, which was developed to establish a relationship between size and activity of ice nuclei, found that most of the ice nuclei in suburban air were greater than 1.0 μm. This is supported by Prodi et al. (1982). According to Byers (1965), 15% of IN were larger than 1.0 μm, 18% were in the range 0.7 to 1.0 μm, 38% in the range 0.4 to 0.7 μm and 29% in the range 0.1 to 0.4 μm. Georgii and Kleinjung (1967) have found IN better correlated with larger particles. Mossop (1963) found the lower limit for the size of natural IN at about 0.02 μm, while Vali (1966, 1968) found evidence, in drop-freezing experiments, that substantial fractions of freezing nuclei were smaller than 0.01 μm. Fig. 4 indicates that large changes in nucleus concentration are associated with local weather conditions. In particular, precipitation, especially snowfall, is able to reduce the nucleus concentration by washout and scavenging of the aerosol particles. The particle concentrations in the size range 0.5 to 5 μm and above also showed a substantial decrease after passage of the front. On 25th May for example the 0.5 to 0.7 μm particles decreased by a factor of six and the 3 to 5 μm particles by a factor of three. The present studies also show a positive correlation of ice nucleus concentration with large particles. These results indicate that the deposition nuclei activated on the filters in these experiments were probably of the order of 1 μm in diameter or were carried on particles of this size.

4. Acknowledgements

This work was supported by the Natural Environment Research Council.

REFERENCES

Allee, P. A., Gerber, H. and Weickmann, H. K. 1968. Determination of sizes of ice nuclei with an aerosol spectrometeor. *J. Rech. Atmos. 1–2*, 167–170.

Bigg, E. K. 1963. Comparisons of concentrations of ice nuclei in different parts of the world. *J. Rech. Atmos. 4*, 41–58.

Byers, H. R. 1965. Identification of ice nuclei in the atmosphere. Pre-prints Int. Conf. Cloud Physics. Tokyo, 126–130.

Choularton, T. W., Fullarton, G. and Gay, M. J. 1982. Some observations of the influence of meteorological variables on the size distribution of natural aerosol particles. *Atmos. Environ. 16*, 315–323.

Elson, D. M. and Chandler, T. J. 1978. Meteorological controls upon ground level concentrations of smoke and sulphur dioxide in two urban areas of the United Kingdom. *Atmos. Environ. 12*, 1543–1554.

Fukuta, N. and Tomlinson, E. M. 1980. A new horizontal gradient, continuous flow chamber and detailed observation of condensation freezing and deposition nucleation. Pre-prints of Cloud Physics Conf. Clermont-Ferrand, 673–676.

Gagin, A. and Aroyo, N. 1969. A thermal diffusion chamber for the measurement of ice nuclei concentrations. *J. Rech. Atmos. 4*, 115–122.

Georgii, H. W. and Kleinjung, E. 1967. Relations between the chemical composition of atmospheric aerosol particles and the concentration of natural ice nuclei. *J. Rech. Atmos. 3*, 145–146.

Huffman, P. J. and Vali, G. 1973. The effect of vapour depletion on ice nucleus measurements with membrane filters. *Nature 12*, 1018–1024.

Hussain, K. and Saunders, C. P. R. 1984. Ice nucleus measurement with a continuous flow chamber. *Q. J. R. Meteorol. Soc. 110*, 75–84.

Magono, C., Endoh, T., Harimaya, T. and Kubota, S. 1974. A measurement of scavenging effect of falling snow crystals on the aerosol concentration. *J. Japan. Meteorol. Soc. 52*, 407–416.

Lala, G. G. and Jiusto, J. E. 1972. Numerical estimates of humidity in a membrane-filter ice nucleus chamber. *J. Appl. Meteorol. 11*, 674–683.

Meyer, D. and Gravenhurst, G. 1976. A low pressure diffusion chamber. Proc. Third Int. Workshop on Ice Nucleus Measurements. Univ. of Wyoming. pp. 128–145.

Mossop, S. C. 1963. Atmosphere Ice Nuclei. *Zamp. 14*, 456–486.

Mossop, S. C. and Thorndike, N. S. C. 1966. The use of membrane filters in measurements of ice nucleus concentrations. I. Effect of sample air volume. *Q. J. R. Meteorol. Soc. 5*, 474–480.

Prodi, F., Santachiara, G. and Prodi, V. 1982. A study of the effect of size on ice nucleation in the aerodynamic range of particles. *J. Appl. Meteorol. 21*, 945–952.

Pruppacher, H. R. and Klett, J. D. 1978. *Microphysics of Clouds and Precipitation*. D. Reidel Publ. Co.

Schnell, R. C. and Vali, G. 1976. Biogenic ice nuclei: Part 1. Terrestrial and Marine Sources. *J. Atmos. Sci. 33*, 1554–1564.

Schnell, R. C., Wrobel, B. and Miller, S. W. 1980. Seasonal changes and terrestrial sources of atmospheric ice nuclei at Boulder, Colorado. Proc. Cloud Phys. Conf. Clermont-Ferrand, 53–56.

Sheih, C. M., Johnson, S. A. and DePaul, F. T. 1983. Case studies of aerosol size distribution and chemistry during passages of a cold and a warm front. *Atmos. Environ. 17*, 1299–1306.

Stevenson, G. M. 1968. An improved millipore filter technique for measuring the concentrations of freezing nuclei in the atmosphere. *Q. J. R. Meteorol. Soc. 94*, 35–44.

Vali, G. 1966. Sizes of atmospheric ice nuclei. *Nature 212*, 384–385.

Vali, G. 1968. The freezing nucleus content of precipitation and its relation to the formation of ice in clouds. Proceedings Int. Conf. Cloud Physics, Toronto, 232–237.

Zamurs, J., Lala, G. and Jiusto, J. E. 1977. Factors affecting ice nucleus concentration measurements with a static vapor-diffusion chamber. *J. Appl. Meteorol. 16*, 419–424.

Zamurs, J. and Jiusto, J. E. 1982. An examination of ice nucleus concentrations in Eastern New York State. *J. Appl. Meteorol. 21*, 431–436.