

Cloud Condensation Nuclei over the Arctic Ocean in Early Spring

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ABSTRACT

Cloud condensation nucleus (CCN) spectral data are presented for the Arctic in spring, which considerably augment the existing meager CCN database for the Arctic. Concurrent measurements of sulfate mass suggest that most of the CCN were commonly not sulfate. Sulfate was more closely associated with particles below the CCN size range. Some measurements of the microphysical structure of Arctic stratus clouds are also described.

1. Introduction

Modeling studies of the earth's climate system suggest that the Arctic is particularly susceptible to climate change. For example, climate models subjected to a doubling of CO₂ show the Arctic warming by approximately 10 K due, in part, to a positive feedback associated with the melting or thawing of sea ice (Schlesinger 1986; Walsh and Crane 1992). Such a large temperature perturbation in the Arctic would produce a significant reduction in the latitudinal temperature gradient of the earth, which would no doubt result in important changes to the global atmospheric general circulation. A strong modulator of the positive feedback in the Arctic, and of the Arctic heat balance in general, is the degree of cloudiness of the arctic atmosphere (Tsay et al. 1989). This cloudiness, in turn, is modulated by the concentration of cloud condensation nuclei (CCN) as discussed, for example, by Twomey (1993).

Little data have been reported on CCN in the Arctic. Hoppel et al. (1973) reported four vertical soundings of CCN taken over the Yukon Valley during February. These soundings showed an increase in CCN with altitude above 2 km, possibly due to the long-range transport of aerosol or to in situ formation aloft as suggested by Clarke (1992). The concentration of CCN active at 1% supersaturation ranged from 90 to 600 cm⁻³. In contrast, measurements made over the Arctic Ocean in June by Saxena and Rathore (1984) showed CCN concentrations at 1% supersaturation in the boundary layer on the order of 10³ cm⁻³, and decreasing with increasing height above the boundary layer. The high levels of CCN (and CN) measured by Saxena and Rathore were likely due to long-range transport of pollutants into the Arctic Basin.

The composition of CCN in the Arctic is poorly characterized. Previous studies have suggested that most submicron particle mass in the Arctic is sulfate (e.g., Radke et al. 1984; Shaw 1986). Shaw (1986) presented data from individual particle analysis that further suggests that most of the particles—in a number sense—contain sulfate. Nevertheless, such data cannot unequivocally support an assertion that Arctic CCN are predominantly sulfate, since CCN are a subset of the total particles and therefore do not necessarily dominate either the number or mass concentration of particles. Even if most CCN were sulfate, it is important to note that particulate sulfate could have two distinct sources over the Arctic Ocean: long-range transport of anthropogenic pollution (cf. Shaw 1986) and in situ production from locally emitted DMS after open water forms in the Arctic Ocean in late spring (Shaw and Yan 1990; Li and Barrie 1993; Ferek et al. 1994). These two sources might be expected to yield different sulfate size distributions and consequently different CCN-sulfate mass relationships. Hence, the overall CCN-sulfate mass relationship could be quite variable. The limited extant work on the origin of sulfate mass in the Arctic (cf. Li and Winchester 1990) has concentrated on the aerosol mass budget rather than the CCN number budget. Therefore, it does not yield useful data on the origins of the CCN, or of possible variability in the CCN-sulfate mass relationship.

2. Database

We obtained measurements of CCN over the Arctic Ocean, within approximately 350 km of Deadhorse, Alaska, in April 1992 during the Arctic Leads Experiment (LEADEX). Our database consists of 41 CCN activation spectra, taken during seven research flights, and simultaneous measurements of SO₄⁻ at altitudes between about 0.1 and 4 km and Aitken nucleus (hereafter CN) concentrations at altitudes below 1.6

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km. (Reliable CN data were not obtained above 1.6 km due to the freezing of a vacuum regulation valve on the CN counter.) Some concurrent measurements of the aerosol size distribution were also made.

The CCN measurements were obtained with the CCN spectrometer described by Radke et al. (1981), which consists of a set of four continuous flow diffusion chambers, each operated at a different supersaturation. The SO_4^- measurements were determined via filtration and postflight ion chromatography analysis, as described by Ferek et al. (1991). The CN concentration was measured with a modified General Electric CN counter, as described by Schnell et al. (1989). Particle size distributions reported here were measured with a differential mobility particle sizing system (DMPS) (Reischl 1991; Hegg et al. 1993). Cloud microphysical measurements were obtained with a Johnson-Williams liquid water meter, a Particle Measuring System (PMS) FSSP-100 cloud droplet spectrometer, a PMS 1-D cloud probe, and an in-house ice particle detector (Turner and Radke 1973).

3. Results

a. Cloud condensation nuclei

The concentration of CCN active at supersaturation S (%) can often be represented by a power function of the form, $\text{CCN} = c(S)^k$ (Twomey 1959), where c is the concentration of CCN active at 1% supersaturation and k is a slope parameter. Values of c and k from our measurements in the Arctic are listed in Table 1. Measurements are, in general, reported only when the data could be regressed to the standard power function with a nonlinear correlation coefficient r^2 exceeding 0.5 (i.e., at least half of the data variance could be explained by the regression function). An exception is made for the CCN data of 27 April ($0.3 \leq r^2 \leq 0.4$) since we will use this data for comparison with cloud microphysical measurements obtained on this flight. However, the CCN data from 27 April are not used in the statistical analyses.

The values of k range from 0.26 to 1.39, with a mean value of 0.71 ± 0.29 . Concentrations of CCN active at 1% supersaturation (c) range from 19.9 to 92.7 cm^{-3} , with a mean of $47 \pm 19 \text{ cm}^{-3}$. The CCN concentrations are quite low compared to the limited previous measurements in the Arctic, but they are consistent with our simultaneous measurements of CN, which are also quite low. The low concentrations are also consistent with the generally low levels of haze present in the Arctic during 1992 (Sheridan et al. 1992).

Below an altitude of 1.6 km, the CN concentrations ranged from 135 to 17 000 cm^{-3} . However, the latter value appears to be an anomaly, possibly associated with a combustion plume. If this anomalously high value is eliminated, the CN concentrations below 1.6 km ranged from 135 to approximately 4600 cm^{-3} , with a mean value of $648 \pm 1185 \text{ cm}^{-3}$. The fraction of CN

that were CCN active at 1% supersaturation (i.e., c/CN) ranged from 0.002 to 0.38, with a mean value of 0.15 ± 0.08 . This fraction is at the low end of the range of values for c/CN in marine air near Australia and off the coast of Washington State summarized by Hegg et al. (1991). Data on the chemical compositions of arctic particles (e.g., Cahill and Eldred 1984; Hoppel et al. 1973) show no systematic differences in solubility compared to particles in other airsheds; therefore, the relatively low fraction of particles that act as CCN over the Arctic Ocean suggests that the mean size of the particles in this region may be smaller than elsewhere. This suggestion is in accord with our light-scattering measurements over the Arctic Ocean, which were low even when CN concentrations were quite high.

The CCN data reported here add substantially to the database on arctic CCN. When compared to previous measurements they also suggest seasonal variations in CCN concentrations. For example, for altitudes below 1.6 km, data collected over the Yukon Valley, Alaska, in February by Hoppel et al. (1973) show CCN concentrations at 1% supersaturation of about 100 cm^{-3} , which is comparable to, although somewhat higher than, the measurements we obtained in April. In contrast, measurements made by Saxena and Rathore (1984) in late June over the Arctic Ocean north of Barrow suggest CCN concentrations at 1% supersaturation of approximately 1000 cm^{-3} . Long-range transport of anthropogenic pollutants into the Arctic is generally thought to be most frequent in the late winter and early spring (cf. Brock et al. 1990 and references therein). The high CCN concentrations in June measured by Saxena and Rathore, together with trajectory analyses reported by them, suggests that there may also be long-range transport of CCN into the Arctic in summer. On the other hand, it is also conceivable that high CCN concentrations in the Arctic in June are associated with high DMS fluxes, as has been found to be the case in other locales (e.g., Ayers and Gras 1991). Recent measurements in the Arctic (Ferek et al. 1994) suggest such DMS emissions and associated particles can come from ice leads. This issue warrants further study.

Another interesting facet of our CCN data is the indication of some vertical structure. Figures 1 and 2 show plots of c and k , respectively, against altitude. Distance-weighted least squares fits (DWLS) to the data points suggest local maxima aloft in both variables. However, the DWLS fits are suspect at altitudes much above 3 km because of a sparsity of data points and it is not clear that a maximum is actually present in either plot. On the other hand, both k and particularly c are significantly correlated with altitude (for c , $p = 0.005$; for k , $p = 0.15$). This is suggestive of in situ particle production aloft (e.g., Hegg and Hobbs 1992) coupled with a surface particle sink such as dry deposition. While, in fact, a surface sink alone could yield profiles similar to those observed, some support for the idea of particle generation aloft is provided by measurements

TABLE 1. CCN measurements and ancillary data from seven research flights over the Arctic Ocean.

Date (1992)	Altitude (km)	k	c (cm ⁻³)	CN (cm ⁻³)	c/CN	SO ₄ ⁻ (μg m ⁻³)
19 April	4.0	0.39	67.2	—	—	0.17
	0.6	0.26	30.4	184	0.17	0.58
21 April (A.M.)	2.2	0.30	47.5	428	0.11	—
	3.2	0.46	32.6	—	—	—
	3.9	0.49	30.3	—	—	—
	0.1	0.33	21.0	135	0.16	0.55
21 April (P.M.)	0.2	0.40	37.5	215	0.17	0.75
23 April	2.8	0.86	43.3	—	—	0.59
	2.8	0.89	40.8	—	—	0.59
	2.8	0.77	69.8	—	—	0.59
	2.8	1.03	51.9	—	—	0.59
	1.5	0.94	53.8	311	0.17	1.25
	0.3	0.74	37.8	231	0.16	—
	0.2	0.98	53.1	2235	0.02	—
	0.2	1.02	48.5	4606	0.01	—
	0.1	0.61	31.1	244	0.13	—
	0.03	1.16	64.2	3146	0.02	—
24 April (A.M.)	3.8	1.38	42.9	—	—	—
	3.0	1.27	46.0	—	—	—
	2.5	1.39	49.0	—	—	0.70
	2.5	0.85	44.7	—	—	0.70
	1.6	0.40	35.0	163	0.21	0.19
	1.6	0.45	37.0	167	0.22	0.19
	1.6	0.91	46.9	186	0.25	—
	1.0	0.97	36.8	241	0.15	—
	0.03	0.43	19.9	138	0.14	0.45
	0.03	0.38	23.8	149	0.16	0.45
24 April (P.M.)	0.43	0.55	31.2	188	0.17	0.89
	0.43	0.52	39.7	190	0.21	0.89
	0.15	0.51	33.1	289	0.11	0.73
	0.15	0.70	26.2	180	0.15	0.73
	0.03	0.56	18.8	193	0.10	0.63
	0.03	0.60	23.4	151	0.15	0.51
26 April	3.3	0.62	76.2	—	—	0.14
	3.3	0.74	82.4	—	—	0.14
	2.2	0.71	67.3	—	—	—
	2.2	0.65	76.0	—	—	—
	2.2	0.60	80.3	—	—	—
	2.2	0.77	78.9	—	—	—
	1.4	0.46	92.7	246	0.38	—
	0.2	0.95	38.0	17 000	0.002	—
27 April*	2.3	0.36	122.2	—	—	—
	1.7	0.34	128.3	—	—	1.26
	0.4	0.24	34.5	100	0.35	0.84

* Not used in correlation analysis; see text.

of the particle size distributions. Figures 3 and 4 show DMPS spectra typical of those encountered during the present study. The spectrum measured at the higher altitude on each flight shows particle concentrations rapidly increasing near the lower bound of the measurement range, which is suggestive of the presence of a nucleation mode. In both cases, the enhanced concentrations of small (compared to large) particles at higher altitudes is consistent with the corresponding increase of k with altitude. However, the dataset is not

definitive and points to the need for more measurements.

Listed in Table 2 are the correlation coefficients between sulfate mass and CCN, CN, and k . For unbiased comparisons, the values of r shown in Table 2 are based on those cases for which measurements of all variables were available (which reduced the number of datasets to 10). This correlation analysis shows that SO₄⁻ mass becomes more poorly correlated with the aerosol number concentration at a given activation supersaturation

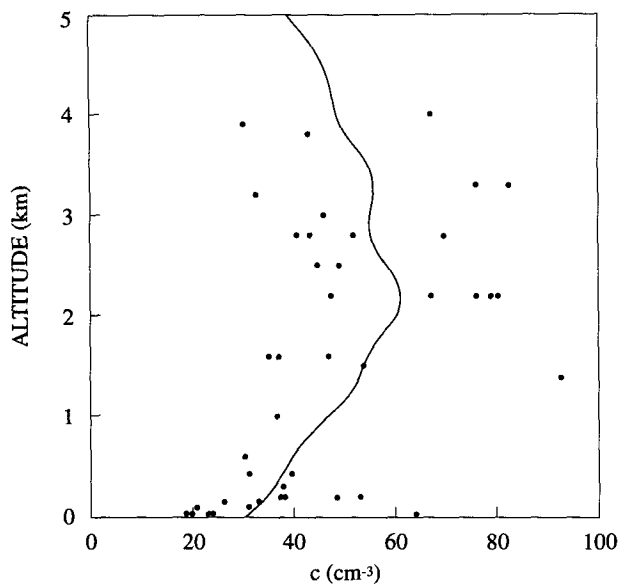


FIG. 1. Concentrations of CCN active at 1% supersaturation c versus altitude. The curve shown is a distance-weighted least squares fit to the data points.

(S) as S decreases (note that CN are simply particles activated at the approximate 400% supersaturation of a CN counter). Hence, the sulfate mass appears to be associated with the smaller particles. From the standpoint of CCN composition, this suggests that most of the CCN active at supersaturations present in Arctic stratus clouds (i.e., less than approximately 1%) are

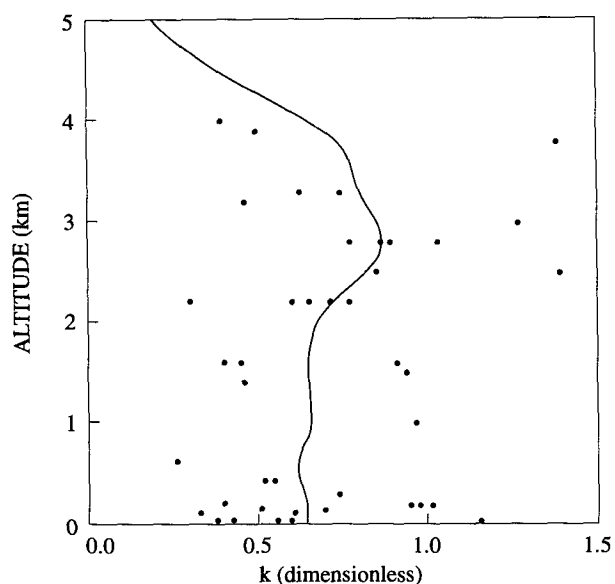


FIG. 2. Values of k in Eq. (1) versus altitude. The curve shown is a distance-weighted least squares fit to the data points.

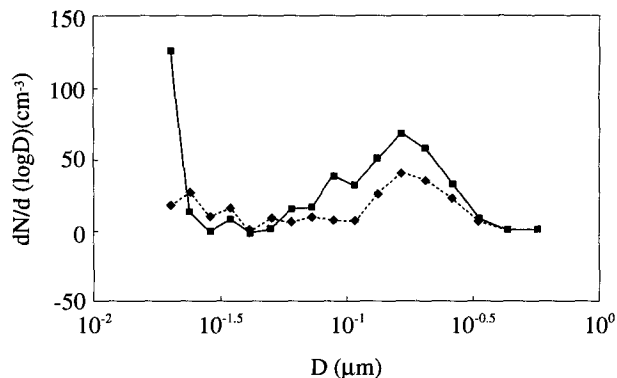


FIG. 3. DMPS size distributions taken on 23 April 1992 at altitudes of 0.1 km (diamonds) and 1.5 km (squares).

not sulfate. For example, a regression analysis of c onto sulfate mass for the 10 cases on which the correlations shown in Table 2 are based yields the following regression equation:

$$c = (23.9 \pm 10.1)SO_4^- + (15.2 \pm 7.2), \quad (r^2 = 0.41)$$

where c is the count per cubic centimeter and SO_4^- is in micrograms per cubic meter. For the sulfate levels encountered in this study ($0.14\text{--}1.25 \mu\text{g m}^{-3}$, with a mean value of $0.58 \pm 0.30 \mu\text{g m}^{-3}$), typically less than one-half of the CCN active at 1% supersaturation would be due to sulfate. Similar results have been obtained in warmer marine environments (cf. Hegg 1994). This suggests that nonsulfate species are of great importance for the CCN budget in the Arctic in spring. One likely such species, or group of species, is the organics. For example, Novakov and Penner (1993) found that organics appear to make a significant contribution to CCN in marine air masses in the Caribbean.

b. Cloud microphysics

During the course of our research flights, Arctic stratus clouds were encountered on several occasions. In

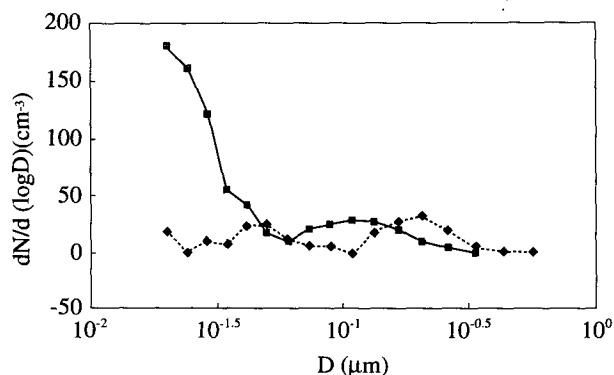


FIG. 4. DMPS size distributions taken on 24 April 1992 at altitudes of 0.03 km (diamonds) and 2.5 km (squares).

TABLE 2. Linear correlation coefficients r between SO_4^- mass and CCN, CN, and k for the airborne measurements over the Arctic Ocean.

Variables	r
SO_4^- and CCN ($S = 0.3\%$)	0.046
SO_4^- and CCN ($S = 1.0\%$) = c	0.640
SO_4^- and CN	0.832
SO_4^- and k	0.697

general, the clouds were very tenuous, with liquid water contents less than 0.1 g m^{-3} . For example, on 24 April 1992 the mean liquid water content was only $0.03 \pm 0.01 \text{ g m}^{-3}$ and the mean cloud drop concentration was $30.2 \pm 6.0 \text{ cm}^{-3}$. These measurements are consistent with the low concentrations of CCN measured on this flight (see Table 1). Indeed, comparison of the CCN spectra with the cloud drop number concentration suggests that the relative lack of CCN resulted in a rather high supersaturation of about 1%. The cloud drop size distribution is shown in Fig. 5.

In contrast to these typical Arctic clouds, on 27 April 1992 a more substantial stratiform deck was encountered. This cloud, with a base at 170 m and a top at 600 m (located at the base of a 5°C temperature inversion), had liquid water contents near cloud top exceeding 0.2 g m^{-3} . Vertical profiles of liquid water content, cloud drop number concentration, cloud drop effective radius, and ice particle (exceeding about $200 \mu\text{m}$ in diameter) concentration through the cloud deck are shown in Figs. 6–9. It is interesting to note that, while the liquid water content increases nearly mono-

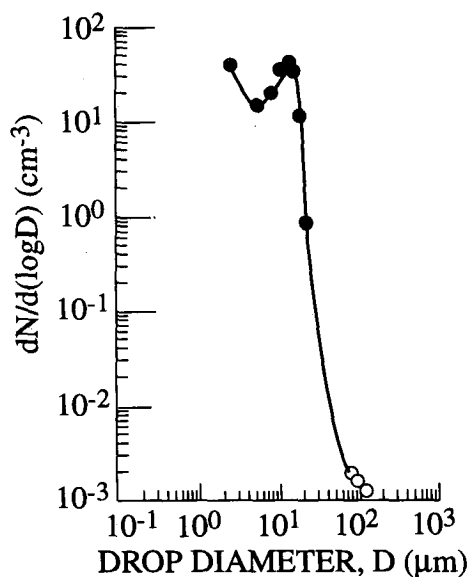


FIG. 5. A cloud drop size distribution measured in arctic stratus cloud. Filled circles are data points from the PMS FSSP-100 probe and open circles from the PMS 1-D cloud probe.

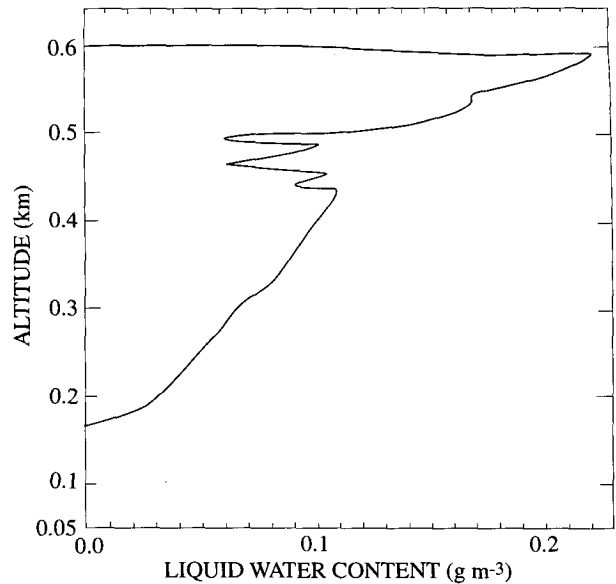


FIG. 6. Vertical profile of the cloud liquid water content for the stratus deck of 27 April 1992.

tonically from cloud base to cloud top, the cloud drop concentration is essentially constant. Hence, the cloud drop effective radius also increases monotonically with altitude (Fig. 8). This structure is similar to the observations of Tsay and Jayaweera (1984), as is the fact that the liquid water content is near adiabatic except for a thin layer ($\sim 40 \text{ m}$ thick) just below 500 m. On the other hand, the cloud droplet size distribution that

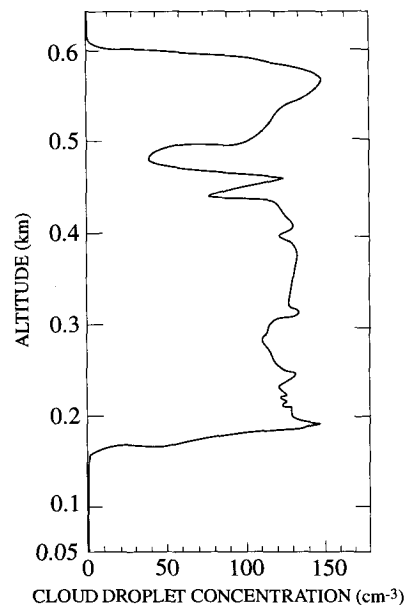


FIG. 7. Vertical profile of the cloud drop number concentration for the stratus deck of 27 April 1992.

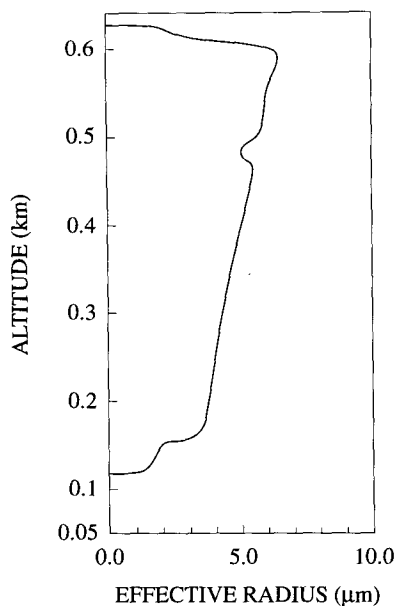


FIG. 8. Vertical profile of the cloud drop effective radius* for the stratus deck of 27 April 1992.

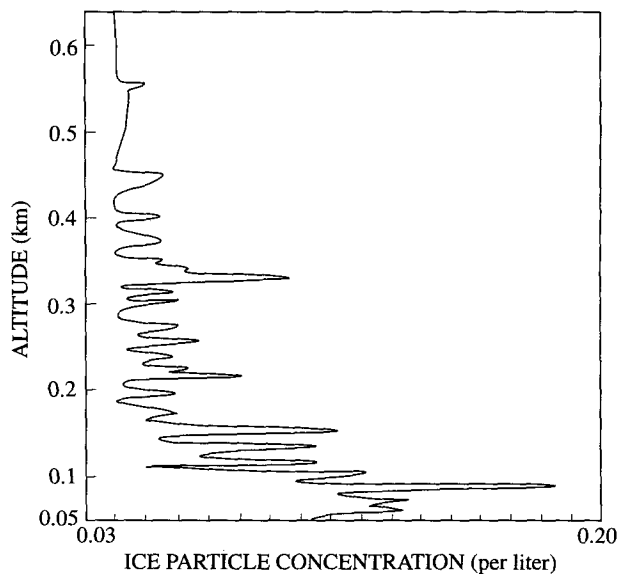


FIG. 9. Vertical profile of the ice particle concentration ($\geq 200\text{-}\mu\text{m}$ diameter) for the case of 27 April 1992.

we measured on 27 April 1992 differs from that reported by Tsay and Jayaweera in that it was monomodal over the entire cloud depth (Fig. 10). This last result is more in accord with the observations of Curry (1986), who found little evidence of inhomogeneous mixing, which Tsay and Jayaweera postulated to explain the bimodal droplet spectra they measured near cloud top.

The anomaly in liquid water content at about 500 m alluded to above is of some interest. Both the liquid water content and droplet number concentration were depressed at this altitude. The phenomenology is suggestive of a breakup of the cloud into two layers due to solar heating in the cloud interior, as hypothesized by Herman and Goody (1976). Curry (1986) documented a more mature case in which the separation into distinct layers had already taken place. The present case appears embryonic.

Another interesting facet of the cloud microphysics is the distribution of ice particles, which were at least partially displaced spatially from the cloud drops, with the highest concentration of ice particles situated below cloud base (as defined by the liquid water content going to zero). Similar structures have been observed in middle latitude clouds, where it has been associated with ice nucleation at cloud top followed by sedimentation and growth of the ice particles (Hobbs and Rangno 1985). However, the ice phase made a negligible contribution to both the mass and number of particles in this cloud.

The relationship between the cloud microstructure and the precursor CCN concentration is noteworthy. The cloud drop concentration of approximately

120 cm^{-3} is roughly equivalent to the values of c (CCN concentration at 1% supersaturation) shown in Table 1 for this flight. This strongly suggests a cloud supersaturation of about 1% was achieved in the 27 April cloud, which is in accord with the 24 April case. Such supersaturations are surprisingly high and no doubt arise, in part, from the scarcity of large particles found in the Arctic atmosphere during this measurement period. This paucity of particles, coupled with the formation mechanism of these clouds, which involves longwave radiative cooling by the cloud drops them-

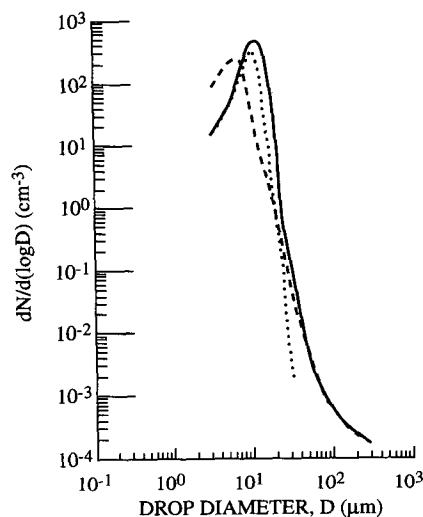


FIG. 10. The cloud drop size distribution in the stratus deck taken on 27 April 1992 at altitudes of 150 m (dashed line), 450–500 m (dotted line), and 500–600 m (solid line).

selves (which is strongly dependent on the number and size of the cloud drops), suggests that these clouds may be unusually sensitive to small perturbations in the CCN concentration (cf. Ackerman et al. 1993).

Although the drop concentration and liquid water contents that we measured in April in Arctic stratus clouds are somewhat lower than those reported by Tsay and Jayaweera (1984) for stratus clouds in the Arctic in summer, the modal drop diameters that we measured ($\sim 11\text{--}13\ \mu\text{m}$) are generally somewhat higher. Also, the values for the concentrations of cloud drops reported here are lower than measurements in an arctic stratus cloud in April reported by Hobbs and Rangno (1985). Curry and Ebert (1992) argued that stratus clouds play a significant role in the radiation balance (based on the small extant database) of the Arctic. The results presented here suggest considerable variability in the microstructure (and therefore optical depth) of arctic stratus clouds. Given the sensitivity of such key parameters as sea-ice thickness to cloud drop radii and number concentrations (Curry and Ebert 1992), the database on the microphysical characteristics of arctic stratus clouds clearly needs to be considerably enhanced.

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