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Sudden changes in arctic atmospheric aerosol concentrations during summer and autumn

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ABSTRACT

The International Arctic Ocean Expedition of 1991 measured number concentrations of condensation nuclei and cloud condensation nuclei from 1 August to 6 October between latitudes 70° N and 90° N. Changes in concentration of more than a factor of 2 in 1 h or less were frequent in spite of the long distances from significant sources and often appeared in groups separated by times of the order of 1 h. These observations provide a unique opportunity to study interacting meteorological mechanisms in the stable marine boundary layer. It is suggested that most of these changes were due to two factors: (1) large vertical concentration gradients caused mainly by interaction of aerosol and clouds and (2) intermittent localised mixing into the shallow surface mixed layer, caused by atmospheric wave motions or roll vortices. Quasi-periodic changes of smaller amplitude were present for about 35 % of the total time with mean periods in the range 60–90 min. About 1/6 of the sudden large changes, usually involving an isolated decrease and recovery in accumulation mode particle number concentrations, is attributed to mixing caused by shear-induced Kelvin–Helmholtz breaking waves. Regular sequences of maxima and minima were about as common but involved 2/3 of all such changes because of the number of events in each sequence. Satellite images, radiosonde wind profiles and the typical mean periods suggested that roll vortices were common in the high Arctic. The associated mixing changes seemed to account adequately for the alternating maxima and minima in aerosol number concentrations. The special conditions of atmospheric stability and high frequencies of low cloud cover in the high Arctic combined with the absence of strong local sources of particles have allowed us to identify processes of mixing that often drastically modified aerosol size distributions and concentrations near the surface which are likely to have a far more general application. These factors have to be added to studies of chemical and physical processes influencing the life cycle of the aerosol and air transport, in order to understand the sources, sinks and to define the radiative forcing by the atmospheric aerosol in the Arctic and elsewhere.

1. Introduction

1.1. Possible biological control of climate

Since the central Arctic basin in summer to a large extent is separated from air masses associated with strong sources of pollution, it is one of the few locations in the northern Hemisphere where the possible biological control of climate proposed by Charlson et al. (1987) can be studied. This hypothesis was based on the demonstration by Twomey (1974) that the albedo of optically thin clouds in regions of low particle number concentration is sensitive to the concentration of cloud condensation nuclei (CCN) and the increasing amount of evidence that over the oceans sulfur-containing gases emitted by phytoplankton contributed strongly to CCN concentrations. The Earth Radiation Budget Experiment described by Ramanathan et al. (1989) showed that the mid-to-high latitude oceans of the summer hemisphere provided the greatest contribution to the global net cooling due to clouds. Consequently, the

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biological production of CCN in these regions is potentially very important.

Leck and Persson (1996a) found evidence in the Arctic of a substantial source of the biogenic aerosol precursor gas, dimethyl sulfide, from the open ocean and along the ice edge zone. In addition, Heintzenberg and Leck (1994) and Leck and Persson (1996b) revealed a strong Arctic marine biological source of both non sea-salt sulfate and methane sulfonate aerosol particles during summer. Further, Leck et al. (1996) have found that the main source of both the small (Aitken mode) and large (accumulation mode) particles and of CCN active at low supersaturations was the open ocean which was also the dominant air mass origin, direct transport from populated regions being rare. The requirements of the Charlson et al. (1987) hypothesis, that there should be (a) a source of particles ultimately of biological origin, and (b) that the region is one that is important to the radiation balance (Ramanathan et al., 1989), and (c) the presence of low liquid water content stratus clouds, is met in the Arctic region maybe better than anywhere else because of the persistent Arctic summer stratus.

The aim of this paper is to show what influences the number concentrations of particles in layer near the surface and to infer what simultaneous changes may occur in the cloudy layers. This information is essential if we are to understand the relative importance of meteorological, physical and chemical processes in affecting the particle number size distributions. It is also necessary if we are to understand the influence of the summer Arctic aerosol on the global radiation balance.

1.2. Previous measurements of the summer Arctic aerosol

Because of the slowly changing mesoscale or synoptic scale meteorological processes and the long distances from continental sources, the number concentration of atmospheric particles in the Arctic Marine Boundary Layer (MBL) might be expected to be relatively stable. Instead, Hogan et al. (1982), Jaenicke and Schütz (1982) and Lannefors et al. (1983) found that near surface particle number concentration is often very changeable, showing an order of magnitude or more change in periods of an hour or less. Jaenicke and Schütz suggested that pollution from isolated Arctic settlements contributed heavily to the particle number concentrations observed (and therefore, presumably, to its variability) but, as mentioned in the previous section, the ocean has been shown to be the main source of particles in this region in the seasons studied. Hogan et al. (1982) and Lannefors et al. (1987) both found an association between fog or precipitation and sharp aerosol minima during which the total particle number concentration fell to less than 10 cm^{-3} for periods ranging from less than one hour to several hours. However, Nilsson and Bigg (1996) have shown that fog droplet concentrations, during the International Arctic Ocean Expedition of 1991 (IAOE-91), were far too low and precipitation far too weak to cause the changes observed.

It therefore seems that the earlier interpretations of sudden changes in the summer Arctic aerosol are inadequate to explain the 1991 observations and this paper will be concerned with examining possible other causes in more detail.

2. Experimental approach

The measurements to be discussed were made between 1 August and 6 October 1991 and included 6 days between latitudes 70°N and 80°N in early August, 22 days between 80°N and 85°N and 27 days north of latitude 85°N. Following winds while the ship was in motion limited the useful aerosol data on some of these days but at the frequent sampling stations almost all data were free from the ship's influence.

A size-segregating inlet for simultaneous sampling of aerosol particles, up to 10 μm diameter, and trace gases was located about 25 m above the sea surface (Leck et al., 1996). The system used to produce complete size distributions of particles every 10 min has been described by Wiedensohler et al. (1994). We divided the particles into 2 groups, those that would usually be affected by cloud nucleation in the size range 80–500 nm diameter (accumulation mode) and those that would usually be unaffected (except by Brownian capture) in the size range 25–80 nm diameter (Aitken mode). Though definitions of these terms differ, as defined above, each contains one of the peaks of the common trimodal fine particle size distributions in number concentration found in the summer Arctic (Covert et al., 1996). The incoming air was warmed so that dry diameters

were measured, the largest particle sized having diameters of 500 nm. If composed of ammonium sulfate, they would only have been rejected by the air inlet at humidities above water saturation.

Cloud condensation nuclei (CCN) were measured with a continuous flow thermal diffusion chamber cycling in supersaturation between about 0.04% and 1% with a period of about 30 min, so that a reading at any supersaturation was obtained twice in that time. Graphical interpolation was used to bring the values to 10-min averages for comparison with condensation nuclei (CN). The overall correlation coefficient between concentrations of CN > 41 nm diameter and CCN at 0.63% supersaturation was 0.94 so that for a discussion on large changes it makes little difference whether we use CN or CCN. For simplicity, the discussion here will be confined to CN except where CCN show additional features of interest.

Radon was estimated from the alpha-emitting decay products of radon attached to particles and caught on a filter. 10-min averages were used to conform to the particle measurements. The method is valuable in giving a relatively fast response to changes and a continuous record but can in certain circumstances underestimate the amount of radon to an extent which depends on aerosol cross-sectional area. Bigg (1996) has shown that on this expedition the problem only appeared on some brief occasions towards the end of the voyage.

Hourly or half-hourly meteorological surface observations of wind speed and direction, air temperature, pressure, relative humidity, visibility and cloud cover were made. Rawinsonde releases at intervals of the order of six hours when the ship was stationary recorded the vertical distribution of relative humidity, temperature, pressure, wind direction and wind speed (Vaisala RS80 and Digicora MW11 rawinsonde). These were all useful in interpreting changes in the boundary layer stability and mixing that could influence particle concentrations. 5-day back-trajectories of the air reaching the ship's position at 1000, 950 and 900 hPa at 6-hourly intervals were calculated with a 3-D model (McGrath, 1989) at the European Centre of Medium-range Weather Forecasts, UK. Calculations from these trajectories of distance from free tropospheric, marine or land sources were specially useful. For a more detailed meteorological description cf. Nilsson (1996).

3. Results and Discussion

3.1. Sudden large changes in aerosol concentration

3.1.1. Definition, and frequencies of events. If we define a "sudden large change" to be more than a factor of 2 in number concentration within 1 h or less in either Aitken or accumulation mode particles, then there were about 130 certain cases during the expedition. There were many other possible cases where suspected shipboard pollution or missing data interfered with interpretation. Of the certain cases, most involved initial decreases in concentration followed by increases. Often these changes occurred in sequences of several maxima and minima, including cases which did not quite meet the above definition. Accumulation mode concentrations usually changed considerably more than those of the Aitken mode.

3.1.2. Example of the simplest common type of sudden change. A typical example encountered at latitude 88.5°N between 00.00 and 06.00 UTC, 6 September 1991 (Julian Day 249) is shown in Fig. 1. Initially accumulation mode number concentrations fell extremely rapidly, then more slowly to a minimum more than a factor of 10 below the initial value while Aitken mode particle number concentrations were relatively unaffected. The dividing line between strongly and weakly affected lay at a diameter of about 80 nm. Air temperature reached a minimum at about the same time as accumulation mode concentrations, but relative humidity remained in the range 95 to 96%. Radon concentrations, too, showed a similar decrease but with a superimposed oscillation.

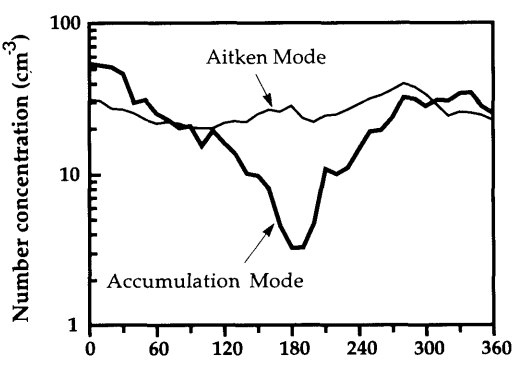

Fig. 1. An example of a sudden change in accumulation mode particle number concentrations at latitude 88.5°N.

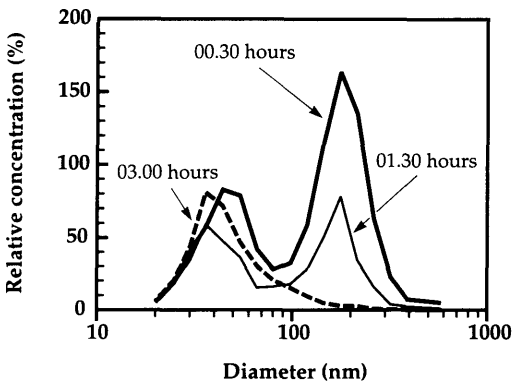

Fig. 2. Number size distribution changes during the episode of Fig. 1 in relative concentrations as percentages of the number concentration between 00.00–03.00 hours.

Fig. 2 shows the size distributions for 00.30, 01.30 and 03.00 UTC, 6 September 1991. The large initial accumulation mode peak, its sudden collapse and final complete disappearance for a period around 03.00 is remarkable. Clearly, particles of 150 nm diameter and larger were the most affected, decreasing by factors of 50 to 100.

3.2. Explanations of sudden changes in aerosol concentration like those of Fig. 1 in terms of vertical mixing

3.2.1. Cloud influences. The fact that large particles were strongly affected while small ones were not, immediately suggests fog or cloud intervention. There was no fog during this and many other similar events of a sudden change. If clouds were involved then it has to be assumed that air from the overlying cloudy layer has somehow been intermittently mixed into the lower part of the MBL.

Chemical analysis of the bulk aerosol has suggested a particle composition roughly corresponding to ammonium bisulfate (Leck and Persson, 1996b). This is supported by the few electron micrographs of individual particles that were obtained. The peak supersaturation required to produce a cloud droplet from all accumulation mode particles, as defined in Section 2, is then about 0.1 %, a reasonable average figure for the sort of clouds formed as a maritime air mass invades the pack ice region.

Summer cloudiness during transport of air from open sea to pack ice has been modelled by Herman and Goody (1976). Cloudiness is likely to be persistent and to be accompanied by advection fogs in the initial stages. These model results were confirmed by observations during the IAOE-91 (Nilsson and Bigg, 1996).

The air mass situation corresponding to Fig. 1 was one of fairly rapid (48 hour) transport from the open waters of the Laptev Sea. Cloud observations at the beginning and end of the period showed 8 octas of Stratus. We therefore need to consider the processes occurring in cloud.

3.2.2. The evolution of vertical gradients in particle concentration. A feature of the clouds of the high Arctic was the absence of drizzle reaching the surface on all but a few occasions. Evidently, the drizzle drops formed in the persistent stratus were usually too small to reach the surface without evaporation. We can therefore envisage the situation where droplets form on the accumulation mode particles growing by condensation of water vapour slowly and perhaps acquiring additional solute phase through oxidation of dissolved gases. Eventually their growth allows them to fall out and they fall towards the surface losing water as they go. There may be many recyclings and the end product will be a relatively high concentration of large particles near the surface and a very strong depletion near and within the cloudy layer. During the Arctic summer, Ottar and Pacyna (1986) found alternating layers of high and low large aerosol concentrations in the lowest kilometre. Curry (1986) found that Arctic stratus clouds occurred in multiple layers ranging from a surface layer fog underlying a stratus deck to two or more closely spaced cloud layers. Thus, if there should be a cloud free and stable layer below the level at which drizzle droplets evaporate a region of relatively high accumulation mode particle concentration may result as hypothesised just above the cloud top in Fig. 3d.

However, Aitken mode particles are inefficiently captured by water drops either by nucleation or diffusion and depletions are likely to be slight. There have been a number of reports of layers of high number concentration of Aitken mode particles just above the tops of stratiform clouds in mid latitude MBL (Hegg et al., 1990, 1991), suggesting that the cloud top may be a favoured region for generation of new particles. If this also

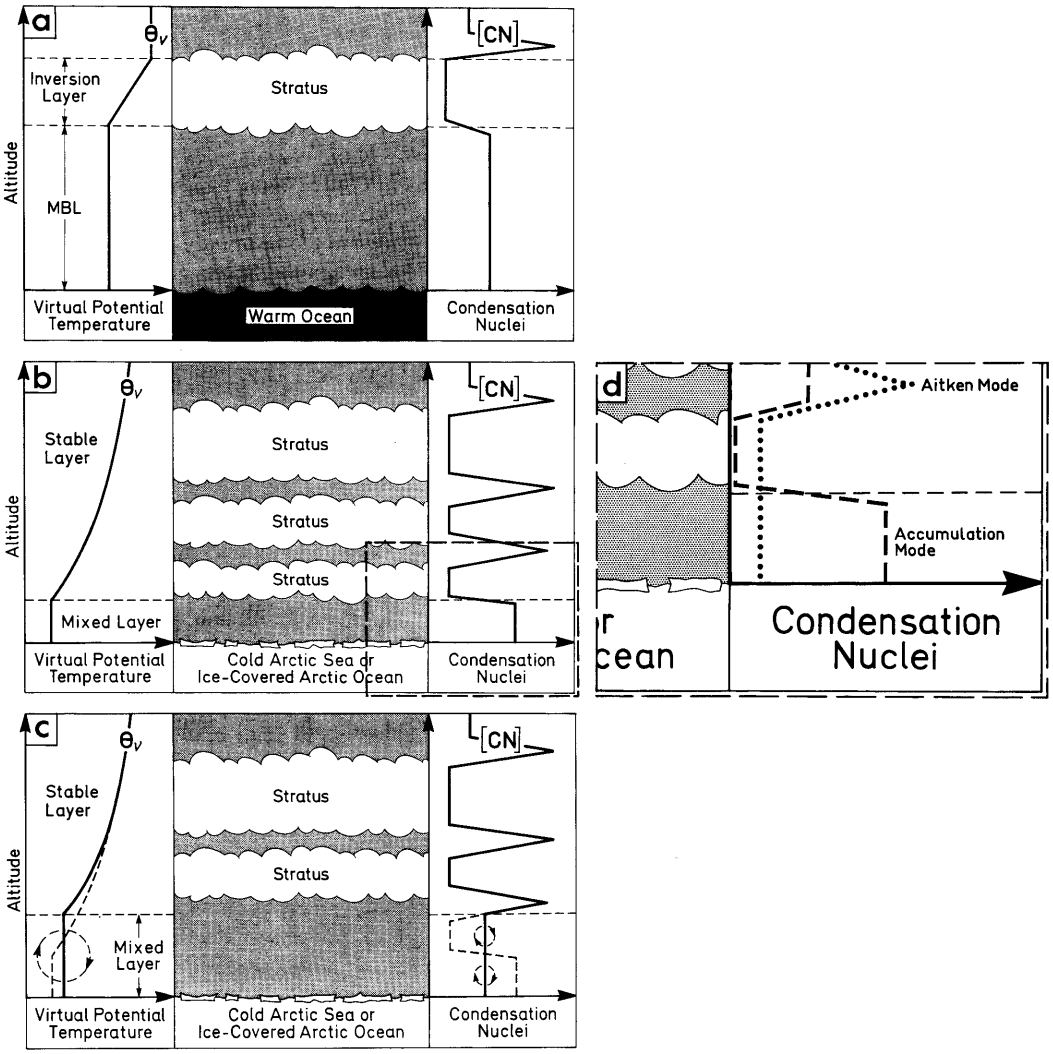

Fig. 3. (a-c) Hypothetical vertical profiles of virtual temperature, θ_v cloudiness and particle number concentration [CN] through marine stratus layers. (d) Enlargement of the part of (b) within the dashed line, illustrating the simplest situation of a single stratus layer. The condensation nuclei profile has been partitioned into Aikten and accumulation modes as indicated. MBL = Marine Boundary Layer.

occurs in the Arctic, profiles of Aitken mode number concentration may look like that of Fig. 3d in the simplest situation of a single cloud layer. This purely hypothetical Aitken mode maximum is consistent with some observed sudden changes during the IAOE-91 having similar decreases in accumulation mode particle number concentrations as of the example shown in Fig. 1 but with strong increases in Aitken mode number concentration. From the mechanisms (processes) suggested above for cloud-aerosol interaction, the multiple layer stratus structure in general probably caused a more complex situation as shown in Figs. 3a-c.

The advection of marine air over a cold surface, that causes the frequent high cloudiness and fogginess in the Arctic summer, is important in transforming the neutral MBL into a stable MBL,

consisting of a mixed layer and a stable layer smoothly blending into the free troposphere, in which any vertical gradient will be preserved (Nilsson, 1996), as illustrated in Fig. 3a, b. The mixed layer was in general shallow compared to the height of the stable layer. In addition to the strong vertical gradients caused by clouds, differences in transport times or source regions for the trajectories arriving at different heights in the stable MBL can also influence profiles of aerosol or other tracers. Radon being an inert gas will not be affected by nucleation as are aerosol particles. It usually showed simultaneous changes in concentration at the time of sudden aerosol changes indicating that transport differential is often an important contributor to vertical concentration gradients.

3.2.3. Mixing processes between the cloud and underlying layers. Now, let us suppose that the stable layer was accumulation mode-depleted in the cloudy layers, as depicted in Fig. 3b, d. If there is some sudden mixing event, there may be a sharp decrease in accumulation mode particles in the near-surface layer, like that of Fig. 1. At the same time, there will be an increase in number concentrations at the former cloud level. Aitken mode concentrations will be relatively unaffected unless mixing proceeds beyond cloud top where the higher concentration layer may exist. In the more complex cloudy situations of Fig. 3b, c the surface changes in concentration will depend very much on the depth of mixing but the same principles will apply. With a different cloud-aerosol vertical profile the mixing may even result in a sudden accumulation mode particle increase, which was sometimes observed. In air that had recently subsided causing evaporation of cloud, mixing events might lead to increases in both Aitken and accumulation mode number concentrations. A few such cases were observed.

The main phenomena known to influence mixing between the adjacent stable layer and the mixed layer have been described in some detail in connection with the planetary boundary layer by Nilsson (1996). Briefly, they are: Kelvin–Helmholtz, gravity, and lee waves, collapse or intermittent breakdown of the boundary layer and roll vortices. We shall discuss these in turn to see if they can provide explanations for any of the sudden changes in particle number concentrations.

3.2.4. Kelvin–Helmholtz instabilities and shear induced gravity waves. During this study, the wind maxima of low level jets were commonly present at the border region of the mixed layer and the stable layer. Therefore, Kelvin–Helmholtz instabilities are a common cause of mixing, developing at the boundaries of regions of different density, wind strength or direction and having periods of the order of one minute. As Kelvin–Helmholtz waves grow in amplitude they may break, enhancing turbulence and mixing at the boundary and tending to destroy the discontinuity that produced them. Cheung (1991) has shown some graphic time-height records of back-scatter from an acoustic sounder in the Arctic that clearly illustrate the processes involved. The periods of wave activity and associated enhanced turbulence varied in length from ten minutes to many hours in the examples shown and turbulence was enhanced down to the minimum usable range of the sodar. Furthermore, the shear of the low level jets can trigger gravity waves which may radiate downward to the surface followed by mixing of air from relatively higher altitudes (Fig. 3b, c). Nilsson (1996) has demonstrated that the conditions during the IAOE-91 were favourable for both Kelvin–Helmholtz and shear induced gravity waves.

Cheung (1991) has suggested that the onset of Kelvin–Helmholtz waves by passing gravity waves was possibly the dominant mechanism of overturning and generation of centimetre-scale turbulence within the near-ground surface layer at Barrow, Alaska. If this occurred upwind, patches of mixed and unmixed air would drift across the observing site. The duration of the sudden change in concentrations of any tracer would be related not to the wave periods but to the length along the trajectory that was affected and surface wind speeds.

None of our measurements was on a sufficiently fast basis to pick up the "braids of turbulence" that Cheung has shown, the wave periods being of the order of one minute. The CN measurements were recorded at intervals of about 35 seconds, but each was for a different size. CCN measurements were at one-minute intervals but at continuously varying supersaturations. Consequently, any variations in particle number concentration due to the mixing caused by these waves will, in our measurements, be the integrated effects for ten-minute intervals and therefore vary with the over-

all wave activity rather than the individual pulses of mixing. The mixing event does not necessarily have to occur overhead but the onset of changes will be sharper when it does. Once mixing ceases, the patch of air with anomalous properties in the near-surface mixed layer will drift downwind, its boundaries becoming more diffuse with time.

A detailed description of the changes shown in Fig. 1 could be interpreted as follows on the basis of Fig. 3b-d. Eddies from a Kelvin–Helmholtz breaking wave just above the mixed layer brought air from the depleted region to the measuring point causing a sudden decrease between 00.20 and 00.30 UTC. Mixing then progressed to successively greater heights in the depleted layer producing further decreases for 3 h. Transport of air from regions of less mixing upwind then allowed concentrations to return their original value.

A radiosonde launch from the accompanying ship, Polarstern, at 00.00 UTC showed an inversion near the surface. The resolution of this rawinsonde was insufficient to establish the depth of the mixed layer but indicated that it must be shallow. This is consistent with the very large decrease in observed concentration.

The recovery of the accumulation mode number concentrations following the minimum could be explained by assuming that simultaneous vertical mixing over about 100 km of the air trajectory had occurred overhead and upwind and was most intense near the centre of the disturbance. With a wind speed of about 7 ms^{-1} the total change would then have the observed duration. If this is the correct explanation, the duration of the mixing event is of less importance than its geographical extent and wind speed in determining the duration of the changes.

3.3. *Quasi-periodic sudden changes in aerosol and radon concentrations*

In contrast to the single decrease and recovery shown in Fig. 1, about 2/3 of all sudden changes formed part of a sequence of maxima and minima. The number of multiple sequences was about the same as the number of single events. We therefore have to try to find some periodic phenomenon in the Arctic atmosphere that could explain these sequences. First it is necessary to establish the mean period.

Because there were no interruptions due to pollution, more accurate statistics could be obtained from the measurements of radon which showed similar variations. First it was necessary to reduce the purely statistical counting fluctuations by applying smoothing which retains as much as possible of the temporal resolution. The smoothed concentration of radon at time t was defined to be:

$$C(t) = (C(t-20) + 2C(t-10) + 4C(t) + 2C(t+10) + C(t+20))/10.$$

The time series of $d(\log(C(t)))/dt$ was then constructed. On most days, sequences of fairly regular maxima and minima were present, sometimes lasting only 2 cycles but typically 5 and occasionally as many as 15. Single cycles were ignored as they could have been due to temporary contact of the air with a land source of radon. The mean period of each sequence was calculated both from maxima and minima and the resulting frequency distribution comprising 49 sequences with a total duration of 18.5 days (35 % of the radon sampling time) is shown in Fig. 4. There is a strong preference for periods in the range 60 to 90 min, which was also found from the available CN and CCN data though in individual cases there were differences in the mean periods deduced from radon and particle concentrations.

Accumulation mode particle number concentration changes (expressed as a percentage change per 10 min) illustrating this type of repetitive sudden change are shown in Fig. 5 for 26 September 1991 (Julian day 269) when the ship was at latitude 83.3° N, longitude 8.0° E. The Aitken mode particle

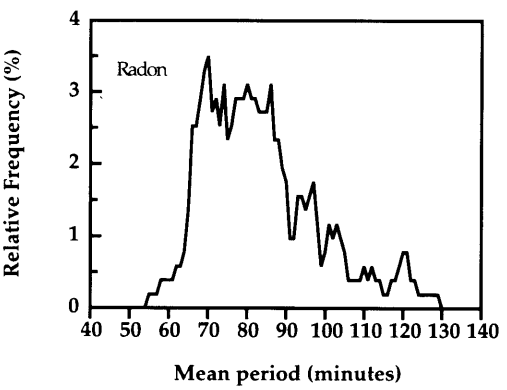

Fig. 4. The relative frequency of occurrence of various mean periods of radon fluctuations.

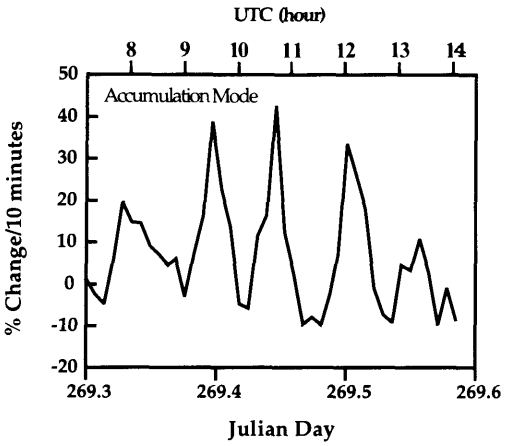

Fig. 5. An example of recurrent changes in accumulation mode particle concentration at latitude 83.3°N with an air trajectory from the North Pole.

number concentrations on this occasion were less than 5 cm^{-3}, the smallest particles present being larger than 100 nm diameter, except near the accumulation mode minimum at day 269.43. The wind in this case was 4 ms^{-1} and at day 269.50 the air trajectory lay across the North Pole.

Again, mixing of air from different levels combined with large changes in particle concentration with height seem to provide the simplest explanation but it is necessary to find repetitive atmospheric phenomena with the appropriate mean periods that could cause the mixing.

3.4. Periodic phenomena in the atmosphere

3.4.1. Mesoscale gravity waves. Of the many periodic phenomena in the atmosphere, mesoscale gravity waves have a range of periods most closely corresponding to those in Fig. 1. According to Koch et al. (1993) these are periodic wave trains or single solitary-like wave disturbances with period of 1 to 3 h and wavelengths of 50–500 km. They state that these are increasingly recognised for their ability to modulate cloud and precipitation fields, to produce and interact with deep convection and turbulence and to transport energy horizontally. Uccellini and Koch (1987) and Koch and Dorian (1988) have reviewed the synoptic setting and possible energy sources for these waves. They suggest that generation occurs through a shedding of a gravity wave, an inertia wave packet in association with a geostrophic adjustment process, a jet stream approaching a stationary ridge in the 300 hPa height field being a favoured location. Occurrence of the waves is usually associated with strong static stability in the lower troposphere. Uccellini and Koch (1987) suggest that this, in conjunction with a critical level aloft where waves speed matches wind speed in the direction of horizontal propagation, may help sustain the waves by acting as a duct to prevent energy loss by vertical leakage.

Such waves could only affect the near surface layers when the local stratification (Brunt-Väisälä frequency) below the ducting layer allows them to propagate downward. Thus, we could imagine a limited region near the surface into which energy from these upper level waves could leak, causing temporary mixing events as each wave crest passes.

Unfortunately, we have no direct evidence that such waves occurred at the very high latitudes reached on this voyage. The probable generating regions would be associated with the polar jet stream well to the south and long distance propagation would usually have been necessary. The possibility that they were responsible for any of the changes must therefore remain speculative until appropriate observations are made on future expeditions and we will confine the discussion to other types of disturbances for which more direct evidence is available.

3.4.2. Lee waves and impulse-generated gravity waves. During the IAOE-91, patterns of low clouds perpendicular to the wind direction were observed on NOAA-11 satellite images. These cloud patterns were probably caused by waves generated in air flowing over the Greenland plateau, or the mountains of Spitsbergen and other Arctic islands. Topographic obstacles are known to cause lee waves and induce long-period gravity waves. According to Gedzelman (1983), such waves have a high coherence, are virtually non-dispersive and can induce large displacements of boundary layer air, thereby modifying shear and stability. There will be enhanced mixing and interaction with, or triggering of, shorter period shear induced gravity and Kelvin–Helmholtz waves. Their mode of propagation resembles that of water waves caused by a pebble thrown into a pond. The satellite images obtained during the expedition showed that these waves retained sufficient amplitude to cause alternating condensation and

evaporation in the cloudy layers about 100 km downwind from their origin. The wavelengths of the observed cloud patterns were of the order of 2 to 6.5 km, which compared to reported figures in the literature correspond to periods of 20 min or less (Ali, 1931; Gossard and Munk 1954; Yamamoto 1957), too short a period in itself to explain the periodic oscillations, either directly or indirectly (for typical wind speeds) through triggering Kelvin-Helmholtz breakdowns which then cause mixed regions to propagate downstream. One possibility is that the wave train produces a sequence of pulses of mixing too short for our ten-minute time base to resolve and that we see the effects of the wave train rather than the individual pulses.

Longer wavelengths can also exist. For example Shütts (1992) described a case of a wave train with a dominant mode wavelength of 16.5 km and a decay scale of 425 km.

3.4.3. Roll vortices. Nilsson (1996) has discussed the frequent occurrences of large areas covered by streets of low clouds, more or less parallel to the wind direction, seen in the satellite images recorded during the expedition. He concluded that they were associated with weak convection in parallel pairs of roll vortices having opposite rotations as illustrated in Fig. 6. The axes of the roll vortices were found to lie at a slight inclination to the geostrophic wind so that a stationary observer would see them moving overhead. Supporting evidence came from vertical distributions of the wind, temperature and humidity which in about 40% of cases suggested a passage through a roll vortex. Roll vortices may therefore be very common in this environment. More details can be found in the paper referred to above.

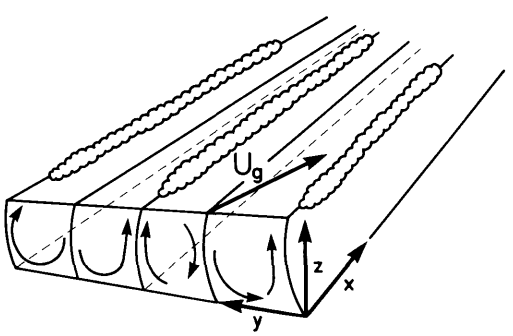

Fig. 6. Schematic diagram of roll vortices (from Le Mone 1973). U_g is the geostrophic wind.

Based on all available satellite photographs with cloud streets visible and all rawinsondes launched through roll vortices during the IAOE-91, Nilsson (1996) has estimated the intervals between passages of successive up or down draft regions at a fixed point to prefer the range 30–90 min. The actually observed most frequent intervals between maxima and minima in tracer concentrations of about 60–90 minutes (cf. Subsection 3.3) lie at the upper end of this range.

If the roll vortices are as common as suggested by the rawinsondes, they might have very substantial effects on the aerosol since it would be subjected to repeated condensation and evaporation cycles as cloud forms on the updraft side and dissipates on the down draft side. Even when the cloud was continuous, the path length within the cloud from the source of the aerosol to the observing point would be much greater than if the flow was direct and would as a result cause decreased concentrations. Ferrare et al. (1991) has related parallel regions of updrafts marked by enhanced aerosol scattering (due to increased relative humidity) revealed by Lidar, to roll vortices.

One occasion (12 September 1991, Julian day 255, latitude 86.8° N, longitude 10.5° E) on which the presence of roll vortices were supported by rawinsonde data, coincided in time with the onset

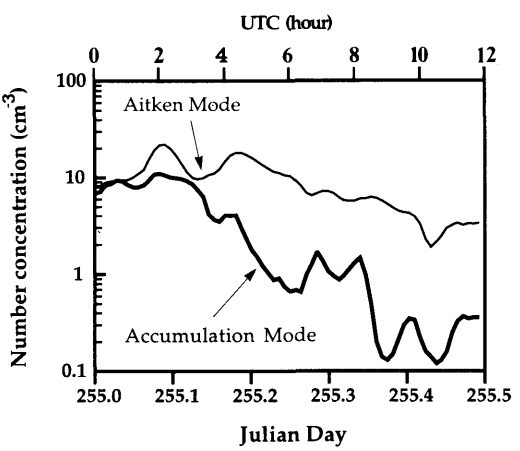

Fig. 7. Changes in Aitken and accumulation mode particle number concentrations during a period when rawinsonde recordings demonstrated that roll vortices were present.

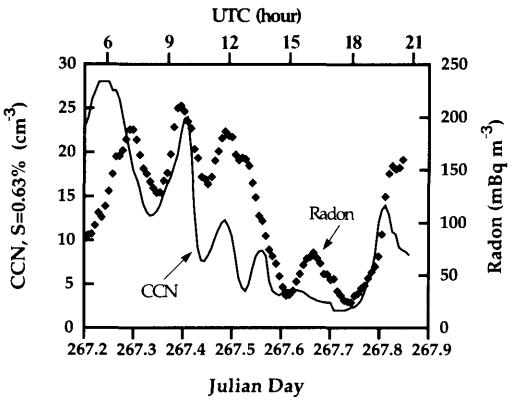

Fig. 8. Changes in CCN and radon concentrations at latitude 83.6° N, suggesting the presence of roll vortices.

of a decrease in particle number concentration and quasi-periodic oscillations. The accumulation and Aitken mode particle number concentrations are shown in Fig. 7. In a similar case as Fig. 5, the 2 corresponding rawinsondes both supported the presence of roll vortices and showed low-level jets and the possibility of shear induced gravity and Kelvin–Helmholtz waves. It is possible that such waves interacted with the rolls and caused mixing in the updraft or down draft regions of the rolls which would explain the strength of the periodic changes. Enhanced turbulence at the passages of the updraft regions in the roll vortices has been observed by Smedman (1991).

To illustrate that these changes are not confined to CN, Fig. 8 shows a similar decrease with oscillations in CCN and radon on 24 September (Julian day 267) at latitude 83.6° N, longitude 4.0° E. The difference in timing of the maxima and minima of CCN and radon is probably due to their differing vertical profiles, radon being unaffected by cloud processes.

3.5. Sudden changes in aerosol and radon concentrations due to other causes

3.5.1. Changing source strengths due to air trajectory changes. Leck and Persson (1996b) have found that particles derived from dimethyl sulfide and sub-micron sea-salt particles, which both originate from the open ocean, dominated the aerosol during IAOE-91. Changes in air trajectory that greatly alter the transport time from open water or marginal ice zone will therefore have a major influence on changes in aerosol concentration. However, alteration in transport time from such diffuse source regions as the seas adjacent to the pack ice are likely to be relatively gradual on most occasions, so that they are unlikely to be a common cause of the sudden changes which we have defined to be a factor of two or more in concentration in one hour or less.

3.5.2. Sudden changes due to anthropogenic sources. The spring studies of "Arctic haze" (Rahn et al., 1980) show that the planetary boundary layer over the central Arctic basin in general is perturbed by anthropogenic inputs from lower latitudes. Nilsson (1996) found that transport from these sources occurred on only a few of the 70 days covered by IAOE-91 and a similar result was found during the Ymer expedition in 1980 (Lannefors et al., 1983). We might therefore expect anthropogenic sources on distant continents to have only a minor and sporadic effect on the summer aerosol of the high Arctic. Jaenicke and Schütz (1982), however, hypothesised that Arctic settlements and their associated aerosol emissions were the main source of condensation nuclei in the region. We therefore need to search the records for occasions when either distant or more local anthropogenic particles might have been present to establish their relative importance.

The influence of a distant but intense source was found on only one occasion (22 September, Julian day 265, latitude 84.7° N longitude 5.8° E) to give rise to increased concentrations large enough and sufficiently rapid to be included in our definition of "sudden change". The air trajectory changed suddenly from an oceanic origin to one coming from the western end of the industrialised Kola Peninsula in Russia at day 265.25, as shown in Fig. 9a. A fog and rising temperature preceded land contact (indicated by increasing radon) but were coincident with the start of rising aerosol concentrations (Fig. 9b). Evidently, offshore production contributed to the aerosol increase although the high (by Arctic standards) concentrations that followed suggested industrial pollution to be the main cause.

Aerosol apparently from the small settlements of Barentsburg and Longyearbyen on the island of Spitsbergen was detected early on 31 August (Julian day 243) at latitude 88.1° N, longitude 84.5° E. Fig. 10a shows the corresponding air tra-

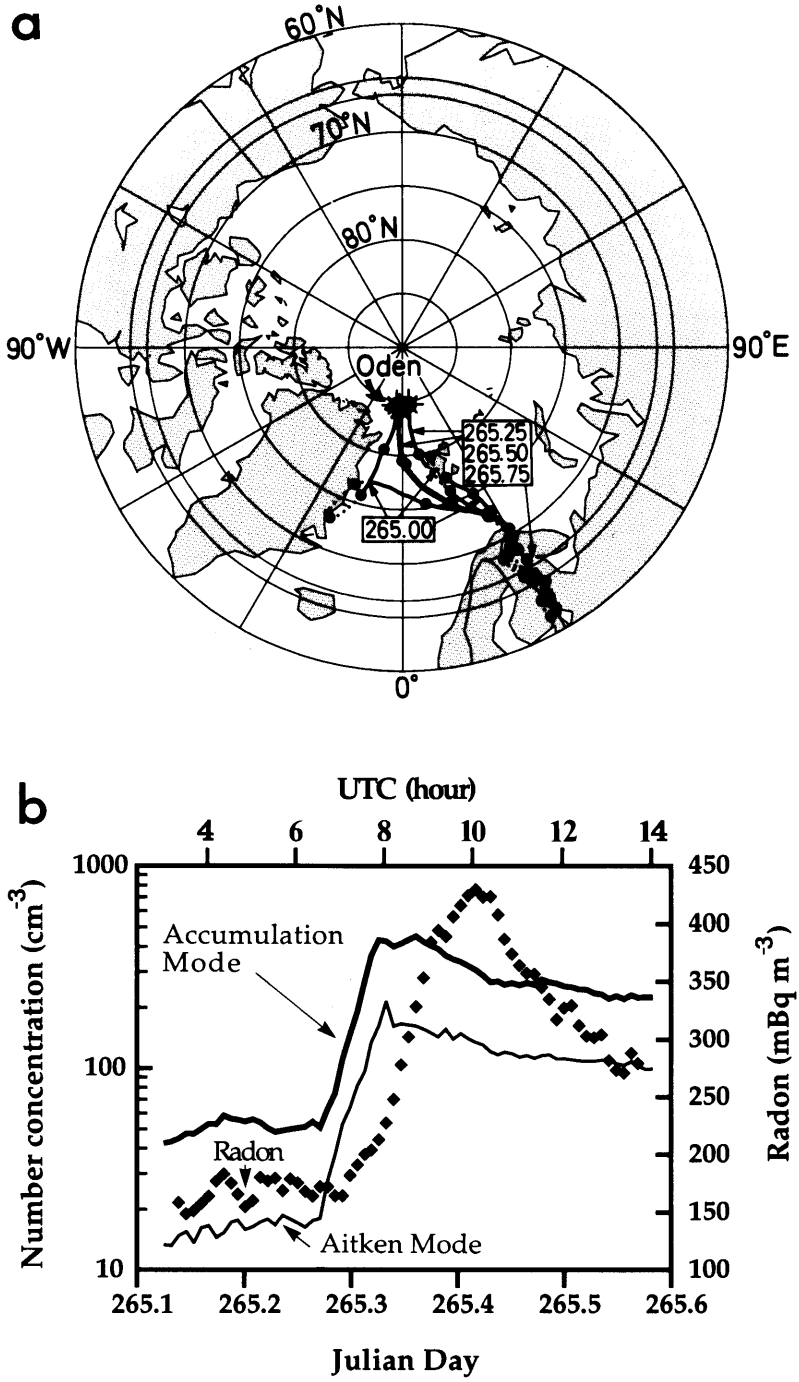

Fig. 9. (a) Trajectories showing a change from oceanic air mass origin to an origin from the industrialised Finland and Kola Peninsula, Russia. (b) Corresponding changes in aerosol concentration.

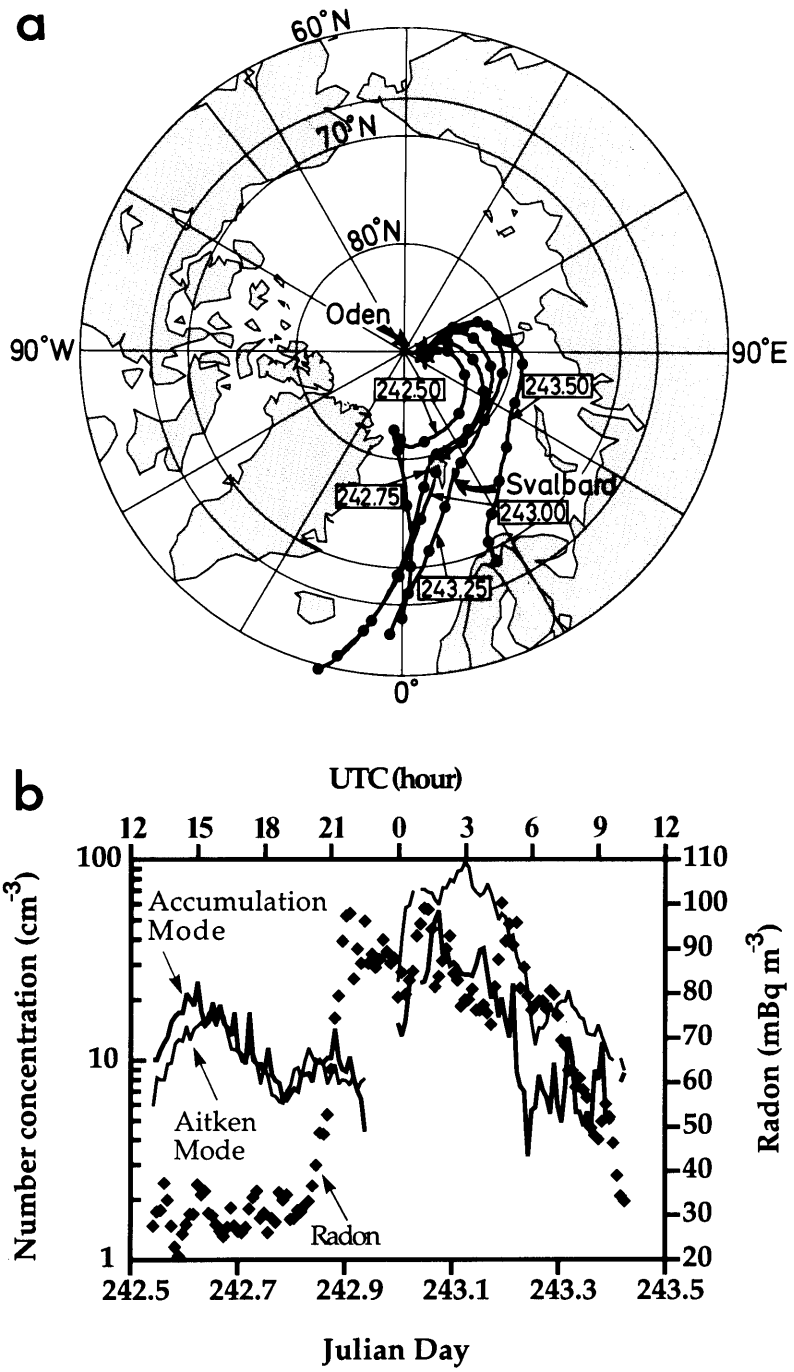

Fig. 10. (a) Trajectories showing a passage of marine air over Spitsbergen, located 2000 km and 70 h transport time upwind from latitude 88.1° N. (b) Corresponding large changes in radon and aerosol concentration of anthropogenic signatures.

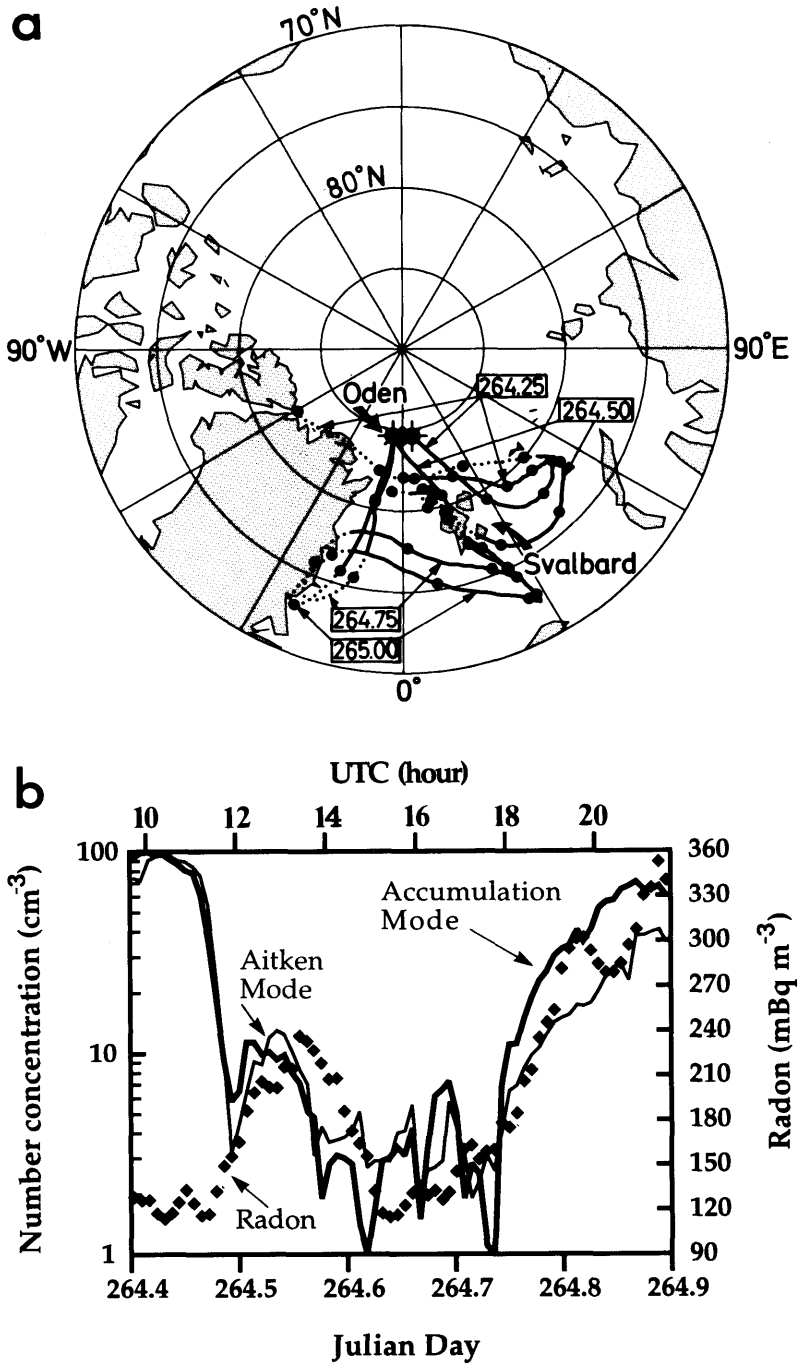

Fig. 11. (a) Trajectories showing a passage of marine air over Spitsbergen, located 500 km and 12 hour transport time upwind from latitude 84.6° N. The mean wind speed was much higher than in the case of Fig. 10, leading to vertical mixing over the mountains. (b) Corresponding changes in radon and aerosol concentration of small anthropogenic signatures.

jectory. In spite of the distance (about 2000 km) and transport time (70 h) the radon signature of the island was very clear and embedded within it was a sharp increase predominantly in Aitken mode number concentrations (shown in Fig. 10b) such as would be expected from an anthropogenic source. For both radon and aerosol to show such large changes there must have been very little mixing along the way.

In contrast, on 21 September (Julian day 264.50) at latitude 84.6° N, longitude 6.2° W the air trajectory of Fig. 11a shows the island to have been only 500 km and 12 h transport time upwind, yet both the radon and particle peaks near 264.53 (Fig. 11b) were relatively small. The very large decrease in particle number concentrations that preceded the 264.53 peaks and the later recovery suggest that there had been mixing to a considerable depth, probably due to the passage of air over the mountains of Spitsbergen. The cause of the difference between this and the previous example almost certainly lies in the mean wind strength deduced from the trajectories, which was considerably higher for Fig. 11 than for Fig. 10. In the former case, it was evidently not strong enough to cause mixing to a great depth while in the latter it was. The comparison provides a graphic illustration of the importance of mixing in the vertical to the aerosol concentrations measured at the surface, and by deduction, at higher altitudes as well. The conclusion is that anthropogenic sources in summer only rarely produce noticeable effects on Arctic aerosol concentrations but their contribution may often be disguised by changes in mixed depth.

3.5.3. Effects of frontal air mass change and mixing. In total, only 5 well-defined fronts passed the ship during this expedition, each associated with sudden large changes. The occluded front of 17 August (day 229.50) was accompanied by a similar decrease in both Aitken and accumulation mode number concentrations by a factor of about 7. An increase in accumulation mode and a larger increase in Aitken mode accompanied the warm front on day 243 but this appears to have been largely due to aerosol from Spitsbergen as discussed in the previous section. The passage of a partly occluded warm front at day 256.25 coincided with a factor of 10 increase in accumulation mode and a small change in Aitken mode. The interpretation of the changes may be complicated by simultaneous changes in mixed depth and air mass although this does not seem to have happened on day 243.

3.6. Summary of the causes of sudden large changes in aerosol and radon concentrations and their implications

Situations with cloudy regions in stable poorly mixed layers were very common during this expedition and would inevitably lead to strong vertical gradients in aerosol concentration as discussed in Subsection 3.2.2. Differing air histories at different heights will have accentuated the aerosol stratification in the MBL and led to stratification in tracers, such as radon, unaffected by cloud processes. Mixing between these layers must tend to average the gradients and sudden changes could result as schematically illustrated in Fig. 3b-c. For isolated changes followed by a recovery to approximately the original conditions, we have seen that turbulent mixing caused by Kelvin–Helmholtz or shear-induced gravity waves provides a ready explanation.

For the equally common phenomenon of recurrent sudden changes, there are several possible causes, but the demonstrable presence of roll vortices on many occasions and the fact that the expected periodicities (30–90 minutes) are consistent with those observed, make it seem likely that they were responsible, as discussed in Subsection 3.4.3. Propagation downwind of mixed patches of air and possible interaction between various forms of wave motion and between Kelvin–Helmholtz or shear-induced gravity waves and roll vortices, makes the interpretation of any particular event potentially very complex. In any of the examples described, the cause assigned to it is likely to have been the dominant rather than the sole cause. Moreover, the recurrent sudden changes in particle number concetrations could be causally related to intermittent breakdown of the planetary boundary layer (ReVelle, 1993).

In addition, air trajectory change and frontal mixing which may also cause sudden changes will have associated with them changes in vertical concentration gradients and wave activity which again make it difficult to assign unique causes.

The changes in particle number concentration at cloud and sub-cloud level resulting from mixing

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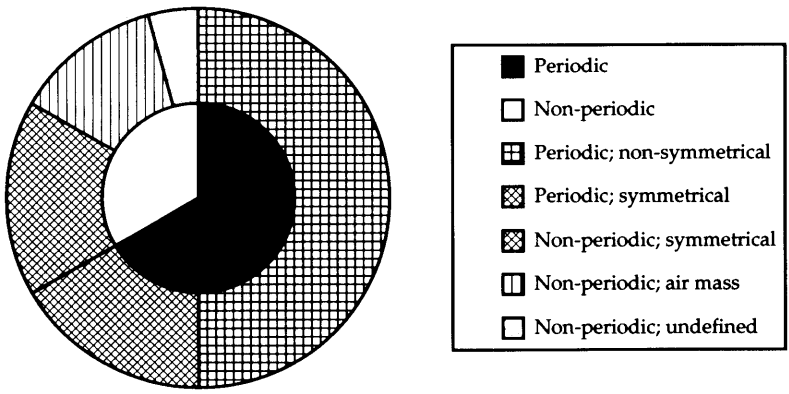

Fig. 12. Distribution of different types of change exceeding a factor of two in number concentration in either Aitken or accumulation mode particles in one hour or less, encountered on IAOE-91.

will probably be less than in the surface mixed layer because of the greater depth. Because of the extremely low concentrations of cloud condensation nuclei that we have observed following these mixing events (sometimes less than 1 cm^{-3}), the mixing events must be very important for maintaining a supply of cloud condensation nuclei for cloud formation. Without them, cloud droplet growth would be rapid, losses by fallout very large and cloud albedo extremely low.

Symmetrical changes, usually leading to a deep minimum in accumulation mode number concentrations, accounted for about 1/6 of all the concentration changes that exceeded a factor of 2 in 1 h. Sequences of alternating maxima and minima accounted for another sixth but for two thirds of all cases because of the number of cycles involved in each sequence. Events associated with fronts, sudden trajectory changes, pollution sources or mechanical mixing combined amounted to about one 1/8, while no obvious cause could be found for the remainder, cf. Fig. 12. Calculation of the proportion of the sampling time during which sudden changes are present is difficult. If we take only the time for which the change exceeds a factor of 2 in 1 h or less, omitting the periods near maxima and minima which did not meet this criterion and parts of a sequence which did not quite meet the factor of 2 requirement, the proportion is 16%. If the definition is made more elastic to include all parts of what appear to be sequences of changes, the proportion is about 40%.

The presence of Kelvin–Helmholtz waves raises an interesting point concerning particle generation. The waves that are the precursors to mixing at shear boundaries must be there a great deal of the time; we know, e.g., that they are present when roll vortices are present. This means that there will be extended periods during which the air near cloud top is subjected to compressions and rarefactions with a period of about 1 min. These, if sufficiently intense, would allow condensation of any supersaturated vapour such as sulfuric acid during the expansion phases which would not completely evaporate during the compressions. The conditions of high humidity that aid such nucleation were present for most of the time during this expedition throughout the Arctic MBL. Since the formation of cloud and stratification of the stable MBL during summer are intimately related we can envisage a possible feedback process between extreme CCN depletion and the replenishing mixing events that results from the stability.

Aerosol radiative forcing is an important component of climate. The direct forcing depends mainly on particle surface or mass, while the indirect forcing (through the rôle of the particles in cloud droplet formation and hence cloud albedo) depends more on particle chemistry and number size distribution. Recent field studies, such as Quinn et al. (1993) and this study have therefore been aimed at obtaining an understanding of the chemical and physical processes that control the properties of the atmospheric aerosol. In this

study, we have shown that interchange of air between the cloud free layers and clouds is important in determining the CCN available for cloud formation and hence cloud albedo in the high Arctic. The causes of that interchange (wave motions and roll vortices) are therefore also important. Thus, in future, field studies aimed at defining the radiative forcing by the atmospheric aerosol need a detailed meteorological description to separate changes caused by the variability in MBL dynamics from processes limiting the life cycle of the atmospheric aerosols.

4. Conclusions and recommendations

The long duration of this expedition and its penetration to 90° N allowed excellent data sets of a large number of variables to be obtained. The interpretation of the changes depended heavily on inference rather than on direct observation and one of the main contributions will have been to indicate the sort of measurements that are needed in the future to provide direct confirmation of the causes of change.

While the open ocean was clearly the dominant source of particles, both large and small in the Arctic summer aerosol, and the mean level was largely determined by the transport time from this source, we have seen that most of the sudden short-lived changes at the surface arise from 2 factors: the stability of the MBL and the presence of multiple layers of stratiform cloud in which micro physical processes lead to intense stratification of aerosols and roll vortices, wave motions or other processes which produce intermittent mixing of some of the cloudy-layer air to the surface. The radon fluctuations show that some of the vertical stratification results simply from trajectory differences for different arrival heights.

It is clear that vertical mixing, from the mechanisms (processes) suggested above, dominates the short-duration changes in aerosol and tracer concentrations over the central Arctic Ocean and will have a strong influence on the chemical and physical processes that control the properties of the Arctic aerosol. Thus, the recommendations for future work are that there should be continuous measurements of boundary layer structure using acoustic sounder or similar techniques, supported by as frequent measurements as possible of tracer concentrations from helicopter or balloon or by Lidar measurements. Measurements of cloud drop concentrations at high latitudes are also essential if we are to understand the way in which vertical concentration gradients of the aerosol develop and the interaction between aerosol and cloud radiative properties. Another essential is to improve the time resolution of all measurements as far as possible so that the periodicity of disturbances can be correctly assessed. For example, CN measurements in a fixed size range, with a short air intake line could be recorded at intervals of one second. The amount of data obtained would be prodigious but could provide valuable insight into the nature of changes. It is more difficult to speed up chemical measurements because of the low mass concentrations of particles < 80 nm diameter, but desirable if we are to provide measurements on homogeneous air samples.

The Arctic experience that mixing events due to wave motions are important in determining aerosol and other tracer concentrations will also apply in lower latitudes. This possibility has been largely ignored in past descriptive work and probably deserves more attention. Of course, because boundary layers are generally much deeper at lower latitudes than in the high Arctic, changes from these causes will often be much less marked but could nevertheless be significant.

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