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Ice forming nuclei in the high Arctic

By E. K. BIGG[1], *The International Meteorological Institute in Stockholm S-106 91, Stockholm, Sweden*

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ABSTRACT

Concentrations of ice forming nuclei detected by the membrane filter technique were measured at temperatures of −12.5, −15 and −17.5°C during a voyage on the icebreaker *Oden* to the North Pole between 1 August and 6 October 1991. Geometric mean concentrations ranged from 13 m^{-3} at −15°C during the first 17 days of the expedition to 2.9 m^{-3} on the last 17 days of the voyage in good agreement with surface measurements made earlier north of latitude 70° N. On average, concentrations increased by a factor of 4.5 for a 5°C fall in temperature, less than is common in continental regions. Although air trajectory analysis showed that land sources occasionally influenced concentrations strongly, the time since the air had been over the open ocean was clearly the most important determining factor other than the seasonal decline. This implies an oceanic origin of the nuclei, the relationship being consistent with a halflife of 48.5 h (*e*-folding time of 70 h), about 60% longer than that of condensation nuclei. An apparent decrease in ice nucleus concentrations with temperature was mainly due to the seasonal change in concentrations but partly to the air trajectories associated with low temperatures. Elapsed time since the air was above the planetary boundary layer or over land also influenced concentrations, suggesting that the upper troposphere was deficient in ice nuclei while land was a weak source. Changes in the mixed depth of the atmosphere appeared to affect ice nucleus concentrations in the same way as condensation nucleus concentrations although poor time resolution limited this to two good examples. North of latitude 80° N, a few ice crystals were present near the surface for a considerable proportion of the total time. Single stellar crystals constituted about 80% of the total, implying growth near −15°C in predominantly supercooled clouds. Their concentrations were usually but not always consistent with surface ice nucleus concentrations.

1. Introduction

The measurements of ice forming nuclei (IFN) to be described were made from the icebreaker *Oden* on the International Arctic Ocean Expedition (IAOE-91) as part of a program to measure features of the atmospheric aerosol and its precursor gases. The measurements covered the period from 1 August to 6 October 1991, starting from about 70° N, 20° E, crossing the North Pole and ending near 79° N, 4° E. It included a period in port in Isfjørd, Spitsbergen from 8–16 August during which measurements continued to be made. Only those IFN that can be detected by the membrane filter technique were recorded.

Kumai and Francis (1962) showed by electron microscopy of residues from ice crystals falling in summer at an altitude of about 2000 m on the Greenland ice cap (latitude 77° N) that ice crystals contained numerous particles, predominantly less than 100 nm in diameter but with some as large as 360 nm in addition to some still larger mineral or clay particles thought to be the nuclei on which the ice crystals formed. They must therefore have been relatively efficient scavengers of the aerosol. Because on IAOE-91 drizzle drops reached the surface very rarely except during fogs, the frequently occurring (though sparse) ice crystals probably also brought with them a significant proportion of the aerosol deposited on the surface. Measurement of the concentrations of the ice nuclei on which these crystals might have formed and attempts to find their origin is therefore

[1] Present address: 12 Wills Ave., Castle Hill 2154, Australia.

relevant to the evolution of the Arctic aerosol and its losses to the surface.

In their study, Kumai and Francis found that particles of clay and related minerals occurred near the centres of 96.3% of all ice crystals and were assumed to be the nuclei on which the crystals originated. This would suggest a land origin of the summer ice nuclei and would appear to pre-empt the need for further work in the same geographical area. However there are a few disturbing features of what was otherwise a remarkable set of observations. It is difficult to determine the centre of a crystal several mm in diameter to better than about 100 μm, particularly after it has evaporated and of the many particles found in the central region there is no guarantee that the largest was the nucleus. One experiment which they performed was to seed a supercooled stratus cloud at the site with dry ice and again search for nuclei in the resulting ice crystals. Holroyd et al. (1978) estimated that 1 gram of dry ice falling through a cloud produced 10^{11} ice crystals mainly by homogeneous nucleation in the air chilled below -40°C. Only the miniscule proportion of those originating from the freezing of the cloud drops could have contained clay particles except by subsequent capture, yet five out of the 11 crystals examined had clay particles "at the centre". It is clear that the estimate that 96.3% of the particles were the nuclei on which the ice crystals formed may have been a gross exaggeration.

Other measurements of IFN in the Arctic were far less ambitious, establishing mean concentrations rather than attempting to find the sources or nature of the aerosol. They include: (1) Radke et al. (1976), who found geometric mean concentrations of ice nuclei active at -20°C at Barrow, Alaska (latitude 71°N) to be 20 m^{-3} in March, (2) Flyger and Heidam (1978) at a coastal site at latitude 82°N in Greenland in June and July who found a geometric mean concentration of 17 m^{-3} active at -18°C, (3) Borys (1983) at latitudes 78–83°N over the ocean during the Ymer-80 expedition, who found arithmetic mean concentrations of 40 m^{-3} active at -16°C. At Ny Ålusund, Spitzbergen in February–March 1980 he found 6 m^{-3} active at -16°C. He has kindly supplied the original data so that geometric means can be calculated for comparison with other measurements. They were 12 and 2.3 m^{-3} respectively. (4) Flyger et al. (1973, 1976) who made summer airborne observations at -20°C of IFN to about latitude 70°N, finding concentrations mainly in the range 10–80 m^{-3} at -20°C, (5) Borys (1989) who also made airborne measurements in "the Arctic Haze" in spring, finding concentrations of IN at -15°C to be in the range 0.3–1.4 m^{-3}. 15 out of the 27 filters did not show higher numbers than the blanks.

Each of these measurments used the membrane filter technique but Radke et al. also used a cloud chamber. Its results, without correction for wall effects appeared to be comparable to those of the filters and because it covered a longer time than the filter measurements its geometric mean is the one quoted. The results should be comparable to those of IAOE-91 once allowance is made for the different processing temperatures. To make this allowance simultaneous measurements have to be made at several temperatures and a mean dependence of log concentration on processing temperature calculated. Once this is available all results from a similar time of year can be reduced to a common temperature. Borys (1983) has already done this for a limited sample finding -0.04°C^{-1} in summer and -0.30°C^{-1} in March. These values will be applied to his results and to Radke et al.'s March results.

Comparison of these measurements requires a knowledge of typical dependence of ice crystal formation on temperature and obtaining this was part of the experimental aims. If the above measurements could be reduced to a common temperature, consistency, seasonal effects and any long-term trends could be assessed. Borys (1983) found the mean logarithm of concentration to change by. -0.04°C^{-1} in summer and -0.30°C^{-1} in March.

2. Relevant measurements made on IAOE-91

2.1. Ice forming nuclei

47 mm diameter "Millipore" cellulose acetate-nitrate membrane filters having a nominal pore size of 0.45 μm were used to collect the particles from 300 to 3000 litres of air drawn from the common air intake from which particles larger than 10 μm diameter had been removed by a cyclone at the entrance. The sample intake was about 25 m above the surface. The filters were then placed on oiled brass discs to seal the pores of the filter and

loaded four at a time onto the cold plate of a thermal diffusion chamber. When thermal equilibrium was attained the humidity was maintained just above 100% so that a film of condensation appeared on the filters. After 25 min the ice crystals on the filters were counted. One filter in ten was unexposed for a blank test. Some of the filters were processed immediately after exposure while others were stored in sealed containers in a freezer at −20°C until they could be attended to.

A full discussion of the problems of this technique has been given by Bigg (1990a). In situations where large hygroscopic particles are numerous, a decrease in apparent IFN concentration with increasing sampled volume is its most serious defect. It is thought to be due mainly to reduced humidity at the surface caused by the growing hygroscopic particles, nucleation of ice from the vapour being less probable than for nuclei immersed in, or contacted by, water drops. There is also a self-depletion effect, the first ice crystals to grow depressing the local humidity thus limiting the number of ice crystals that can be formed. On this expedition maximum sample volumes of 3000 litres did not experience serious problems from hygroscopic particles because of their relatively low concentrations, while the coldest temperature used of −17.5°C kept crystal numbers low enough for self-depletion losses to be rare. Another problem is the variable number of "background" ice nucleating sites on the blank filters. These averaged 0.3 per filter at −12.5°C, 1 per filter at −15°C and 3 per filter at −17.5°C. Subtraction of this average value when in fact it is variable and unpredictable causes errors when counts are low. So, too, do the statistics of sampling and measurements in the last days of the expedition suffered from these problems.

When the ship was moving through ice it had to change direction often to follow open leads and self-pollution episodes were frequent. With the continuous measurements of CN (condensation nuclei) and CCN (cloud condensation nuclei) the intervening unaffected measurements often allowed temporal changes to be followed but with the long sampling periods necessary for IFN this was prevented. Concentrations obtained using the pollution free periods only, provided by the CN operated control system, were compared with those of the whole time. No detectable change due to the ship's emissions were found. This had been sufficiently established by 22 August, after which some samples were run continuously regardless of pollution. Shipboard pollution was usually from activities such as cooking and cigarette smoking rather than from the smoke stack.

In planning the measurements it was assumed that north of 80° N long path lengths would ensure only slow variations in concentration and that three filters exposed in parallel for periods of 8 to 24 h would be sufficient to define mean concentrations at 3 different temperatures to be associated with particular air trajectories. The frequent, sudden and often very large changes in CN and CCN concentrations soon suggested that such long averaging times might produce misleading information if IFN concentrations behaved similarly. Accordingly, after 22 August single filters were exposed for the minimum time necessary to obtain a 500 litre sample (about 1 h) as frequently as possible during the special study periods when the ship was stationary and facing into the wind. Most were processed at −15°C. Altogether 72 filters were processed at −12.5°C, 175 at −15°C and 39 at −17.5°C.

2.2. *Associated measurements*

IFN measurements were only a small part of the atmospheric programme. The associated measurements will be described by those who made them in companion papers now in precipitation. They include simultaneous size distributions and concentrations of condensation nuclei (CN) measured from 3 to 500 nm diameter. Those larger than 20 nm diameter which will be referred to in this paper were measured with a programmed differential mobility analyser followed by a Thermosystems Inc. counter, model TSI3025.

Cloud condensation nucleus (CCN) concentrations were found using a continuous flow horizontal plate thermal diffusion chamber in which the lower plate's temperature was cycled to produce supersaturations between about 0.04% and 1% with a 30-min period. The correlation coefficient between particles larger than 41 nm diameter and CCN active at 0.63% supersaturation (equivalent if all particles were composed wholly of ammonium sulfate) was 0.94, so high that for the purposes of this paper it is not necessarily to treat them separately. The CN measurements were more complete and thought to be more accurate.

Counts of alpha particle emissions from radon daughters caught on a filter were monitored at 1 minute intervals and appeared to provide a satisfactory estimate of radon concentrations. Because emissions from a sudden influx of radon daughtors continue with an exponential decay having a half-life of around 50 min, short-lived peaks become broadened. Ozone was measured continuously with a commercial Dasibi system. Dimethylsulfide concentrations in air were obtained from gas chromatography of cryogenically trapped DMS at intervals ranging from 20 min to 1 h when the ship was stationary and facing into the wind.

Half hourly to hourly meteorological observations at a height of 27 m included wind speed and direction, temperature, pressure, relative humidity and notes were made on clouds and precipitation. Radiosonde releases at intervals of about 6 hours while the ship was stationary (Vaisala RS80 and Digicora MW 11 rawinsonde) recorded winds, temperature and humidity aloft, allowing estimates of boundary layer depth and state of mixing.

5-day back trajectories of the air reaching the ship's position at 1000, 950 and 900 hPa at 6-hourly intervals were calculated using a 3-D isentropic model (McGrath, 1989) at the European Centre for Medium-range Weather Forecast, UK.

3. Results

3.1. Changes in IFN concentrations with processing temperature

Few filters exposed in parallel were processed at different temperatures after 15 September, a more continuous record at -15°C being preferred. Those that were made towards the end of the voyage at -12.5°C had so few ice crystals that the results were unreliable.

Mean logarithms L_m of concentration will therefore be considered only for the period 1 August to 15 September and only on occasions of simultaneous measurements at different temperatures T.

From -12.5 to -15°C, $dL_m/dT = -0.11$ and from -15 to -17.5°C, $dL_m/dT = -0.13$. The difference in these two independent estimates is a better guide to their probable errors than standard deviations of individual slopes which are enhanced by the measurement errors on occasions of low

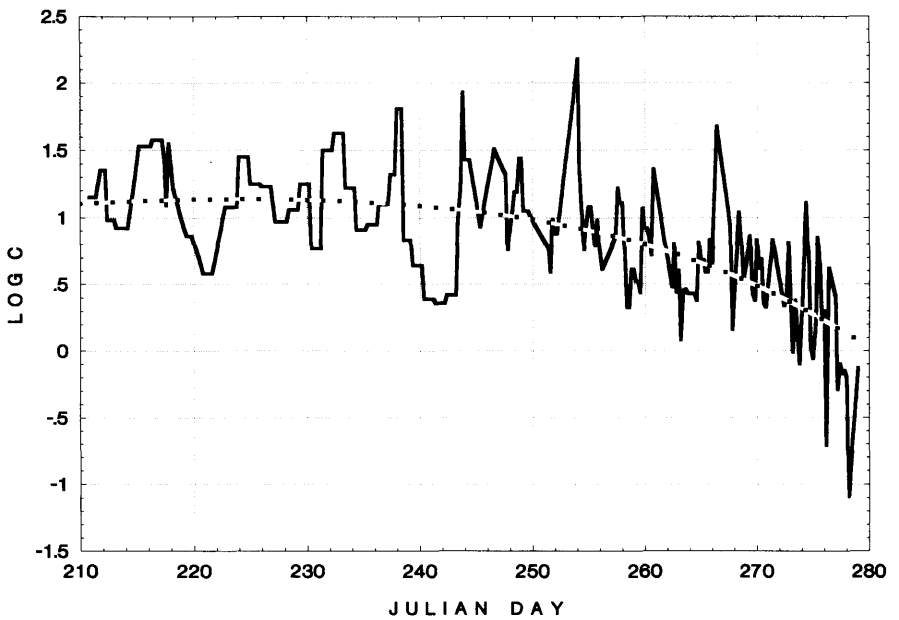

Fig. 1. Time series of mean 0.2 day logarithms of concentrations (C) m^{-3} of IFN active at -15°C during IAOE-91. The dotted line is a least squares fit.

concentrations. Of the two, that for -15 to $-17.5°C$ will be the more reliable because of higher ice nucleus concentrations. It is higher than Borys's (1983) summer slope of $-0.04°C^{-1}$ but lower than typical continental values of about $0.2°C^{-1}$ shown by the collection of graphs given by Pruppacher and Klett (1978).

3.2. *A composite time series of IFN concentrations*

The above values of slope can be used to convert each measurement to an equivalent concentration at $-15°C$. Because filter exposures often overlapped in time and were for very variable lengths of time an artificial series was constructed in which the geometric mean values for each 0.2 day were entered. The penalty lies in introducing errors due to real fluctuations in temperature dependence but this is offset by the benefits of averaging random errors and obtaining a continuous time series for comparison with other parameters.

Fig. 1 shows the time series, the dotted line representing a least squares fit. The overall geometric mean was $(8.2 \pm 2.9)\,m^{-3}$ at $-15°C$. The highest measured concentration of $250\,m^{-3}$ was found on 10 September at latitude 88°N. Several less than $1\,m^{-3}$ were recorded, mainly in October.

3.3. *Seasonal variability*

Fig. 1 shows that during the whole of August, little systematic change in IFN concentrations was apparent but an accelerating decrease began in mid-September. These changes are compared in Table 1 with the geometric means of the few surface observations made north of 70°N listed in the Introduction. They have been converted to $-15°C$ equivalent concentrations using the slope of $-0.13°C^{-1}$ found in Subsection 3.1. Bory's measurements however have been adjusted by the slopes that he found and his March factor of $-0.3°C^{-1}$ applied also to Radke et al.'s measurements which covered much the same period.

Table 1

Observer	Month	Conc. (m^{-3})
Borys (1983)	28 Feb.–11 Mar.	1
Radke et al. (1976)	11 Mar.–25 Mar.	0.6
Flyger and Heidam (1978)	June–July	9
Present work	1 Aug.–3 Sept.	13
Borys (1983)	11 Aug.–1 Sept.	12
Present work	4 Sept.–20 Sept.	10
Present work	20 Sept.–6 Oct.	3

Some order now begins to appear in the very disparate observations, with a midsummer maximum and much lower concentrations in the colder part of the year. For IFN measurements they are surprisingly consistent, probably because the same technique was used by all observers. Concentrations were at all times low compared with values found at 44 lower latitude sites by Bigg and Stevenson (1970) in January to March 1969 which averaged around $60\,m^{-3}$ at $-15°C$. Because interference from hygroscopic aerosols would have been generally greater at these sites, the real difference was probably larger.

3.4. *Effects of ambient air temperature*

Borys (1983) noted an apparent dependence of IFN concentrations on ambient temperature at his Arctic and near-Arctic sites but only at positive temperatures. Fig. 2 investigates this relationship with a least squares fit (dotted) between log concentration (C) m^{-3} of IFN active at $-15°C$ and air temperature at the time of sampling. There is an obvious change in slope near 0°C but unlike Bory's observation it appears to be steep for negative temperatures and slight (or absent) at positive temperatures. Given the seasonal trend evident from Table 1 and Fig. 1, we might suppose this to be a purely seasonal influence. To test this the IFN concentration on day d was multiplied by the ratio of the best fit line of Fig. 1 on day 220 to that on day d and the new values plotted as a function of air temperature. A significant correlation still remained (probability of a change relationship = 0.0003).

3.5. *Evidence of sources of IFN from air trajectory analysis*

The five-day back trajectories of air arriving at the ship's position at the 1000 hPa level were used to calculate the elapsed time since the air was last above the planetary boundary layer, in contact with open ocean, or over land. The values shown in Fig. 1 of the logarithm of the concentration of IFN active at $-15°C$ were then correlated with these elapsed times. For each the correlation was highly significant (probability of a chance rela-

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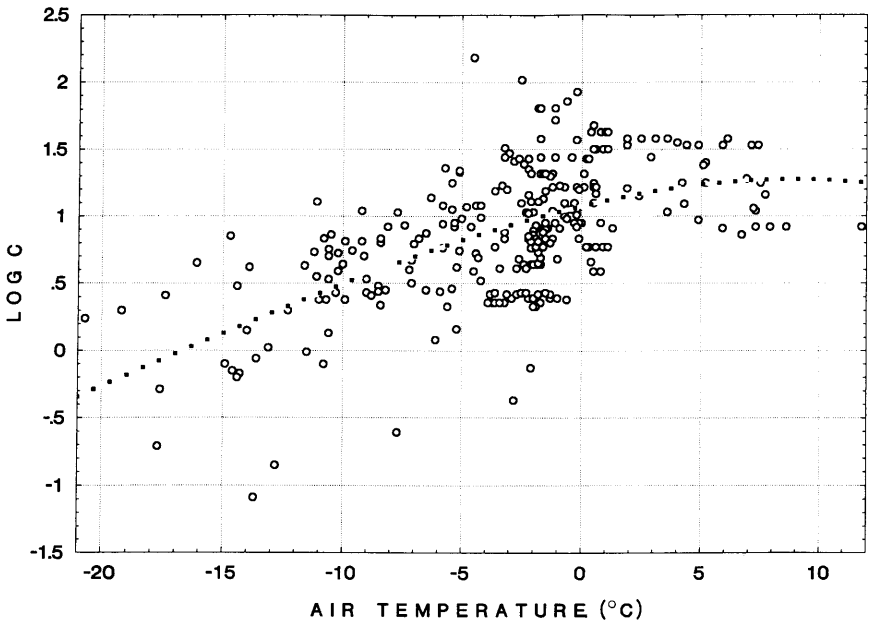

Fig. 2. Logarithms of IFN concentrations (C) m^{-3} active at $-15°C$ as a function of air temperature. The dotted line is a least squares fit.

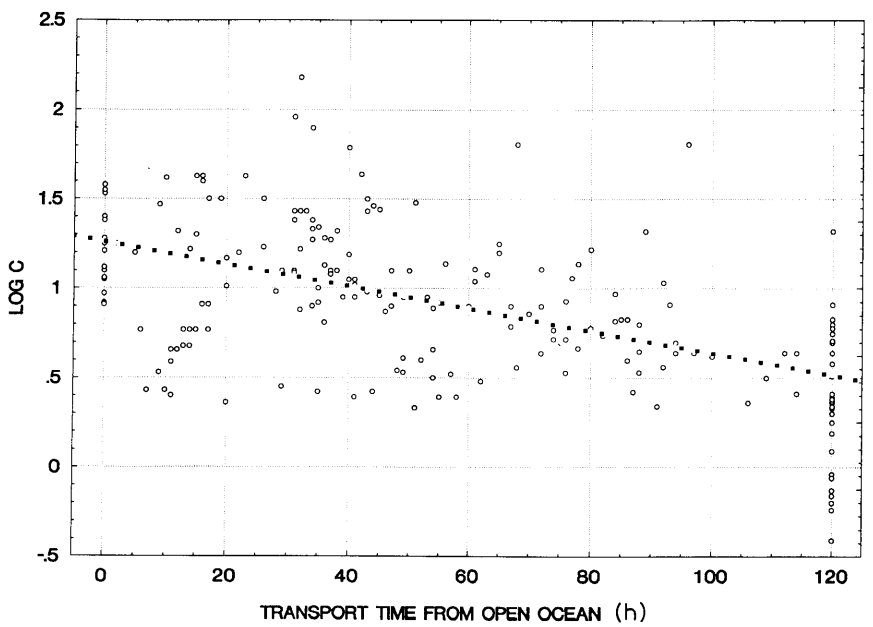

Fig. 3. Logarithms of IFN concentrations (C) m^{-3} active at $-15°C$ as a function of transport time from the open ocean. The dotted line is a linear fit.

tionship $<10^{-4}$) the correlation coefficients being 0.30, −0.54 and −0.28 respectively for 216 pairs of values. The first implies a lower concentration above the boundary layer, the second a strong oceanic source and the third a weaker land source. Fig. 3 shows the relationship between transport time from a marine source and log C, together with a linear fit.

3.6. *Specific events*

On 31 August and 21 September 1991 during special study periods when the ship was stationary and facing into the wind, frequent measurements of IFN were being made at times when very large changes in concentration of condensation nuclei (CN) and cloud condensation nuclei (CCN) were observed. In both cases, the changes in IFN were similar to those in the other particles. IFN and accumulation mode CN (diameters >80 nm) concentrations are shown for the 21 September event in Fig. 4.

Radiosondes released during this period showed an inversion to a height of 80 m at the beginning of this event, then its disappearance with mixing to the unusually high level of 500 m, followed by a restabilization of the lower layers. The simple interpretation is that the cloudy layers not far above the measuring point at the start of the observations were depleted in CN, CCN and IFN and the mixing caused the sudden decrease in concentrations. A similar interpretation appears appropriate for the 31 August event. The time resolution of IFN measurements was insufficient to follow the more common sudden changes in CN and CCN concentrations that lasted only an hour or two.

The only event for which a particular land source was strongly indicated was on 22 September (the day following Fig. 4) when 7 successive filters registered concentrations above 20 m^{-3} at a time when Fig. 1 indicates a mean of about 4 m^{-3}. At the same time CN, CCN and radon concentrations climbed to their highest values since early August. Air trajectories suggested that rapid transport of air from the industrialised Kola Peninsula of the Russian Arctic was involved.

While air that had recently been above the planetary boundary layer was usually deficient in

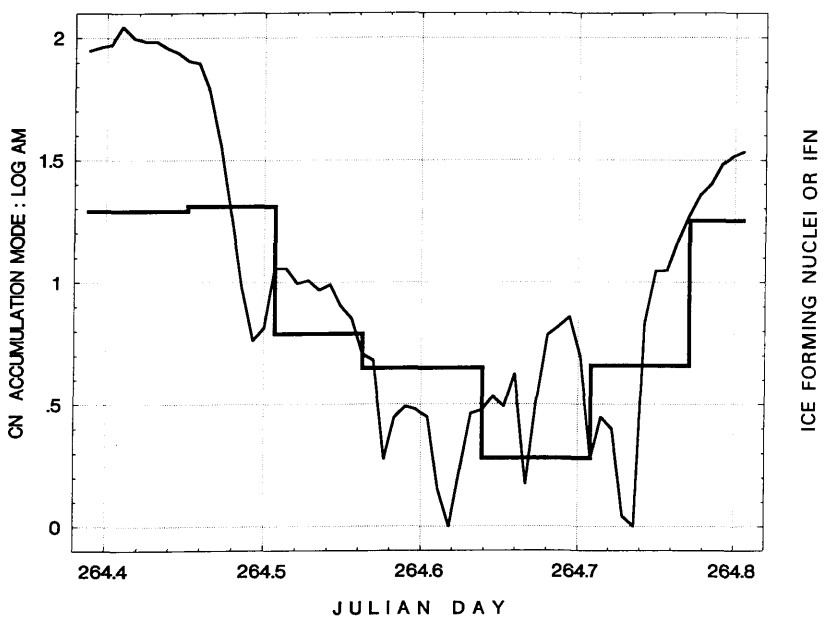

Fig. 4. Changes in the logarithm of concentrations (AM) of accumulation mode condensation nuclei (diameters >80 nm) and IFN (C) m^{-3} active at −15°C (stepped values) during an event in which the depth of mixing increased on 21 September 1991. The ship's position was 85° N, 6° W.

IFN, there was a notable exception on 1 October when four successive filters showed concentrations 5 times the least squares fit for that date shown in Fig. 1. Marine and land sources were more than 80 h distant, while it was only 30 h since the air had subsided into the planetary boundary layer.

4. Discussion

4.1. The open ocean as a source of ice nuclei

The highly significant negative correlation ($r = -0.54$) of IFN concentrations with time since the air was over open ocean (Fig. 3) implies a strong oceanic source for high Arctic ice nuclei in the summer season. The source of radon is unequivocally the land, yet correlation between radon concentrations and the elapsed time since the air was over land was less ($r = -0.45$). The slope of the regression line in this correlation can be used to calculate the halflife of radon. It turns out to be 84 h, about 9% less than the known value. We can calculate the halflife of IFN from the regression line in Fig. 3 in the same way with expectations of a similar error. A value of 48.5 h, equivalent to an e-folding time of 70 h is obtained, about 60% longer than the similarly derived halflives of CN between 25 and 80 nm in diameter (31 h) or accumulation mode CN of 80–330 nm diameter (32 h). The difference, if real, might imply either that loss mechanisms are less for IFN (for example, they may not be involved in condensation) or that there are continuing production mechanisms. The fact that IFN concentrations varied less than those of accumulation mode CN in the mixing event illustrated in Fig. 4 is consistent with this difference in mean residence times.

4.2. Some possible sources of oceanic ice nuclei

Because no attempt was made to examine the nuclei in ice crystals the nature of the oceanic source of ice nuclei on this expedition cannot be determined. It may however be useful to list some possibly relevant published observations even though their applicability to the present observations is purely speculative.

(1) The observations of Schnell and Vali (1975, 1976) and Schnell (1977) of strong ice nucleating ability of seawater containing phytoplankton potentially provide a very direct source if we assume that the bubble bursting mechanism discussed by Cipriano and Blanchard (1981) is capable of making some of the ice nucleating particles airborne. They also showed that the southern hemisphere oceanic distribution of IFN reported by Bigg (1973) was consistent with regions of high biological productivity. Rosinski et al. (1986) also examined the oceanic distribution of IFN confirming this finding and showing in addition that the airborne IFN in these regions were smaller than 0.5 μm in diameter and seemed to consist mainly of organic matter. Very low concentrations were found over less productive regions. The concurrent measurements of dimethylsulfide in air during IAOE-91 (Leck and Persson, 1996) indicated that phytoplankton were prolific near the ice edge in August. DMS concentrations also showed a seasonal decline but it started earlier than that of IFN. There was, of course, a strong correlation between DMS and IFN concentrations because of their common oceanic origin and seasonal trends.

(2) Kumai and Francis (1962), as discussed in Section 1 established that clay or mineral particles were present in most ice crystals in Greenland, though it was not clear what proportion of these were the IFN. It was obvious from inspection of sea ice near the ice edge that Siberian rivers had added a considerable amount of mud to the Arctic seas. It would be possible to invoke the bursting bubble mechanism again to explain at the one time the oceanic source indicated by IAOE-91 and the Kumai and Francis result. We could further use Schnell's (1981) suggestion that some clays owe their ice nucleating ability to attached biological debris to reconcile the results with (1) above.

(3) Beard (1992) has reviewed the evidence for "evaporation nuclei", concluding that evaporation residues of cloud droplets may nucleate ice because of an organic shell which allows hydrogen bonding with anchor sites. The almost permanent cloud cover during IAOE-91 would certainly have allowed the opportunity for such evaporation nuclei to occur given the presence of suitable organic material.

(4) A specific mechanism for (3) is suggested by the discovery of Gavish et al. (1990) that monolayers of long chain alcohols and to a lesser extent, acids, have the ability to nucleate ice at temperatures quite close to zero. The nucleating

temperature is lower for shorter chain alcohols or acids. We could speculate that such compounds may occur in the surface film on the ocean and become airborne through the bursting bubble mechanism. Long chain alcohols and acids have been found in detectable quantities in marine atmospheres both in the vapour phase and attached to aerosols (for example, by Duce et al., 1983).

4.3. The continental source

The high IFN concentrations associated with rapid transport of air from an industrial source (Subsection 3.6) shows that continental sources can at times provide a substantial contribution to the IFN population. The fact that the correlation coefficient between IFN concentrations and transport time from land was considerably less than that from the open ocean suggests that the land is at most a weak source of particles except for infrequent influxes such as that on 22 September. Correlations between land-sourced radon and IFN concentrations were barely significant. Both of course will be influenced by mixing events due to vertical gradients in concentration but probably in different ways since gaseous radon will not be lost to the surface by precipitation processes.

4.4. Secular changes

Bigg (1990b) noted an apparent large decrease in mean IFN concentrations at $-15°C$ in sub-Antarctic waters and continental Antarctica in 1988–1989 from those found between 1961 and 1973. Further unpublished measurements made on two voyages in 1989–1990 showed the same difference. Table 1 suggests that there has been no similar change in the Arctic.

4.5. Effects of air temperature

The residual relationship described in Subsection 3.4 between IFN concentrations and air temperature once the results have been adjusted for the mean seasonal change (Fig. 1) is probably related to the air trajectories that cause low temperatures. After polar sunset, air coming from the vicinity of the pole was very cold. It had also spent a long time without contacting open ocean or ice-free land so that its IFN content would have been greatly depleted.

5. Observations of ice crystals

North of about latitude 80°N, individual ice crystals were observed frequently, often in such low concentrations that they would generally have been overlooked. Sufficient snow to cover the decks occurred only occasionally and consisted of crystal aggregates. The forms of individual crystals were examined on 43 occasions on 23 separate days after collection on glass slides or a black cloth and examined at $10\times$ magnification. 79% were found to be stellar, the great majority being non-dendritic with diameters 2 to 10 mm, 6 mm being typical. This implies that most of the crystals grew in water clouds at a temperature close to $-15°C$ and their usual very low concentration was consistent with the generally low ice nucleus concentrations. A snowfall of aggregated stellar crystals occurred on 10 September, the day with the highest IN concentration of the entire voyage ($250\ m^{-3}$).

On other days with snowfall the ice nucleus concentration was far too low to explain the observed concentration of ice crystals. Possible reasons are: (1) the IFN activated by the membrane filter method were not representative of those active in the cloud; (2) a strong vertical gradient in IFN concentration was present; (3) the lower clouds were seeded by crystals from colder clouds; (4) Ice multiplication mechanisms were operative.

On only one occasion was it possible to make an IFN measurement in a cloud cold enough for ice crystals to form. A thin fog formed at about 16 h on 2 October at a temperature of about $-18°C$, shortly after a measurement of IFN at $-17.5°C$ had yielded a concentration of $4.5\ m^{-3}$. By 17 h, a few hexagonal plate crystals had become visible, certainly not in greater concentrations than this.

The conclusion is that on most occasions the ice nucleus measurements were consistent with ice crystal concentrations in this environment but there were exceptions, when ice crystals were far more numerous than expected from IFN concentrations in the near-surface air.

6. Conclusions and recommendations for future research

The most interesting results of this work concerned the origin of the summer Arctic IFN, but as is usual with IFN experiments, the actual nature of

the nuclei remains elusive. The dominance of the open ocean source was surprising in view of the Kumai and Francis (1962) demonstration that clay or mineral particles were present in most ice crystals. However, there is a possibility that the two observations can be reconciled and testing this should be one aim of future research.

The deduction that residence times of IFN in the atmosphere are longer than for CN or CCN is interesting and needs confirmation because it could provide some indirect information on the nature of the nuclei, for example whether they were hygroscopic or hydrophobic.

Comparison of the present results with early work has shown a feature unusual in IFN studies: a good degree of consistency in mean concentrations. This must in part be due to the fact that all used a similar technique and does not necessarily mean that the results will give an accurate indication of the number of ice crystals that would form in clouds. However, the observations of ice crystals falling to the surface suggested that the measurement was usually (but not always) of the right order of magnitude. That has not been a common experience in lower latitudes. Changes in concentration of CN and CCN induced by sudden mixing events in the Arctic and the demonstration that IFN concentrations react similarly show up one great need for future research, better time resolution. Collection of the particles on a continuously moving membrane filter tape is probably an easier option than the use of a continuous flow cold chamber for this purpose because of the problems of ice build-up and spurious crystals in cold chambers. Vertical profiles of IFN would be highly desirable but difficult to organize because of the time required to sample volumes of the order of $1\ m^{-3}$.

It would be very valuable to repeat the experiments of Kumai and Francis (1962) and to search specifically for the sort of mineral particles that they found to be so common in ice crystals and to look at their ice nucleating properties. The frequent occurrence of large individual non-dendritic stellar crystals would make collection relatively easy in this environment.

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