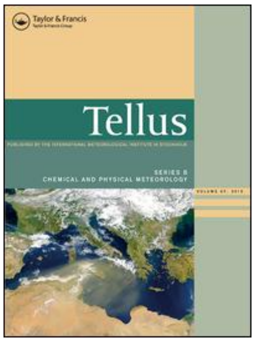

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Overview of the atmospheric research program during the International Arctic Ocean Expedition of 1991 (IAOE-91) and its scientific results

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ABSTRACT

The broad aim of the Atmospheric program of the International Arctic Ocean Expedition (IAOE-91) was to test the hypothesis that marine biogenically produced dimethyl sulfide (DMS) gas can exert a significant global climatic control. The hypothesis states that DMS is transferred to the atmosphere and is oxidised to form airborne particles. Some of these grow large enough to act as cloud condensation nuclei (CCN) which help determine cloud droplet concentration. The latter has a strong influence on cloud albedo and hence on the radiation balance of the area affected. In summer, the central Arctic is a specially favourable region for studying the natural sulfur cycle in that the open waters surrounding the pack ice are the only significant sources of DMS and there are almost no anthropogenic particle sources. Concentrations of seawater and atmospheric DMS decreased at about the same rate during the period of measurements, (1 August to 6 October, latitudes 75°N to 90°N) spanning about three orders of magnitude. Methane sulfonate and non-sea salt sulfate in the submicrometer particles, which may be derived from atmospheric DMS, also decreased similarly, suggesting that the first part of the hypothesis under test was true. Influences on cloud droplet concentration and radiation balance could not be measured. Size-resolved aerosol chemistry showed a much lower proportion of methane sulfonate to be associated with supermicrometer particles than has been found elsewhere. Its molar ratio to non-sea salt sulfate suggested that the processes controlling the particulate chemistry do not exhibit a net temperature dependence. Elemental analysis of the aerosol also revealed the interesting possibility that debris from Siberian rivers transported on the moving ice represent a fairly widespread source of supermicrometer crustal material within the pack ice. Highly resolved measurements of aerosol number size distributions were made in the diameter range 3 nm to 500 nm. 3 distinct modal sizes were usually present, the "ultrafine", "Aitken" and "accumulation" modes centred on 14, 45 and 170 nm diameter, respectively. The presence of ultrafine particles, implying recent production, was more frequent than has been found in lower latitude remote marine areas. Evidence suggests that they were mixed to the surface from higher levels. Sudden and often drastic changes in aerosol concentration and size distribution were surprisingly frequent in view of the relatively slowly changing meteorology of the central Arctic during the study period and the absence of strong pollution sources. They were most common in particles likely to have taken part in cloud formation (> 80 nm diameter). 2 factors appear to have been involved in these sudden changes. The 1st was the formation of vertical gradients in

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aerosol concentration due to interactions between particles and clouds or favoured regions for new particle production during periods of stability. The 2nd was sporadic localised breakdowns of the stability, bringing changed particle concentrations to the measurement level. Probable reasons for these sporadic mixing events were indicated by the structure of the Marine Boundary Layer (MBL) investigated with high resolution rawinsondes. Low level jets were present about 60% of the time, producing conditions conductive to turbulence and shear-induced waves. It is concluded that an even more detailed study of meteorological processes in the MBL in conjunction with more highly time-resolved measurements of gas-aerosol physics and chemistry appears to be essential in any future research aimed at studying the indirect, cloud mediated, effect of aerosol particles.

1. Description of the expedition

1.1. *Objectives*

The possibility that global warming might occur (caused by increased atmospheric loadings of carbon dioxide and other gases) has been of concern for several decades. The cause of the warming is the influence of the gases on the planet's radiation balance (IPCC, 1994). It has recently been realised that aerosols may have comparable effects on the radiation balance. One is a direct effect through scattering of sunlight in cloud-free air which can result in a net cooling at the surface in the more polluted parts of the planet that may be as large as half the warming due to increased concentration of trace gases (Charlson et al., 1991; Kiehl and Briegleb, 1993). The other is an indirect effect due to the influence of the larger hygroscopic particles (cloud condensation nuclei, CCN) on cloud droplet concentration, which for a given liquid water content controls cloud albedo (Twomey, 1974). Boucher and Lohman, (1995) have estimated the indirect effect to be comparable to the direct effect but having a large uncertainty.

Charlson et al. (1987) used Twomey's theory to show that marine biogenic production of dimethyl sulfide (DMS) could have a major effect on the control of the planet's climate through its ability to form CCN after its oxidation in the atmosphere.

2 other classes of particle may also influence cloud radiation balance. Twomey (1974), pointed out that optically absorbing particles (such as elemental carbon) if immersed in cloud drops could decrease cloud albedo. The formation of ice crystals in supercooled clouds, which are common at high latitudes, concentrates the water into fewer, larger hydrometeors. Ice forming nuclei, which are particles having the ability to produce ice crystals in such clouds, therefore may also affect cloud radiation. The possible effects are complicated by the production of precipitation and consequent resulting changes in cloud area and lifetime.

Although aerosol particles have this large potential climatic impact, a lack of globally distributed data and understanding of some of the processes of particle generation and growth and of their interactions with clouds make it difficult to incorporate their effects in global climatic models. Some of the reasons why the central Arctic Ocean in summer is a specially favourable region for research into these complex but very important problems are as follows.

(1) It represents an appreciable proportion of the world's surface area and because of the relatively thin cover of highly reflecting ice is important for the radiation balance. It is a region in which very few studies of aerosol concentrations, size distributions and the properties of the particles that might affect clouds have been made in summer, when effects on the radiation balance are greatest. Most of the previous effort has been directed towards the "Arctic haze" of early spring (Rahn et al., 1980).

(2) Because the atmosphere over the central Arctic Ocean in summer is to a large extent separated from airmasses associated with strong sources of pollution (Liljequist, 1970) it is a very favourable area in which to explore the natural sulfur cycle of biogenic origin. Studies of seawater DMS, gas phase DMS and its transformation to particles, the growth of the particles that leads to them becoming potential CCN and finally the way in which the particles can be cleansed from the atmosphere can all be made with a minimum of interference from particles having other sources.

(3) Cloudiness has a maximum in summer (Schweigger and Key, 1992; Curry and Ebert, 1992) with optically thin stratus layers predominating. Concentrations of particles available for creating cloud drops are also usually low, so that

conditions favour a large effect of CCN concentration on the radiative properties of the clouds.

To improve understanding of the cloud mediated effect of natural aerosol on radiative transfer and climate, measurements were planned for the International Arctic Ocean Expedition of 1991 (IAOE-91) that would follow the sources of atmospheric sulfur, its transformation to particulates, the growth of the particulates to cloud-active size and their removal from the atmosphere. The starting point was to find the geographical distribution of, and temporal changes in, reduced sulfur compounds in the ocean and their relation to biological activity. Atmospheric measurements would show the extent to which these were transferred to the air and lost due to oxidation or other processes. Observations of particle concentrations down to 3-nm diameter were expected to reveal conditions for the production of new particles. By forming complete number size distributions up to 500-nm diameter and making measurements of CCN it was hoped that the growth of the particles to cloud-active size and their interactions with clouds could be interpreted. Other particles that have the potential for influencing cloud radiative properties, ice forming nuclei (IFN) and elemental carbon (EC) were to be included in the study. Finally, the chemistry of the particles segregated by size was to be studied in order to find whether the seasonal changes of the sulfur-containing components followed those of the seawater sources.

This overview aims to describe the extent to which the objectives of that ambitious program were met, some of the theories suggested to explain the observations and the problems remaining to be solved. Details of the sampling manifold, an outline of the instrumental set-up and a statistical summary of the data (cf. Tables 1a, b) will be found in the Appendix.

1.2. Conditions encountered during the expedition

The icebreaker *Oden*, chartered from the Swedish National Maritime Administration by the Swedish Polar Research Secretariat, was of recent construction and incorporated many novel design features that allowed it to become the first conventionally powered ship to reach the North Pole.

Oden left Tromsö in northern Norway on the 1 August (Julian day 213), crossed the Barents Sea to Fram Strait and the Greenland Sea and then operated west and north of Spitsbergen into the marginal ice zone from 4–7 August. The German polar research ship *Polarstern* also left Tromsö on the 1 August but proceeded to the east of Spitsbergen, rejoining *Oden* on 4 September. *Oden* stayed in Isfjorden, offshore from Longyearbyen in Spitsbergen, from 8–17 August for mechanical repairs. It then operated over the ice-covered central Arctic Ocean, passing over the Nansen and Amundsen basins, with a short visit to the Makarov basin. The geographic North Pole was reached on 7 September, together with *Polarstern.* On 5 October (Julian day 279), *Oden* again reached the open sea in the same area as on 4–7 August, allowing a comparison of the various quantities measured in two very different seasons. Fig. 1 presents the cruise track and location of stations for atmospheric research during which the ship was stationary (between 12 and 24 h) and headed into the wind.

Fig. 1 also shows the distribution of maxima and minima in the extent of the pack ice encountered. Within the pack ice of the central Arctic Ocean, there was an average of 10% of open water during August and open leads existed right to the Pole through the 2-m to 4-m thick perennial ice. All open leads became covered with thin new ice as the water temperature decreased to below freezing after mid-September. Further details of ice cover are given by Leck and Persson (1996a) and Liljeström (1992). During the expedition, the sun was continuously above the horizon until 13 September.

From August to mid-September, weather conditions were typical for the central Arctic Ocean summer. Advection of airmasses from the Atlantic sector and open seas north of Siberia dominated over shorter periods of air being either advected from continents or subsiding from the free troposphere. In general this gave weather conditions of high relative humidity (90–100%) and air temperatures in the vicinity of 0°C, as the relatively warmer maritime air was cooled when advected over the pack ice. An optically thin, low cloud cover of stratus was present most of the time. There were frequent fog episodes. After mid-September, advection of airmasses which had followed a long path over the ice and airmasses subsided from the free troposphere became more frequent. Thus, the air sampled was generally cooler (as cold as −23°C) and drier with less cloud cover and fog.

Winds ranged from an occasional calm period

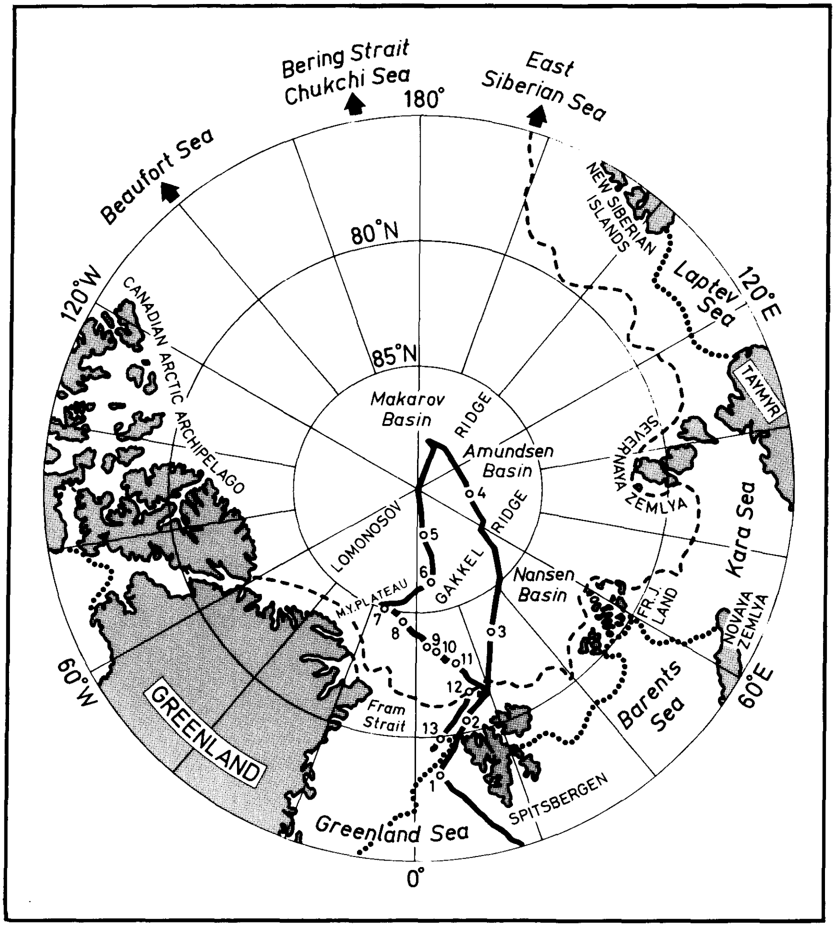

Fig. 1. Map of the study area, showing the cruise track (——) during the International Arctic Ocean Expedition, 1991 (IAOE-91). Stations with between 12 to 24 hours continuous sampling are marked. Also illustrated is the geographical distribution of the most southerly (••••) and the most northerly (– – – –) location of the ice edge zone during the expedition.

to 21 ms^{-1} with an average value of 6 ms^{-1}. Significant precipitation was rare, being confined to a few light snow showers. Fog droplets however often became large enough to have appreciable sedimentation rates while widely spaced ice crystals fell much of the time when air temperatures were 0°C or below.

Of crucial importance to many of the measurements and their interpretations was the history of each of the airmasses sampled. It was derived from 5-day receptor-oriented trajectories at three arrival heights, calculated using the 1991 version of McGrath's (1989) three dimensional model. The latter used the analysed horizontal and vertical wind components and mass fields of the European Centre for Medium Range Weather Forecasts. The expectation that the central Arctic in summer is largely decoupled from the bordering continents was well verified by these analyses, as only 6% of the 5-day trajectories included any significant pollution sources.

Only 5 well-defined fronts passed the ship during the 67 days of the expedition and this slowly changing mesoscale or synoptic scale meteorology appears to be typical of the region at these times of the year. Correspondingly, 7 consecutive periods were identified which had meteorological similarities but trajectories within those periods often showed considerable variations. This variability was useful for estimating the effects on measured parameters of such factors as transport time from the open ocean, from land which could provide sources of particles or from the free troposphere.

2. Observed variations in seawater and atmospheric constituents on three time scales

The first measurements were made at the height of summer while towards the end of the expedition many of the measurements were made in conditions approaching those of winter. Changes were observed in all the measured quantities on long, medium and short time scales. The long time scale changes can easily be associated with seasonal changes over nearly 10 weeks and the medium time scale changes with airmass or trajectory changes typically ranging from about a day to a week. The short-term variations ranging from tens of minutes to a few hours in length were unexpectedly frequent, often very large and most had no obvious interpretation.

The separation into three time scales forms a convenient basis for a description of the changes observed and how they have led to some interpretations of the processes involved, even though the time scales are likely to be linked to some extent.

2.1. *Seasonal effects*

If it is assumed that biological production of DMS provides a major contribution to aerosol generation, then seasonally changing biological productivity should be reflected not only in its concentration in seawater and in air but also in the mass of methane sulfonate ($CH_3SO_3^-$, MSA, which is derived from DMS by oxidation in the atmosphere) in the aerosol. If it is also a major contributor to non-sea salt sulfate (nss-SO_4^{2-}) in the aerosol or to sulfur dioxide (SO_2) concentrations, these too should show similar seasonal changes.

Surface seawater DMS did indeed show a clear seasonal progression, decreasing by about three orders of magnitude in concentration between August and October. On average DMS fell about 30% per week in the waters south of and within the ice edge zone whereas a higher seasonal decline, about 45% per week, was observed in the pack ice during the period of re-freezing (Leck and Persson, 1996a). The measurements demonstrated the importance to DMS concentrations in seawater of the phytoplankton bloom and zooplankton abundance in the open water near the ice edge, in the wake of the receding ice and in the pack ice during summer ice-melts. However, they failed to find a clear relation between DMS concentration and phytoplankton standing stock, although there was a probable relationship to degraded phytoplankton material (Phaeopigments). This agrees with the result of earlier studies (Nguyen et al., 1988; Turner et al., 1988; Leck et al., 1990).

The apparent mean seasonal decline in the total atmospheric DMS data agreed closely with that in the open waters south of and within the ice edge zone during summer ice-melt. The decline was reduced to 20% per week if mean losses during advection over the pack ice were taken into account by normalising the data to zero transport time. This assumed that the observed rate of decline in seawater DMS at the ice edge was the same at lower latitudes. It is quite likely that the DMS source strength in the seas south of the ice edge does not change as rapidly with time, so the discrepancy may not be real.

To study the relationship between seasonal changes in seawater DMS and particulates and their chemistry, it was necessary to remove as far as possible interference from other possible sources such as continental pollution and subsidence from the free troposphere. On the basis of air trajectories a "marine biogenic" subset of the total data was constructed (80% of the total data). This subset excluded cases where air had subsided from the free troposphere within the previous 48 h, or in which significant anthropogenic influences may have been present. As illustrated in Fig. 2, it showed that submicrometer masses of both MSA and nss-SO_4^{2-} declined at much the same rate as DMS in air and seawater, although there are again some unknown factors in calculating the true rates which have been discussed by Leck and Persson (1996b). A very similar seasonal change was found for the submicrometer ammonium (NH_4^+). This indicates links not only between the different sulfur

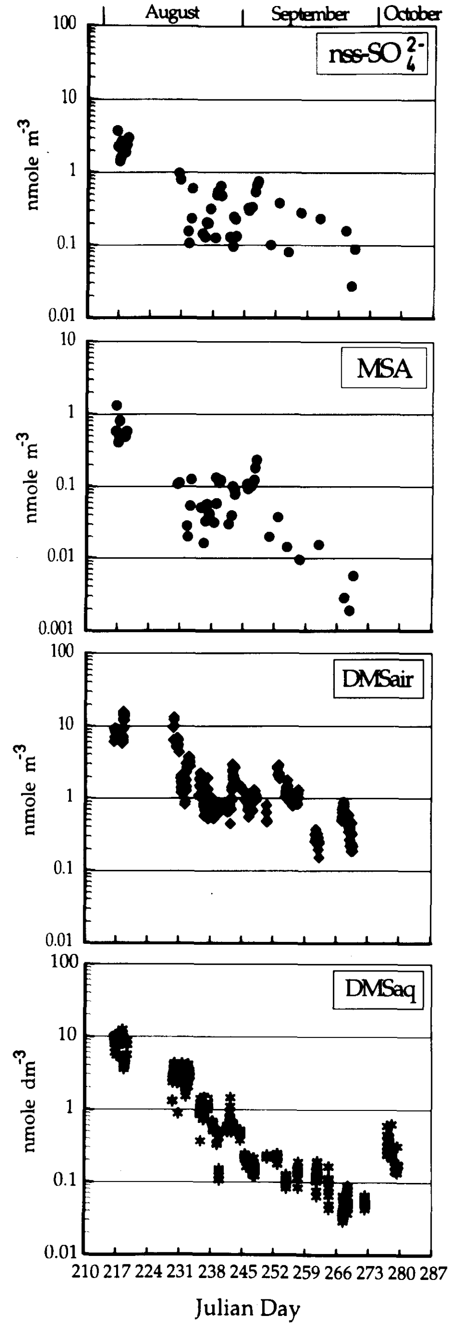

Fig. 2. Temporal distribution of seawater DMS, atmospheric DMS, and submicrometer aerosol MSA and nss-SO_4^{2-} in the "marine biogenic" sub population between August and October, 1991. All values are at STP. Between Julian Day 230 to 273 sampling was performed in the pack ice area.

compounds but also between the biogenic sulfur and reduced nitrogen cycles.

Similar mean seasonal declines in concentration occurred for the "marine biogenic" data set in Aitken mode (25 to 80 nm diameter), accumulation mode particles (80 to 500 nm diameter) and CCN concentrations (anywhere within the measured supersaturation range of 0.1 to 0.65%).

While SO_2 concentrations only showed about half the apparent seasonal rate of decline of the other sulfur species, the uncertainty in the estimate was much wider than for them.

The agreement between apparent seasonal changes in seawater DMS, atmospheric DMS, MSA and nss-SO_4^{2-} in submicrometer particulates and in CCN suggests strongly that the particulates in this area, during the periods selected on the basis of air trajectories to be relatively free from non-biogenic sources, were ultimately derived from the seawater DMS. As the selected periods represent 80% of the total data we can say that for a major proportion of the time between August and mid-September submicrometer particulates are predominantly derived from seawater DMS. In terms of mass of sulfate this may not be true, as other sulfur sources may contribute a comparable or even larger total mass in the 20% of cases excluded.

The basic premise of the Charlson et al. (1987) hypothesis that DMS emanating from marine organisms and their products can affect the concentrations of particles large enough to act as CCN in clouds is therefore supported for the central Arctic in summer.

The next step in the Charlson et al. (1987) hypothesis, that the DMS-derived particles could therefore substantially affect the radiation balance due to their influence on cloud drop concentration, is much more difficult to verify. Curry (1995) has discussed some of the considerable complexities of radiation balance in a cloudy Arctic environment. However, some general statements are possible. Particulate concentrations were generally so low after mid-September that no direct radiational effects on the environment are likely. Indirect effects after that time would also be minor because of the rapid decline in cloud cover and the low sun. Thus if there are any substantial indirect effects of DMS-derived aerosols on the radiation balance, they will be mainly confined to the summer period.

Elemental carbon, which may also affect cloud radiational properties, does not appear to be of great importance in the summer Arctic. Its concentrations were too low for there to be any significant effects on cloud albedo and no obvious seasonal changes in concentration were observed. This absence of seasonal change did indicate that the apparent seasonal trends in sulfur compounds were not due to a changing anthropogenic input.

The remaining cloud-active particles, the IFN, did show a seasonal change. Concentrations remained approximately constant from August to mid-September, then declined at an increasing rate totalling an order of magnitude by 6 October. During the summer the marine boundary layer (MBL) clouds were too warm for ice crystal production. Ice crystal production was therefore limited to middle or high level clouds, crystal habit indicating a preferred growth temperature of −15°C. It is doubtful whether concentrations measured near the surface would be relevant to such clouds. After mid-September, the decreasing cloudiness reduced the chances that IFN could significantly affect radiation. The main environmental role of IFN may therefore have been in the formation of precipitation and consequent aerosol scavenging.

2.2. *Changes in aerosol and gas concentrations associated with airmass or trajectory changes*

As mentioned earlier, the synoptic scale meteorology changed slowly relative to that at mid latitudes but large trajectory changes on a scale of hours occurred when they passed near centres of circulation. Where a trajectory changed sufficiently to alter substantially the time of transport of the air from open water, bare land or the free troposphere, there were usually conspicuous changes in concentrations of trace gases such as DMS, ozone and radon and of particulates. Such changes were useful in determining mean rates of loss from the source regions. For example, considering a short period (so as to reduce seasonal effects) having very variable transport times from open water, DMS and submicrometer aerosol concentrations showed a mean exponential loss rate of about 35% per day of transport over the pack ice as shown in Fig. 3 (Leck and Persson, 1996b).

Although the mass median concentration of submicrometer aerosols declined with time of transport the ionic ratios stayed approximately constant, indicating an approximately uniform loss of all constituents. It is consistent with the fact that the rate of decrease in concentration with transport time from the ice edge of the smaller Aitken mode particles was much the same as that of larger submicrometer particles. This is perhaps surprising in view of very different sinks and sources.

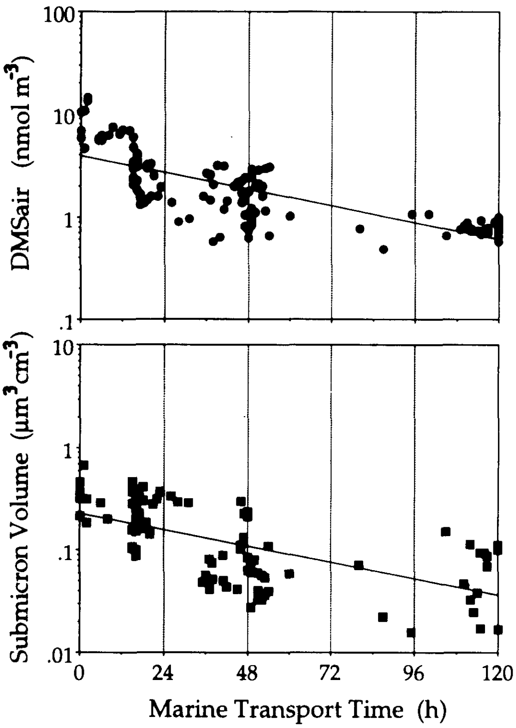

Fig. 3. Plot of atmospheric DMS (top panel) and submicrometer aerosol by volume (bottom panel) concentrations against marine transport time. Reprinted from Leck and Persson (1996b).

2.3. *Short-term changes*

Sudden very large changes in aerosol number size distributions were an unexpected feature of the results. Although they had been noted before in Greenland (Hogan et al., 1982) and on the Ymer-80 expedition (Lannefors et al., 1983), they had been linked to such events as precipitation or relatively local pollution, neither of which was a likely cause on IAOE-91 during stations when the

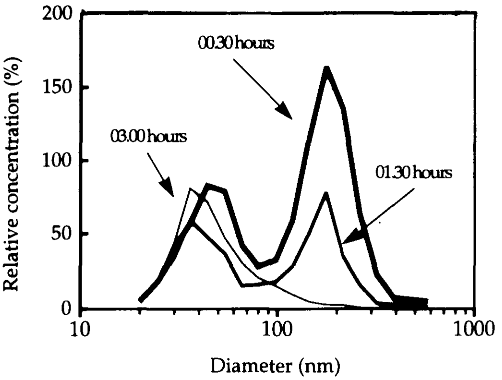

Fig. 4. An example of a sudden change in accumulation mode particle number concentrations at latitude 88.5° N. Units are in relative concentrations as percentages of the number concentration between 00.00–03.00 h. Reprinted from Bigg et al. (1996).

ship was correctly oriented into the wind and far from any other human activity. Such changes were also linked by Lannefors et al. to fog events but Nilsson and Bigg (1996) have shown that during IAOE-91 fogs had only a small mean effect on particle concentrations.

Defining a "sudden change" to be more than a factor of two change in either Aitken mode or accumulation mode number concentration within one hour or less there were about 130 cases during the expedition (Bigg et al., 1996). A typical example is shown in Fig. 4. There were far more occasions of sudden changes in ultrafine particle (3 to 25 nm diameter) concentrations. Many such events would of course have been missed because there was little pollution-free time when the ship was moving. Concentrations sometimes changed by more than two orders of magnitude and usually returned to near their level before the change after an hour or two. On occasions the changes were quasi-periodic with a succession of maxima and minima.

These changes were sufficiently large and short-lived that at times they could introduce wide scatter into measurements which required an hour or more to complete. Since they have not been reported as a common phenomenon in other remote marine areas, conditions in the summer Arctic may be specially favourable for observing them. It is evident that such events must be explained if the way in which the aerosol evolves is to be understood. The problem will be discussed further in Section 5 after introducing the necessary background of aerosol formation, size distribution, removal processes and boundary layer meteorology.

3. Aerosol formation, growth and chemistry

Complete number size distributions from diameters of 3 nm to 500 nm were produced every ten minutes allowing studies to be made of the formation of new particles and of the way in which the particles became concentrated into size groups. The aerosol statistics and size distributions were as follows and are discussed in more detail in Covert et al. (1996) and Wiedensohler et al. (1996).

(1) A measurable number of particles smaller than about 25 nm diameter, indicating relatively recent formation, was present for a surprisingly large proportion of the time (65%).

(2) 3 distinct and clearly separated size groups were nearly always present having modal diameters centred around 14, 45 and 170 nm, already referred to as the ultrafine, Aitken and accumulation modes. Well-defined minima occurred at 20–30 nm and 80–100 nm. On average the accumulation mode accounted for about 45% of the total number of particles and the Aitken mode 40%.

(3) The geometric mean diameters and standard deviations of the modes were relatively constant, the greatest variability being in the ultrafine mode, and independent of concentration and airmass source.

To define the controlling factors in the formation of particles, relationships were sought between number concentration, surface area and geometric mean diameter of the ultrafine mode and the same properties of the larger particle modes and also concentrations of radon, ozone, DMS and absolute humidity. Theories of new particle formation and experience elsewhere require that there should be an anti-correlation between new particle formation and accumulation mode surface area (Hegg et al., 1992). No such anti-correlation was found. If ultrafine particle production occurs only in regions having low particle surface area then they must be produced in such large numbers that a very slight mixing to the measuring point results in a measurable increase in their concentration without a large accompanying decrease in particle surface area.

The geometric mean diameter of the ultrafine mode seemed to be correlated with DMS and absolute humidity, indicating that these factors are involved in their growth as well as in their formation. The number concentration anti-correlated with the geometric mean diameter, implying that concentrations decrease as the particles age and grow.

While production of both ultrafine and Aitken mode particles may be possible within the MBL in particular circumstances, the most likely place for new particles to form in the MBL was considered by Hegg et al. (1990) to be in the region just above the top of clouds where cloud drops are evaporating into the drier air. Here scavenging and coalescence of drops will have reduced the available surface area for condensation of gaseous oxidation products, unoxidised precursor gases in the drops will be released to the atmosphere and ultraviolet radiation is strong. Each of these factors favours the nucleation of new particles. Since cloud top is usually defined by a temperature inversion or stable layer, the formation of new particles will often be confined to a thin region with very strong concentration gradients. The relatively high concentrations of the new particles, their continued production and the atmospheric stability all favour growth to Aitken mode size.

The conspicuous and stable separation of the Aitken and accumulation modes appears to be adequately explained by the processes described by Hoppel et al. (1991, 1994). They proposed that particles larger than about 80nm diameter were involved in cloud drop formation and subsequently grew by uptake of gases such as SO_2, followed by aqueous phase oxidation. Evaporation of cloud droplets then released these larger particles to the air. Within the cloud, they also found that concentrations of interstitial accumulation mode particles were extremely low.

The presence of very strong vertical concentration gradients in all three particle modes is therefore to be expected when clouds are present. Because the processes are different and act on different time scales, the profiles of concentration may also be very different. Accumulation mode profiles are likely to vary to a greater extent than Aitken mode profiles because of in-cloud losses, while the ultrafine mode profiles are likely to have greater variations than either because of more localised production regions and probably higher maximum concentrations. These points are essential to the explanation given in Section 5 of the sudden changes in concentration that were observed.

3.1. *Chemistry of the aerosol*

The common ions in the aerosol were studied using filter packs ($<1\ \mu$m aerodynamic diameter (EAD)) and impactors ($<10\ \mu$m EAD) having a time resolution of 4 to 24 h depending on the concentration of particles present (Leck and Persson, 1996b). A much more detailed elemental analysis, also size segregated but with lower time resolution was also made (Maenhaut et al., 1996).

Over the open ocean, nss-SO_4^{2-} and NH_4^+ ions showed a bimodal distribution with about 70% of their mass in the submicrometer particle fraction. Only 15% of the distributions over the pack ice were bimodal but the proportion of the mass contained in the submicrometer size fraction was much the same as over the open ocean. In view of many previous observations (Hubert et al., 1993; Ayers et al., 1991) which have shown much of the MSA to be associated with supermicrometer particles, the most surprising result was that 80 ± 8% of the mass over the open ocean and 70 ± 13% over the pack ice was associated with the smallest particles ($0.16 < Dp < 0.53\ \mu$m EAD) and only 8 ± 3% with the largest size fraction ($1.6 < Dp < 5.3\ \mu$m EAD).

MSA and nss-SO_4^{2-} were highly correlated with an average molar ratio of 22% in the "marine biogenic" data set. At all stations in the pack ice the molar ratios did not vary with particle size. From the open ocean samples however, molar ratios of less than 5% occurred in the largest size fraction. The fact that MSA was found predominantly on smaller particles than nss-SO_4^{2-} suggests that the sulfate formed on the basic sea salt particles by both reaction (SO_2 oxidation) and deposition, while MSA arose only by deposition. However it does not explain why the molar ratio on the large particles was different in the open ocean and pack ice subsets.

The MSA to nss-SO_4^{2-} molar ratio did not correlate with observed temperatures showing that the processes controlling the particulate chemistry do not exhibit a net temperature dependence, in contrast to laboratory studies by Hynes et al.

(1986) and a field study by Bates et al. (1992) but in agreement with a study by Heintzenberg and Leck (1994).

In the same "marine biogenic" selection, the ammonium to non-sea salt sulfate molar ratio was close to 1 for the smallest particles (also verified by single particle analysis) but was closer to 0.5 for the largest particles.

The multi-elemental analyses showed only 3 elements associated predominantly with the fine particle fraction: S, C, and I. The iodine was evidently formed from a gaseous precursor and from changes in concentration with transport time was evidently from a marine source. Previous Arctic investigations by Barrie and Barrie (1990) had arrived at the same conclusions. With the exceptions of S, C, and I, all elements measured including the anthropogenic elements such as Zn, As and Sb were predominantly associated with particles $>2\,\mu$m EAD.

Comparison of total suspended particulate mass (TSP: $<10\,\mu$m EAD) obtained by weighing filters and total suspended particular mass obtained from ion chromatography of the common ions showed the samples clustered in two groups. In one of these up to 100% of the mass was explained by the ions sodium, ammonium, potassium, chloride, nitrate, sulfate and methane sulfonate. In the other only 20–40% was explained by those ions. The samples showing the largest difference were collected in an area influenced by river run-off from the Siberian coast. The multi-element analyses showed a concentration of crustal material high enough to explain a large fraction of the unidentified residue in the ionic TSP determinations.

In view of the aerosol size distributions it is clear from the analyses that the great majority of submicrometer particles must be predominantly ammonium bisulfate with a significant component of MSA. While much of the total mass resides in larger particles having either a sea salt or crustal origin, when possible radiative effects on clouds and climate are being considered, they can be ignored.

4. Boundary-layer meteorology

The concentration of any atmospheric component produced at the surface, such as DMS, depends critically on the depth of the surface mixed layer. In the absence of mixing at cloud levels, strong gradients in concentration of particles will develop as described in Subsection 3.1. Air interchanges between surface and cloud levels, or between anywhere in the MBL and the free troposphere may therefore lead to large changes in concentration of these components. Correct interpretation of such changes requires a detailed knowledge of boundary layer structure and the possible air interchanges that may occur.

While the program certainly did not solve all the problems, considerable progress was made towards defining the boundary layer and its evolution, listing the types of mixing events between levels that the structure indicated to be possible and finding evidence that at least some of them had occurred.

In outline, the observations (Nilsson, 1996) showed that a stable layer grew in height from 300 to 1700 m in proportion to the square root of the distance from the ice edge up to a distance of 3000 km. After the first 400 km an underlying mixed layer formed, typically 100 to 250 m thick, which was related to the presence of low level jets (wind maxima) in 60 to 90% of the profiles. Layers above and below the jet centres having Richardson numbers that would allow the development of mixing were present in 65 to 80% of the profiles.

Of particular interest to the interpretation of aerosol growth and possibly formation and removal from the atmosphere, was the observation that cloud streets and roll vortices occurred. Their presence was associated with air that had been advected for 2000 km or more over the ice and was apparently due to the cold air advection and weak convection. The rotational motion within such clouds gives the air a much longer path over the pack ice than if it were proceeding in a straight line and may involve many cycles of condensation and partial or complete evaporation of cloud droplets.

The meteorological studies therefore turned out to be of far more scientific value than would have been predicted from the knowledge that mesoscale and synoptic meteorology change in the Arctic summer more slowly and generally involve weaker systems than those commonly found at lower latitudes.

4.1. *Clouds, fogs and aerosol removal processes*

Stratus or stratocumulus clouds were present almost continuously until mid-September. The rawinsondes usually suggested that cloud base was only a few hundred meters from the surface and that there were multiple relatively thin layers of cloud separated by regions where the humidity remained high. Satellite photographs showed that middle- and high-level clouds were often present but not nearly as persistent as the lower clouds. After the MBL clouds diminished in mid-September altocumulus and cirrus clouds were often seen to be present.

The role of clouds in determining the aerosol size distribution and vertical profiles has already been discussed. Their other influences include particle removal from the atmosphere and sites for aqueous oxidation processes.

Significant precipitation from the clouds was rare and because ice crystal habit showed that the crystals had usually grown at levels where the temperature was near $-15°C$, must predominantly have come from mid-level clouds. While ice crystals were rarely present in sufficient numbers to be classed as precipitation, widely separated single crystals were very frequently present when the air temperature was below 0°C. With their stellar shape and relatively large sizes (2 to 10 mm diameter, typically 6 mm) their fall velocity was quite high and their passage through the boundary layer would have caused a slight but continuous drain on the aerosol population.

Fogs were found to produce small drizzle drops. Again concentrations were very low (much less than $1\ \mathrm{cm}^{-3}$) but since fogs were frequent there would be a slight drain on accumulation mode particles. Because measurements of fog droplet size distributions and concentrations were only made well within the pack ice region, their ability to deplete the aerosol may have been underestimated. Fog formation is likely to have been more frequent and more vigorous near the ice edge.

5. Causes of sudden changes in aerosol concentration

The sudden changes in aerosol concentrations, more frequent in the accumulation mode particles than in the Aitken mode and more frequent in the ultrafine mode than in either, were described in Subsection 2.3 and a typical example shown in Fig. 4. In view of the expectation that independent vertical gradients in concentration of all 3 modes would become established during periods of stability, the obvious explanation of the changes is that sudden vertical mixing events brought air from higher levels to near the surface.

A good indication of how it occurs has been given by facsimile records of turbulence in the lower part of the Arctic MBL obtained using acoustic sounding (sodar). Cheung (1991), for example, has published some excellent examples of the impermanence of the stable non-turbulent layers. While turbulent layers may remain separated for hours, sooner or later Kelvin-Helmholtz wave instabilities will build up, the waves will break and there is a sudden overturning of the air that may extend to the surface. Such events are not confined to the Arctic but are common anywhere there is a thermally stable layer with subcritical Richardson number and wind shear. Ottersten et al. (1973) show many examples from the early literature on the subject. Typically such events last from 10 to 15 min but as they are triggered by gravity waves (Finnigan, 1988), they may recur and last for varying times.

Concentrations of other tracers such as radon, ozone, or DMS often correlated well with changes in aerosol concentration but the correlations were sometimes positive, sometimes negative and sometimes changed sign within the event. This observation was at first puzzling because the correlations were often too good to be mere coincidence. However, when we consider that each tracer probably develops its own profile of concentration related to its sinks and sources and the changes of air trajectories with arrival height (often appreciable for small changes in arrival heights), it is precisely the result that would be expected when air from some higher layer suddenly reaches the surface.

As the boundary layer meteorology showed, the Kelvin-Helmholtz breaking waves at interfaces are not the only means of mixing air between levels. Roll vortices provide another path and may produce quite a different result because the mixing processes and cloud interactions are very different.

If air is indeed being brought to the surface from above the boundary layer, then obviously air from the surface must also be transferred out of the boundary layer. This may represent a sink for all atmospheric components produced at the surface or in the boundary layer and be responsible for at least part of the losses at surface level as the air progresses over the pack ice. It is obvious that these mixing processes are fundamental to an understanding of the evolution of the Arctic aerosol and of all the observations made at the surface and will have to receive far more attention in any future expedition.

The MBL studies showed that the surface mixed layer was much thinner, the successive cloud layers closer together and the lower ones much closer to the surface than is common at lower latitude remote marine sites. On the above hypothesis, this would lead to more pronounced and more frequent changes in aerosol size distributions and bursts of ultrafine particles in the Arctic, as was observed. The important point to note is that the same explanation of changes and bursts of ultrafine particles may apply equally to the Arctic and to other remote marine sites.

6. Directions for future research suggested by the results of the expedition

6.1. Mixing processes in the boundary layer

While excellent progress was made in documenting concentrations and seasonal variations of particulates, chemical species, gases and seawater DMS, it became apparent when interpreting the results that atmospheric mixing processes were far more important than had been expected. The mesoscale and synoptic scale meteorology in the central Arctic may change slowly but the small scale mixing processes, while intermittent and patchy in their operation, have a large influence on all parameters measured near the surface. Ideally, sampling should be 3-dimensional, extending to beyond the MBL. From the point of view of expense and logistics this may be impractical except perhaps for limited measurements. Profiles could be inferred however from a knowledge of changes that occur at times when mixing to the sampling level takes place providing that these occasions can be identified and the height to which mixing occurs specified. Published Arctic time-height records of turbulence in the boundary layer using sodar such as those of Cheung (1991) suggests that acoustic sounders can provide that information.

Because the sudden fluctuations that occurred had not been expected to be so frequent or to have such large effects, temperature, pressure, humidity and wind speed and direction were not logged continuously. If they had been, it would have been easier to identify features such as roll vortices and to deduce their effects on surface layer constituents.

If we are to understand the evolution of the aerosol, the initial stage of nucleation has to be much better defined than it is at present. The conclusion that formation occurs somewhere above the sampling level is another strong motive for understanding the time dependence of the small scale mixing processes of the MBL.

The most important addition to research suggested by the IAOE-91 measurements is therefore a study of the causes and temporal changes in the small scale local mixing processes of the Arctic, how they vary with distance from the ice edge or with season, and how they affect the constituents measured near the surface. Therefore, a continuation of the rawinsonde and other meteorological measurements (including turbulence at the surface) would be needed to interpret the sodar turbulence profiles as frequently as possible.

6.2. The influence of aerosols on clouds and radiation

Since one of the justifications for research in the central Arctic Ocean is to evaluate the region's role in global climatic change, the effects of aerosols on clouds and the effects of clouds (including fogs) on the radiation balance were listed as objectives. However, because it was not possible to sample clouds above the measuring level and fog properties were studied only on a casual basis without a detailed knowledge of mixing processes from surface to cloud and cloud to surface, the influence of aerosols on cloud properties could not be established with certainty. To complement the measurements suggested above to deduce profiles of aerosol concentration and to make a start on the aerosol-cloud-radiation relationships, it would be very useful to continuously record fog droplet size distributions (assuming that higher clouds will remain inaccessible) and relate these to changes in radiation at the ice surface. In addition, the evolution of the fog droplet spectrum would be valuable in estimating aerosol losses through droplet sedimentation. Such observations should be accompanied by an array of radiation measuring instruments.

6.3. Some other problems in need of further research

Almost every measurement made, although adding considerably to the knowledge of central Arctic Ocean conditions, raised further problems that need to be answered.

For example, the measurements of atmospheric DMS and of DMS in open leads within the pack ice were made long after the summer solstice. Would the conclusion that this large area was a negligible source have been the same if measurements had been made earlier in the season?

The seasonal decline in seawater DMS was slightly greater than that in atmospheric DMS when corrections were made to the latter for transport time from open waters. The implicit assumption that the seawater source declines at the same rate in all bordering seas as it did in Fram Strait requires justification.

It was found that although atmospheric DMS concentrations were high near the ice edge and in the early part of the season remained appreciable during transport to higher latitudes (>80°N), sulfur dioxide concentrations were very low. Since SO_2 is thought to be a major oxidation product for DMS, either this is untrue or losses are larger than expected. If the former is the case, it provides a constraint on DMS oxidation schemes. However, because clouds fill a larger proportion of the boundary layer than at most lower latitude remote marine sites and there are intermittent mixing processes between surface and cloud levels, the sink strength due to oxidation in cloud drops may be large. It is clearly important to both theory and observation that the high DMS to SO_2 ratio should be explained.

The very low sulfur dioxide concentrations necessitated long sampling times with the method used. If a sudden severe depletion of the accumulation mode implies the arrival of air from cloud level, as suggested in Section 5, then a sulfur dioxide measuring instrument with good time resolution would allow an estimate to be made of the extent to which it was depleted in the cloud layer. This information would be very useful in determining sink rates for the SO_2, for modelling its aqueous phase oxidation and the resultant growth of the particles involved in condensation.

The aerosol number size distributions obtained on IAOE-91 were excellent and it would be essential to repeat them in any future research as they are needed for an understanding of many of the processes that are occurring. However, many unknowns remain. For example, the methods of formation and maintenance of the three modes of particles have been inferred rather than observed. It is not at all obvious why the modal diameters of the Aitken and accumulation mode particles are so close to those in lower latitude remote MBLs when cloudiness, mixed depth and precursor gas concentrations are probably often quite different.

The chemical composition of the aerosol contains even more unknowns that are very important for a full understanding of the processes of aerosol formation an evolution. What is the chemistry of the ultrafine and Aitken mode particles? This will be a very difficult question to answer because of the very small mass involved. Even with the larger particles many questions remain. For example, what is the organic content of the aerosol? Why is MSA in the Arctic almost exclusively on small particles when elsewhere it tends to be distributed over all sizes? Is the fine fraction MSA associated with a particular type of particle, or can it be (in spite of vapour pressure considerations) in a pure or almost pure form?

One of the interesting possibilities derived from elemental chemical measurements was that there is a wind-produced coarse aerosol source on the pack ice itself, derived from the drift of contaminated ice formed adjacent to Siberian river outlets. This hypothesis certainly needs verification.

Removal processes in the atmosphere would be another profitable area of research as it was not adequately studied during this expedition. Growth and sedimentation of fog droplets need to be monitored continuously as does the number, size and level of origin of ice crystals. While the open ocean source of the ice nuclei was suggested by the expedition results, their nature has still not been established and deserves further study.

7. Conclusions

The integration of so many facets of the complex processes relating to sources of sulfur in the atmosphere, its conversion to particulates, cloud interactions and dependence on meteorological processes and the ultimate environmental effects presented a large challenge. The results exceeded expectations in that they not only produced documentation on concentrations and seasonal changes of so many constituents of the central Arctic Ocean atmosphere (and water) in regions where no previous measurements had been made but provided new insights into the processes involved in regulating them. Part of the reason for the success was the long duration of the expedition which allowed seasonal changes to be explored.

The demonstration that central Arctic Ocean summer airborne sulfur concentrations are largely controlled by biogenic DMS produced in seas adjacent to the ice edge forms an interesting contrast with the situation in spring, where the aerosol is known to be controlled by distant anthropogenic sources. This statement, however, is based on the "marine biogenic" data set that we have chosen, the region studied and the period August to mid September. It is possible that other sources may have contributed substantially to the sulfate of the region in the excluded cases in other periods or in other places.

The submicrometer aerosol measurements represent the first highly size resolved and lengthy observations of aerosol in the Arctic MBL and should be particularly useful for modelling purposes. Because CCN concentrations were found to be so closely linked to total particle concentrations larger than a certain size (assuming the dominant chemical nature determined by measurement) the measurements would also be useful for modelling clouds in the area.

Another notable achievement was a detailed description of the atmospheric boundary layer of the region and a recognition of its extreme importance to interpretation of measurements made near the surface. It leads to the suggestion that the role of MBL processes should be a necessary part of programs studying formation, growth and fate of aerosols at other sites.

While the gas and aerosol measurements provide an excellent basis for modelling cloud radiative processes, there were no direct observations made with which to compare the models. This was a result of the logistic difficulties and expense of such an undertaking which will probably also limit future research in this area.

8. Acknowledgements

One of the reasons for the success of the program was the excellent planning of the logistics by the Swedish Polar Research Secretariat. Another was that the friendly and helpful crew on the ship did everything possible to make the voyage a pleasant and productive one. It is a pleasure to record the generous support given by Professor Henning Rodhe to the IAOE-91 program and the penetrating and helpful comments he made on this manuscript. Special thanks must go to Leif Bäcklin and Sune Moritz for their invaluable help with equipment and logistics. Madelein Westin we thank for help with the meteorological observations and Robert Tryzell for help with the chemical measurements. We are also grateful to Ulla Jonsson for help in drafting the figures and to S. Whittlestone for providing the radon counter. We also express our deepest thanks to everyone at the Department of Meteorology, Stockholm University, Sweden who had helped us in making this work possible. This research was funded by the Swedish Natural Science Research Council (contract E-EG/GU 9906-30/E-EG 3922-313), the International Meteorological Institute and The Knut and Alice Wallenberg Foundation.

9. Appendix

Experimental details

9.1. Sampling manifold

The atmospheric program was housed in two laboratory containers (one of them connected to a pump house) on the 4th deck and a small container above the bridge containing the rawinsonde equipment. Gases and aerosol particles were to be sampled with a minimum interference from the ship and from the sea/ice surface surrounding the ship. For that purpose, facing into the forward direction, an inlet and air sampling system was set up on top of one of the laboratory containers such as to maximize both distance from the sea level and from the superstructure of the ship.

The inlet system consisted of three masts that extended at an angle of 45° to about 3 m above the container roof so that the height of the inlet points above sea level was about 25 m. For service and/or sample changes, the masts could be felled backwards onto the container roof. Mast one was used for simultaneous measurements of SO_2 and submicrometer filter samples at ambient conditions. It had a system of cyclones which excluded all particles above one micron aerodynamic diameter (EAD), at ambient relative humidity (RH).

Mast two conducted sampled air through a 4 cm i.d. pipe into the laboratory downstream of another combination of 1 μm EAD cyclones operating at ambient RH. The total flow was set to 550 litres/min. Inside the container, 6 different gas and particle samplers and instruments were connected to this sample stream through isokinetic take off lines from the main pipe of mast two.

On mast 3, an Andersen impactor (Andersen Inc. Atlanta, GA) excluded particles larger than 10 μm EAD at ambient RH from the sample stream which was then conducted through a 9 cm i.d. pipe into the laboratory. Because of the high flow rate through this inlet, (1130 liters/min), the residence time between inlet point and the first air take off from this inlet was about 0.2 s. Thus the pollution sensor, (see below), for the whole sampling system was connected to this sample point in order to be able to sense contaminating air before it arrived at any of the samplers. Downstream of this pollution sensor, 13 aerosol instruments and samplers were connected to the main line through isokinetic take offs.

The waste flow of all intakes and instruments was conducted to the pump house, through dry air gas meters and pumps to a plenum. A particle filter connected to this plenum filtered all exhaust air. Fig. 5 gives a schematic view of the complete arrangement.

The pollution sensor [CNCC], of the sampling system consisted of a condensation particle counter (TSI 3760, TSI Inc., St. Paul, MN) which was connected to an intelligent controller (Ogren and Heintzenberg, 1990). Experiments were carried out early in the expedition to determine wind sectors safe from shipboard pollution by measuring condensation nucleus concentrations while releasing massive amounts of smoke from various parts of the ship which was successively oriented in different directions relative to the wind. The set up showed that sampling of air was uncontaminated by ship borne emissions provided that the wind was within the sector ±70° of the bow. While the laboratory container described in detail above served as an inlet and air sampling system for the various gas and aerosol parameters measured, the second laboratory container housed two Gas and Ion-Chromatographs for their chemical analyses. It also contained a glove box which enabled preparation and change of the filter substrates in air free from particles, SO_2 and ammonia. Deionised water with a conductivity of 18 $M\Omega^{-1}$ cm^{-1} was produce by a Millipore Alfa-Q system attached to the water supply inside the container.

9.2. *Instrumentation for sampling and size distribution and determination of aerosol particles*

The complete set of instruments used during the expedition is described in detail in each of the contributed manuscripts appearing in this issue. Below follows a brief summary. The instruments descried were connected to the sampling inlets as shown in Fig. 5.

9.2.1. Size distribution by number. One differential mobility analyser (DMA) followed by a condensation particle counter (CPC, TSI 3760) as sensor scanned upwards from 20 nm in 11 equal log mobility steps per decade every 10 min

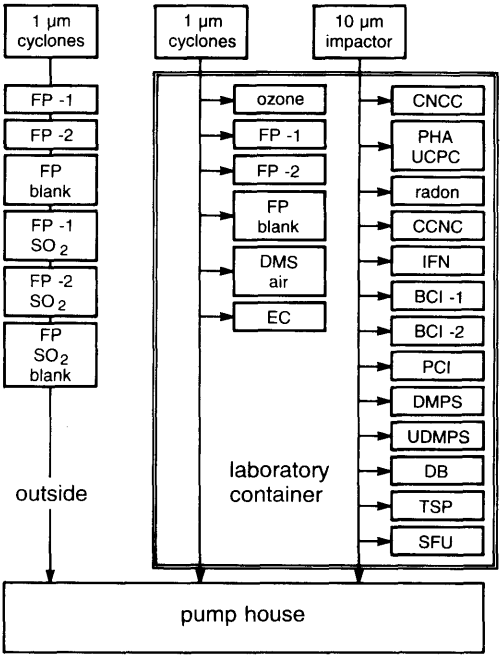

Fig. 5. Schematic overview of the sampling system for gases and particles, (cf. text for details).

Table 1a. *Concentration statistics for seawater and gas phase DMS, SO_2, submicrometer particulate phase MSA, nss-SO_4^{2-}, NH_4^+ and EC, and IFN and total airborne suspended matter (TSP: <10 μm EAD) samples collected during IAOE-91; all values are at STP; 273.15 K, 1013.2 hPa*

	DMS_{sea} (nmol m^{-3})	DMS_{air} (nmol m^{-3})	SO_2 (nmol m^{-3})	MSA (nmol m^{-3})	nss-SO_4^{2-} (nmol m^{-3})	NH_4^+ (nmol m^{-3})	EC (nmol m^{-3})	IFN* (m^{-3}, −15°C)	TSP (μg m^{-3})
			Total	Julian Day 216–280					
no. samples	405	392	22	58	59	58	18	316	25
50th percentile (median)	0.29	1.1	0.27	0.06	0.55	0.49	0.11	8	1.3
25th percentile	0.13	0.48	0.22	0.04	0.21	0.26	0.06	4	0.69
75th percentile	2.0	2.3	0.45	1.1	1.3	1.2	0.22	17	2.2
min	0.04	0.05	0.04	0.002	0.03	0.03	0.009	0.08	0.10
max	12	17	1.7	1.4	6.9	3.9	0.51	151	3.8
			Ocean August	Julian Day 216–229					
no. samples	57	56	4	10	10	10	4	70	5
50th percentile (median)	8.3	8.8	0.79	0.57	2.5	1.6	0.15	12	3.1
25th percentile	4.0	7.2	0.57	0.51	2.0	1.2	0.09	9	1.8
75th percentile	9.6	10	1.3	0.67	3.2	3.6	0.23	18	3.3
min	0.84	1.1	0.45	0.07	1.3	1.0	0.06	4	1.1
max	12	17	1.7	1.4	6.9	3.9	0.26	38	3.5
			Ocean October	Julian Day 278–280					
no. samples	29	18	1	3	3	3	1	6	1
50th percentile (median)	0.25	0.31	0.28	0.05	2.3	1.8	0.11	0.3	2.0
25th percentile	0.17	0.26		0.05	2.2	1.6		0.1	
75th percentile	0.29	0.33		0.06	2.5	2.1		0.6	
min	0.33	0.20		0.05	2.2	1.5		0.08	
max	0.14	0.56		0.07	2.6	2.2		0.7	
			Pack ice	Julian Day 230–273					
no. samples	302	318	17	45	46	45	12	220	18
50th percentile (median)	0.20	0.91	0.24	0.06	0.31	0.41	0.11	8	0.87
25th percentile	0.11	0.45	0.17	0.03	0.16	0.23	0.06	4	0.53
75th percentile	0.89	1.5	0.31	0.11	0.67	0.76	0.24	16	1.7
min	0.04	0.05	0.04	0.002	0.03	0.03	0.009	0.08	0.10
max	4.3	7.5	1.1	0.33	1.4	2.2	0.51	151	3.8

* 0.2 day geometric average.

Table 1b. *Concentration statistics for ΔT (air-aq), T_{air}, RH, wind speed, radon, ozone, submicrometer aerosol by number and CCN samples collected during IAOE-91; all values are at STP (273.15 K, 1013.2 hPa); only averages for periods corresponding to atmospheric DMS sampling periods have been used*

	ΔT (air-aq) (°C)	T_{air} (°C)	RH (%)	Wind speed (ms^{-1})	Radon ($mBq\,m^{-3}$)	Ozone (ppb(v))	N_{tot} (cm^{-3})	$N_{ult.}$ (cm^{-3})	$N_{Ait.}$ (cm^{-3})	$N_{acc.}$ (cm^{-3})	CCN at 0.6% (cm^{-3})	CNN at 0.1% (cm^{-3})
Total Julian Day 216–280												
no. samples	406	406	406	406	354	352	352	352	352	352	286	291
50th percentile (median)	−1.0	−1.8	94	5.0	86	34.8	81	16	19	34	40	15
25th percentile	−4.5	−4.9	91	3.2	58	32.1	42	7.1	9.2	10	13	4.9
75th percentile	0.6	0.6	96	7.2	161	38.0	178	55	56	58	77	25
min	−16	−18	67	0	9.8	23.8	0.7	0.2	0.03	0.1	0.9	0.3
max	3.9	8.0	99	20	611	49.6	1240	926	627	229	313	94
Ocean August Julian Day 216–229												
no. samples	57	57	57	57	45	46	45	45	45	45	0	0
50th percentile (median)	−1.3	3.3	92	6.2	190	38.0	338	63	142	112		
25th percentile	−3.0	2.5	90	3.0	175	33.6	240	22	83	93		
75th percentile	0.5	4.2	94	8.0	430	44.9	572	122	238	146		
min	−5.2	0.9	67	0	131	32.3	38	5.8	9.5	18		
max	3.9	8.0	97	15	611	47.6	1240	697	627	229		
Ocean October Julian Day 278–280												
no. samples	30	30	30	30	30	28	30	30	30	30	29	29
50th percentile (median)	−6.6	−2.2	85	4.2	66	41.0	81	11	19	54	91	27
25th percentile	−11	−13	83	3.3	59	40.7	76	7.4	14	44	69	25
75th percentile	−4.6	0.4	87	4.9	106	41.9	98	15	22	64	112	40
min	−13	−15	75	1.3	52	40.1	48	3.7	12	27	29	9.3
max	−2.9	1.2	92	6.5	140	44.9	162	83	56	80	207	78
Pack ice Julian Day 230–273												
no. samples	303	303	303	303	263	261	263	263	263	263	243	228
50th percentile (median)	−0.5	−2.1	95	5.1	80	33.9	64	18	17	22	35	12
25th percentile	−3.2	−4.8	94	3.1	53	30.8	31	7.0	7.3	7.0	11	4.1
75th percentile	0.9	−0.7	97	7.3	132	36.5	137	52	45	43	65	22
min	−9.6	−11	86	0	9.8	23.8	0.7	0.2	0.03	0.1	0.9	0.3
max	2.8	1.2	99	20	362	48.2	1090	926	360	115	313	94

[DMPS]. A second DMA followed by an ultrafine CPC (TSI 3025) simultaneously stepped downwards in size from 20 nm to 8 nm [UDMPS]. The data were inverted from a mobility distribution to a number size distribution assuming a bipolar charging distribution. Together with the two DMA particle sizers an ultrafine counter with pulse height analysis [PHA-UCPC] provided the best definition of particles concentration for particles smaller than 5 nm. The result was a complete number size distribution from 3 nm to 500 nm in 34 intervals of diameter every 10 min. Another ultrafine CPC was also used to give an independent measure of total particle concentrations greater than 3 nm, sampling for 2 min out of every 10. While other instruments, such as a diffusion battery [DB], were used to study the ultrafine particles, the systems described above provided most of the published data.

9.2.2. Size distribution by mass. Several sampling systems were used to collect and size segregate the aerosol by mass. One group used 3 parallel filter packs on the inside sampling line, [FP; one serving as a blank], a further 3 outside to collect particles below this size and further 1 filter pack for collection of the total suspended particulate matter [TSP]. In addition a special filter stack was used for collecting submicrometer elemental carbon [EC]. To obtain further size resolution two three-stage Berner Cascade impactors [BCI] were used. These filter packs and impactors collected enough material for analysis of the common ions in 4 h in the early stages of the voyage but sampling times had to be increased to as long as 24 h later in the voyage. Analysis of the filter extracts was performed using chemically suppressed ion chromatography (IC).

The 2nd group used a pair of samplers designed for elemental analysis and more detailed size segregation but required much longer sampling times of 60–70 h so that only 10 sampling periods were obtained. A pair of stacked filter units [SFU] separating the aerosol into less or greater than 2 μm EAD was analysed for elemental mass by particle-induced X-ray emission (PIXE) and by instrumental neutron activation analysis. A 7-stage cascade impactor [PCI] with back-up filter allowed good size resolution in the samples and PIXE was used to analyse them.

9.2.3. Cloud condensation nuclei. A parallel plate thermal diffusion chamber cloud chamber with continuous airflow was used to measure cloud condensation nuclei [CCNC] at supersaturations between about 0.1 and 0.7% by controlling the lower plate temperature in equal steps with a half hour period. The emerging droplets were detected by a photocell using forward scattering from a helium-neon laser beam.

Time series of the concentrations at 5 fixed supersaturations were interpolated to 10-min intervals to synchronize with the particle number size distribution measurements. Tests with monodisperse ammonium sulfate aerosols produced by a differential mobility analyser showed some corrections to be necessary for thermal lag and slightly lower effective supersaturations than calculated. Corrections for this and counting efficiency were applied to the time series.

The main source of inaccuracy in the measurements was sudden changes in temperature of the container as people entered or left, or the heating was changed. While the overall correlation coefficient between 4700 pairs of 10-min CCN ($S = 0.63\%$) and CN ($N > 41$ nm) measurements was 0.94, comparison on some specific occasions suggested the CCN concentrations to be as much as 20% too high. (Undercounting also probably occurred but cannot be estimated since some of the condensation nuclei might have been non-hygroscopic).

9.3. Instrumentation for sampling and determination of gasphase chemistry

9.3.1. Atmospheric DMS. For determination of atmospheric DMS air was sampled from the 1 μm EAD inlet of the sampling system inside the laboratory container. The DMS samples were preconcentrated for 20 to 60 min in a Pyrex glass cold trap which had been placed in liquid argon. For scavenging of oxidants a cotton scrubber was combined with a Nafion drier. The samples were analysed using gas chromatography with flame photometric detection.

9.3.2. Sulfur dioxide. Measurements of SO_2 was performed as a part of the outside stacked sampling system (1 μm EAD inlet). Each of the three SO_2 samplers [FP SO_2; one serving as a blank] consisted of a 47 mm Millipore Teflon filter followed by four Whatman 41 filters coated with 0.1 M K_2CO_3 glycerol solution. Analysis of the filter extracts was made using an IC.

9.3.3. Radon and ozone. Radon was estimated from alpha-emitting radon daughters captured on a filter with a flow rate of 20 $\mathrm{dm}^{-3}\,\mathrm{min}^{-1}$. Alpha particles emitted from the filter struck a scintillator, a photomultiplier recorded the resulting flashes of light and a data logger totalled the pulses every minute. The sensitivity of the system had been calibrated using a standard radon source and a delay tank filled with artificially created particles.

Surface ozone was measured every minute with a recently calibrated Dasibi ozone monitor. 10-min averages of both radon and ozone were used to conform with the aerosol data.

9.4. Ancillary measurements

9.4.1. Ice forming nuclei. Ice forming nuclei were detected by capturing all the particles from about 500 dm^{-3} of air on a cellulose ester membrane filter and sealing the pores of the filter with oil (from the unsampled side). The filter was then cooled on the base plate of a thermal gradient diffusion chamber and the humidity raised to just above water saturation by warming the ice-covered top plate. After 25 min the ice crystals that had formed were large enough to be easily counted. The main defect of the method in most applications is interference from hygroscopic particles which limits the effective humidity on the filter. In the Arctic, particle concentrations were usually so low that this was unimportant.

9.4.2. Meteorology. Hourly or half-hourly meteorological surface observations of wind speed and direction, air temperature, pressure, RH, 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 RH, temperature, pressure, wind speed and direction (Vaisala RS80-15N and Digicora MW11 rawinsonde).

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