# An Examination of Ice Nucleus Concentrations in Eastern New York State

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ABSTRACT

This work reports the results from a five-station (remote to highly urbanized) sampling network operated in 1976 in eastern New York State in which daily measurements were made of ice nucleus and condensation nucleus concentrations among the stations at most of the test relative humidities. Generally, semi-urban Albany had the highest concentration of ice nuclei and remote Whiteface Mountain the lowest. Albany also typically had the highest slope values (ice nucleus concentration-supersaturation spectrum) and the highest concentrations of condensation nuclei, while Whiteface Mountain typically had the lowest values for these two parameters. Lower ice nucleus concentrations in the New York City area suggest that the degree of urbanization there did not have a major impact on ice nucleus concentrations.

## 1. Introduction

Most ice nucleus (IN) concentration measurements have been made by individuals at single sites and thus the characterization of the IN distribution over a large geographical extent and its spatial and temporal variation have received relatively little attention. The attempts that have been made to establish IN concentration measurement networks have met with varying degrees of success.

Kline (1963) reported the results of a 15-station network across the United States, sampling sporadically from 1959 to 1962. Bigg and Stevenson (1970) established a 44-station global IN sampling network. Each author prepared the samples by a different method which resulted in a factor of three difference in the IN concentrations. In addition, Bigg and Miles (1964) operated a 24-station network in Australia and New Zealand, and Allee (1973) established a 19-station network in the western United States.

Certain advances in IN concentration measurements were described at the Third International Workshop on Ice Nucleus Measurements (Vali, 1976), and the introduction of supersaturation spectra of ice nuclei suggested that another network study might be appropriate. Continued interest in the question of whether urban areas constitute a source of anthropogenic ice nuclei was another motivating factor.

It should be reiterated that a direct correlation between ice nucleus concentrations and initial ice particle concentrations in clouds has only been demonstrated in a few cases, e.g. Gagin and Neumann (1974). Obviously, the better method to discern such a relationship would involve aircraft sampling of IN just below cloud base. However, in terms of examining potential sources of surface ice

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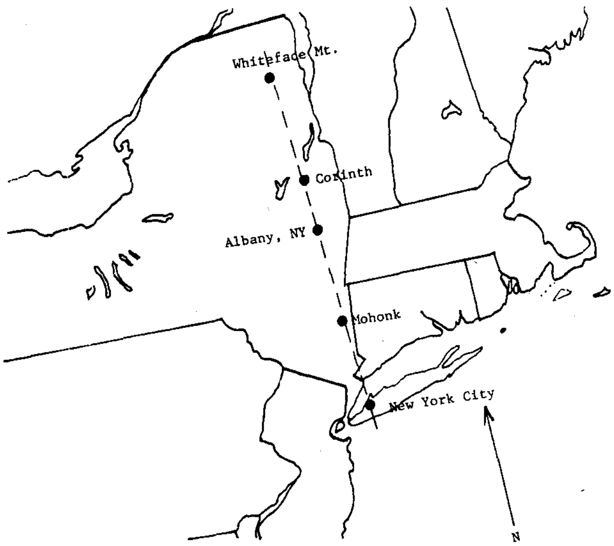


FIG. 1. 1976 ice nucleus sampling network stations.

nuclei and their activity spectra, without prohibitive cost, a ground network has considerable merit.

## 2. New York State ice nucleus measurement network

An IN sampling network was established in the summer of 1976 to obtain more information on IN concentrations and their variation in New York State over a path with exceptionally diverse air quality characteristics. Five sites comprised the network (see Fig. 1), as follows: Albany, New York; Whiteface Mountain in the Adirondack Mountains, ~169 km NNW of Albany; Corinth, New York, ~56 km NNW of Albany; Mohonk Resort, near New Paltz, New York, ~104 km SSW of Albany; and Greenlawn, New York, ~202 km SSE of Albany. These stations formed a general north-south axis through eastern New York State. Sites were chosen to represent conditions and distinctly different locales which could be characterized as rural (Whiteface), semi-urban (Albany), or urban (Greenlawn/New York City). The remote "rural" nature of Whiteface Mt. is graphically illustrated by the low condensation nucleus (CN) concentrations reported in Table 1, by contrast to the "urban" CN character of New York City (Table 2).

The ice nucleus detection method used for this study was the filter technique and the principal instrument used was the static vapor diffusion chamber. Its description, the filter preparation method, the filter processing procedure, and the factors which affect the performance and operation of this technique can be found in Lala (1972), Zamurs (1975), Jiusto *et al.* (1976) and Zamurs *et al.* (1977). Briefly summarizing, much of the volume effect and vapor sink difficulties can be reduced by the use of hydrophobic filters, modest volumetric air samples (typically 100 L, to avoid excessively large concentrations of particles on the filter surface) and shallow processing-chamber heights of 0.5–1 cm. When these corrective procedures are applied, IN concentration measurements made to −20°C with the filter method show acceptable reproducibility and reasonable agreement with other IN counting devices (Jiusto *et al.*, 1976).

Daily sampling commenced 1 July 1976 and continued for two months. (The July samples required about two years to analyze in detail with the results presented here.) The samples were taken six days a week (Monday–Saturday) at 0900 EDT (1300 Z) at all five stations. Sartorius hydrophobic filters with 0.45 um pore size (type SM 13306) were used. At virtually all the sites, four filters (eight at Albany) were sampled simultaneously at a rate of 5 L min$^{-1}$ for 20 min. In addition to the daily filter samples, CN concentration measurements were taken at sampling time at all the stations to obtain a measure of the total aerosol concentration; small particle detectors (Gardner Associates, Inc., model CN) were employed.

TABLE 1. Sampling network condensation nucleus and ice nucleus concentrations and slope values (median and range).

| Station | Condensation nuclei (cm$^3$) | 95% | 98% | Ice nuclei (L$^{-1}$) 100% | ($T$ = −20°C) 102% | 105% | Slope $k$ |
|---|---|---|---|---|---|---|---|
| Albany | 42100<br>14400–95500 | 0.36<br>0.17–0.90 | 0.70<br>0.31–1.05 | 1.28<br>0.67–1.90 | 1.62<br>1.02–2.53 | 2.22<br>1.47–3.66 | 3.30<br>1.69–4.47 |
| Whiteface | 1600<br>310–5600 | 0.27<br>0.15–0.93 | 0.42<br>0.23–0.93 | 0.70<br>0.33–1.26 | 0.86<br>0.42–1.54 | 1.10<br>0.58–1.82 | 2.47<br>1.19–3.58 |
| Corinth | 7200<br>2100–110000 | 0.27<br>0.10–0.75 | 0.40<br>0.20–0.86 | 0.70<br>0.29–1.33 | 0.98<br>0.46–1.76 | 1.37<br>0.56–2.41 | 3.04<br>1.51–4.79 |
| Mohonk | 6100<br>1750–124400 | 0.29<br>0.15–0.77 | 0.55<br>0.23–1.28 | 0.93<br>0.37–1.80 | 1.23<br>0.63–2.47 | 1.71<br>0.81–3.18 | 3.03<br>2.08–4.92 |
| Greenlawn | 16500<br>6000–39000 | 0.27<br>0.10–0.56 | 0.47<br>0.21–0.83 | 0.84<br>0.37–1.40 | 1.10<br>0.46–1.98 | 1.51<br>0.87–2.87 | 3.11<br>1.97–4.95 |

TABLE 2. New York City-Greenlawn experiments.

| | Average ice nucleus concentration ($L^{-1}$) | | | | | Average derived slope and constant | | Average condensation nucleus concentration ($\times 10^3$ $cm^{-3}$) |
|---|---|---|---|---|---|---|---|---|
| | 95% | 98% | 100% | 102% | 105% | K | c | |
| New York City | | | | | | | | |
| 9/12/77 | 0.21 | 0.38 | 0.53 | 0.72 | 0.93 | 2.61 | 1.74 (−4) | 78.2 |
| 10/31/77 | 0.22 | 0.33 | 0.49 | 0.69 | 1.08 | 2.77 | 1.03 (−4) | 48.8 |
| 5/15/78 | 0.42 | 1.02 | 1.48 | 1.75 | 2.11 | 2.79 | 2.42 (−4) | 47.8 |
| 5/22/78 | 0.29 | 0.54 | 0.88 | 1.17 | 1.75 | 3.14 | 5.47 (−5) | 84.1 |
| Overall average | 0.29 | 0.56 | 0.84 | 1.08 | 1.47 | 2.83 | 1.34 (−4) | 65.9 |
| Greenlawn | | | | | | | | |
| 9/12/77 | 0.18 | 0.32 | 0.49 | 0.68 | 0.85 | 2.77 | 9.38 (−5) | 14.2 |
| 10/31/77 | 0.24 | 0.36 | 0.53 | 0.66 | 0.97 | 2.42 | 3.06 (−4) | 40.9 |
| 5/15/78 | 0.34 | 1.11 | 1.62 | 1.92 | 2.49 | 3.38 | 4.21 (−5) | 23.3 |
| 5/22/78 | 0.20 | 0.46 | 0.72 | 0.99 | 1.46 | 3.44 | 1.76 (−5) | 92.0 |
| Overall average | 0.24 | 0.56 | 0.84 | 1.06 | 1.44 | 3.09 | 5.77 (−5) | 42.6 |

The four-filter set from each daily measurement was subsequently processed in the static vapor diffusion chamber at the State University of New York at Albany. Each set, constituting one sample point average, was processed at a temperature of −20°C and at five relative humidities, ranging from 95 to 105% (i.e., 95, 98, 100, 102 and 105%). Details of the reprocessing procedure, involving crystal sublimation and re-cooling of the filters, are given elsewhere (Zamurs *et al.*, 1977). In addition to IN concentration at five relative humidity (RH) values, a supersaturation spectrum was determined. Supersaturation spectra are sometimes expressed in the form

$$N = cS_i^k, \tag{1}$$

where $N$ is the IN concentration, $S_i$ is the calculated supersaturation with respect to ice, and $c$ and $k$ are empirical constants. This relationship, analogous to that for CCN spectra, was first introduced for IN by Huffman (1973) and Gagin (1973).

## 3. Results of the IN measurement network

Table 1 synthesizes the results of the IN measurement network. The median IN concentration for each humidity, the median spectrum slope, and median CN concentration are presented along with the observed range of each of these variables.

The most notable development was consistently high IN concentrations observed at Albany. Albany had the highest median IN concentration at all five test humidities and the greatest number of days (60% average) when it registered the highest count of the five stations, especially at the higher relative humidities. Conversely, Whiteface had the lowest median concentration at most test humidities. This site rarely experienced the highest daily average IN concentration. After Albany, Mohonk experienced the next highest concentration and was the site with the next highest daily frequency of high-count occurrence. From Table 1, Greenlawn can be considered the site with the next highest (3rd) IN concentration, followed by Corinth and then Whiteface.

These results show that a considerable range existed in IN concentrations over a 370 km line in eastern New York State. The most striking difference was between Albany and Whiteface. Albany's median concentration at 105% RH was twice that (2.2 vs. 1.1 $L^{-1}$) of Whiteface's; indeed Albany's median concentration at 100% RH was greater than Whiteface's median concentration at 105% RH. At the other sites as well, the difference in the median concentration was sizeable. Some of these interstation differences at a given RH value were about one-third of the total IN concentration change over the entire supersaturation spectrum (i.e., from 95 to 105% RH). Thus typical summer IN concentrations along the sampling network were quite variable.

IN concentrations fluctuated markedly over a period of one or two days. For example, the average daily change in IN concentrations at Albany and Whiteface Mountain were 0.3 $L^{-1}$ (23% change) and 0.2 $L^{-1}$ (29%), respectively at 100% RH. However, oscillations of 150 to 300% were not uncommon. The large daily variability in IN concentrations found in this study questions processing procedures such as Allee's (1973), wherein filters were processed at −15 and −20°C on alternate days. The day-to-day variations in IN concentrations may have been due in part to airmass changes (or air trajectory variations) as previously observed by Kline and Brier (1961).

Table 1 indicates that a single station's daily IN concentration range declined as processing humidity increased—from a factor of 5 to 6 at 95% RH to about a factor of 2.4 to 4 at 105% RH. This reduction in concentration variability is attributed to vapor competition effects which become more pronounced at the higher humidities.

The pattern or supersaturation spectrum slope ($k$, Eq. 1) is reminiscent of the daily IN concentration patterns. Albany had the highest $k$ value most frequently, as well as the highest median slope (Table 1). Whiteface had the lowest median slope and lowest frequency of high slope occurrence. The slope values also occasionally exhibited large daily fluctuations, as large as a factor of $\sim$2.6.

The most striking feature of the CN concentration data (Table 1) is the dominance of Albany as the site with the highest concentration of condensation nuclei (42 100 $cm^{-3}$ average), with Whiteface over an order of magnitude lower (1600 $cm^{-3}$). Of the 27 sampling days, Albany had the highest CN concentration on 25 days! The CN measurements showed that Albany had the highest atmospheric aerosol loading, Whiteface the lowest, and intermediate atmospheric loading at the remaining three sites. CN concentrations exhibited daily fluctuations that were much greater than those for ice nuclei. Factors of 5 to 10 changes over a period of one or two days were not uncommon.

To summarize, Albany had the greatest concentration of ice nuclei and condensation nuclei and the greatest slope values $k$ (of the five sampling stations) and Whiteface had the lowest respective values. The remaining three sites were ranked intermediately for the three parameters. This suggests that the aerosol of Albany and Whiteface was considerably different, with Albany's aerosol having a much larger ice nucleating activity with increasing moisture availability. This could be due to differences in aerosol chemistry and the number of ice nucleating sites per particle; a difference in the partial solubility of mixed nuclei; or a lower number of total particles (same chemistry) available for ice nucleation at Whiteface Mountain (analogous to lower CN concentrations). Additionally, all parameters varied considerably from one sampling period to the next. Thus, the aerosol composition, in terms of some combinatorial factor of amount, makeup and ice nucleating activity, apparently varied appreciably in eastern New York State.

To evaluate the results of Table 1 and the conclusions drawn from it, a statistical analysis was performed on the data to determine if the differences in IN concentration among the stations were statistically significant. The Wilcoxon Rank-Sum Test was used.

Table 3 supports many of the conclusions drawn from Table 1. Using a probability level of 0.05 or less as statistically significant, it can be seen that the concentrations of ice nuclei measured at Albany were significantly different from and, therefore, greater than those at all the other stations for every humidity level. The differences in IN concentrations among the other stations became statistically significant only at 98–100% RH and higher, if at all. At 95% RH for the given sampling and processing conditions, we conclude that the "noise" level of the filter technique is being approached. Generally, for humidity levels $\geq$ 100%, Whiteface's IN data were statistically different from the other stations, reflecting its low levels of IN concentration and supporting the designation of Whiteface as the site with the lowest IN concentration. Table 3 indicates that the differences in IN concentrations among Corinth, Mohonk, and Greenlawn, for the most part, are not significant. These sites, therefore, may be said to have generally the same IN concentrations.

In examining possible explanations for this observed pattern, a rather interesting parallel appeared with the soil composition of the various sampling sites and may suggest that the composition of the soil (or associated surface constituents) directly influence the ice nucleation activity of the ambient aerosol. Historically, certain soil particles have been shown to be effective ice nuclei (Schaefer, 1949; Mason and Maybank, 1958; Mason, 1960; and Roberts and Hallett, 1967).

The soil found in the Albany area is moderately to highly basic. It has a brown or grey coloration and a thin sub-surface accumulation of clay. Whiteface's terrain is very rocky and the soil is low in base supply and has sub-surface accumulations of organic matter, aluminum and iron. The soils of the remaining three sites are essentially similar. These soils have been formed by crystalline clay minerals, are gray or light colored, and have no well-defined sub-surface layer (*National Atlas of the United States*, 1970). Thus, the composition of the soil at Albany and at Whiteface is different from each other and from the three remaining sites of Corinth, Mohonk, and Greenlawn, where the soils are generally of the same composition. This parallels the findings of the sampling network in terms of three distinct ice nuclei regimes. We cannot necessarily preclude some possible role of distinctive surface biogenic nuclei (Schnell and Vali, 1976) at the respective sites.

The correlation between IN concentrations and CN concentrations was examined at each site (and for all stations combined) on a daily basis. No statistically significant correlation between these two variables was evident.

The Greenlawn site yielded some additional interesting details. This site was chosen to represent the air of the New York City region; however, the

TABLE 3. Probability levels from the Wilcoxon Rank-Sum Test (ice nucleus concentration differences between stations).

| | Albany | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | to Whiteface | to Corinth | to Mohonk | to Greenlawn | to Corinth | to Mohonk | to Greenlawn | to Mohonk | to Greenlawn | to Greenlawn |
| RH 95% | 0.03 | 0.04 | 0.04 | 0.01 | 0.2 | 0.4 | 0.3 | 0.4 | 0.4 | 0.3 |
| 98% | <0.001 | 0.001 | 0.006 | <0.001 | 0.4 | 0.06 | 0.2 | 0.14 | 0.4 | 0.08 |
| 100% | <0.001 | <0.001 | 0.001 | <0.001 | 0.2 | 0.004 | 0.03 | 0.06 | 0.3 | 0.1 |
| 102% | <0.001 | <0.001 | <0.001 | <0.001 | 0.09 | <0.001 | 0.02 | 0.05 | 0.4 | 0.1 |
| 105% | <0.001 | <0.001 | <0.001 | <0.001 | 0.02 | <0.001 | 0.004 | 0.05 | 0.4 | 0.2 |

relatively low IN and CN concentrations measured there brought this supposition into question. Therefore, further measurements were performed on four different occasions at the Greenlawn site and in New York City (on the campus of Fordham University in the Bronx)—50 km apart. This additional sampling was performed during generally similar meteorological conditions, in the late morning or early afternoon with light wind speeds.

These comparisons, summarized in Table 3, showed virtually identical average IN concentrations. The minor differences in IN concentrations at the two sites on individual days canceled one another in the overall average. The CN concentrations, however, were substantially greater in New York City than in Greenlawn. The $k$ values were only slightly larger for the Greenlawn samples in three out of four cases, as well as in the overall average.

In addition, the IN sampling network data for Greenlawn were stratified according to wind direction. On those days with a predominately WSW to westerly flow for the entire metropolitan New York City region, Greenlawn would be sure to receive transported aerosols from the immediate New York City area; the IN concentrations on these "common" days were compared to those for the entire period of network operations to see if significant differences occurred.

Six of 27 days in July had the prescribed flow from New York City to Greenlawn. The average IN concentrations from the two sample populations were virtually identical (e.g., 0.84 $L^{-1}$ at 100% RH in both cases and <3% differences at higher humidities). As might be expected, CN concentrations were higher on the six days with direct city trajectories, averaging 26 200 $cm^{-3}$ vs. 16 500 $cm^{-3}$ for the overall Greenlawn data sample.

The additional sampling and the wind direction–IN concentration analysis supports the hypothesis that Greenlawn did represent New York City air, in terms of IN concentration at least; and the relatively low IN concentration at Greenlawn reflected the actual ambient IN concentration of that urbanized area of the state.

Several investigators have shown that certain industries, such as steel mills, aluminum works, and some power plants, are rich sources of IN (see review in Pruppacher and Klett, 1978). Urban areas overall, however, are not necessarily high source regions and in fact may be lower in IN concentration than nearby rural or less-urbanized regions.These results support the latter viewpoint. If automobile exhaust were a significant source of ice nuclei as has sometimes been hypothesized, one would expect the New York City, southern end of the network to have registered much higher IN concentrations. Relatively low concentrations in urbanized areas are sometimes attributed to IN poisoning by other industrial emissions (Georgii and Kleinjung, 1967; Braham and Spyers-Duran, 1974). Another possibility stemming from these observations is that large, sprawling urban complexes have effectively paved over the IN component associated with local surface soils.

## 4. Concluding remarks

Several findings have come from an ice nucleus sampling network, spanning a distance of 370 km from a remote mountain location to urban New York-Long Island. One result was the clear differences in IN concentrations at the five sampling stations, with a semi-urban site (Albany) consistently having the highest IN concentration, and Whiteface Mountain typically the lowest. The other three sites were intermediate and statistically similar, despite the great difference in urbanization represented. Some of the observations that ensued were as follows:

1) IN and CN concentrations and ice-supersaturation $k$ values varied markedly along the sampling network, with large daily oscillations frequently observed in these parameters.

2) Heavy urbanization in a region alone does not necessarily lead to high IN production, but the latter can sometimes be related to the presence of rather specific anthropogenic sources.

3) The soil composition at the sampling sites offers one possible explanation for the IN concentration differences reported.

4) There was a lack of a statistical correlation between IN and CN.

5) The filter method for processing ambient ice nuclei becomes unreliable at relative humidities $\leq$95%.

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