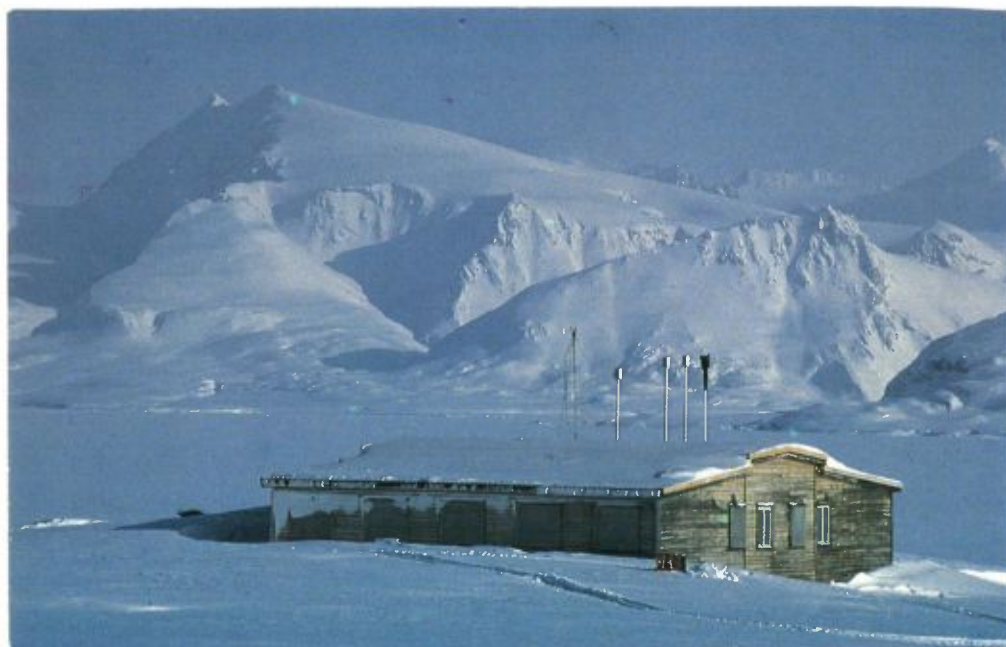


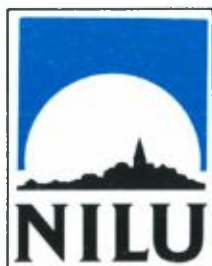
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# AIR POLLUTANTS IN THE ARCTIC



**FINAL REPORT OF A RESEARCH PROGRAMME  
CONDUCTED ON BEHALF OF BRITISH PETROLEUM, LTD.**

B. Ottar	M. Oehme
Y. Gotaas	J. Pacyna
Ø. Hov	A. Semb
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E. Joranger	V. Vitols



**Norwegian Institute For Air Research**  
POSTBOKS 64 — N-2001 LILLESTRØM — NORWAY

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## SUMMARY AND CONCLUSIONS

*The wintertime arctic haze, with concentration levels of man-made pollutants which are comparable to average concentrations over the industrialized continents, is due to pollutants emitted from sources within the arctic air mass. In late winter and spring, this cold and stable air mass, which is characterized by very low deposition rates and absence of photochemical activity, may engulf large parts of northern Eurasia.*

*Aircraft measurements show that the vertical extension of this haze is typically less than 1000 metres. The presence of further haze layers at elevations up to 5000 metres or more is due to sources outside the arctic air mass. Also natural aerosols are present at high altitudes, in the form of soil dust, which may have originated from the large deserts in Asia or Africa.*

*Atmospheric deposition in the Arctic is generally slight, and in areas with high precipitation rates, much of the precipitation is derived from relatively clean, marine air masses.*

*Sulphate and hydrocarbon pollutants in the arctic air mass may be partly responsible for the background concentration levels of sulphate and ozone in "clean air" at lower latitudes, particularly in spring and early summer.*

*A large-scale multilayer atmospheric dispersion model has been formulated, utilizing the concept of transport along isentropic surfaces. Calculations, using available meteorological data and a spatial emission survey for the northern hemisphere, show that the model is capable of simulating the advection of pollutants from different source areas into the Arctic at different elevations.*

*The model calculations show that sources in the USSR contribute most to the high sulphur dioxide and sulphate aerosol concentrations at low altitudes during winter and spring (~ 80% in March 1983), while other European sources contribute more to the concentrations at higher altitudes (~ 60% above 2-3 km). At the highest levels there is also a contribution from sources in North America.*

*The programme also included an experimental study of the atmospheric dispersion over sea surfaces under different stability conditions, and a statistical evaluation of turbulent energy spectra from wind observations at platforms in the North Sea, in order to provide basic input data for evaluations of hazard areas in connection with accidental gas releases.*

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The first indications that arctic haze was due to man-made pollutants, were obtained less than 10 years ago. The BP programme has substantiated and extended these findings. More than 8 years of continuous measurements of sulphur dioxide and sulphate at Bjørnøya and Ny-Ålesund, have been supplemented with measurements from other sites in the Norwegian Arctic and Northern Norway to provide a data base which is sufficient to give long-term and short-term characteristic concentration variations as well as spatial gradients.

These data show: a seasonal variation with maximum concentrations of sulphur dioxide and sulphate in the periode February-April and very low concentrations in July-November; a spatial gradient with the highest concentrations in the cold arctic air mass of continental origin; and evidence of episodic transport events with typical duration 5-10 days.

Detailed measurements show that the winter arctic pollution aerosol consists of ammonium sulphate and sulphuric acid droplets with a narrow size distribution between 0.1 and 0.5  $\mu\text{m}$ . This is typical of an aged atmospheric aerosol which has been produced by condensation and coagulation from gaseous precursors. In addition the aerosol contains soil particles and various trace metals, which can be used as chemical fingerprints to pinpoint emission sources.

The aircraft measurements have shown that the major part of the wintertime arctic haze is generally confined to a shallow boundary layer. Concentrations decline sharply with height, from a maximum 100-300 m above the surface, to about 1200 m. Elevated haze layers, sometimes with several distinct maxima in the vertical, were found to occur at different altitudes between 3 and 5 km above the surface.

Chemical analyses of air filters, and aerosol size distributions, indicate that both man-made and natural aerosols are present in these elevated haze layers, which were also observed during summer sampling flights.

In summer, total aerosol concentrations near the surface are an order of magnitude lower than in winter, and in addition, soil dust particles above 2  $\mu\text{m}$  size constitute a significant fraction of the total aerosol volume. The occurrence of Aitken particles less than 0.1  $\mu\text{m}$  in size, indicates gas to particle conversion, possibly photochemically induced oxidation of reduced sulphur compounds.

Polyurethane foam samplers have been used to determine concentrations of chlorinated hydrocarbons in ambient air. Polychlorinated biphenyls, hexachlorobenzene, and hexachlorocyclohexanes, were identified in nearly all samples, in concentrations ranging from a few picogrammes to nanogrammes per cubic metre. Although average concentration levels are comparable to those found in other remote locations, the highest concentrations were measured in situations with advection of polluted air masses to the Arctic.

Light hydrocarbons, with up to seven carbon atoms, are of particular importance in the photochemical production of ozone and other photo-oxidants in the troposphere. The different hydrocarbons cover a wide range of reactivities, which are reflected in their latitudinal atmospheric distribution. Concentrations of hydrocarbons in the air samples from Spitsbergen were generally one order of magnitude higher in March than in July. It has been shown, through photochemical model calculations, that the high concentrations of hydrocarbons in the Arctic can be a significant source of ozone production at mid-latitudes in spring and early summer.

It has been shown that the atmospheric transport of air pollutants into the Arctic is connected with large scale atmospheric circulation patterns that lead to an extended exchange of air in the meridional direction. These circulations must be of large extension and last for long periods of time. The air currents created in the vicinity of the moving lows and highs at middle latitudes are normally not sufficient. Meridional exchange is at a maximum when large amplitude waves are ap-

parent in the jet stream. When this occurs, the normal eastward propagation of cyclones is halted, and the situation is called blocking.

Two indices indicative of blocking-like flows were correlated with the logarithmic residual of the measured sulphate concentrations at Ny-Ålesund and Bjørnøya, which remained after removing the systematic seasonal variations. For winter and spring there was a close correspondence between arctic air quality and persistent south to north flow over western Eurasia. During summer there was very little correlation, suggesting that the observed concentration variations have little to do with long range transport.

When low level air approaches the polar front from the warm side and penetrates into the Arctic, it will rise to a higher level above the ground, while cooling as a result of adiabatic expansion. If the air is dry, the cooling rate is close to  $1^{\circ}\text{C}$  per 100 m rise. In wet air, heat will be released by condensation, and the cooling rate is reduced to  $0.6^{\circ}\text{C}$  per 100 m rise. Air flows as described, will follow isentropic surfaces with nearly constant potential temperature. Knowing the height and temperature of an observed polluted layer, an analysis along isentropic surfaces can be used to estimate the original ground level temperature and latitude of the emissions.

This concept of quasi-isentropic transport of air has been utilized in the formulation of a new multi-layer atmospheric transport model for the entire Northern hemisphere down to  $30^{\circ}\text{N}$ . Model simulations have been made for periods in March and July, 1983, for which observed arctic pollutant concentrations were available. Using meteorological data and gridded  $\text{SO}_2$  emission intensities, it was possible to simulate the formation of discrete, polluted haze layers in the Arctic, and to calculate the concentrations of sulphur dioxide and sulphate aerosol arising from different source areas, at different heights above the surface.

Low deposition rates is another condition for the atmospheric transport of aerosols over long distances. Precipitation in the Arctic is generally slight, with a summer maximum. However, high wintertime snowfall amounts may occur in mountaineous and glaciated areas due to orographic effects. At Spitsbergen, winter precipitation is also en-

hanced because of the strong cyclonic activity in the Norwegian Sea. In these cases, however, much of the precipitation is derived from clouds formed above the relatively shallow air layer close to the surface which is influenced by the arctic haze and pollution aerosols. In accordance with this, comparison of precipitation chemistry data with ambient air concentrations at the surface, indicate very low apparent wintertime scavenging ratios.

Dry deposition rates for snow-covered surfaces are also small. In consequence, the high winter-time concentrations of pollutants are associated with low deposition rates. This conclusion is supported by analyses of lichens and other vegetation samples in the Arctic.

In summer, precipitation occurs by fog or drizzle from low stratus clouds, resulting in rapid removal of aerosols and water-soluble gases, with high scavenging ratios. A special feature is the high concentrations of ammonium in precipitation samples from sites close to bird colonies, which demonstrate the close link between marine and terrestrial ecosystems in the Arctic. Thus, the deposition studies also demonstrate the importance of the vertical structure of the atmosphere, and that atmospheric residence times of aerosols and reactive gases can be highly variable, depending both on time and location of release.

The difference between winter and summer conditions in the Arctic provides excellent conditions for the investigation and monitoring of atmospheric trace substances which are of significance to the entire northern hemisphere. A key for the interpretation and evaluation of measurement results is provided by the quasi-isentropic atmospheric dispersion model which was developed in the BP programme. This model represents an extension of the regional long-range transport models, and a first step towards a global atmospheric dispersion model, which is not climatological, but based on real time observations of meteorological parameters.





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COVER PHOTOGRAPH: NILU measurement station at Ny Ålesund  
(H. Willoch).



## 1 INTRODUCTION

While up to the middle of this century, air pollution was generally regarded as a local problem in large cities and industrial areas, it is today evident that man-made emissions are gradually changing the chemistry of the whole atmosphere, even on continental and global scales. This development started with the industrial revolution and the large-scale utilization of fossil fuels as energy sources. It became a matter of public concern for the first time when Odèn (1968), and others, indicated that the emission of sulphur dioxide from fossil fuel combustion was the main cause for the observed acidification of rivers and lakes in Scandinavia. The long range transport of air pollutants in Europe, and the acid precipitation problem were verified and substantiated by the findings of the OECD study (OECD, 1977) and subsequent studies of the effects on soil, surface water chemistry, and freshwater biota (Overrein et al., 1980). Today, acid precipitation is recognized as a serious environmental problem both in Europe and in North America.

Another class of air pollutants, the photochemical oxidants, was first studied as a regional air pollution problem in California, but subsequent studies (e.g. Cox et al., 1975) showed that ozone and oxidizing radicals, formed by the photochemical reactions of nitrogen oxides and hydrocarbons, were playing an important part in tropospheric chemistry both on regional, continental and global scales. Increased ozone concentrations due to emissions of hydrocarbons and nitrogen oxides are now found over large areas in both North America and Europe (Grennfelt and Schjoldager, 1984).

For a long time the Arctic was supposed to be one of the last places still undisturbed by man's activities. An unexplained arctic haze observed in the winter north of Alaska in the early 1950's (Mitchell, 1956) did not receive much attention until Shaw and Rahn brought up the question 20 years later (Rahn et al., 1977). At an early stage, windblown dust from the great Asian deserts was believed to be a major component, but continued studies (Kerr, 1979; Rahn and McCaffrey, 1980) left no doubt that the arctic haze consists mainly of man-made air pollutants.

The OECD study had indicated that approximately 20% of the sulphur

dioxide and sulphates emitted, were advected out of the 18.5 mill. km<sup>2</sup> European study area, mainly in a north-easterly direction. To investigate this, measurements of sulphate and sulphur dioxide were initiated by NILU in 1977 at Bjørnøya and Ny Ålesund. Over the years, these measurements have shown that there is a pronounced transport of air pollutants into the Arctic during the winter, while the air is very clean in summer (Larssen and Hanssen, 1980).

However, the concentrations of sulphur dioxide and sulphate were much higher than expected, and could not be accounted for in terms of known emissions and atmospheric transport, without assuming very low transformation and deposition rates (Rahn et al., 1980). Thus, the atmospheric transport of air pollutants over distances of several thousand kilometres gives rise to a number of questions with respect to transport pathways and atmospheric lifetime of pollutants under arctic conditions.

While the main source areas of man-made emissions to the atmosphere are located between 30° and 60°N on the Eurasian and North American continents, emissions of sulphur dioxide and various trace elements also occur from metal smelters and other industrial activities on the Kola peninsula and around Norilsk in the northern USSR. All these areas lie in the westerlies, with a mean atmospheric transport from west to east. Transport from south to north is generally associated with ascending warm air at lower latitudes and a slow subsidence over the polar cap, governed by the radiative heat loss. This cannot, however, explain the strong seasonal variations of the concentrations of aerosols and sulphur dioxide in the Arctic. These are obviously connected with ground level transport and different atmospheric lifetimes of the pollutants under winter and summer conditions. Also the low level circulation and the meridional exchange of air is more intense in the winter-spring season.

The Svalbard islands are close both to the intense cyclonic activity in the North Atlantic and to the Polar Basin. Observations in this area are therefore particularly valuable in studies of the meteorological conditions governing the air pollution transport into the Arctic. In this connection aircraft measurements are essential in order to investigate the vertical distribution of the air pollutants

in the stable polar air mass.

In the boundary layer at middle latitudes sulphur dioxide and aerosols have atmospheric half-lives of 1-2 and 5-10 days, respectively. Dry deposition and precipitation are the main sinks. Generally, there is very little precipitation in the Arctic, as an average about 120 mm/yr (Orvig, 1970), and most of this occurs in summer. Due to orographic effects, the amounts of precipitation are much higher on the Svalbard islands and on arctic glaciers and icecaps generally. The deposition of air pollutants in the Arctic is therefore of considerable interest, not only as a sink mechanism, but also in connection with the historical information which can be extracted from chemical analyses of ice cores from Greenland and other arctic glaciers.

Other air pollutants, e.g. light hydrocarbons, have lifetimes which are determined by photochemical reactions in the atmosphere, ranging from days to several years. Seasonal variations in the concentrations of these substances may be used as a further check on the atmospheric circulation process, and on their emissions and atmospheric decay. This also applies to the heavy chlorinated hydrocarbons, such as polychlorinated biphenyls, hexachlorobenzene, and hexachlorocyclohexanes, which are also transmitted through the atmosphere on a global scale.

Towards the end of the 1970's, the oil exploration on the continental shelves started to move further north, and it was realized that the Barents Sea probably contains large reserves of oil, which may be exploited towards the end of the century. In order to establish a basis for evaluation of the concurrent pollution problems, a comprehensive study of the air pollution situation in the Norwegian sector of the Arctic was suggested by NILU. During a 5-year period the programme was to examine the chemical composition, occurrence and possible sources of the observed air pollution. The programme which started in 1981, was financed by British Petroleum Ltd., under an agreement between the Norwegian Government and the oil companies working on the continental shelf.

Under this programme a network of measuring stations was established, including four stations in the high Arctic, in addition to the sta-



Figure 1.1: Location of measuring stations in the BP programme.

tions on the Norwegian mainland, see Figure 1.1. An extended measurement programme at the station in Ny Ålesund was supported by additional measurements at the other stations.

The field measurements started in the summer of 1982 and lasted until the end of 1984. A routine programme was carried out at all the ground stations, which included daily measurements of sulphur dioxide, particulate sulphate and high-volume aerosol samples for trace element determination. Collection of precipitation samples on a weekly basis was also included.

The BP programme for the first time provided means for extensive aircraft measurements in the Norwegian Arctic. The measurements were carried out during selected periods of one month duration each summer and winter, and focused on studies of the aerosol with recording instruments. Aerosol samples were also collected for chemical analysis. During these periods, extended measurement programmes were carried out at the ground stations, including size-fractionated sampling and chemical analysis of the aerosol, and measurements of polychlorinated and light hydrocarbons.

An extensive collection of snow samples was carried out on Spitsbergen in the spring of 1983. Also samples of mosses, lichens, and soils have been collected and analysed.

An important objective of the BP programme has been to identify the sources of the air pollutants in the Arctic. Based on analyses of the meteorological transport conditions, the measured concentrations have been interpreted in terms of emissions, wind trajectories, and deposition processes. In this connection the significance of the very long atmospheric quasi-stationary waves for the poleward transport of air pollutants was examined, and an isentropic dispersion model has been developed to describe the large scale atmospheric transport into the Arctic.

The BP programme also included a special study of the atmospheric dispersion over sea surfaces, including tracer ( $\text{SF}_6$ ) experiments in the Oslofjord and a statistical analysis of wind data collected at oil platforms in the North Sea. This research is related to other studies at NILU of the dispersion of condensed gases in "blow-outs" and other accidental releases (Eidsvik 1985; Gotaas 1986).

During the BP programme there has been extensive cooperation with research institutions in other countries on special subjects. An event of particular importance was the coordinated arctic aircraft measurement campaign, which was organized in cooperation with the U.S. National Oceanographic and Atmospheric Administration (NOAA), Max-Planck-Institut für Chemie, Mainz, and Deutsche Versuchsanstalt für Luft- und Raumfahrt in Oberpfaffenhofen, FRG, in March/April 1983.

The BP research programme has provided much new information in the form of measurement data, and a better insight into the atmospheric transport processes both in the Arctic and in the northern hemisphere. The main results are summed up in this report.



## 2 AMBIENT AIR MEASUREMENTS

The air pollution measurements in the Norwegian Arctic started in 1977 at Bjørnøya and Ny Ålesund with sulphur dioxide and particulate sulphate as the main subjects. (Larssen and Hanssen, 1980; Rahn et al., 1980; Heintzenberg and Larssen, 1983). Under the BP programme the number of ground stations was expanded, and aircraft measurements, primarily of the aerosol, were taken up. In addition, sampling and analysis of polychlorinated and other hydrocarbons were undertaken at the ground stations and to a limited extent from the aircraft. In the present chapter a survey of the results from this work is given.

### 2.1 THE SULPHUR POLLUTANTS

During the BP programme regular measurements of sulphur dioxide and particulate sulphate were made at 4 arctic and 4 sub-arctic stations (Figure 1.1) during 1982-1984 (Joranger et al., 1986). Daily samples were collected at the EMEP stations Bjørnøya, Jergul, and Kårvatn, while the sampling was limited to a 2+2+3 day weekly schedule at Ny Ålesund, Jan Mayen, Hopen, Skrova, and Vardø.

In the high Arctic the concentrations of sulphur dioxide and particulate sulphate show pronounced seasonal variations, with a maximum from February through April, as shown for Ny Ålesund and Bjørnøya in Figure 2.1. Daily measurements of lead and cadmium at the two stations in 1977-1979 showed similar seasonal variations (Larssen and Hanssen, 1980). During the summer months, the concentrations were very low and for sulphur dioxide mostly below the detection limit of  $0.1 \mu\text{g}/\text{m}^3$ . The annual mean concentrations show no particular trend.

During the winter season the sulphate concentrations at Bjørnøya were persistently higher than at Ny Ålesund, while the concentrations of sulphur dioxide were higher at Ny Ålesund. This systematic difference is explained by the different physical and chemical properties of the two components. Absorption at the sea surface is an important sink for sulphur dioxide, while sulphate aerosol is mainly removed by

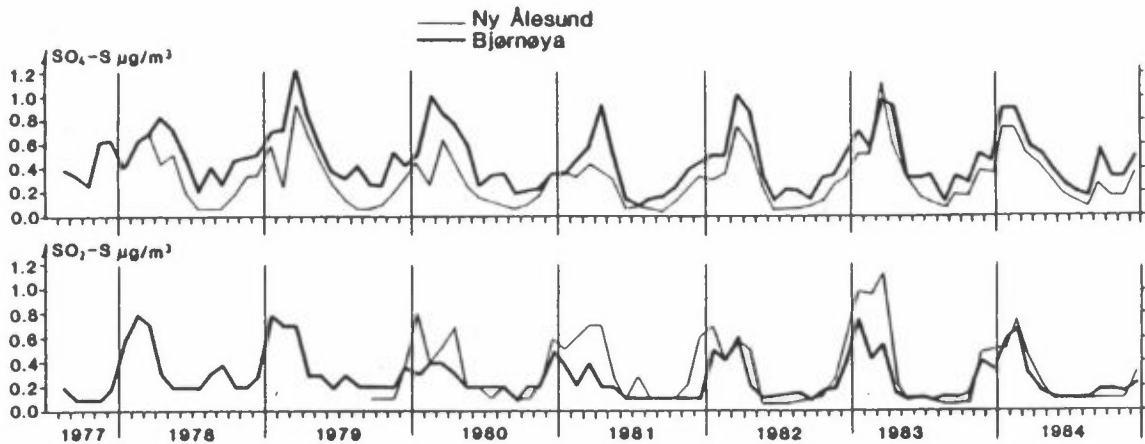


Figure 2.1: The variation of the monthly mean concentrations of  $\text{SO}_2$  and  $\text{SO}_4^{-2}$  at the arctic stations Bjørnøya and Ny Ålesund during<sup>4</sup> 1977 to 1984.

precipitation. Transport of polluted air masses to Ny Ålesund in winter occurs to a large extent from east to south-east over ice-covered areas, while Bjørnøya is mostly surrounded by open waters, also in winter. Bjørnøya is, however, closer to the emission in Europa, hence the sulphate concentrations are higher at this location.

These differences in behaviour and sinks are also evident in Figure 2.2, which shows the estimated spatial variations of sulphur dioxide and particulate sulphate during the periods of maximum (winter) and minimum (summer) concentrations (Joranger and Ottar, 1984; Iversen and Joranger, 1985). In winter the sulphur dioxide values have a minimum zone in the area between Svalbard and Norway, which is not found for sulphate. As mentioned above, the probable explanation is that air arriving at Bjørnøya and Hopen has travelled more often over the open sea than polluted air arriving at Ny Ålesund. The mean spatial distribution during summer-fall is different, with minimum concentrations in the Arctic (Figure 2.2). Generally, the mean concentrations of both sulphur dioxide and sulphate decrease with the distance from the Eurasian emission sources.

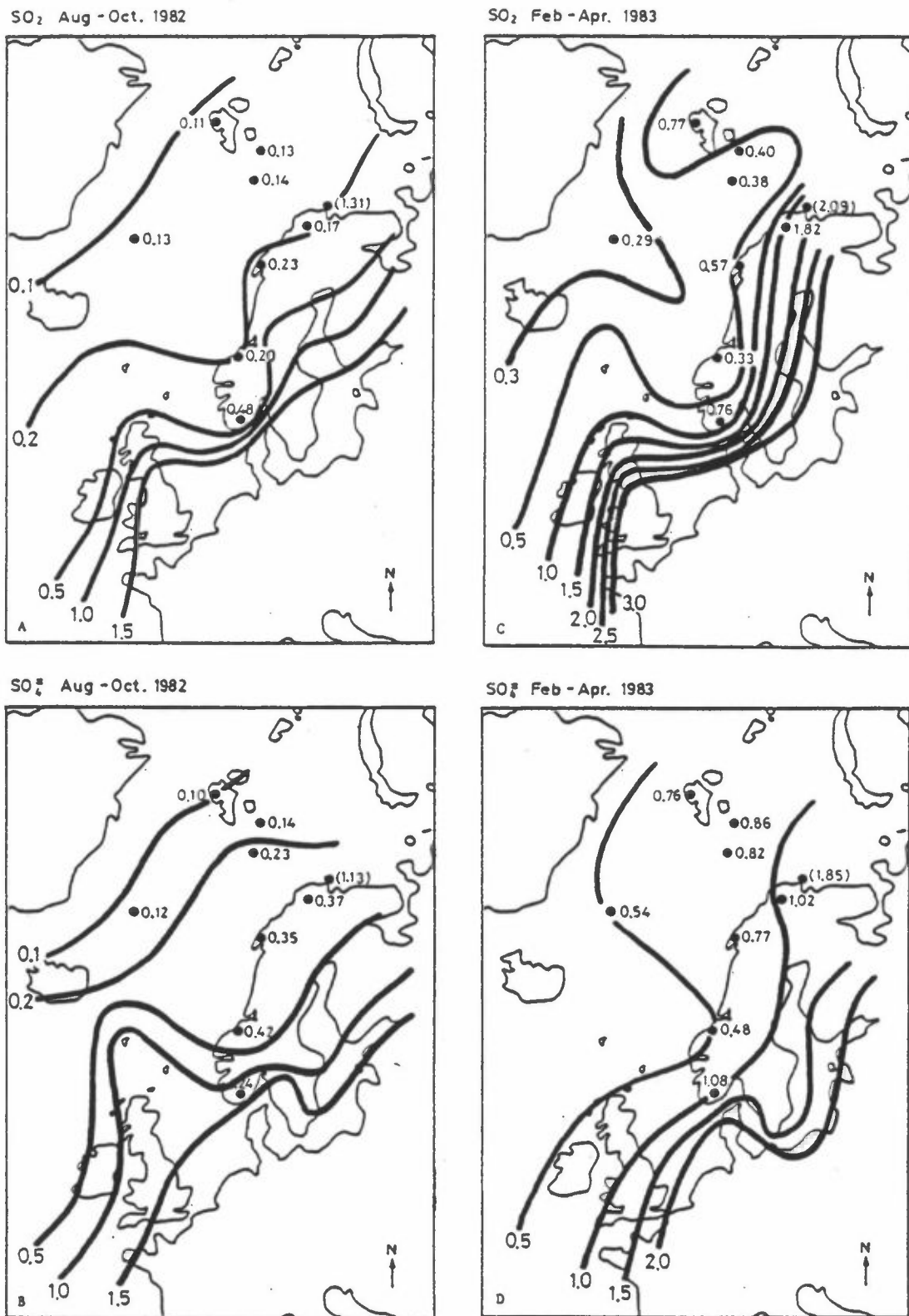


Figure 2.2: Isolines for  $\text{SO}_2$  and  $\text{SO}_4^{-2}$  subjectively analysed on the basis of quarterly averaged observations.

The measurements of the sulphate aerosol at the arctic stations show that there is an alternation between polluted and relatively clean air masses with episode lengths of typically 5-10 days. In Figure 2.3 this episodic character is illustrated by the sulphate measurements at Ny Ålesund and Jan Mayen in 1981. Three definite features are apparent: the annual cycle, the short term variability, and a variance approximately proportional to the mean value. It may also be noted that the peak concentrations in summer are higher at Jan Mayen than at Bjørnøya, which is due to long range transport of polluted air masses from Western Europe, cfr. Figure 2.2.

To study the variability of these data, the systematic seasonal variations in mean and variance were removed from the long time-series at Ny Ålesund and Bjørnøya. As the measured concentrations,  $c$ , are log-normally distributed, the logarithm of the time series was Fourier-analyzed, and the systematic part  $c_s$  of the measured concentrations, which contains periods longer than approximately 100 days, is shown in Figure 2.4. Also shown is the high-pass residual of the logarithmically transformed series,  $\log(c/c_s)$  (Iversen, 1985).

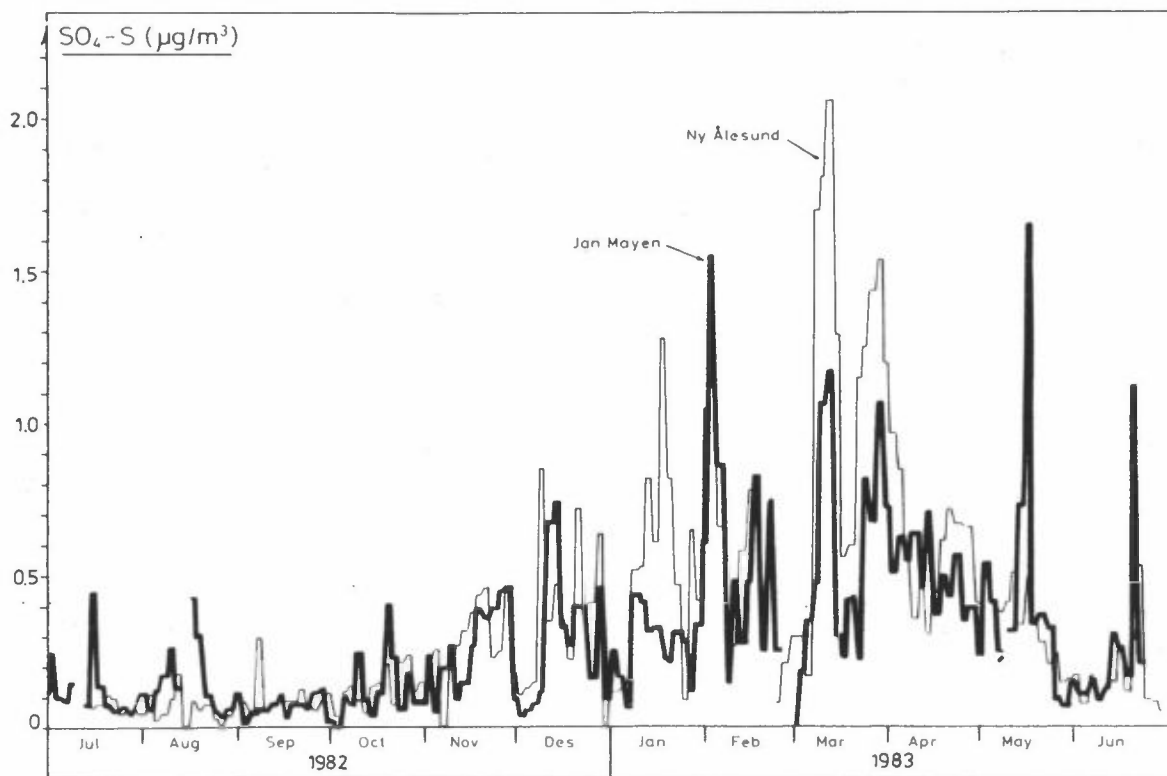


Figure 2.3: Measured concentrations of  $\text{SO}_4^{-2}$  at Ny Ålesund and Jan Mayen 1982/83.

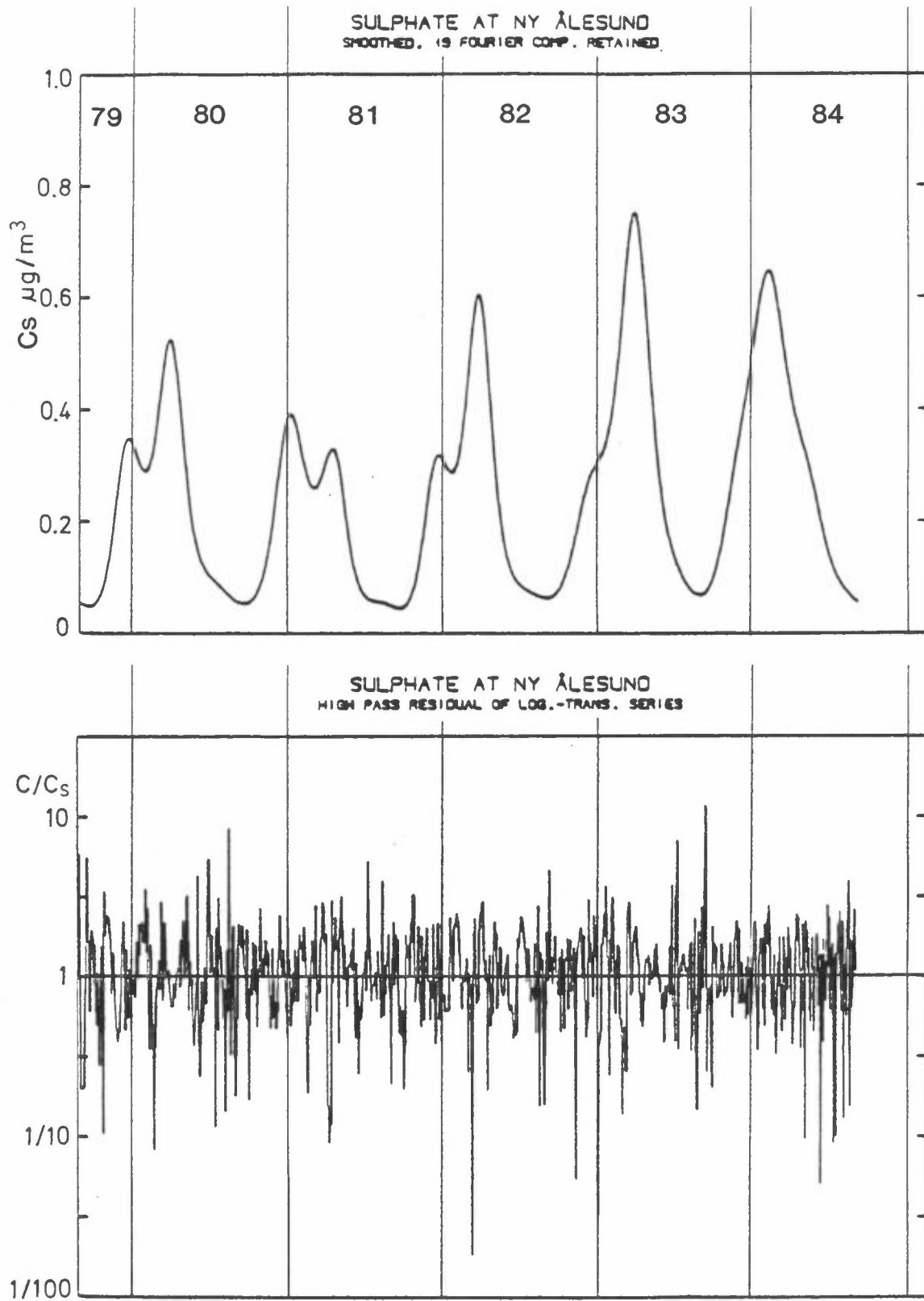


Figure 2.4: The systematic part and the residual of the logarithmically transformed time-series of the air concentrations of  $\text{SO}_4^{2-}$  at Ny Ålesund during 1979 to 1984.

This non-dimensional residual, which shows no systematic seasonal variation of the mean or the variance, may be tentatively interpreted as a seasonally adjusted index of the relative degree of pollution. It is utilized further in a statistical study of the correlation of the sulphate concentrations with large-scale meteorological phenomena in Section 3.2. However, of the low concentration levels in summer and fall high values of the residual in the period June-October reflect only moderate absolute values. Actually, the highest positive value of the residual is due to a 3-day period in September 1983, with a mean concentration of sulphate aerosol of only  $0.9 \mu\text{g S}/\text{m}^3$ .

The measured ambient concentration levels of sulphur dioxide and sulphate aerosol in the Norwegian Arctic may be compared with measurements which have been carried out in the arctic part of Canada and Alaska (Barrie, 1986). The concentrations of sulphur dioxide and sulphate aerosol at these sites show similar seasonal variations as in the Norwegian Arctic, but the concentration levels in the periods January-April 1980 and January-April 1981 were 10-30% lower in Arctic Canada. The spatial concentration pattern is strongly suggestive of transport across the Polar Basin towards Alaska and Canada.

## 2.2 AIRCRAFT MEASUREMENTS

Most of the information on arctic air pollution up to 1983 was obtained from measurements at ground sampling sites on Svalbard, Bjørnøya, Greenland, Arctic Canada, and Alaska. The aircraft measurements in the BP programme were intended to survey the vertical distribution of the pollutants in the arctic haze, and to assess the representativity of the ground sampling stations. Four measurement campaigns were carried out, in March and August, 1983; and in February-March and July, 1984; with a total number of 35 sampling flights.

The aircraft, a twin-engine Piper Navajo, was equipped with: 1) a high-sensitivity integrating nephelometer (IN), for measuring the particle light scattering coefficient,  $\sigma_{sp}$ ; 2) a continuous flow condensation nuclei counter (CNC); 3) a laser optical particle counter (LAS), for measuring aerosol particle numbers in 60 size intervals; 4) low- and high-volume filter samplers for particles; and 5) instruments

for altitude, air temperature, and dew point observations. The various parameters measured during the flights, as well as the aircraft position, were recorded and stored by a microprocessor-controlled data logger onboard the aircraft. A detailed description of the aircraft instrumentation is given by Berg and Willoch (1986). Diagrams and tables giving all data from the individual sampling flights have been collected in a separate report (Pacyna et al., 1986b), and the results have been discussed by Pacyna et al. (1985b).

The measurement campaign in March 1983 coincided with an "episode" with high concentrations of sulphate aerosols at Ny Ålesund during the period 20-25 March. Isobaric 850 mb trajectories indicated transport of air from the northern USSR.

The highest aerosol concentrations occurred at, or near ground level, as shown by the integrating nephelometer data presented in Figure 2.5. Concentrations declined sharply with height, from a maximum 100-300 m above the ground, to about 1200 m. Another haze layer with distinctly increased haze and aerosol concentrations appeared about 3.5 km above the ground. Similar elevated layers were also observed from the NOAA aircraft in March-April 1983 (Schnell and Raatz, 1984), and from the two German aircrafts participating in the March 1983 campaign at Spitsbergen (Wendling et al., 1985; Werhahn et al., 1986).

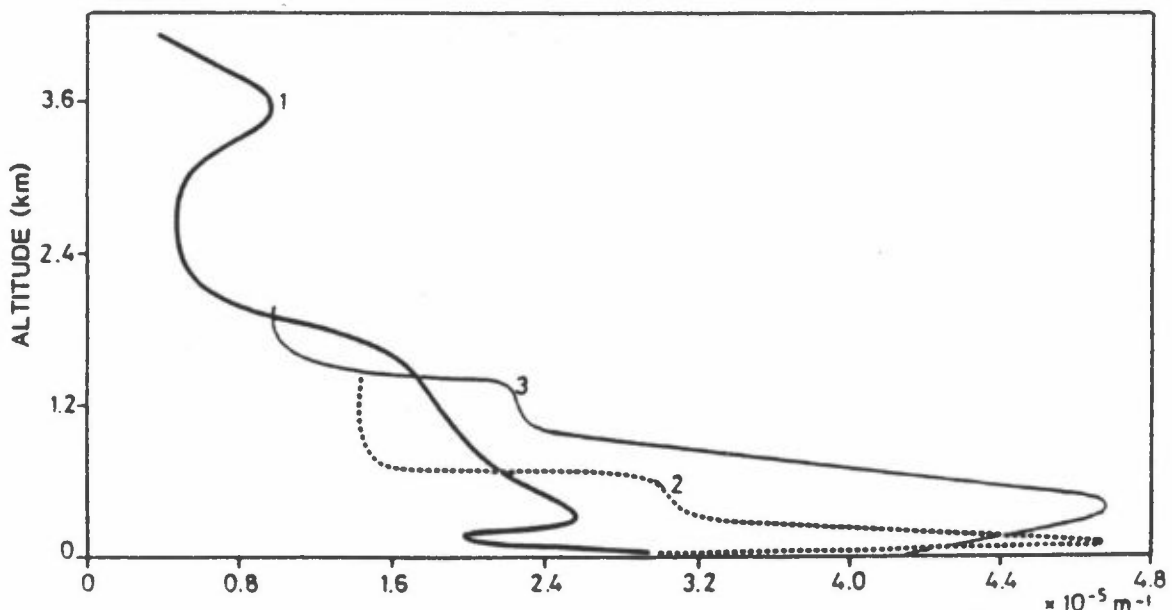


Figure 2.5: Nephelometer values  $\sigma_{sp}$  vs. flight altitude during Flight A-3 on 1983-03-21-I (1), A-4 on 1983-03-21-(2), and A-5 on 1983-03-22 (3).

The concentrations of aerosols at ground level were considerably lower during the second winter aircraft measurement campaign in March 1984. This time the highest concentrations occurred at different altitudes below 2400 m, sometimes with several distinct maxima. Elevated haze layers with maximum concentrations between 3000 and 5000 m were also found during the March 1984 measurement flights (Figure 2.6). The 850 mb trajectories and isentropic trajectories indicated transport of air from Eastern Europe over the Kola Peninsula.

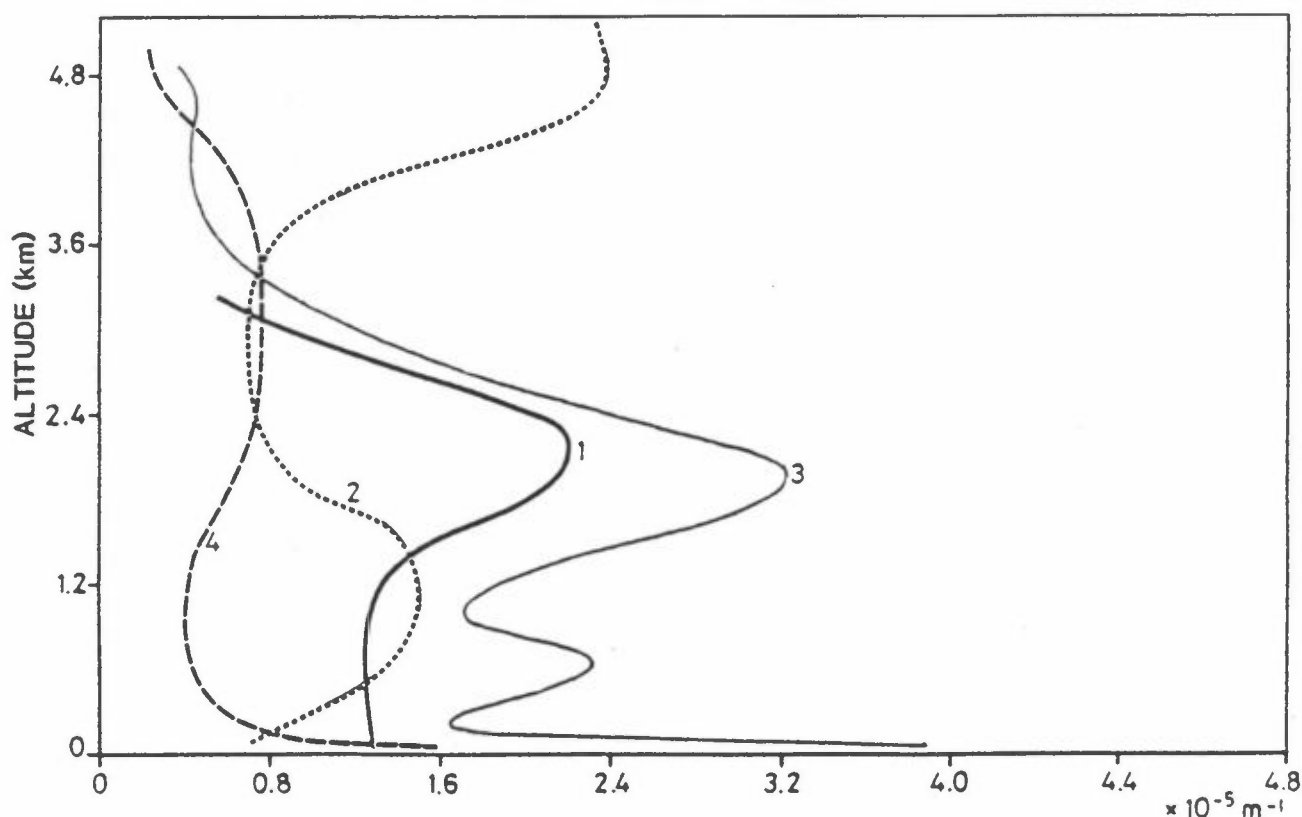


Figure 2.6: The  $\sigma_{sp}$  values vs. flight altitude during Flights C-3 on 1984-03-01 (1), C-4 on 1984-03-02 (2), C-5 on 1984-03-02 (3), and C-7 on 1984-03-07 (4).

During the summer measurement campaigns, a typical episode with high concentrations of aerosols occurred at Ny Ålesund on August 18, 1985, but in general the summer measurement campaigns indicated very clean air up to about 2 km, in confirmation of the findings at the ground sampling stations. However, elevated haze layers were also found during the summer measurement flights, at elevations above 2400 m, as shown in Figure 2.7.



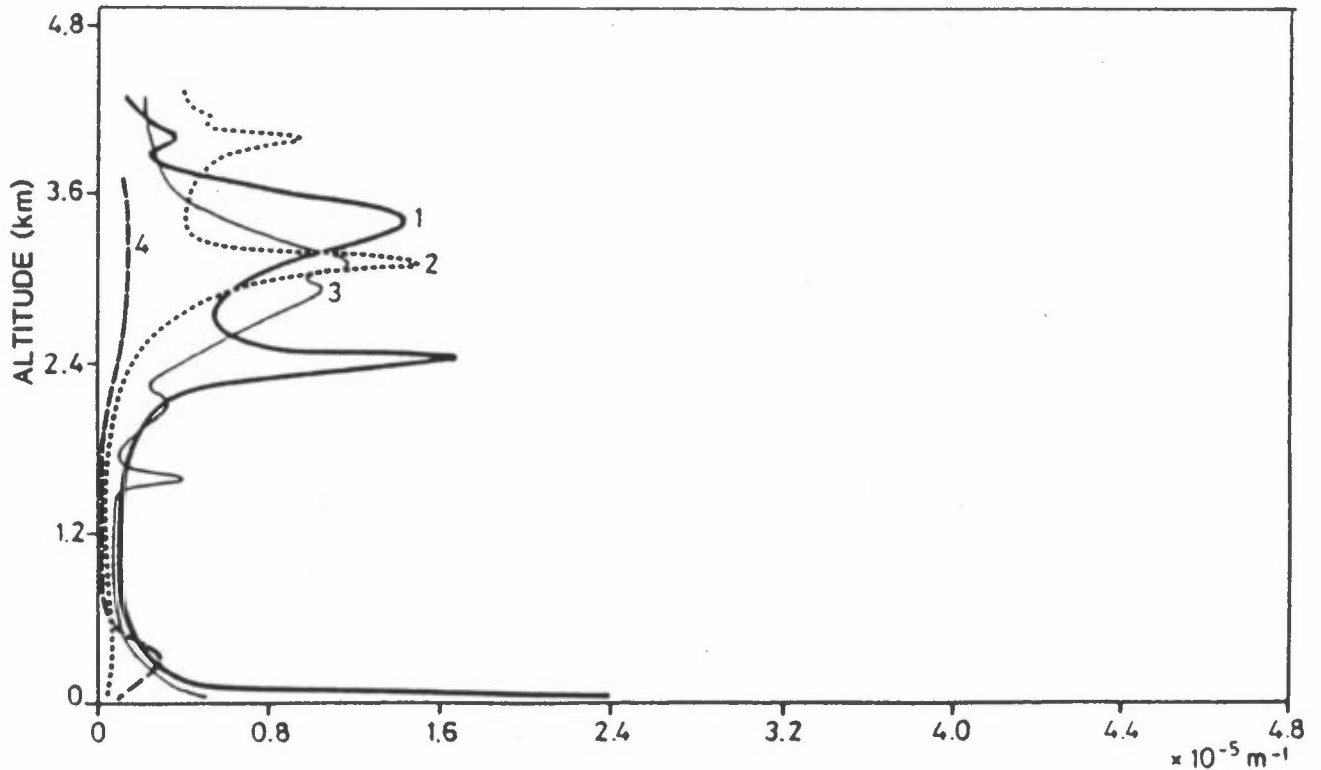


Figure 2.7: The  $\sigma_{SP}$  values vs. flight altitude during Flights B-3 on 1983-08-18 (1), D-4 on 1984-07-08 (2), D-8 on 1984-07-13 (3), and B-5 on 1983-08-21 (4).

The size distributions of the low level aerosols from the winter measurement campaigns are characterized by maximum volume concentrations in the size range 0.2 to 0.3  $\mu\text{m}$  (Figure 2.8). There were very few particles larger than 0.5  $\mu\text{m}$ , and the low CN particle counts indicated that particles smaller than 0.1  $\mu\text{m}$  were also scarce.

The size distributions in the elevated haze layers, particularly those found during the summer campaigns, are distinctly different. Here, particles larger than 1-2  $\mu\text{m}$  constitute a large fraction of the total aerosol volume (Figure 2.9). It is reasonable to assume that these particles originate from other sources and processes than the sub-micron size particles. This is supported by chemical analyses of collected samples which show a composition mainly of major earth crust elements. This indicates that the origin may be wind erosion at lower latitudes.

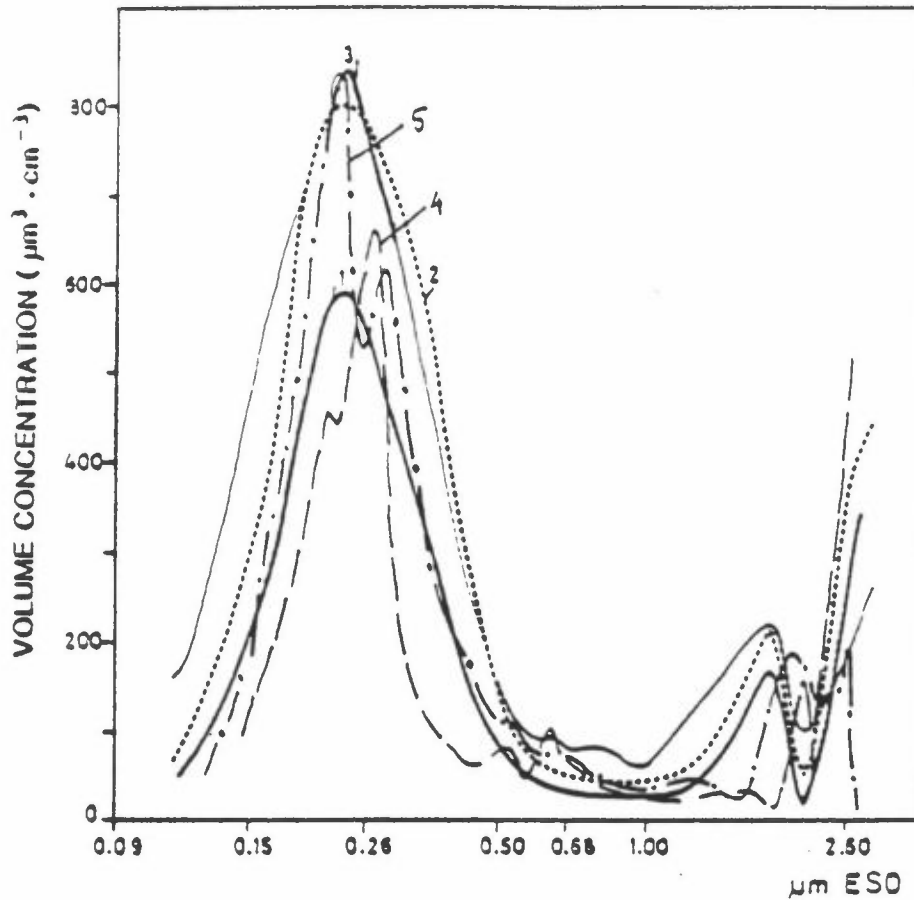


Figure 2.8: Volume-size distribution of particles in the polluted air below 2000 m during Flights A-3 on 1983-03-21-I (1), A-4 on 1983-03-21-II (2), A-5 on 1983-03-22 (3), C-3 on 1984-03-01 (4), and C-5 on 1984-03-03 (5). (ESD, equivalent scattering diameter.)

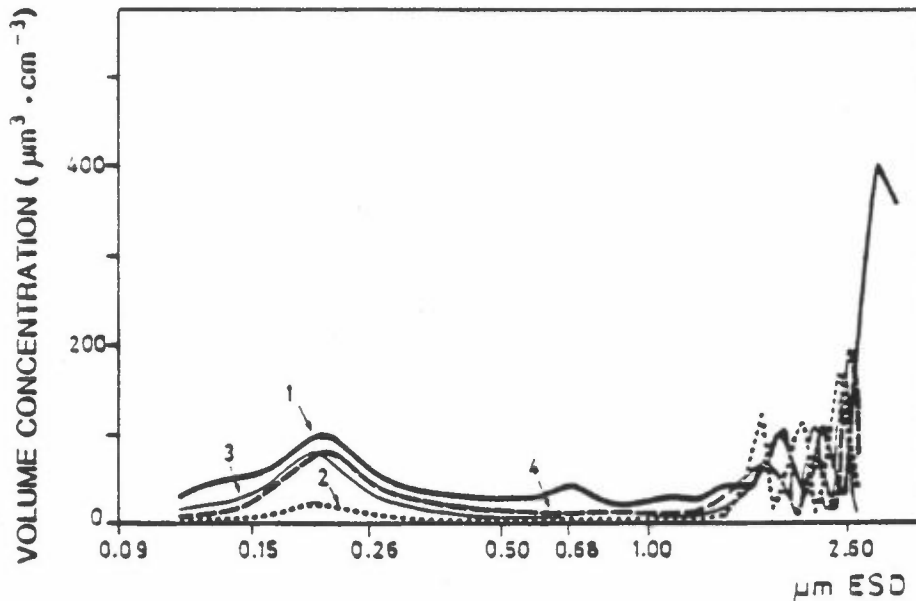


Figure 2.9: Volume-size distribution of particles in the air layer between 2000-3000 m during Flights B-3 on 1983-08-18 (1), D-4 on 1984-07-08 (2), D-8 on 1984-07-13 (3), and B-5 on 1983-08-21 (4). (ESD, equivalent scattering diameter.)

The lower layer "pollution aerosols" are enriched in several trace elements (V, As, Cu, Sb, Au, Cl), which are typical of aerosols from fossil fuel combustion and non-ferrous metal smelters, cf. Figure 2.10. Variations in the ratios of the various trace elements support the hypothesis of different origins of arctic aerosols measured at altitudes above and below 2500 m during the winter measurement campaigns in 1983 and 1984.

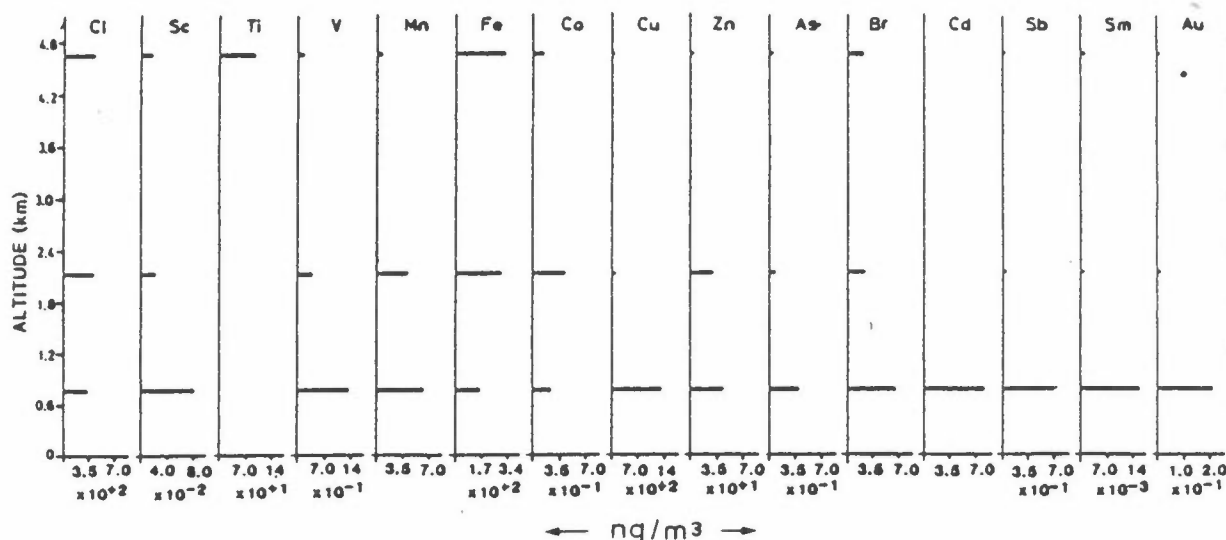


Figure 2.10: Chemical composition of aerosols measured at different altitudes during Flight C-5 on 1984-03-03.

Local air pollution sources in Spitsbergen are not generally expected to generate enough particulate air pollutants to disturb the sampling of the arctic air. The flight on August 25, 1983, was specially designed to sample emissions from Longyearbyen and other mining settlements around Isfjorden. Several haze layers were observed at various heights under 1 km, apparently originating from the mining communities of Pyramiden, Longyearbyen, and Barentsburg. The particle size distribution in Figure 2.11 shows a large contribution of the finest particles (<0.1  $\mu\text{m}$ ) from fresh local discharges. For particles larger than 0.1  $\mu\text{m}$  the size distribution is similar to the size distributions for particles collected at higher levels during summer (Figure 2.9).

It is difficult to assess the contribution of sea-salt particles to the aerosol size distribution, as most of these particles are larger than those measured by our equipment, viz. >3.0  $\mu\text{m}$ . The filter samples, however, show that seasalt is present in all samples, also those collected at high altitudes.

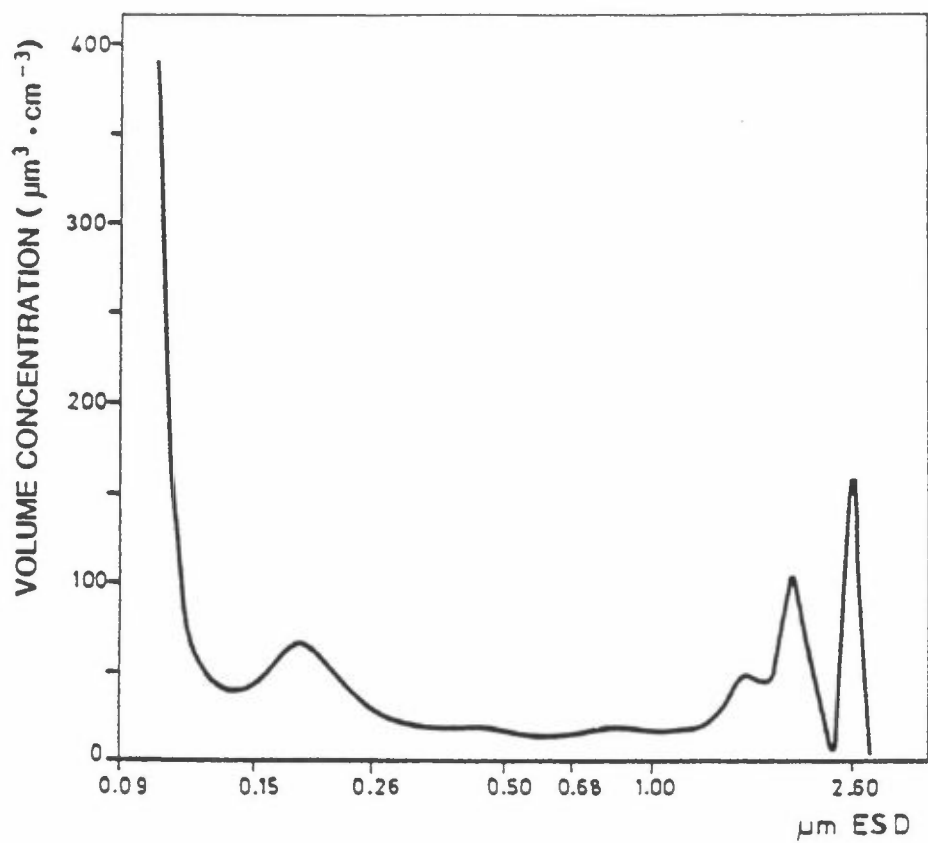


Figure 2.11: Volume-size distribution of particles at 350 m over Isfjorden during Flight B-8 on 1983-08-25. (ESD, equivalent scattering diameter.)

### 2.3 AEROSOL CHARACTERIZATION

During the intensive measurement campaigns in August-September 1982, March-April and August-September 1983, February-March and June-July 1984, measurements were carried out at Ny Ålesund with various instrumental methods to investigate the size-distribution and chemical composition of the arctic aerosol. These measurements were partly performed in cooperation with the Meteorological Institute of the University of Stockholm (MISU) and the Nuclear Physics Department of the University of Lund.

The measurements included counting of cloud condensation nuclei (CCN), optical scattering by aerosols using a sensitive integrating nephelometer and a size-differentiating optical particle counter (Royco), and size fractionated sampling of aerosols for subsequent chemical analyses by particle-induced x-ray emission (PIXE). The results are described in a separate report (Vitols and Pacyna, 1985).

One of the most striking results is the high correlation between the number of CCN and the optical scattering during the winter campaigns (Figure 2.12). This remarkable correlation is, however, consistent with the sizedistributions obtained from the aircraft measurements, which show that the bulk of the aerosols occurs within a narrow size range between ca 0.1 and 0.5  $\mu\text{m}$  ESD, and the low measured CCN counts.

This conclusion is further supported by the other measurements. The Royco optical particle counter has a lower size limit of detection at about 0.3  $\mu\text{m}$  equivalent scattering diameter (ESD), and the number concentrations of particles in the lowest size range between 0.3 and 0.5  $\mu\text{m}$  were only 5-20  $\text{cm}^{-3}$  in March 1983, which is insufficient to account for the concentrations of sulphate collected on the filters. Since the number of particles larger than 0.5  $\mu\text{m}$  represents an even smaller contribution to the total aerosol volume, it follows that the volume-weighted mean aerosol diameter must be less than 0.3  $\mu\text{m}$ .

The Battelle-type cascade impactor, which was used to collect size-fractionated aerosol samples for chemical analysis by the PIXE method, separates the aerosol particles according to their equivalent



Sulphate is the main constituent of the arctic winter aerosol, and it occurs predominantly in the  $<0.25 \mu\text{m}$  EAD size fraction (Figure 2.13). This suggests that most of the sulphates originate from gas-phase oxidation of sulphur dioxide in the atmosphere. This leads to a

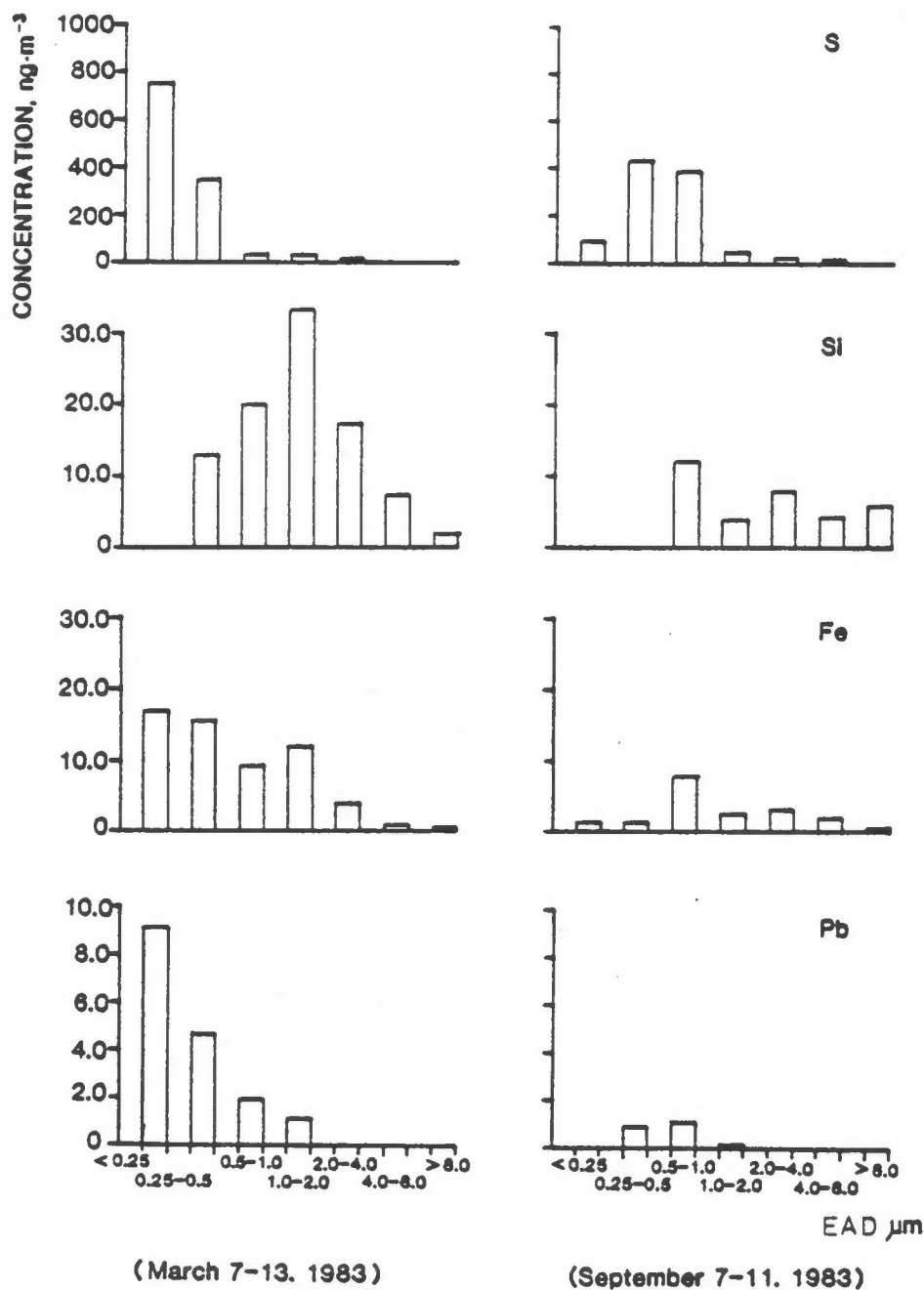


Figure 2.13: Concentrations of particle-associated elements in air samples at Ny Ålesund, divided in aerosol size fractions with a Battelle cascade impactor: a) March 7-13, 1983; b) September 7-11, 1983.

production of very small "embryo" particles, which agglomerate rapidly by coagulation to particles in the size range 0.1-0.2  $\mu\text{m}$ . The other elements are associated with aerosols which originate from various high-temperature and other processes, including soil erosion, and the size distributions are a function both of the production processes and the size-dependent depletion of particles larger than 0.5-1  $\mu\text{m}$  during the transport from the source area to Ny Ålesund.

The summer arctic aerosol has a much more variable size distribution. The CCN counts, measured within the boundary layer in summer are not always correlated with  $\sigma_{\text{sp}}$ , as seen in Figure 2.14. Actually, the CCN counts are almost as high in summer as during

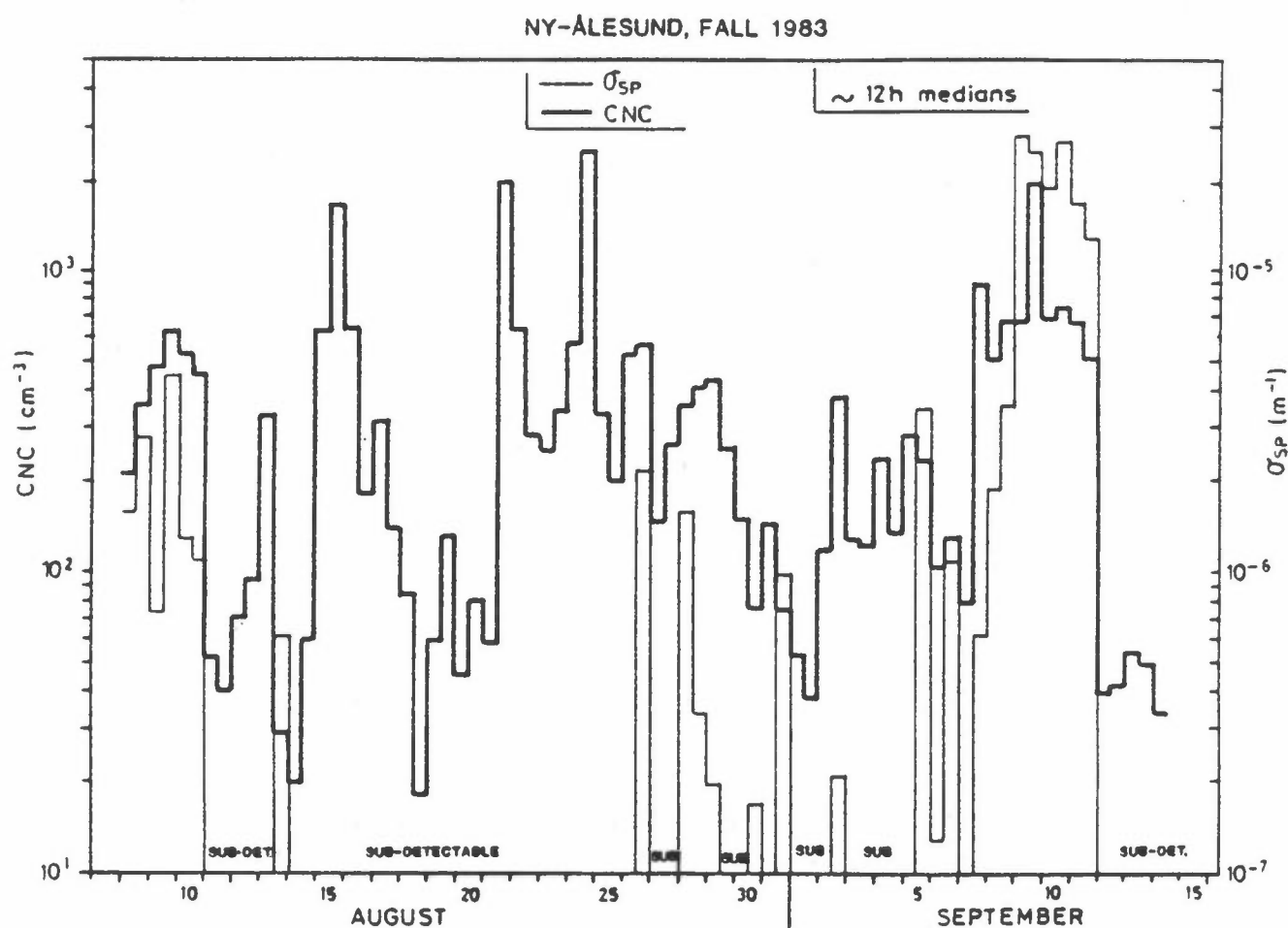


Figure 2.14: 12-hour CN concentration (CNC) and aerosol light scattering coefficients ( $\sigma_{\text{sp}}$ ) at Ny Ålesund during the summer/fall 1983 campaign.

"Sub-detectable" means below the detection limit of the nephelometer.



winter "pollution" episodes, indicating the presence of small "embryo" aerosol particles larger than  $0.02 \mu\text{m}$ , which is the limit of detection at the super-saturation level reached by the CCN counter, but significantly smaller than  $0.1 \mu\text{m}$ . Because of the rapid agglomeration of these small particles, their presence indicate gas-to-particle conversion at the site, possibly oxidation of reduced sulphur compounds by photochemically-induced reactions. Aerosol particles larger than  $1 \mu\text{m}$  EAD, and with chemical composition typical of silicates, are also a relatively more important part of the total aerosol mass in the arctic summer aerosol. Figure 2.13(b) shows the concentration, in different size ranges of S, Si, Fe, and Pb in air samples collected during an episode of long range transport air pollution in September 1983. The concentrations during this period were considerably higher than usual during the summer and fall seasons. However, the size distributions are distinctly different from the winter pollution episodes, particularly for sulphur. In this case, the trajectories indicated transport of air from the south, over the Kola peninsula.

Trace element concentrations are also available from analyses of high-volume filter samples. In many cases, variations in elemental ratios can be related to source characteristics, in particular emissions of Cu, Ni, and other elements which occur in connection with non-ferrous metal smelters (Ottar and Pacyna, 1984; Pacyna et al., 1984; Pacyna and Ottar, 1985). Tables 2.1 and 2.2 give mean concentrations of some characteristic trace elements at Ny Ålesund during the winter and summer measurement campaigns. In addition, high-volume filter samples were collected at Hopen, Jan Mayen, and Bjørnøya, and at 5 stations on the Norwegian mainland. These were analysed by neutron activation analysis and atomic absorption spectroscopy.

Table 2.1: Concentrations ( $\text{ng}/\text{m}^3$ ), of trace elements at Ny Ålesund during March 1979 (Heintzenberg et al., 1981), and during the BP measuring campaigns in March 1983 and February-March 1984.

	March 1979	March 1983	February-March 1984
Mn	1.5	1.73	0.57
V	-	2.44	0.50
Ni	0.7	0.71	0.22
Zn	3.2	9.08	2.88
Pb	<4.9	7.01	1.81

The winter concentration levels are comparable, within a factor of 2-3, to mean concentrations observed in southern Scandinavia (Hanssen et al., 1980; Heintzenberg et al., 1981). The summer concentrations are about an order of magnitude lower. The source-receptor relationships which can be inferred from these trace element concentrations and their elemental ratios, are discussed in Chapter 4.

Table 2.2: Concentrations ( $\mu\text{g}/\text{m}^3$ ) of trace elements at Ny Ålesund during the summer measurement campaigns.

	August-September 1982	August-September 1983	June-July 1984
Mn	0.08	0.27	0.05
V	-	-	-
Ni	0.02	0.04	-
Zn	0.21	1.67	0.62
Pb	0.22	0.40	0.11

#### 2.4 CHLORINATED AND OTHER HYDROCARBONS

The substances which are dispersed globally through the atmosphere also include polychlorinated biphenyls (PCBs), DDT and other chlorinated pesticides and industrial chemicals (Harvey and Steinhauer, 1974; Bidleman et al., 1981). These compounds, because of their persistence, also occur in the Arctic, and it is of interest to determine the concentration levels, and possible interrelationships with atmospheric transport and seasonal variations.

In order to study the transport of these substances to the Arctic and the seasonal variation of their concentration level, samples of particles and gaseous high-molecular weight organic compounds were collected with glass fibre filters and polyurethane foam samplers at stations in the Norwegian Arctic and Norway during 1982-1984. The concentrations of  $\alpha$ - and  $\gamma$ -hexachlorocyclohexane (HCH), hexachlorobenzene (HCB), pentachlorobiphenyls (5-PCB),  $\alpha$ -chlordane, fluoranthene, phenanthroquinones, and anthraquinones were determined in these samples by a combination of liquid chromatography, gas chromatography, and mass spectrometry (Oehme, 1982). In general, more than 98% of the chlorinated hydrocarbons were found on the polyurethane plugs, i.e., in the gaseous phase.

Table 2.3: Concentration ranges and mean values measured at Ny Ålesund during 5 measurement campaigns (in  $\text{pg} \cdot \text{m}^{-3}$ ).

Campaign	$\alpha$ -HCH		$\gamma$ -HCH		HCB		5-PCB		$\alpha$ -chlordane		Fluoranthene		Quinones	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Fall 1982	757 -1416	1192	0.1 -9.4	2.1	75 -169	123	2.3 -47	15	0.6 -3.2	1.2	8.2 -134	43	74 -364	217
Fall 1983	407 -695	548	34 -67	46	158 -277	186								
Summer 1984	260 -774	188	24 -100	47	20 -201	154	0.9 -43	21	1.7 -5.4	2.8	0.4 -46	20	52 -610	217
Winter/ Spring 1983	151 -734	485	21 -102	66	112 -187	144	32 -145	62	1.1 -2.3	1.7	13 -145	57	38 -952	325
Winter/ Spring 1984	121 -787	273	12 -70	29	29 -389	151	4 -51	16	0.6 -5.1	2.0	12 -142	48	21 -1040	267

Table 2.4: Concentration ranges and mean values measured at Bjørnøya during 4 campaigns (in  $\text{pg} \cdot \text{m}^{-3}$ ).

Campaign	$\alpha$ -HCH		$\gamma$ -HCH		HCB		5-PCB		$\alpha$ -chlordane		Fluoranthene		Quinones	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Fall 1982	420 -1550	774	0.1 -29	4.2	78 -200	120	6.3 -38	17	0.5 -1.7	0.8	18 -143	47	67 -236	145
Fall 1983	277 -477	326	14 -32	22	88 -180	134								
Summer 1984	38 -305	155	5 -41	18	42 -149	57	4.4 -14	8	0.6 -2.1	1.3	12 -43	17	68 -458	185
Winter/ Spring 1983	110 -469	282	23 -80	47	87 -201	124	8.5 -66	23	1.2 -3.1	1.7	42 -359	139	107 -1646	590

The wide range of the measured 2- or 3-days mean concentrations are related to episodic transport situations. Sometimes peak concentrations are observed at all stations simultaneously or with phase lags corresponding to the distance between the stations. The most pronounced episode occurred during the period March 6-13, 1983, when a situation with transport of air masses from northern USSR gave rise to the highest concentrations of  $\gamma$ -HCH, 5-PCB, fluoranthene and quinones measured at Bjørnøya and Ny Ålesund, as seen in Figure 2.15 (Oehme and Ottar, 1984).

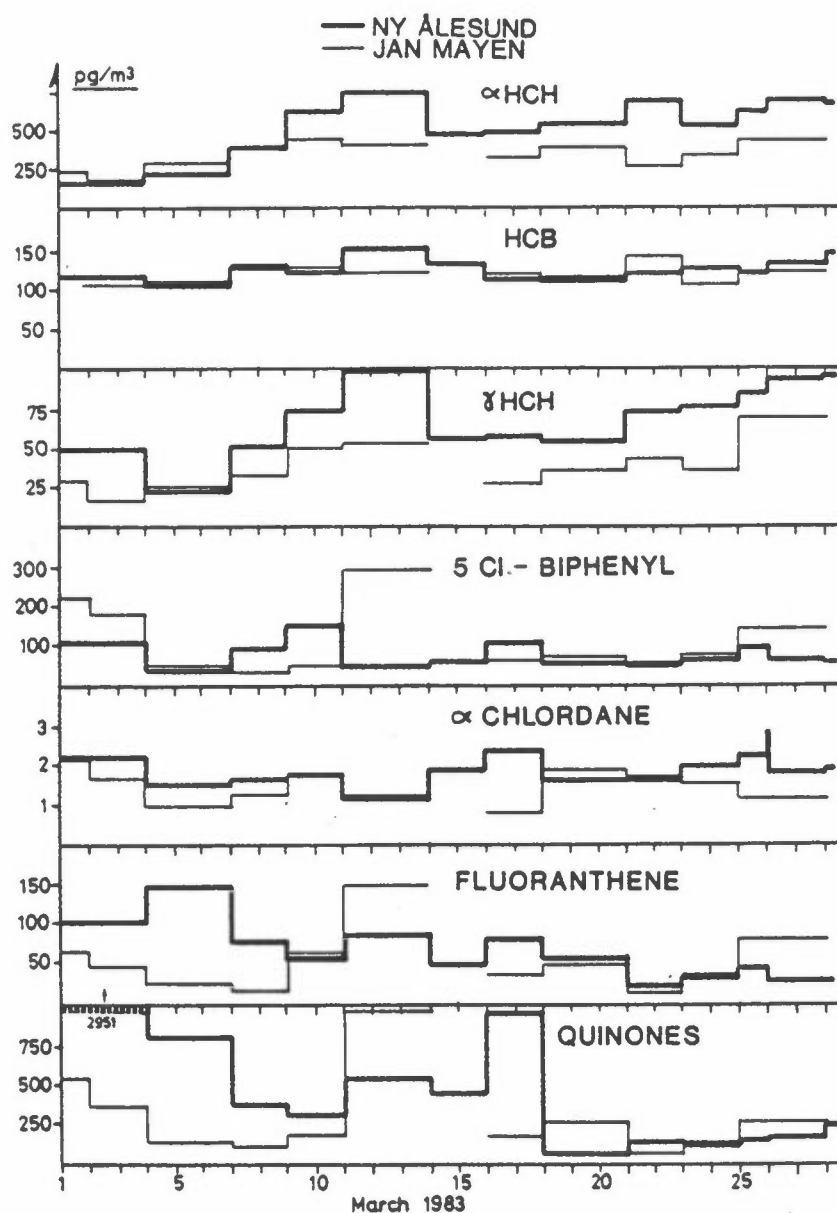


Figure 2.15: Time series of 2- and 3-day mean concentrations of  $\alpha$ -HCH, HCB  $\gamma$ -HCH, 5Cl-biphenyl,  $\alpha$ -chlordane, fluoranthene, and quinones at Ny Ålesund and Hopen during the winter/spring 1983 campaign.

The seasonal variation is most pronounced for fluoranthene and quinone, which are fossil combustion products and exhibit higher concentrations in winter. This may partly be due to emissions from local sources. The other components show little seasonal variation, except that  $\alpha$ -HCH is more abundant in summer. This can be due both to more efficient isomerisation of  $\gamma$ -HCH to  $\alpha$ -HCH during summer, and to preferential scavenging of the more water-soluble  $\gamma$ -isomer. Variations of the  $\gamma/\alpha$  ratio occur also on a shortterm basis, indicating different age histories and/or contributions from different source areas.

For comparison, measured concentrations of  $\alpha$ -HCH and  $\gamma$ -HCH, and HCB from different remote locations in the world are listed in Table 2.5. This shows that the measured concentrations in the Arctic are comparable to values from other remote locations in the northern hemisphere.

Table 2.5: Concentrations of  $\alpha$ -HCH and  $\gamma$ -HCH and HCB in various places of the world (in  $\text{pg} \cdot \text{m}^{-3}$ ).

Location	$\alpha$ -HCH	$\gamma$ -HCH	HCB
1. USA			
Denver, CO (Billings and Bidleman, 1983)	300*		240
New Bedford, MA (Billings and Bidleman, 1983)	1000*		180
Columbia SC (Bidleman, 1981)	370	180	200
College Station, TX (Atlas and Giam, 1981)	420		200
2. NORWAY			
Lillestrøm (Oehme and Ottar, 1984)	875	318	162
3. NORTH ATLANTIC (Atlas and Giam, 1981)	390		150
4. NORTHERN PACIFIC OCEAN Enewetak Atoll (Atlas and Giam, 1981)	250	15	100
5. WESTERN PACIFIC OCEAN (Tanabe et al., 1982)	1000	100	
6. EASTERN INDIAN OCEAN (Tanabe et al., 1982)	500	64	
7. ANTARCTIC OCEAN (Tanabe et al., 1982)	390		150

\*  $\alpha$ -HCH and  $\gamma$ -HCH.

### 3 ATMOSPHERIC TRANSPORT PROCESSES

An early conceptual model for the long range transport into the Arctic, was presented by Rahn and McCaffrey (1980). This model related the annual variation of the arctic air pollution at ground level to the seasonal variations of the surface position of the polar front, which was considered to function as a barrier preventing surface transport into the Arctic from source regions situated south of this front. On this basis a winter transport of polluted air masses from the Eurasian area was considered the more likely source of the arctic haze, a conclusion supported by studies of surface weather maps and available measurements of sulphates and aerosol trace elements, particularly vanadium and manganese. For similar reasons the highly industrialized regions in Eastern USA and the Far East were considered to be of little importance for the arctic haze.

The technique of subjectively identifying transport pathways on the basis of weather maps, has since been developed further by Raatz and Shaw (1984), who applied both surface and 500 mb weather maps. Nine types of transport from important source regions to Barrow, Alaska, were defined and by comparison with V/Mn ratios measured at Barrow, it was concluded that Central Eurasian sources contribute predominantly during winter and Western Eurasian sources during spring.

#### 3.1 VERTICAL DISPLACEMENT

In the first reported observations of the arctic haze, Mitchell (1956) describes an unexplained haze layer 2 km above the ground. When the arctic haze was again brought up for studies 20 years later, the haze was also visible at ground level, and the studies concentrated on ground level observations and transport within the boundary layer.

However, the aircraft measurements which were carried out by several institutions in March 1983 showed that haze layers also occurred above the boundary layer, at altitudes up to more than 5 km above the surface (Joranger and Ottar, 1985; Schnell and Raatz, 1985; Werhahn et al., 1986). These observations may be taken as experimental evidence of advection of polluted air into the Arctic above the boundary layer,

indicating that the polar front may not always function as an impenetrable barrier.

When low level air approaches the polar front from the warm side, it will rise to a higher level above the ground while being cooled down by adiabatic expansion. If the humidity is sufficiently high, cooling will result in condensation of water vapour and precipitation, which will remove most of the aerosols and the water soluble gases. On the other hand, dry air may be subject to considerable adiabatic expansion without leading to precipitation.

Following the arguments presented by Carlson (1981) and Iversen (1984), pollutant transport into the Arctic occurs along quasi-isentropic trajectories, with approximate conservation of potential temperature. The most important modifications are the release of heat by water vapour condensation, and radiative heat loss.

Figure 3.1 shows the longitude-averaged potential temperature profiles for the winter and summer seasons (Lorenz, 1967). The polar front may be recognized in these diagrams as a zone of inclining isentropic surfaces between  $30^{\circ}\text{N}$  and  $50^{\circ}\text{N}$  in winter, and between  $40^{\circ}\text{N}$  and  $60^{\circ}\text{N}$  in summer. It should be remembered, however, that the position of the polar front is highly variable, both as a function of time and longitude.

In winter, the cold and stable arctic boundary layer may extend far southwards into the USSR, and may occasionally include areas with large industrial sources south of  $60^{\circ}\text{N}$ . In summer the arctic boundary layer is most of the time less stable and confined to the region of the North Polar Ocean. Also apparent in Fig. 3.1 is the different vertical stability of the arctic atmosphere in winter and summer. It is seen that the range of potential temperature from the surface to 500 mb is  $40^{\circ}$  in winter, but only  $25^{\circ}$  in summer. The potential for creating several distinct haze layers is therefore larger in winter.

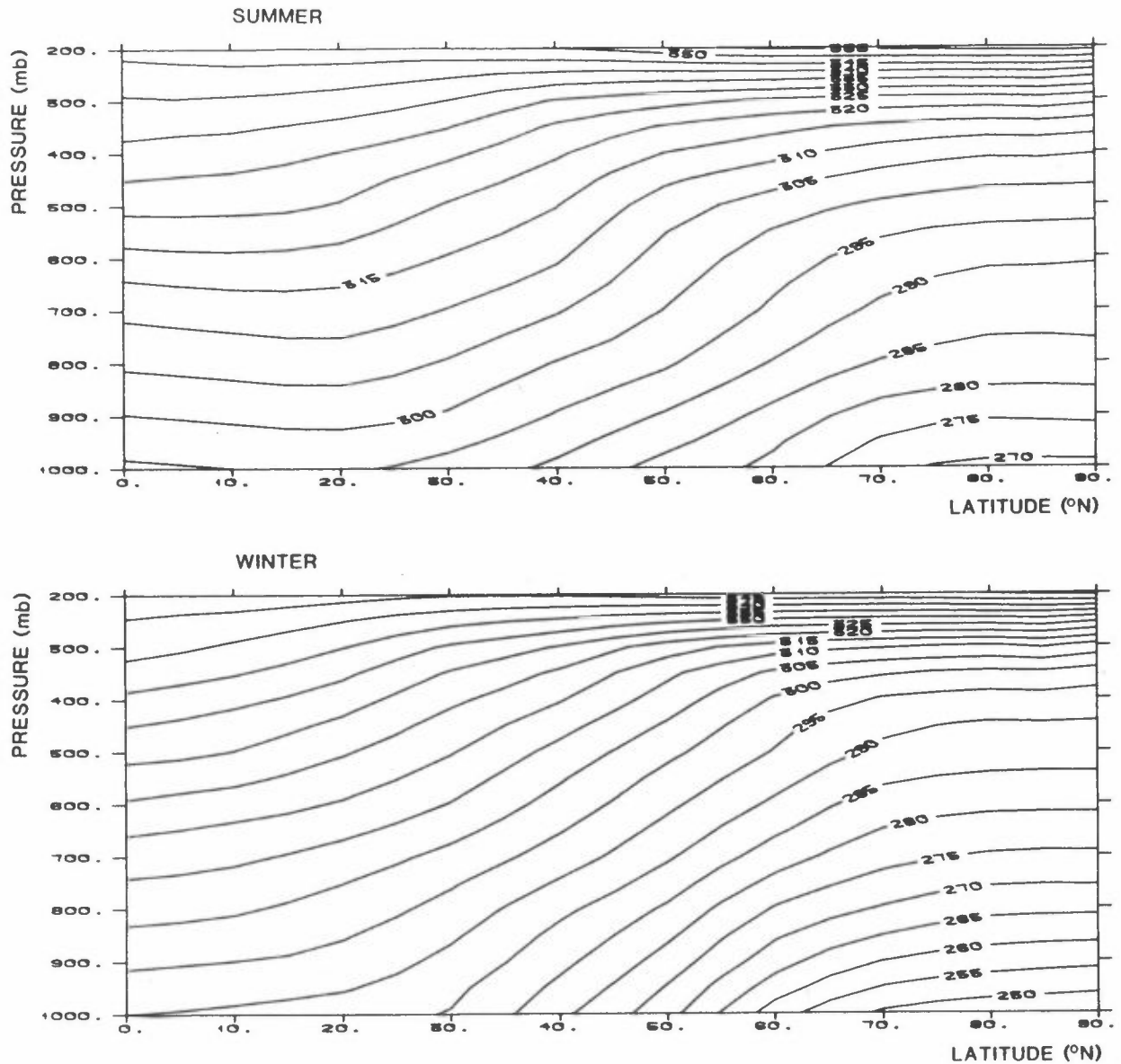


Figure 3.1: Mean potential temperature averaged over longitudes and seasons.  
(Summer = April-September; Winter = October-March).

The southern border of the cold, stable arctic boundary layer is frequently called the arctic front. This low level frontal zone and the polar front system separates the northern hemisphere into three main source areas. Pollutants from sources north of the arctic front will generally be confined within the arctic boundary layer. Sources between the arctic front and the polar front may pollute the arctic atmosphere above the boundary layer, while the third group of sources, south of the polar front, may be able to pollute the arctic troposphere at still higher levels, if precipitation scavenging is not



complete.

This crude separation should be used with care, because both the position of the front systems and the stability of the arctic boundary layer are very variable. The concept of isentropic surfaces has been utilized in the transport model for sulphur oxides which is described in Chapter 4.

### 3.2 OBJECTIVE METHODS FOR IDENTIFICATION OF PATHWAYS

An attempt to relate episodes of high and low concentrations of non-crustal vanadium in the air at Point Barrow, Alaska, with conditions of the large-scale planetary wave system was made by Reiter (1981). This work did not result in any definite conclusions. However, further work along this line has been carried out within the BP-programme (Iversen and Joranger, 1985; Iversen, 1985).

The main sources of air pollution in the northern hemisphere are generally situated between  $30^{\circ}\text{N}$  and  $60^{\circ}\text{N}$ . Notable exceptions are only a few large sources in the northern USSR, just south of  $70^{\circ}\text{N}$ . Longrange transport of air pollutants must therefore be associated with situations which lead to extended exchange of air in the meridional direction. The wind systems in connection with the ordinary low pressure cyclons in the westerlies are normally not sufficiently extensive in this connection.

The general atmospheric circulation system may be envisaged as a large heat engine, driven by the temperature difference between the low and high latitudes. Between these heat reservoirs work is carried out by means of air currents. Since the earth rotates, the upper level current does not head directly towards the poles, but deflects towards the east, creating westerly jet currents in both hemispheres, along which travelling cyclones and anticyclones are formed due to hydrodynamic instabilities. In the northern hemisphere, oceans and continents occupy alternating meridional sectors. There are also a few extensive mountain ridges. These features represent forcings on the jet stream in the northern hemisphere. Long meanders on the jet stream, with large amplitudes, are occasionally formed as a consequence of these

forcing elements, and when the atmosphere is in this mode of circulation, the meridional exchange of air is at its maximum.

Large amplitude meanders have a tendency to develop only in a certain meridional sector at a time, and since the normal eastward propagation of cyclones is halted, the situation is often called blocking. Figure 3.2 shows a typical case of blocking over Europe. The blocking region is normally dominated by a large warm ridge, and there is often a cold trough in the southern part. The jet current often splits in two branches, one heading northwards and the other southwards. These two branches rejoin again  $40^{\circ}$ - $90^{\circ}$  further east. It is the northern branch of the splitted jet current which provides the conditions for transport of polluted air from lower latitudes into the Arctic.

Two criteria have been used to identify blocking-like situations. The L $\emptyset$ -index is due to Lejenäs and Økland (1983), who used the geopotential height difference of the 500 mb pressure level between  $40^{\circ}$  N and  $60^{\circ}$  N to identify cases similar to the one shown in Fig. 3.2.

The second index was based on summation of the four largest planetary wave components. This "meridional index" is positive if the atmospheric flow is directed northwards between  $60^{\circ}$  N and  $70^{\circ}$  N. The two indices were calculated for each  $10^{\circ}$  longitude for each day and correlated with the short-term variability of the observed sulphate concentrations at Ny-Ålesund and Bjørnøya, cf. Section 2.

The results for Ny-Ålesund are shown in Fig. 3.3. It is seen that there is close correspondence between pollution "episodes" in the Arctic during winter and spring and the occurrence of a persistent meridional flow over Western Eurasia, as defined by the meridional index. Correspondingly, clean air "episodes" are correlated with transport of clean air from the North Atlantic.

Blocking situations, as defined by the L $\emptyset$ -index, are connected with S-N meridional flows  $20$ - $45^{\circ}$  west of the location of the blocking dipole. In the sector  $20^{\circ}$  N- $60^{\circ}$  E this index is negatively correlated with the sulphate concentrations at Ny-Ålesund, because of the associated inflow of clean air from the North Atlantic.

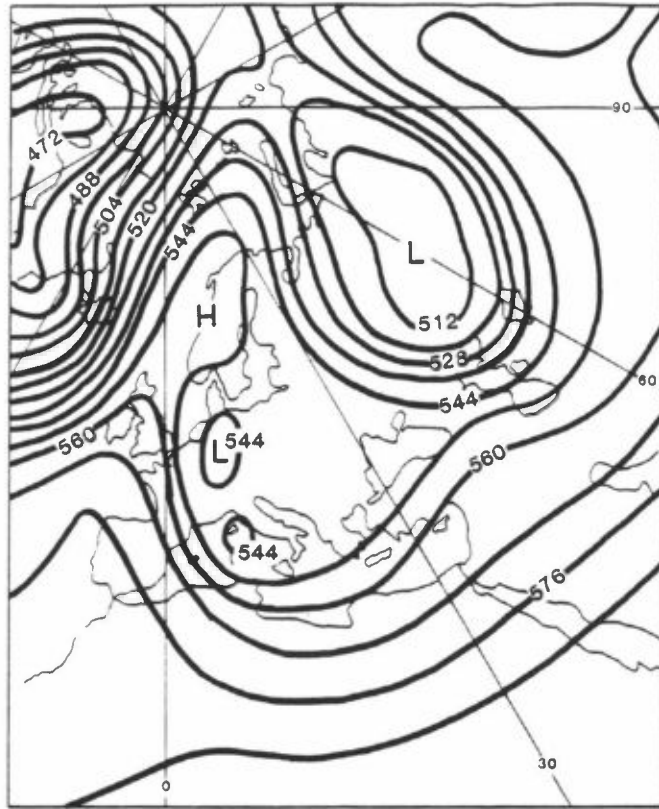


Figure 3.2: Geopotential height of 500 mb surface on 2 March, 1983, 00 GMT (European Meteorological Bulletin).

The sulphate episodes at Ny-Ålesund are not, however, significantly correlated with the  $L\emptyset$ -index in the sector  $60^{\circ}$  E to  $120^{\circ}$ . A reason for this may be that the  $L\emptyset$ -index defines blocking and meridional flow at lower latitudes than the "meridional index" defined above. The  $L\emptyset$ -index is therefore not representative of meridional transport to Ny-Ålesund within the boundary layer.

Neither of the two indices are correlated with the sulphate concentrations in Ny-Ålesund during the summer period, indicating that the observed fluctuations in the low-level sulphate concentrations during this season have little to do with large-scale atmospheric transport. Precipitation scavenging, in connection with the high frequency of fog and low stratiform clouds, may be an important factor.

The frequency of blocking situations, as well as the strength of the meridional air exchange associated with these oscillations of the jet stream, are both at a maximum during the winter and spring seasons. This corresponds well with the observed seasonal variations in the

concentrations of air pollutants in the Arctic. The strong correlation between short-term concentration variations and meridional transport in the sector  $20^{\circ}\text{E}$  and  $60^{\circ}\text{E}$  shows that there is a main pathway of polluted air from the Western Eurasian continent to the Arctic during winter and spring.

### CORRELATIONS, NY ÅLESUND

OPEN BARS: LØ-INDEX, SOLID BARS: MERID. INDEX

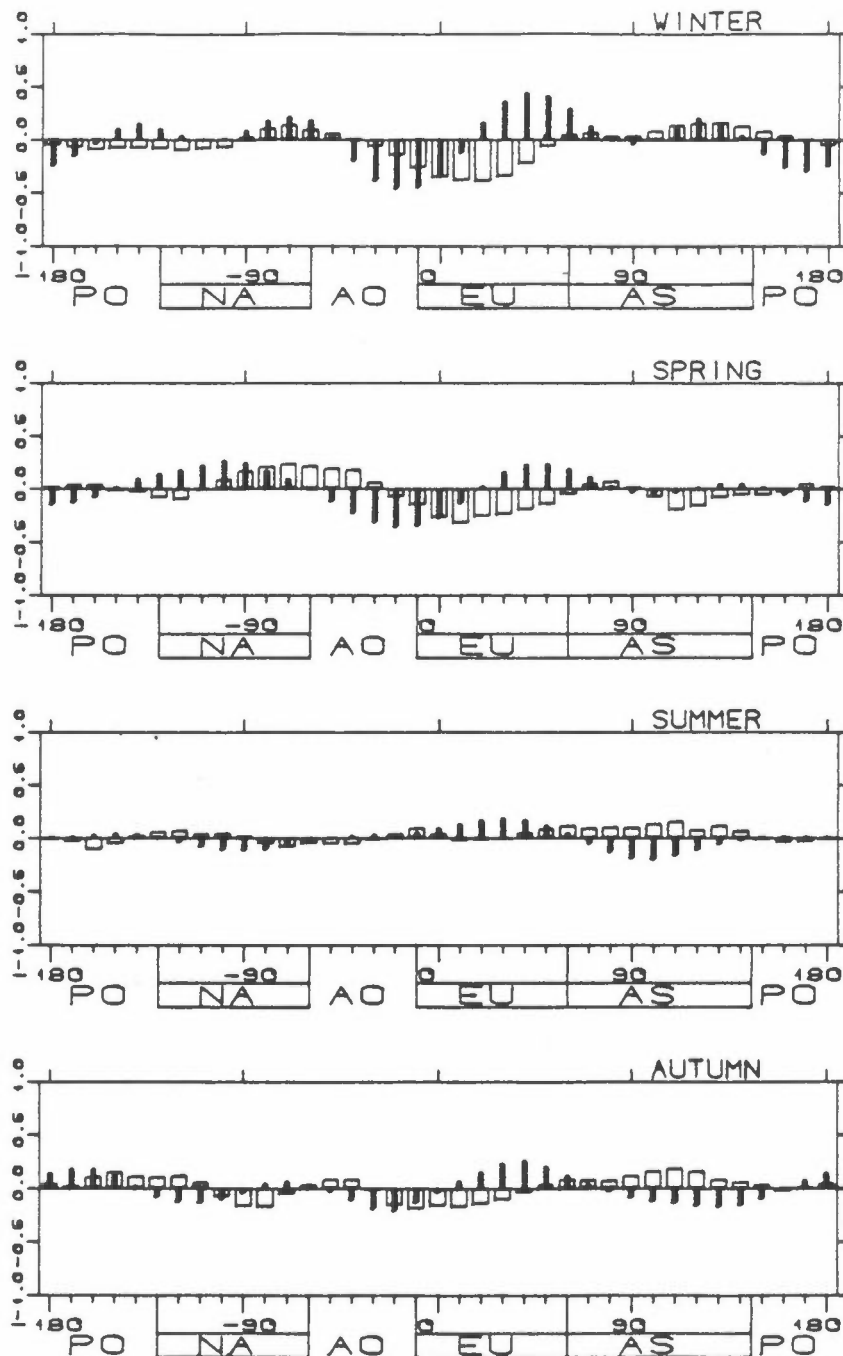


Figure 3.3: Correlations for each ten degree longitude, between residual concentrations at Ny-Ålesund and LØ-index (open, wide bars) and meridional index (solid bars).

## 4 SOURCES OF ARCTIC AIR POLLUTION

### 4.1 MODEL CALCULATIONS OF SULPHUR DIOXIDE AND SULPHATE CONCENTRATIONS

In order to carry out quantitative model estimates of the transport of airborne sulphur into the Arctic, available information on the emission of sulphur dioxide in Europe (Dovland and Saltbones, 1979, 1986), and in North America (Barrie and Hales, 1984), has been transferred to a grid system with 300 km x 300 km squares. Emissions in the Far East, and in the USSR outside the EMEP grid, were estimated and positioned on the basis of data on fossil fuel consumption, population density, industrial production and other relevant information. The resulting emission intensities are shown in Figure 4.1 (Semb, 1985).

As explained in Sec. 3.2, the airborne transport into the Arctic will, as a first approximation, follow isentropic trajectories. The developed atmospheric dispersion model (Iversen, 1986a) utilizes atmospheric isentropic surfaces as coordinates. However, release of latent heat by condensation, turbulent heat conduction, and radiation are also included. Parameters describing the oxidation of SO<sub>2</sub> to sulphate, precipitation scavenging and dry deposition have been adapted from the work of Eliassen and Saltbones (1982). A summary of some relevant model parameters is given in Table 4.1. Further details are given in the technical description by Iversen (1986a).

Meteorological input data were obtained from the National Center for Atmospheric Research (NCAR), Boulder, Colorado.

The simulations of airborne sulphur dioxide and sulphate concentrations (Iversen, 1986b) were carried out for two periods, 5-30 March 1983 and 20 June - 10 July, 1983). The simulations were initiated by starting with estimated "background" concentrations, and let the model run for a 5-day starting up period.

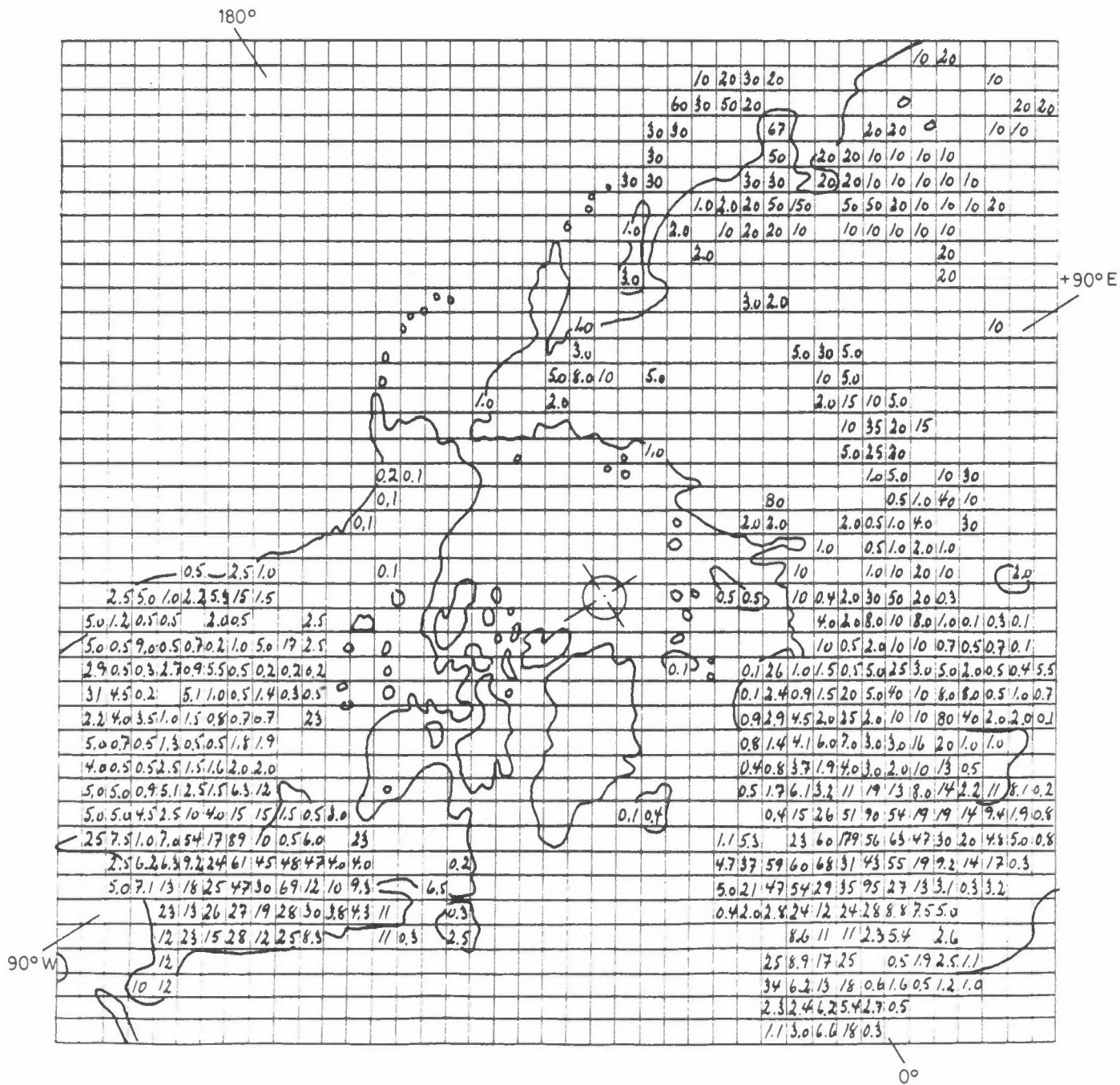


Figure 4.1: Estimated SO<sub>2</sub>-emissions for individual grid squares (≈ 300 km x 300 km)<sup>2</sup>. Unit: 10<sup>4</sup> t.y<sup>-1</sup> (as S).

Table 4.1: Choice of parameters.

	PARAMETER	VALUE
	Number of grid points in x-direction, I	41
G	Number of grid points in y-direction, J	43
R	Number of isentropic surfaces, L	10
I	X-coordinate of north pole, $l_p$	23
D	Y-coordinate of north pole, $J_p$	21
	Distance north pole - equator, grid coord., R	39.5
	Radius of earth	6371 km
	"Background" $SO_2$ -concentrations	$0.03 \mu g(S)m^{-3}$
	"Background" $SO_4$ -concentrations	$0.05 \mu g(S)m^{-3}$
	Dry dep. velocity, $SO_2$ , normal conditions	$8 \cdot 10^{-3} ms^{-1}$
P	Dry dep. velocity, $SO_2$ , snowcovered open landscape	$10^{-3} ms^{-1}$
R	Dry deposition velocity, $SO_4$	$10^{-3} ms^{-1}$
O	Part of emissions deposited locally, $\alpha$	0.15
C	Part of emissions as $SO_4$ , $\beta$	0.05
E	Scavenging ratio, $SO_2$ , $W_q$	$5 \cdot 10^4$
S	Scavenging ratio $SO_4$ , clouds $W_s$ (cloud)	$10^6$
S	Scavenging ratio, $SO_4$ , subclouds $W_s$ (subcloud)	$5 \cdot 10^4$
E	Effective scavenging depth, h	$10^3 m$
S	Density of water, $\rho_w$	$10^3 kg m^{-3}$
	Oxidation rate of $SO_2$ at equator: $k_{c,ekv}$	$4 \cdot 10^{-6} s^{-1}$
	Oxidation rate at north pole, period I, $k_{c,pole}^{(I)}$	$0.6 \cdot 10^{-6} s^{-1}$
	Oxidation rate at north pole, period II, $k_{c,pole}^{(II)}$	$2 \cdot 10^{-6} s^{-1}$

Time averaged, computed concentration fields at different heights for the period 5-30 March are presented in Fig. 4.2. It is seen that the sulphur dioxide concentrations are highest close to major source areas and at relatively low heights above the surface, while sulphate has a more widespread occurrence. The ratio of sulphate to sulphur dioxide concentrations increases with time because of the slow transformation of sulphur dioxide to sulphate, and the different rates of dry deposition. The precipitation scavenging process has been assumed more efficient for sulphate than for sulphur dioxide, however. Table 4.2 gives estimated mean concentrations at different heights above the surface, for the Arctic north of  $72.5^{\circ}N$ . It is seen that the calculated concen-

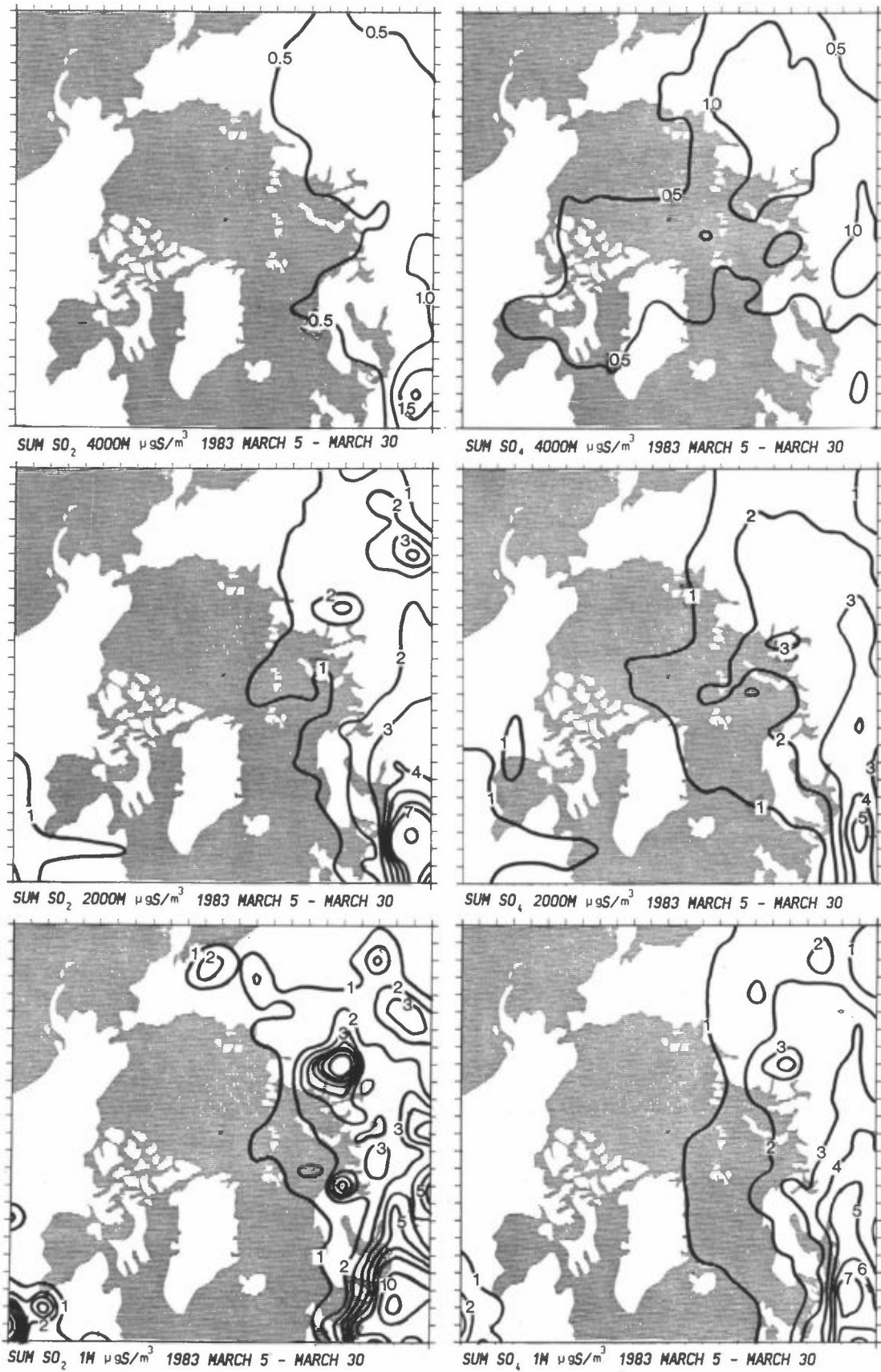


Figure 4.2: Calculated average concentrations of sulphur dioxide and sulphate at different heights, March 5-30, 1983.



trations at ground level are in reasonable agreement with measured values in the Norwegian Arctic during the same period (Ch. 2). The calculations indicate a vertical distribution with maximum concentrations between 1000 and 3000 m above the surface, while the nephelometer vertical profiles obtained during the aircraft sampling campaigns in March 1983 show marked decreases in aerosol concentrations above 1000 m. However, since the aircraft sampling flights took place within a rather limited time period around 20 March, and were centered around Spitsbergen, these vertical profiles are not directly comparable with the model estimates as presented in Table 4.2. In addition, the model calculations show considerable day-to-day variability in the vertical distribution of the sulphur compounds (Iversen, 1986b). Other measured vertical profiles, particularly those obtained in March 1984, indicate higher concentrations of aerosols aloft.

Table 4.2: Mean estimated concentrations over the area north of  $72.5^{\circ}$  N, Unit:  $\mu\text{g S/m}^3$ . Period: 5-30 March 1983.

	Estimated	
	SO <sub>2</sub> -S	SO <sub>4</sub> -S
5000 m	0.19	0.35
4000 m	0.28	0.60
3000 m	0.40	0.91
2000 m	0.59	1.10
1000 m	0.69	1.04
1 m	0.60	0.76

Average sulphur dioxide and sulphate concentrations for the summer period 20 June - 10 July 1983 are shown in Fig. 4.3. The most striking difference, when compared with the March 1983 simulations, is that transport into the Arctic is much more restricted, both at ground level and in the upper troposphere. Mean concentrations for the area north of  $72.5^{\circ}$  N are given in Table 4.3.

Comparison with winter conditions (Table 4.2) shows less pollution and increased ratio  $\text{SO}_4/\text{SO}_2$  in summer especially in the boundary layer.

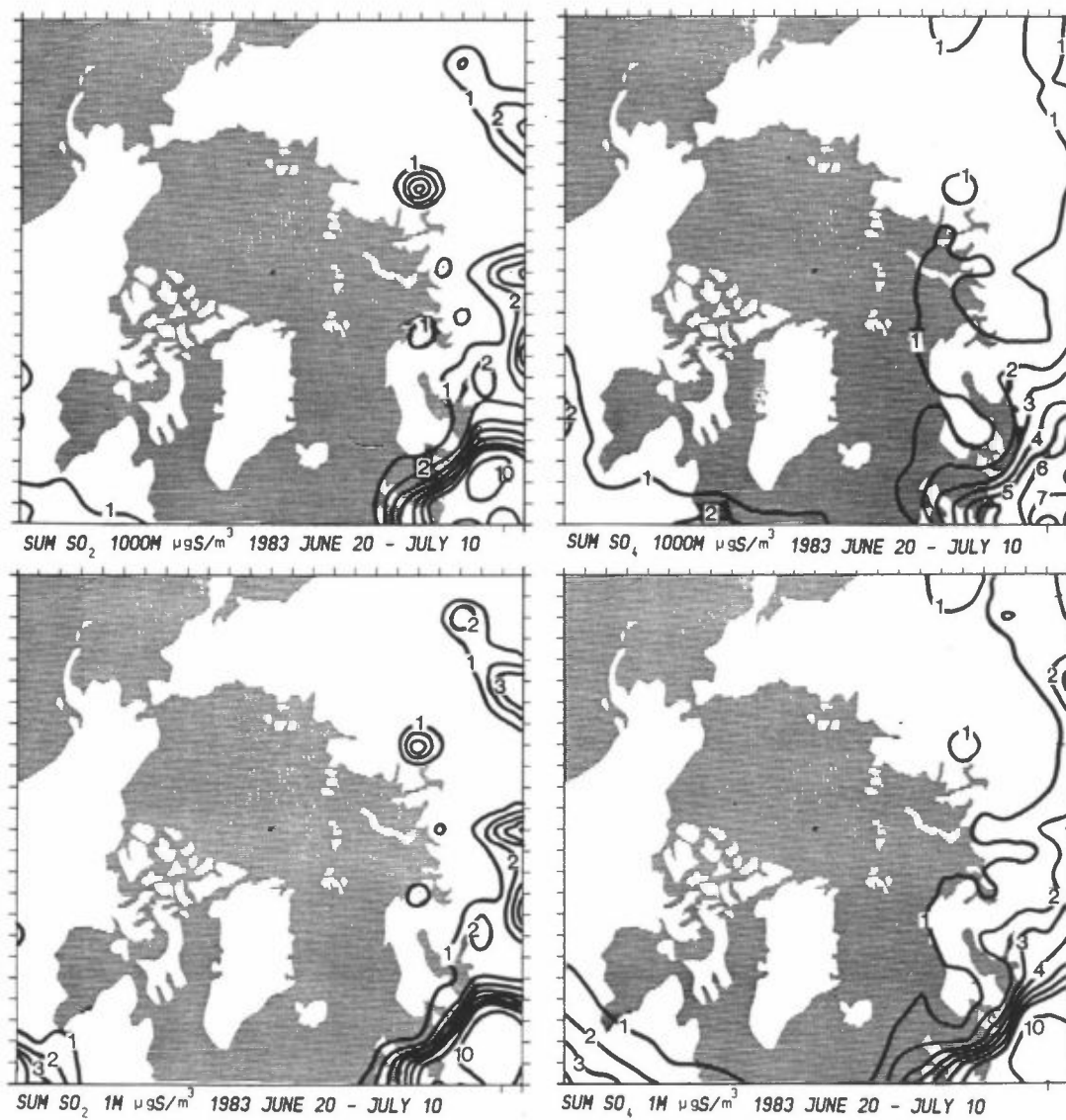


Figure 4.3: Calculated average concentrations of sulphur dioxide and sulphate at different levels June 20 - July 10, 1983.

An important feature of the model is its ability to determine separately the contributions of sulphur dioxide and sulphate, due to emissions in particular source areas: i.e., USSR, Europe outside the USSR, North America, and the Far East. The percentage contributions to the concentrations of sulphur dioxide and sulphate at different height levels, for the two simulation periods, are given in Tables 4.4 and 4.5.

Table 4.3: Mean concentrations of sulphur dioxide and sulphate north of  $72.5^{\circ}$  N, 20 June-10 July 1983. Unit:  $\mu\text{g S/m}^3$ .

Height	SO <sub>2</sub>	SO <sub>4</sub>
5000 m	0.01	0.03
4000 m	0.02	0.07
3000 m	0.05	0.16
2000 m	0.08	0.23
1000 m	0.07	0.31
1 m	0.01	0.13

Table 4.4: Percentage contributions from the 4 different source regions to the concentration levels north of  $72.5^{\circ}$  N during 5-30 March 1983.

	EUROPE		USSR		NORTH AMERICA		FAR EAST	
	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>
5000 m	62.1	64.2	16.0	10.0	20.4	23.1	1.6	2.7
4000 m	61.4	68.1	28.0	17.6	9.3	12.3	1.2	2.0
3000 m	43.3	58.8	52.1	33.7	4.0	6.3	0.7	1.2
2000 m	20.5	40.8	77.8	55.4	1.4	3.2	0.3	0.6
1000 m	10.9	30.1	88.4	67.4	0.6	2.1	0.1	0.4
1 m	6.2	23.5	93.4	74.9	0.3	1.4	0.0	0.2

Table 4.5: Percentage contributions from the 4 different source regions to the concentration levels north of 72.5°N during 20 June-10 July 1983.

	EUROPE		USSR		NORTH AMERICA		FAR EAST	
	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>	SO <sub>2</sub>	SO <sub>4</sub>
5000 m	4.9	7.7	62.2	51.0	32.8	41.0	0.1	0.3
4000 m	9.5	11.9	67.0	51.3	23.3	36.0	0.2	0.8
3000 m	20.7	23.2	66.3	51.6	12.8	24.0	0.2	1.2
2000 m	24.2	30.3	69.5	52.6	6.1	16.3	0.1	0.9
1000 m	18.8	31.4	78.2	56.6	2.9	11.3	0.1	0.6
1 m	9.4	34.2	90.5	59.8	0.0	5.5	0.0	0.4

These results may be seen as a quantitative confirmation of the assumptions made in Ch.3. Generally, the main sources of air pollutants in the lowest layers in the Arctic are in the USSR, particularly in winter. The relative contribution of sources in Europe increases with altitude, and sources in North America may contribute to concentrations at altitudes above 3-4000 m.

The simulated precipitation scavenging and dry deposition can also be quantified and assigned to the respective source areas. In general, dry deposition occurs from the lowest layer, while snow and rain also contain contributions from layers above 2000 m, particularly in winter and spring. More work, including comparisons between measured precipitation and model predictions, should be undertaken in order to explore this further.

It should be realised, that the model calculations involve simulation of advection over very large areas and with correspondingly crude spatial resolution. This, and the quality of the meteorological input data available, to some extent limit the possibility of detailed comparisons with measured data. On the other hand, the model represents a step towards a global atmospheric dispersion model based on real time observations of meteorological parameters.

#### 4.2 TRACE ELEMENTS

Because of the different elemental composition of aerosols from different sources, trace element ratios can be used as chemical finger-

prints for emissions from given source areas. The estimates of trace element emissions were based on emission factors and statistical information on fuel consumption, industrial production, and waste incineration (Pacyna, 1986a, NILU, 1984). Of the elements, Be, Co, Mn, Sb and Se are emitted mainly from coal combustion, Ni and V from fuel oil, and Pb from leaded gasoline. Primary and secondary nonferrous smelters account for the main part of the As, Cd, Cu and Zn emissions to the atmosphere, while Cr, and Mn are emitted mainly from iron, steel and ferroalloy productions.

Spatial distributions of estimated As, Cu, Cr, Mn, Cd, and Zn emissions in Europe are shown in Fig. 4.4 It is seen that there are considerable variations in the spatial distribution patterns for these elements in Europe. In the northern USSR (Fig. 4.5), both the Kola peninsula and the Norilsk area have large copper-nickel smelters. These smelters, and fuel combustion sources in the Pechora basin are relatively close to Spitsbergen. Other areas in USSR are characterized by manufacturing industries and general consumption (Leningrad and Moscow), petroleum refineries and chemical industries (Ural), iron and steel (Donbas and Black Sea, area 7).

The average trace element composition of aerosol samples collected at Ny-Ålesund indicates contributions from a variety of sources. Comparison with the elemental composition of aerosol filter samples collected in Southern Norway (Pacyna et al., 1984) show that the general chemical composition is rather similar, but relatively higher concentrations of As, Cd and Cu at Ny-Ålesund may indicate a larger contribution from nonferrous smelters.

More detailed comparisons of estimated and measured airborne trace element concentrations at Ny-Ålesund were made for a specific pollution episode, 6-13 March 1983 (Pacyna et al., 1985a). Concentrations were estimated on the basis of 96-h, 850 mb trajectories, assuming constant, element specific, dry deposition rates and no precipitation scavenging. The results are shown in Fig. 4.6. Sources contributing to the estimated airborne concentrations were located mainly in Western USSR, and, although the concentration levels of some elements, such as Pb, Se, V and Mn, appear to have been overestimated, the observed

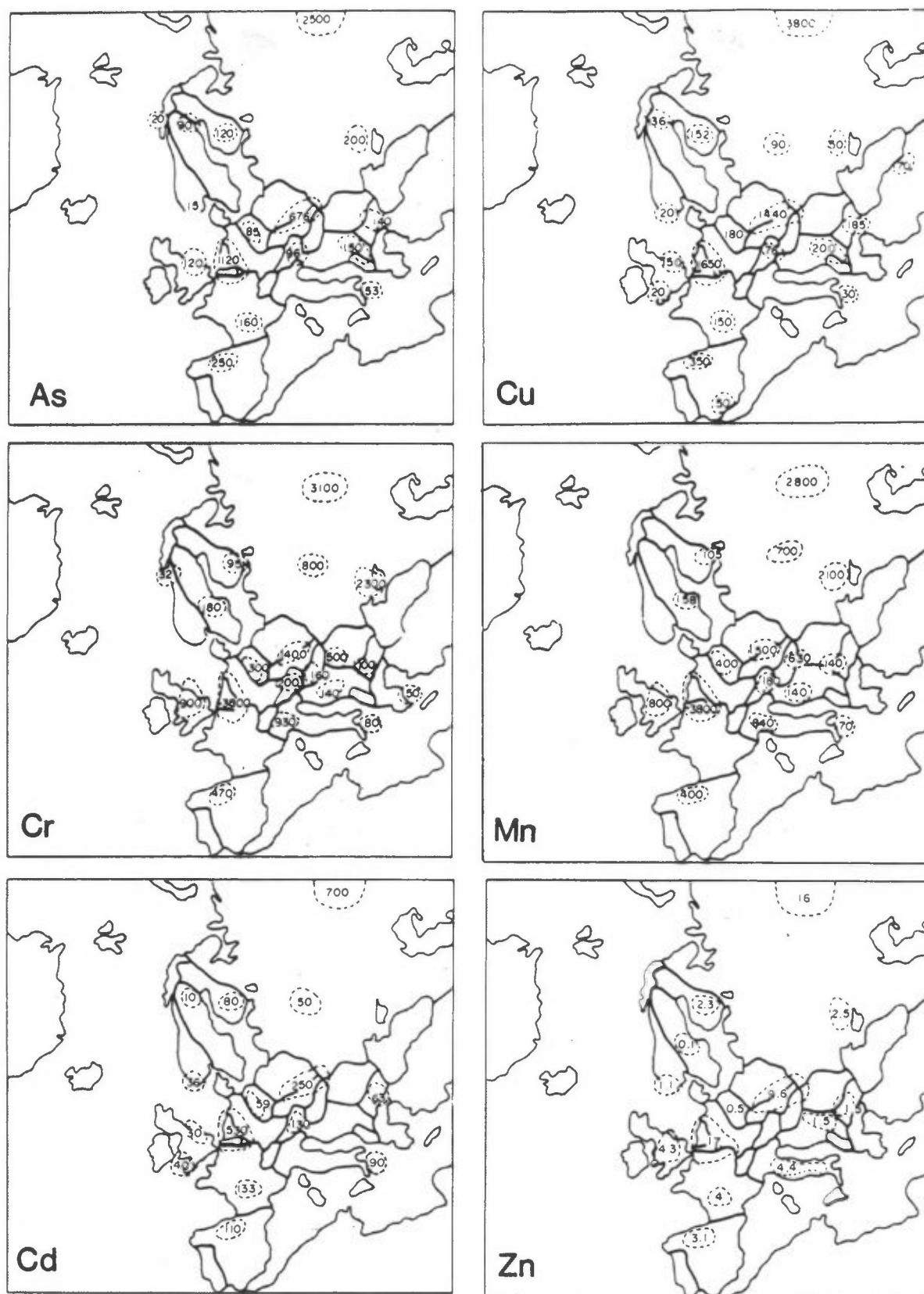


Figure 4.4: Spatial distributions of the European emissions of arsenic, copper, chromium, manganese, cadmium, and zinc. The As, Cu, Cr, Mn, and Cd emissions are in tonnes/yr and the Zn emission in  $10^3$  tonnes/yr.

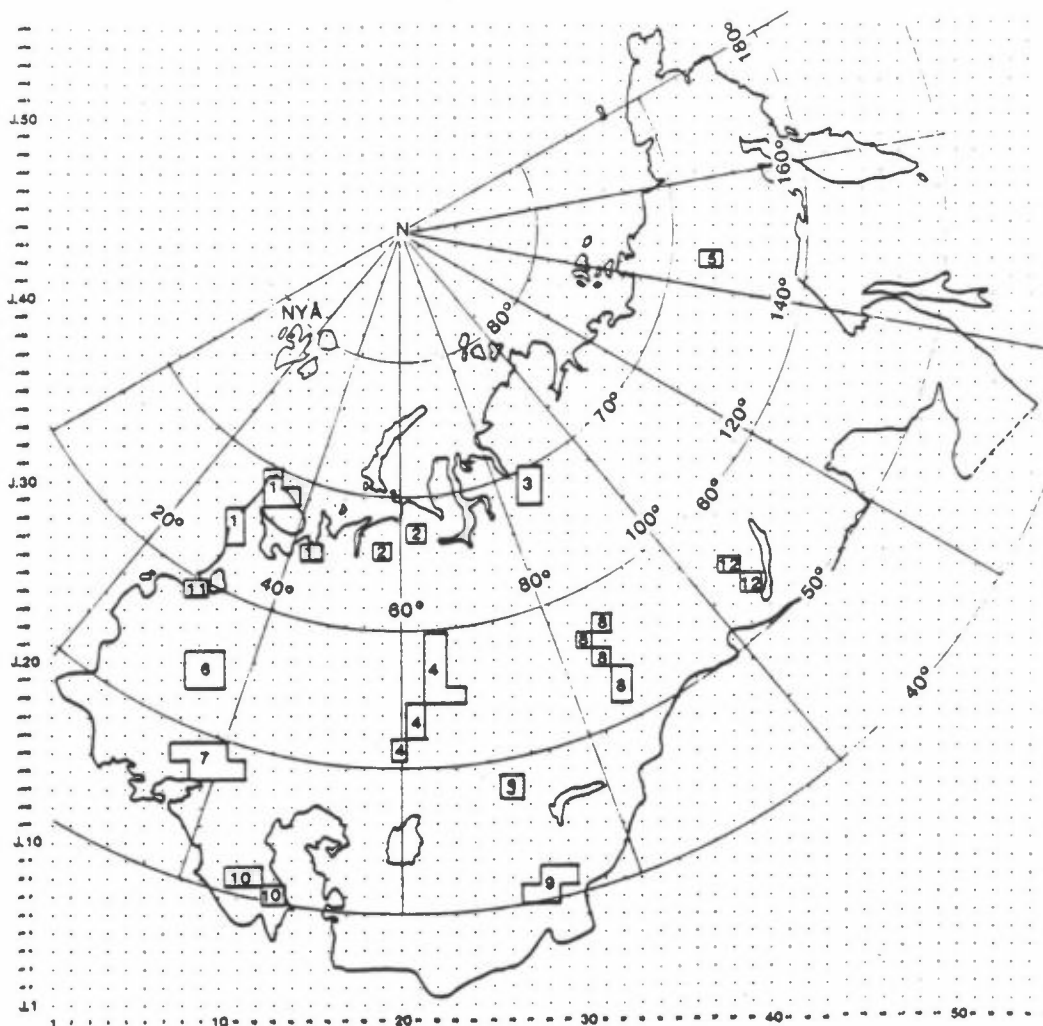


Figure 4.5: Location of emission sources considered in this report: (1) The Kola Peninsula, (2) The Pechora basin, (3) The Norilsk area, (4) The Urals, (5) The Yakutsk area, (6) The Moscow area, (7) The Donetsk area, (8) The Kuznetsk area, (9) The Fergana area, (10) Caucasus, (11) The Leningrad area and (12) The Baikhal area.

concentrations are not explainable in terms only of emissions in the northernmost parts of the USSR.

A statistical comparison of the chemical composition of samples collected simultaneously at Vardø and in Ny-Ålesund during March 1983 (Pacyna, 1986b) showed very close similarity between the two sets of samples. Since Vardø is very close to the large copper nickel smelters on the Kola peninsula, the most likely explanation seems to be that the ambient aerosol at both Ny-Ålesund and Vardø is due to the combined emissions in large areas of the Eurasian continent. Relative to this extended source area, the two measurement sites are close together.

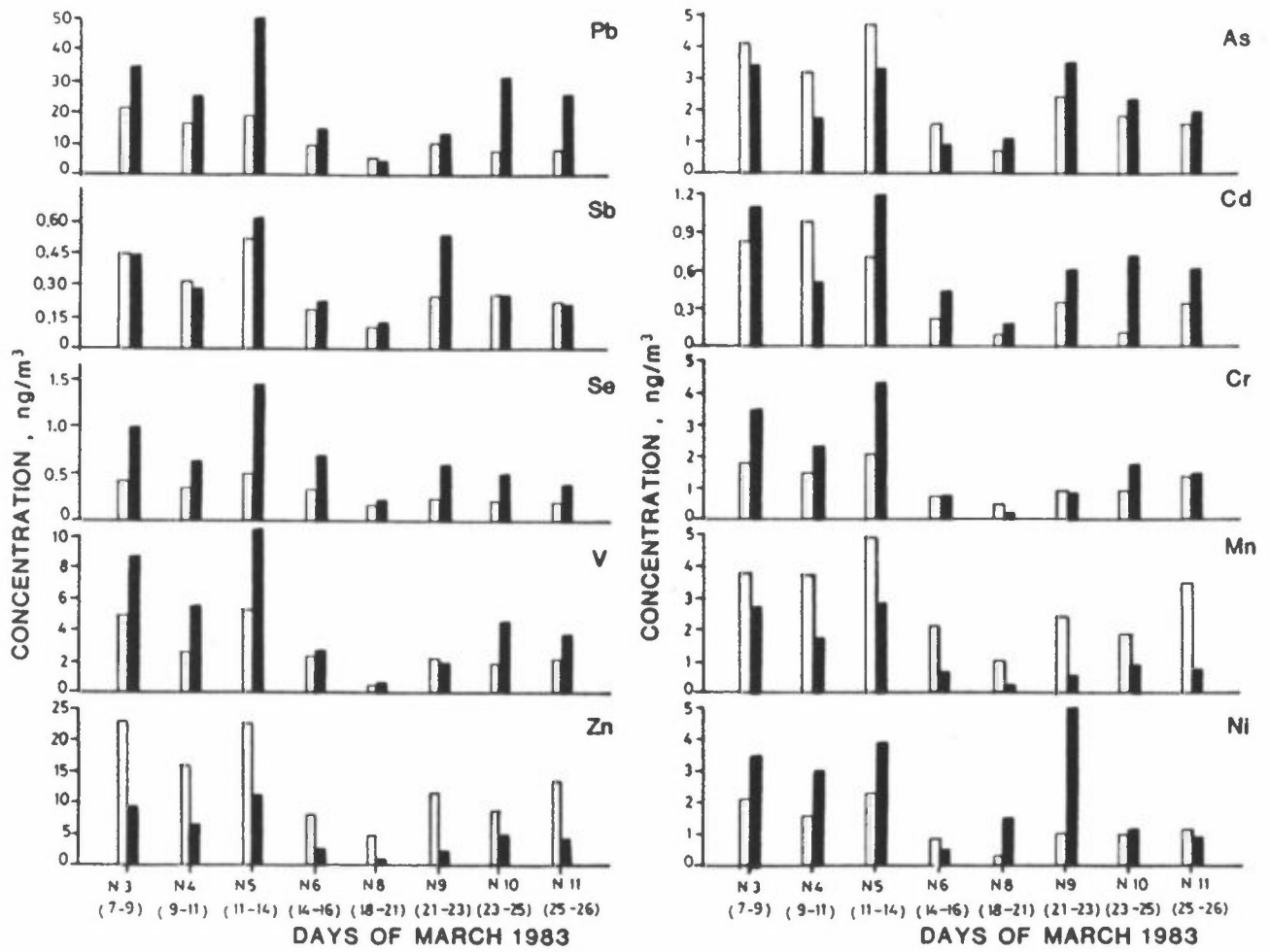


Figure 4.6: Measured (open bars) and calculated concentrations (full bars) of trace elements at Ny-Ålesund in March 1983.



## 5 LIGHT HYDROCARBONS IN THE ARCTIC ATMOSPHERE

The atmospheric concentration of light hydrocarbons ( $C_1-C_7$ ) is an essential factor in controlling the production rate of ozone and other photochemical oxidants in the atmosphere. Indirectly light hydrocarbons determine the rate of sulphate and nitrate formation through their control of the oxidation mechanisms. Sulphur dioxide is oxidized to sulphuric acid by hydrogen peroxide or ozone dissolved in cloud droplets and in gas phase reaction with the hydroxyl radical, and nitrogen dioxide also reacts with hydroxyl to form nitric acid.

The increasing emissions of nitrogen oxides and hydrocarbons have resulted in elevated concentrations of photochemical oxidants over regions like Central Europe and the eastern U.S.A. in summer in anti-cyclonic weather. During these photochemical episodes the light hydrocarbons are consumed. In winter the photochemical activity is less, and the hydrocarbon concentrations correspondingly higher.

The different hydrocarbons cover a wide range of reactivities, and many of them react so slowly that they have a hemispheric or global distribution. The main sources for non-methane hydrocarbons are automobile exhaust, refining and distribution of oil products and natural gas, and the use of solvents. Each of these source categories have different characteristic compositions. This can be used to trace the sources contributing to the hydrocarbon concentration in episodes with one prevailing atmospheric flow direction, or to determine global emission distributions and trends in time, if average concentrations over months, seasons or years are measured over long periods of time.

### 5.1 MEASUREMENTS AND INTERPRETATION

Samples of compressed or liquified air were taken in cooperation with Atomic Energy Research Establishment at Harwell, UK, during several periods in the spring and summer at Ny Ålesund and at other arctic locations (Hov et al., 1984). The samples were later analysed by gas chromatography at Harwell, and it was found that with the exception of propene and ethene, the hydrocarbon concentration levels were generally one order of magnitude higher during spring (March 1983) than in

summer (July 1982). The spring samples at Spitsbergen were taken in air masses coming off the Soviet Union, and they were more abundant in alkanes and less abundant in some of the solvents than air sampled at Barrow in March 1982 by U.S. scientists (Rasmussen, et al., 1983). This may indicate a systematic difference in the source distribution of hydrocarbons in the Soviet Union compared to North America.

The arctic measurements of light hydrocarbons were analysed theoretically in cooperation with the University of Oslo, applying a global, 2-dimensional model of the atmosphere describing the combined effect of transport and chemistry in a transect of the atmosphere from pole to pole and from the ground to a height of 15 km (Isaksen et al., 1985). The global sources of light hydrocarbons were estimated and distributed with latitude. Uniformity in the east-west direction was assumed. As an example, the calculated, annual variations of ethane, propane and propene are shown in Figure 5.1 together with the measured concentrations. One standard deviation is indicated by half of the length of the vertical bar. The shape of the measured annual variation is correctly reproduced by the model for ethane and propane, while it is opposite for propene in a calculation where pollution sources only are assumed to be important. For propene, this suggests that the measured concentration is dominated by local natural (oceanic) sources, while for ethane and propane pollution sources seem to be the dominant ones. However, the measured concentrations were higher than calculated, and this may point to a larger than average contribution of alkanes from the source areas of the polluted air sampled in March 1983.

Less is known about the seasonal variation of secondary species like ozone and peroxyacetylnitrate in the Arctic, than about the light hydrocarbons. Figure 5.2 shows the calculated annual behaviour of the concentration of ozone in the lowest 2 km of the atmosphere for  $80^{\circ}\text{N}$  and  $50^{\circ}\text{N}$  (Isaksen et al., 1985). There is a winter minimum and summer maximum at both latitudes. The direct influence of the high spring concentration of light hydrocarbons was investigated by reducing them to zero in March. It can be seen that at  $80^{\circ}\text{N}$  this had virtually no impact on the ozone concentrations later in the year, while at  $50^{\circ}\text{N}$  a 20% lower ozone concentration was calculated at mid-summer compared to

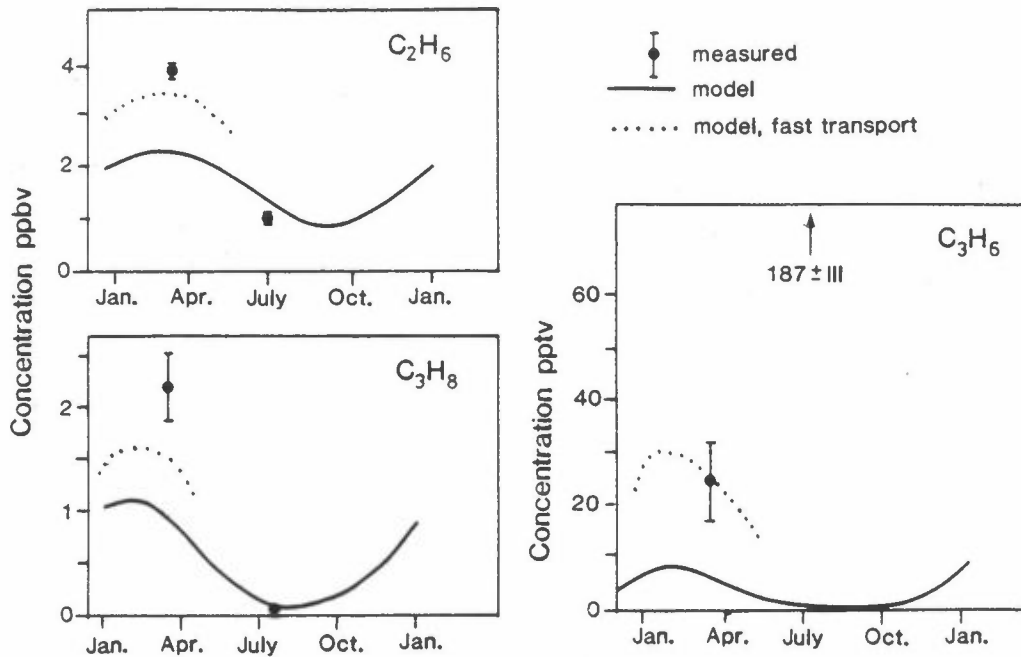


Figure 5.1: Calculated annual variation of ethane, propane and propene at 80°N latitude at 1250 m height compared with average measured concentrations on Spitsbergen, during July 1982 and March/April 1983.

The standard deviation of the measured concentration is indicated. The calculated concentration during winter with a particularly efficient poleward transport is also indicated.

the first calculation. The high concentration of light hydrocarbons during the spring at northerly latitudes, thus seems to contribute to a pulse in the ozone production in the northern hemisphere during late spring and summer. This supports the view that some of the spring increase in the ozone concentration observed in the northern hemisphere could be due to tropospheric ozone production rather than stratospheric injection.

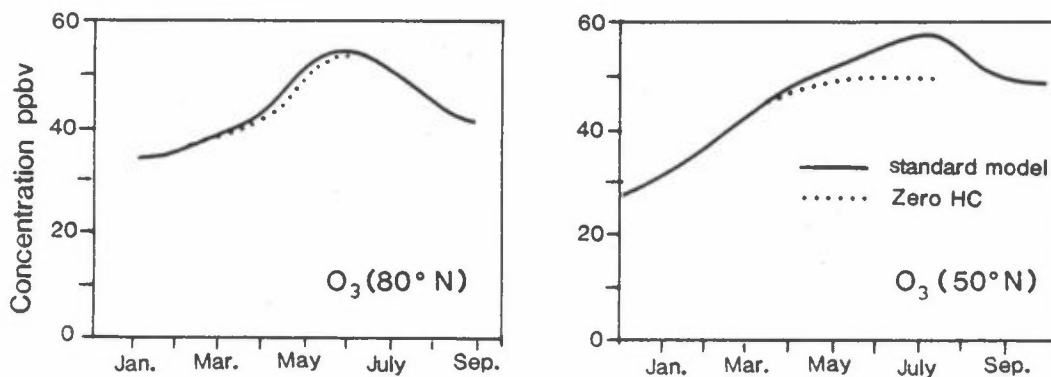


Figure 5.2: Calculated annual concentration variation for ozone at 80°N and 50°N latitude at 1.25 km height in the atmospheric boundary layer. Also indicated is the development of the ozone concentration when starting with the hydrocarbon concentration equal to zero, in February/March.

Several investigators have reported that the concentration of ozone is increasing in the troposphere over the northern hemisphere. The hydrocarbon measurements in the Arctic support the view that there is an anthropogenic contribution to the springtime maximum in background ozone (Isaksen and Hov, 1986), see Figure 5.3. An increase in ozone in the troposphere is very important because ozone generated in photochemical smog episodes in the atmospheric boundary layer is superimposed on a background concentration which actually is increasing. Ozone in the troposphere is also an important infra-red absorbing gas and may play a climatological role approaching that of  $\text{CO}_2$  in significance. The hints that arctic hydrocarbon measurements have given about the controlling processes for the global distribution of a vital trace gas like ozone, really points out the need for continued measurements of organic gases and to extend the measurements to include nitrogen oxides and other gases at a representative arctic site like Ny Ålesund.

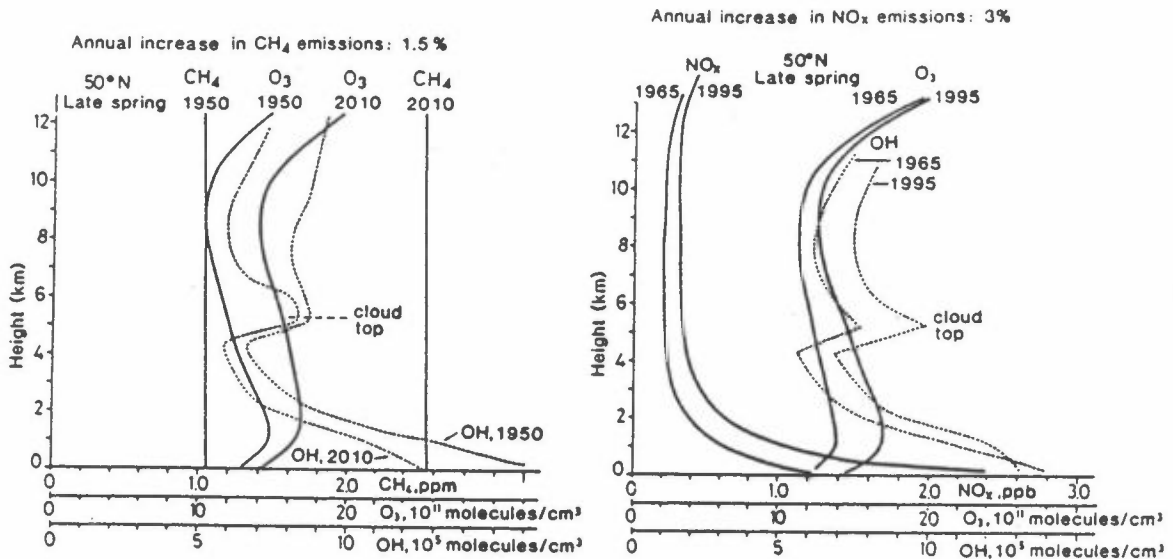


Figure 5.3: Calculated vertical profiles at 50°N latitude for  $\text{O}_3$ ,  $\text{CH}_4$  and  $\text{OH}$  for 1950 and 2010 with a  $\text{CH}_4$ -emission increase of 1.5%/yr (left), and for  $\text{O}_3$ ,  $\text{OH}$  and  $\text{NO}_x$  for 1965 and 1995 with a  $\text{NO}_x$  emission increase of 3%/yr (right). A 2-dimensional, meridional model of the global troposphere was used (Isaksen and Hov, 1986).

## 6 DEPOSITION

### 6.1 SNOW AND PRECIPITATION CHEMISTRY

Snow and precipitation chemistry data for the Arctic are relatively scarce, except for Greenland, where chemical analyses of ice cores from the inland ice cap have been used to infer changes in atmospheric chemistry due to anthropogenic emissions of sulphur dioxide, lead, and mercury (Koide and Goldberg, 1971; Boutron and Delmas, 1980; Herron, 1982). Little is known, however, with respect to the atmospheric transport and origin of this snow chemistry.

Precipitation in the Arctic is generally slight, averaging only about 90 mm/y (Orvig, 1970). In the Svalbard area, the annual amounts of precipitation measured with gauges are about 300-400 mm/y (Steffensen, 1982). Considerably larger precipitation amounts are found from mass balance studies of the glaciers on Spitsbergen.

Precipitation chemistry data are available from precipitation gauge samplers at Bjørnøya and Jergul for the period 1978-1984. For Ny-Ålesund, Jan Mayen, Hopen and Vardø data are available for 1982-1984 (Joranger et al., 1986). A summary of the data for Jergul and Bjørnøya are given in Figure 6.1. The conspicuously high ammonium concentrations at Bjørnøya during the summer months are an effect of the 2-5 million seabirds nesting on this little island, demonstrating the importance of the sea-bird colonies in providing an abundant source of nitrogen for the terrestrial vegetation in the Arctic. The mean concentrations of nitrate in precipitation at both stations are small, and show no systematic seasonal variations.

The seasonal variation in the concentrations of non-marine sulphate in the arctic precipitation samples is smaller than the corresponding seasonal variation of the sulphate concentrations in the air. This may partly be due to the scavenging mechanisms.

Scott (1978) has discussed scavenging ratios ( $W$ : concentration of sulphate in precipitation/concentration of sulphate in air) for different types of precipitation and varying water contents in the precipitating

clouds. The lowest W-values were estimated for cold frontal precipitation, while much higher scavenging ratios were estimated for summer convective storms.

It is seen from Figure 6.2, that scavenging ratios in the Arctic increase strongly in the summer season, while Birkenes in southern Norway shows no such clear seasonal trend in the W-values. Precipitation in the high Arctic during summer occurs mainly as fog or fine drizzle from low stratiform clouds, which may partly explain the high scavenging ratios in summer.

During winter, the relatively high concentrations of sulphate in the arctic air are confined to a shallow (about 1 km) layer of air next to the ground, while a significant part of the precipitation is derived from cleaner air above this layer during frontal precipitation events.

A large number of snow samples were collected from Spitsbergen in April-May 1983 (Semb et al., 1984). Snow cores were taken from glaciers or snowfields down to the icy crust corresponding to the summer melting of last year, and divided into sections which were melted in clean polyethylene bags and transferred to the laboratory for analysis.

The spatial gradients of the excess sulphate concentrations, shown in Figure 6.3, indicate that the deposition of these components occurs with transport of air from south-east and east. The very high values of snow accumulation on the glaciers along the east coast are due to orographic effects. Figures 6.4 and 6.5 show that the variation of the sulphate concentrations in the snow cores was nearly parallel to the variation of the concentration of sulphate in air at Ny-Ålesund during the snow accumulation period. A low scavenging ratio ( $W = 0.2 \times 10^{-6}$ ) suggests, again, that while the sulphate and other pollutants in the snow samples originate from a relatively shallow layer of air close to the surface, most of the precipitation water comes from cleaner air above this layer.

The concentrations of nitrate and ammonium ions in the snow samples were surprisingly low. Somewhat higher concentrations of nitrate were,

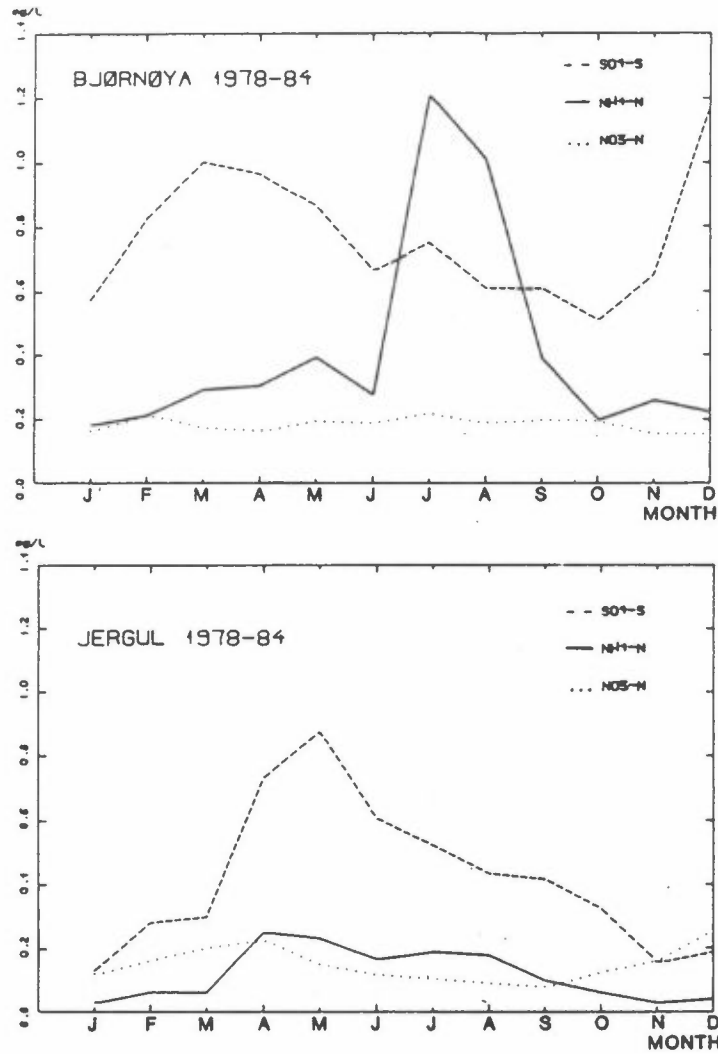


Figure 6.1: Monthly mean concentrations of SO<sub>4</sub><sup>-2</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> in precipitation at Bjørnøya and Jergul during 1978 to 1984.

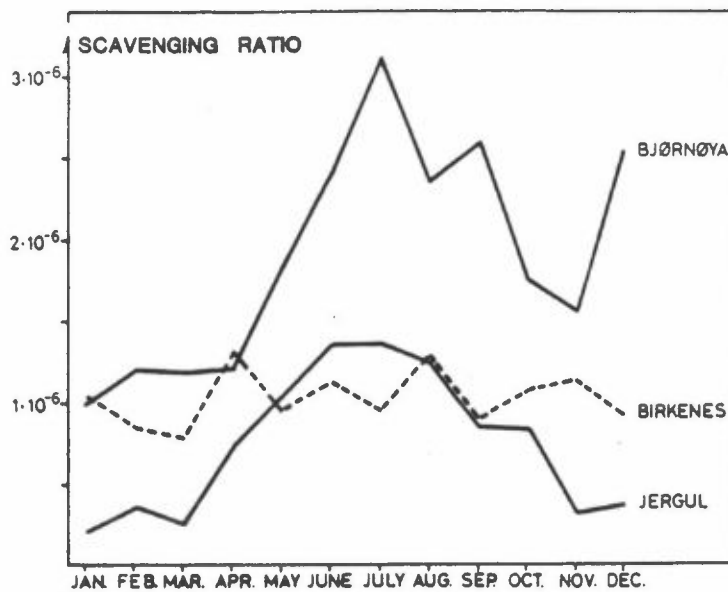


Figure 6.2: Annual variations of the scavenging ratios of SO<sub>4</sub><sup>-2</sup> at Bjørnøya, Jergul and Birkenes. Calculated from monthly mean concentrations in precipitation and air during 1978 to 1984.

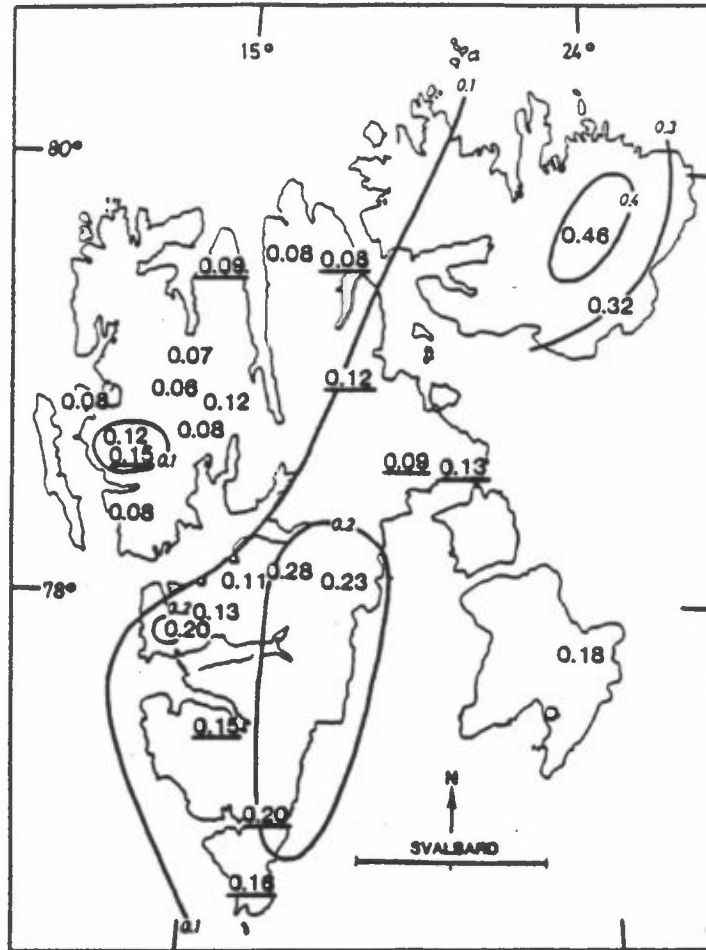


Figure 6.3: Mean weighted concentrations of excess (non-marine) sulphate (mg SO<sub>4</sub>-S/litre) in snow at Spitsbergen, April to May 1983. Underlined figures mark the sites of detailed vertical profiles.

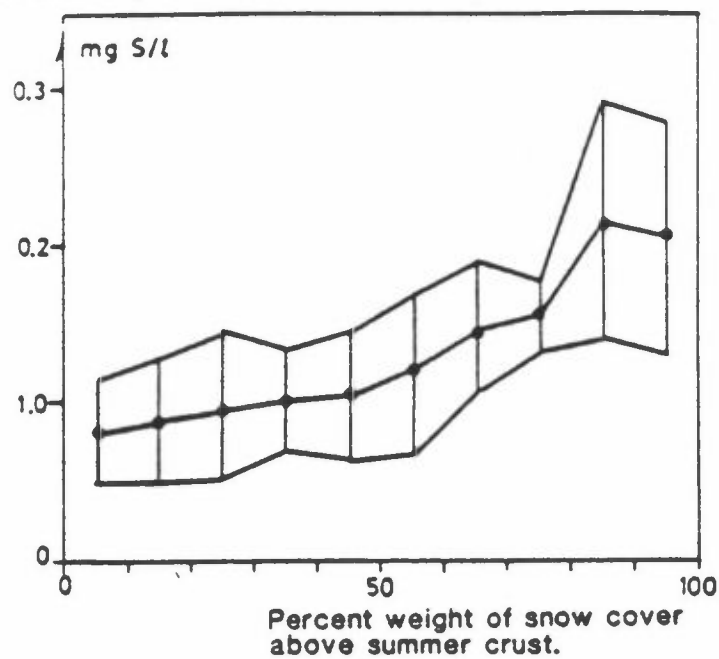


Figure 6.4: Variation of the excess sulphate concentrations in snow cores with height above the summer layer. Arithmetic mean of 9 profiles from sites on West Spitsbergen, with standard deviation.



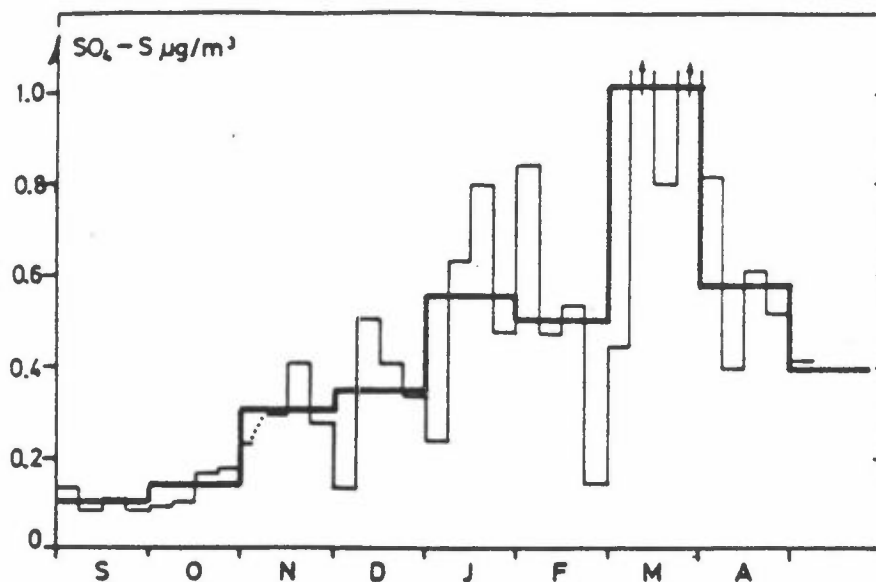


Figure 6.5: Monthly and weekly mean concentrations of sulphate aerosol in air samples at Ny Ålesund during the snow accumulation period.

however, found in snow samples collected at Kongsøya (Kong Karls Land) in 1982. High ammonium ion concentrations probably occur only in the neighbourhood of bird colonies.

## 6.2 SOIL AND VEGETATION ANALYSES

Analyses of woodland mosses have been extensively used to assess the deposition of trace elements from the atmosphere, giving an integrated value of the deposited amounts during the preceding 2-4 years. This approach can be extended, by analyses of extractable organic species, to give information also on the deposition of chlorinated hydrocarbons and polycyclic aromatic hydrocarbons from the atmosphere to terrestrial ecosystems.

As a part of the BP programme, and in cooperation with the University of Bayreuth, the concentrations of selected heavy metals, major ions, PAHs and polychlorinated hydrocarbons, were determined in samples of vegetation and surface soil collected on Spitsbergen in August 1983. In order to obtain geographical concentration profiles, samples were collected along a transect from the Federal Republic of Germany, across Denmark and Norway. The sampling locations are indicated in Figure 6.6.

The vegetation samples included the mosses *Hypnum cupressiforme* (Germany and Denmark), *Hylocomium splendens*, and *Racomitrium lanuginosum*. Two lichen species, viz. *Cetraria nivalis* and *Cetraria delisei* were also sampled on Spitsbergen. Trace elements were determined by atomic absorption spectrometry, and the polychlorinated hydrocarbons by high resolution gas chromatography combined with electron capture detection. Alternatively, negative ion mass spectrometry was used. Polycyclic aromatic hydrocarbons were determined by thin layer chromatography using a fluorescence circular scanning spectrometer for quantification. Sampling, sample pretreatment and cleanup, and analysis procedures, are described in detail by Thomas (1984, 1986) and Oehme et al. (1985).

The mean concentrations of Cd, Pb, Cu, and Ni in moss samples collected at the various locations are shown in Figure 6.7, and the mean concentrations of benzo-a-pyrene, polychlorinated biphenyls (PCB), and  $\gamma$ -hexachlorocyclohexane ( $\gamma$ -HCH) are shown in Figure 6.8. Generally the concentrations decrease when approaching the Norwegian Arctic, and the low variances observed for the concentrations at different locations in the Arctic suggest that long range transport of air pollutants is the main contributor to these concentrations. Concentrations of some extractable inorganic components in surface samples (2 cm) of Spitsbergen soil samples are given in Table 6.1 below.

Table 6.1: Concentrations of heavy metals and some major ions in soil samples from Spitsbergen (mg/kg dry material).

Location	Concentration							
	Zn	Pb	Ni	Fe	Mn	Ca	K	Na
1	66	7	25	7400	630	17160	3550	200
3	63	32	26	7400	45	9615	2210	104
4	93	10	33	7800	676	4090	4150	37

The soils in Spitsbergen are poorly developed and soil development is confined to small areas and very thin layers. Trace element concentrations in soil samples are, however, significantly higher than in moss and vegetation samples. The natural content of metals in the parent minerals, and the long-term accumulation of material deposited from the atmosphere, are both reflected in the analysis results.

A comparison of the levels of Zn, Cu, fluoranthene (one of the PAHs), and  $\gamma$ -HCH in samples of different vegetation species and soil samples at Ny Ålesund is shown in Figure 6.9 (Thomas, 1986). The levels of Zn and Cu are enhanced in the soil, but similar in all plant species. Lichens appear to accumulate fluoranthene, which also occurs in the soil samples.

The concentration level of  $\gamma$ -HCH in the *Cassiope tetragona* samples is very much higher than in any of the moss or lichen samples. This may be related to the content of cuticular wax or terpenes in this miniature heath plant.

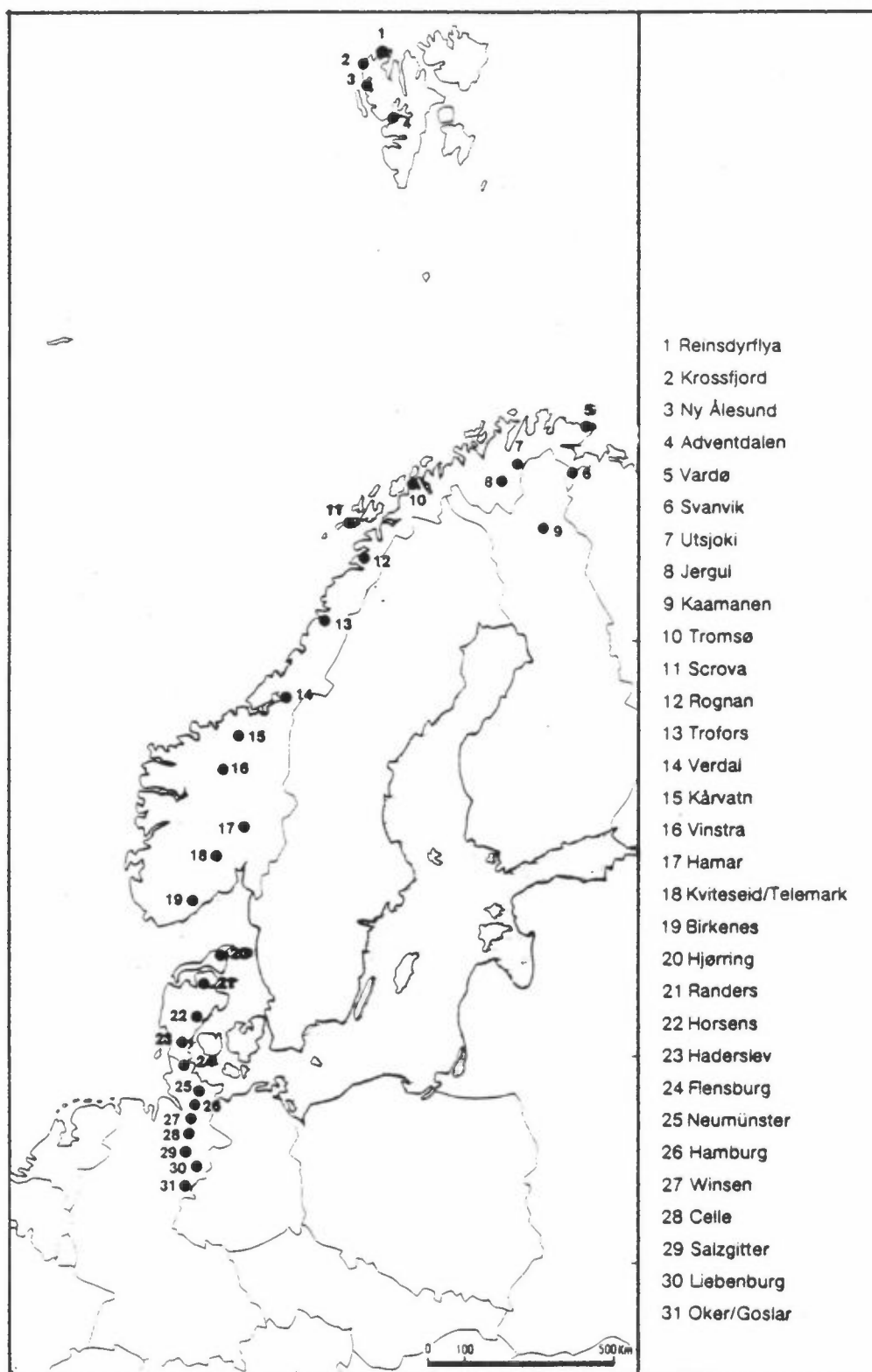


Figure 6.6: Location of the moss sampling sites.

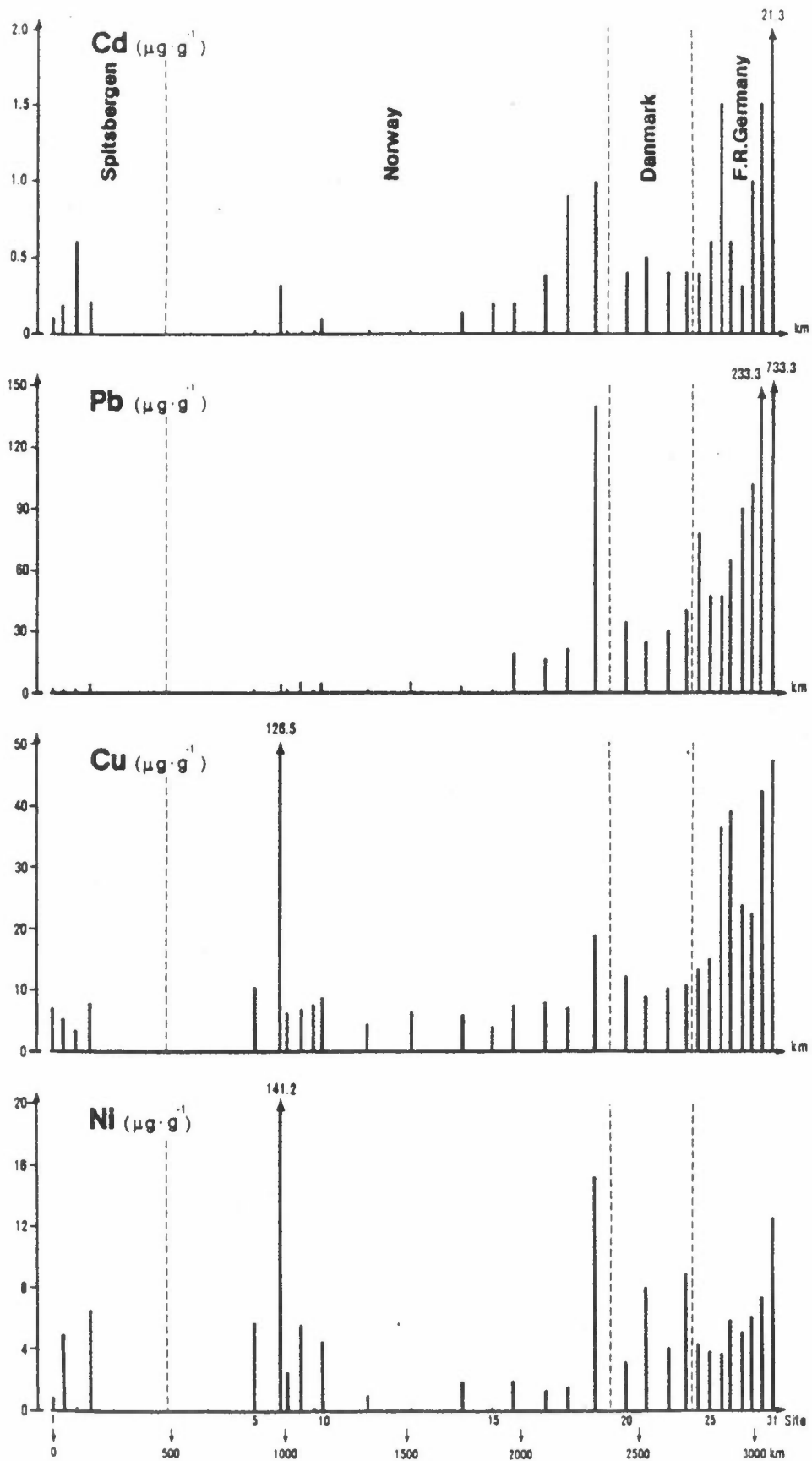


Figure 6.7: Concentration profiles for heavy metal found in the collected moss samples.

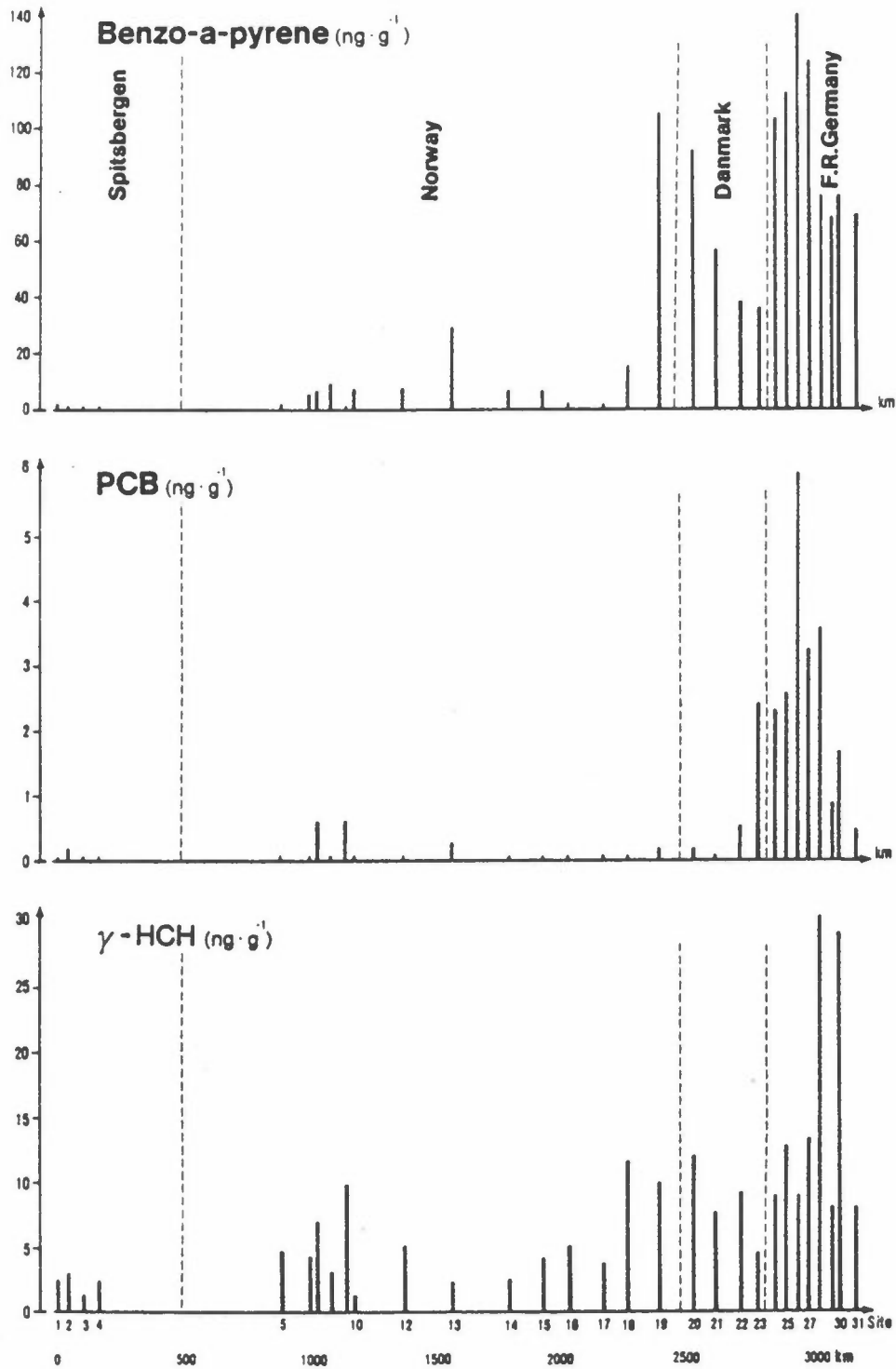


Figure 6.8: Concentration profiles for organic pollutants in the collected moss samples.  
 PCB : Polychlorinated biphenyls.  
 $\gamma$ -HCH:  $\gamma$ -hexachlorocyclohexane.

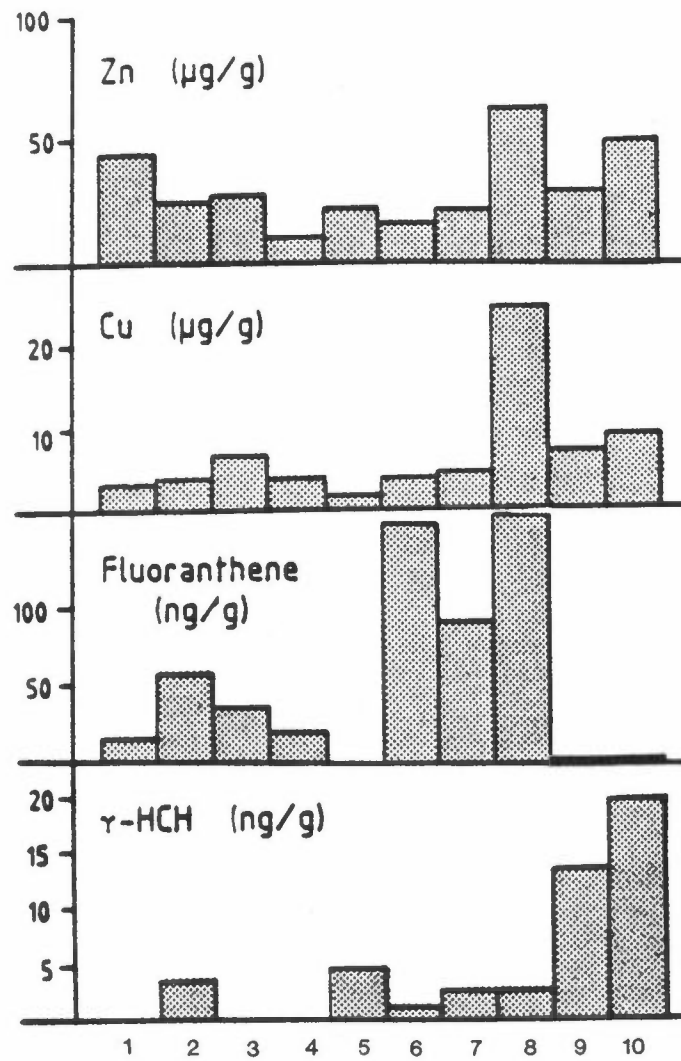


Figure 6.9: Comparison of trace substance concentrations in mosses (1-4), lichens (5,6,7), soil (8) and higher plants (9,10) at Ny Ålesund.

- |                                    |                                 |
|------------------------------------|---------------------------------|
| 1 = <i>Hylocomium splendens</i>    | 6 = <i>Cetraria delisei</i>     |
| 2 = <i>Tomenthypnum nitens</i>     | 7 = <i>Stereocaulon alpinum</i> |
| 3 = <i>Drepanocladus uncinatus</i> | 8 = Organic humus layer         |
| 4 = <i>Racomitrium lanuginosum</i> | 9 = <i>Cassiope tetragona</i>   |
| 5 = <i>Cetraria nivalis</i>        | 10 = <i>Cassiope tetragona</i>  |

## 7 DISPERSION OVER SEA SURFACES

In offshore operations there is a need to estimate danger distances and extension of hazard areas following an accidental release of explosive gases. The immediate danger area depends on the distribution and fluctuation of instantaneous concentrations. They are determined by the small scale turbulence. Rescue operations and steps to stop a blow out require estimates of the location and extension of possible danger areas in the immediate future. These depend on larger scale wind fluctuations. The aim of the two following studies is to provide basic information which is required for such calculations.

In order to examine the turbulence over a surface, a number of tracer diffusion experiments were carried out over Bundefjorden, a side arm to the inner Oslofjord. A tracer gas was released from a sunken rock and sampled by a boat crossing the plume at a distance of 400 m. The sampling took place every 2 seconds at three levels along the mast. The tracer gas ( $SF_6$ ) was released from an instrumented platform on the sunken rock, which is shown in Figure 7.1, together with the boat, syringes used for sampling and smoke released to visualize the plume.

The results are reported by Gotaas (1986). One of the observations made was a tendency of travelling lumps to form in the upper part of the cloud. The result is a slower decrease with increasing height of the maximum concentration than of the mean concentration. The mean crosswind distributions were close to normal (Gaussian), individual distributions were not. The influence of the temperature difference between the air and the water surface was less than expected from over land experiments. Statistical examination of the fluctuating concentration distributions relative to the center of gravity show the most prominent feature to be that when fluctuations are large, they are largest at the cloud center. Either the concentrations are high throughout the whole cross section of the cloud, or they are all low. The second most important distribution is a plume split in two, with low concentrations near the plume center and high concentrations near the boundary.



In order to provide data for calculation of hazard areas, also the wind fluctuations over the ocean was examined (Eidsvik, 1985). This study, which was also founded by Norsk Hydro and Statoil, was conducted at the Division of Port and Ocean Engineering at NTH in Trondheim, their motivation being that platforms for deep water oil exploration may have resonance periods of the order of minutes so that wind may be a most important forcing function.

The best quality data from the Norwegian sector of the North Sea have been analyzed. They constitute 3662 time series of wind speed from the top of the Statfjord A derrick. The high frequency spectra follow the usual laws of the inertial sub-range. The small scale turbulence intensity is very low. The drag and the aerodynamic roughness of the sea is very small, and the friction velocity coefficient is equal to  $1.5 \times 10^{-3}$ . The low frequency fluctuations are considerably more energetic than expected. Figure 7.2 shows the spectrum at frequency  $10^{-2} \text{ s}^{-1}$  in different wind classes.

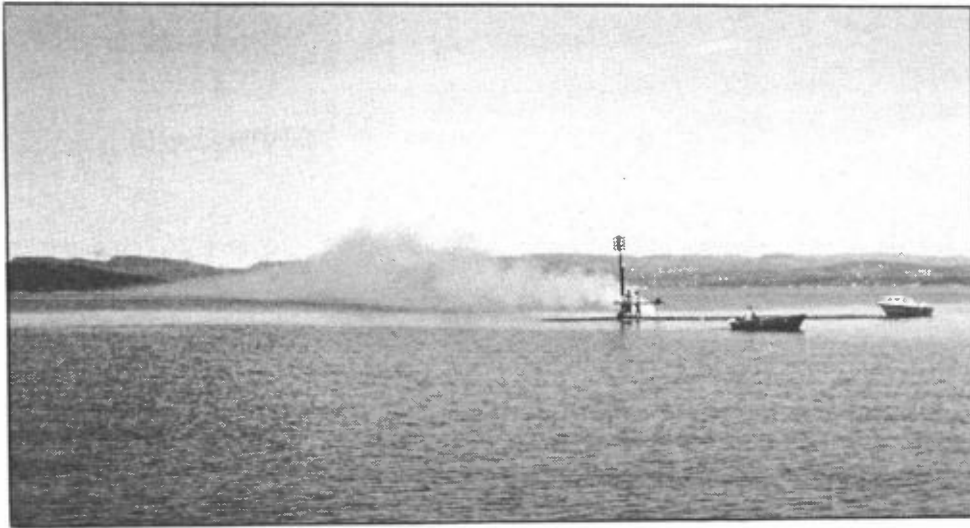


Figure 7.1: Tracer experiments in the Oslofjord in 1984.

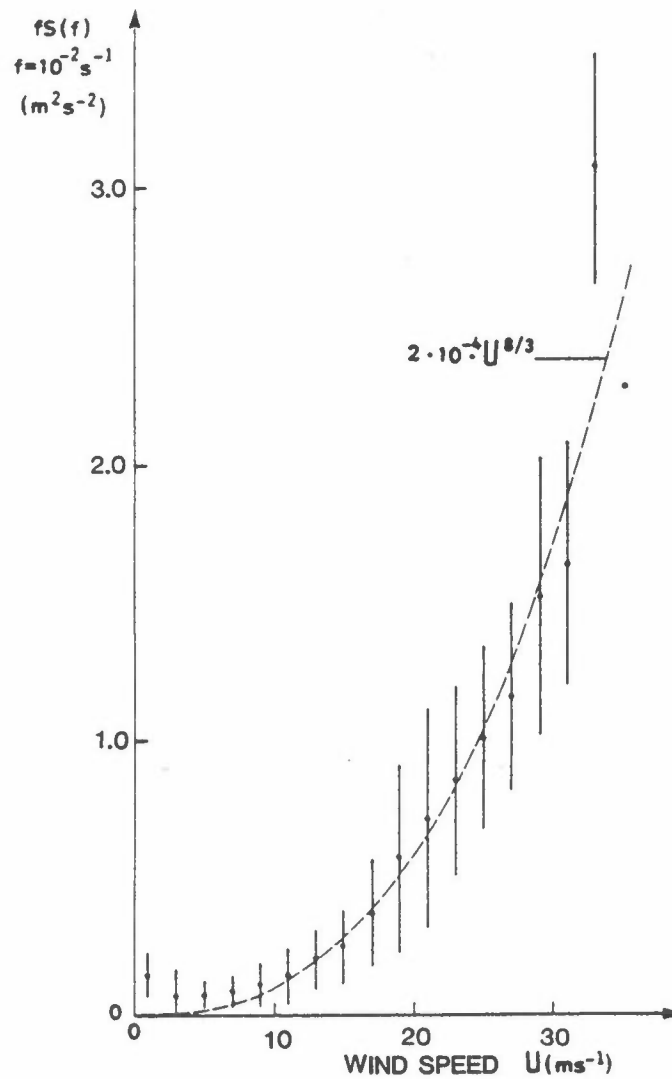


Figure 7.2: Average estimated spectrum at  $f=10^{-1} \cdot s^{-1}$  in different mean wind classes. Vertical lines are root-mean-square values.

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3 STIKKORD (à maks. 20 anslag) Arctic                                      Air pollutants                                      Air transport			
REFERAT (maks. 300 anslag, 7 linjer) Rapporten gir en oversikt over arbeid utført under det arktiske forskningsprogram finansiert av Britisk Petroleum i perioden 1981-86. Sammensetning og opprinnelse av den arktiske luftforurensning er undersøkt og en har pekt ut viktige områder for fremtidig forskning. I sin alminnelighet har Arktis vist seg å representere en sentral observasjonspost for undersøkelser av luftforurensningssituasjonens utvikling på den nordlige halvkule.			

TITLE Air pollutants in the Arctic.
ABSTRACT (max. 300 characters, 7 lines) The present report gives a survey of the work carried out under an arctic research programme financed by British Petroleum Ltd. during the period 1981-86. The composition and origin of the arctic air pollution has been examined and important areas of continued research are pointed out. In general, the Arctic has turned out to represent a key observation post for studies of the air pollution developments in the northern hemisphere.

\* Kategorier: Åpen - kan bestilles fra NILU                                      A  
 Må bestilles gjennom oppdragsgiver                                      B  
 Kan ikke utleveres                                      C